COMMENTS RECEIVED FROM REVIEWER #1

General comments

The study under review applied in-situ instrumental data from an aircraft campaign with the aim to seperate cloud microphysical properties for conditions of clean/background air and for air polluted from anthropogenic aerosol. Therefore the aircraft measurements were taken inside and outside of a plume created from the city Manaus during the wet season over the Amazon regions. While studies for the dry period exist, the study seems to be one of the first to investigate the wet season, which has the advantage that background conditions are comparable to oceanic conditions, i.e. thermodynamic variables do not vary much with distance from the city. The study clearly outlines and justifies the methods that are applied. The results are presented in a clear way and the discussion is conclusive and supports the current understanding of aerosol-cloud interactions. The data is unique and the results should definitely be of interest for the scientific community. Therefore I would suggest to publish the manuscript after some minor corrections, which are listed in the following.

Authors answer

Dear Reviewer #1, we would like to express our gratitude for your efforts to review the submitted manuscript. We found your comments very important to improve the quality of the manuscript. Following you will find a detailed explanation of our approach regarding your questions. Your concerns are numbered and answered individually in order in the next section of this document.

Thank you and best regards,

Micael A. Cecchini and coauthors

Minor revisions:

1.
    a. **(Question)** You could consider to separate the results section into topic related subsections, as the section is quite long in general (E.g. differences in LWC for background and polluted clouds, effect of updraft speed, vertical development of clouds). This would make it easier for the reader to find the relevant information in the results.
    b. **(Answer)** We agree that it would be clearer to separate the results into subsections. We divided into two separate subsections, labeled “Bulk DSD

properties for polluted and background clouds" and "Vertical DSD development and the role of the vertical wind speed".

2.

a. **(Question)**The summary and conclusions section ends quite abrupt. Consider to add a short outlook. What are the remaining open questions? Are further field studies planned? You already mention that the effects on ice-clouds is one focus for future endeavors in the motivation.

b. **(Answer)** We added a new paragraph in the end of the "Summary and conclusions" section, which is reproduced below:

"While the effects of aerosol particles in the warm layer of the clouds is relatively straightforward, this may not be the case for the mixed and frozen portions. An aspect that was not directly addressed in this work is the impacts that warm layer characteristics have on the formation of the mixed phase (above the 0°C isotherm). Given that aerosols alter the properties of the whole warm phase, it is reasonable to assume that this would have an impact on the initial formation of the mixed layer. Such impacts can be in the form of the timing and physical characteristics of the first ice particles and the maximum altitude with supercooled droplets above the freezing level. This issue will be addressed in future studies, taking advantage of data provided by the HALO (High Altitude and Long Range Aircraft) airplane that operated in the second GoAmazon2014/5 IOP between September and October, 2014."

3.

a. **(Question)** p.4, l.13: Can you tell more about the uncertainties of the instruments? E.g. what is the accuracy of the particle concentrations from the CPC?

b. **(Answer)** We added the requested information about the accuracy of the instruments. The accuracy for the CPC is ±10%, while for the FCDP it is around 3 μm.

4.

a. **(Question)** p.6, l.27: You name one factor is commonly cited in literature but do not add any references. I suggest to add some references at this point.

b. **(Answer)** Added a citation to Abrecht's (1989) work: Albrecht, B.A.: Aerosols, cloud microphysics, and fractional cloudiness. Science 245, 1227–1230, 1989.

5.

a. **(Question)** p.7, l.18: Calculations show that… -> It would be nice if you shortly can present how you did this estimation.

b. **(Answer)** This affirmative is based on calculations of the averaged second moment in polluted and background clouds. We found that the average second moment for polluted clouds is around twice as the background one. This ratio between the second moment in the polluted/background DSDs is representative of the ratio of the overall surface areas. The text was updated to reflect this change.

6.
   a. **(Question)** p.8.,l. 21: While your statement seems to be true for the background clouds, especially for the polluted clouds there seems to be an increase in the last LWC bin. Also the spread is increased. Can the latter be explained by a larger LWC bin size?
   b. **(Answer)** Added the sentence at the end of the paragraph: "This effect is clearer in background clouds given the limited aerosol availability". This should make the matter clearer. We believe that the LWC bin size should not have as big of an impact here. Under polluted conditions, new droplets may form even if the LWC is big.

7.
   a. **(Question)** Table 2: Add the definition of bottom, mid and top layer to the Table caption.
   b. **(Answer)** Added the requested information.

8.
   a. **(Question)** Figure 2: This figure looks a bit clumsy. My suggestion would be to create a plot with subfigures with the individual flight plans and add the estimated plume area and the average wind direction for each flight.
   b. **(Answer)** We tried several approaches to improve this figure, taking into account the clumsiness and the message we want to get through. The most important thing to show with this figure is that most of the flights had a similar trajectory, which enables the plume classification. The updated figure separates each flight in a subplot, with the plume angular section. We changed the text in order to describe this figure. The last 5 lines of the second paragraph of section 2 is now:

"Figure 2 shows the trajectories for all flights, where the dashed grey lines represent the plume angular section considered from the airplane data. Note that the plume usually disperses from Manaus to the T3 site, with relatively small variations on the direction based on the wind field. Two flights (4 and 6) had low sampling on the plume given the trajectories and the grey lines may not represent the overall region of the plume. However, the directions identified presented higher CN concentrations than the other ones".

Phrasing / spelling corrections:

All phrasing and spelling corrections were addressed. Thank you for taking the time to highlight these issues. The specific corrections you suggested are listed below.

p.1, l.11: in terms *of* aerosol conditions

p.1, l.17: split the sentence after the brackets -> The cloud droplets observed are in the range…

p.1, l.24: correct the superscript of km-1

p.1, l.25: Why you use e.g. for the definition of larger droplets? In my opinion, you can just omit this.

p.1, l.25: to the cloud base

p.1., l.26: change sentence structure to: The overall shape of the droplet size distribution (DSD) does not appear to be : : :

p.1, l.31: initiation of the collision-coalescence

p.2, l.4: maintains

p.2, l.8: for the Amazon by Martin et al.

p.2, l.11: that a city like Manaus has on atmospheric conditions

p.2, l.22: Amazonian cloud properties

p.3, l.7: suggests -> suggest

p.3, l.8: over the Manaus area

p.3, l.9: stronger wind component -> dominant wind component

p.3, l.20: clouds microphysical properties -> cloud microphysical properties

p.3, l.25: add comma after background air reference

p.4, l.1: consider to change pollution-aerosols to anthropogenic aerosols

p.4, l.2: are almost only urban, while biomass-burning contribution is very exceptional

p.4, l.6: omit numbered before chronologically

p.4, l.29: what is meant by true airspeed? I guess you mean the speed of the aircraft?

p.6, l.22: by effective size you refer to effective diameter D_e?

p.7, l.29: omit *profiles*

p.7, l.30: updraft speeds levels -> updraft speed levels

p.8, l.18: relationships De x LWC and DNC x LWC -> relationships of De and LWC, and of DNC and LWC

p.8, l.20: omit e.g.
p. 9, l.4: omit brackets. Instead write: for each layer, as there are more measurements for lower levels.

p.9, l.14: its mass -> their mass

p.9, l.19: omit e.g.
p.9, l.32: and l.33: once you write plume and once polluted. Try to be consistent.

p.10, l.10: justifies -> explains

p.10, l.10: vertical velocities region -> vertical velocity region

p.10, l.25: the updraft regions DSD -> DSDs in the updraft region

p.11, l.6: Polluted clouds had 10
p.11, l.16: omit e.g.
p.11, l.17: bi-modality *favors* the efficiency

p. 11, l.20: aerosols conditions -> aerosol conditions

Figure 5 caption: affected or not -> affected and unaffected

Figure 5 caption: units of LWC should be gm^{-3}

Figure 6 caption: add the information that this is for clouds lower than 1000 m only.

COMMENTS RECEIVED FROM REVIEWER #2

MAJOR CONCERNS

The manuscript describes unique aircraft measurements of polluted and pristine clouds over the Amazon region during the wet season of 2014. The results are of potential interest not only for those involved in the GoAmazon experiment but to the general ACP audience.

However, the manuscript is poorly written and needs substantial revision to meet ACP standards. Part of the methodology should be better explained and some of the results needs further investigation. Moreover, I have serious concerns about some methods and the interpretation of results. Hence, I cannot recommend its publication without a major revision.

I have annotated the author's PDF file with many comments and questions to the authors, which I hope will help to improve the manuscript. Here, I only list my major concerns.

Authors answer

Dear Reviewer #2, we would like to extend our gratitude for the effort to revise this paper and ultimately helping it be improved. We worked hard to address each one of your concerns individually and this document will detail how we approached it. Following you will find our answers to the major comments you made, while next section will detail the specific comments from the supplement material you provided.

Best regards,

Micael A. Cecchini and co-authors

1.
    a. **(Question)** Introduction needs to be throughly revised. Lines discussing this work are mixed with paragraphs discussing the current state of the art, making it hard to follow for those not part of GoAmazon.
    b. **(Answer)** We revised the introduction with all you recommendations. There are several aspects that were covered by the introduction (i.e. effects of Manaus pollution on air chemistry and cloud physics, differences between wet and dry season, etc…).
2.
    a. **(Question)** Session about the instrumentation should explain what corrections or data processing were performed for the different instruments / probes used. Alternatively, other papers describing that should be cited.
    b. **(Answer)** We updated the text with new information about the instruments – see comments 34-39 below.
3.
    a. **(Question)** Authors used CN to identify if clouds probed under each circunstance were or were not beeing affected by the plume of pollution. I beleive CCN would be better to indicate the influence of the plume on the clouds for 2 reasons. Firstly, because most of the initial pollution particles

emitted will be too small to become CCN, hence the initial plume will not affect much the cloud formation. Secondly, as the plume is chemically and physically transformed downwind of Manaus, the extra aerosols will grow, be oxidized, and thus will interfere more and more with the CCN population. See for instance previous results from Kuhn et al (2010). Hence, as the G1 flight legs are at different distances from Manaus, CCN would be a better indicator than CN.

b. **(Answer)** There are both technical and conceptual reasons why we chose to use CN instead of CCN. The technical one is that there were no CCN measurements for 2 flights, which would make it harder to have a single reference for all flights. Besides, the supersaturations would have to be taken into account, and small differences in supersaturation can add more complexity to our analysis. Our idea was to provide a binary classification, in or out of the plume, with a simple, but strong, criterion adequate to our flights. The conceptual reasons are related to the type of classification we wanted to produce. In this work, we are not analyzing the way the plume changes as it ages and its consequences for cloud formation. The intent is just to locate the plume in order to compile characteristics of clouds growing under its presence and compare to clouds formed under background conditions. The added requirements for the plume classification (i.e. the angular section and the CN concentrations have to be higher than the 90% percentile) further contribute to make sure that the DSD measurements are indeed inside the pollution.

   Additionally, from our classification, it is possible to observe that the CCN concentrations are higher for the plume regions even though the classification itself did not take it into account directly. See Figure R.1, for example ("R" refers to revision, so no confusion is made to the paper figures). It shows mean CCN concentrations in different SS for the plume and background regions (for altitudes lower than 1000 m). The CCN concentrations in the plume are more than double the background concentrations for SS=0.23%, while this difference increases with supersaturation. In this way, it is possible to conclude that the plume is able to increase the CCN concentrations and, thus, to affect cloud formation. The second-to-last paragraph of section 2.2 was revised:

"The final result of the classification scheme for March 10 is shown in Figure 4. A visual inspection of radiosonde (released from the Ponta Pelada airport located on southern Manaus) trajectory plots confirmed the overall direction of the plume for each flight. Given the nature of the meteorology in the Amazonian wet season, i.e. its similarities with oceanic conditions concerning horizontal homogeneity, there should be no significant difference between the thermodynamic conditions inside and outside the plume region for the G-1 flights. In this way, differences observed in pollution-affected clouds are primarily due to the urban aerosol effects. It should be noted that even though the plume classification is defined from the CN measurements, there are also observable differences regarding CCN concentrations. The in-plume CCN concentrations (for altitudes lower than 1000 m) averages at 257 cm^{-3} for a 0.23% supersaturation, while the respective background concentration is 107 cm^{-3} (Figure 5). Note the overall low concentrations, representative of the wet season. In that case, the plume more than doubles the CCN concentrations. For higher supersaturations (which can be achieved in strong updrafts), the differences are even more pronounced. At 0.5% supersaturation, the average CCN concentration inside the plume is 564 cm^{-3}, while outside it is 148 cm^{-3}. This shows that the plume increases the concentration of aerosol particles that are able to form cloud droplets under reasonable supersaturation conditions, even though they are less efficient than the particles in the background air".

Figure R.1 was added to the manuscript as Figure 5.

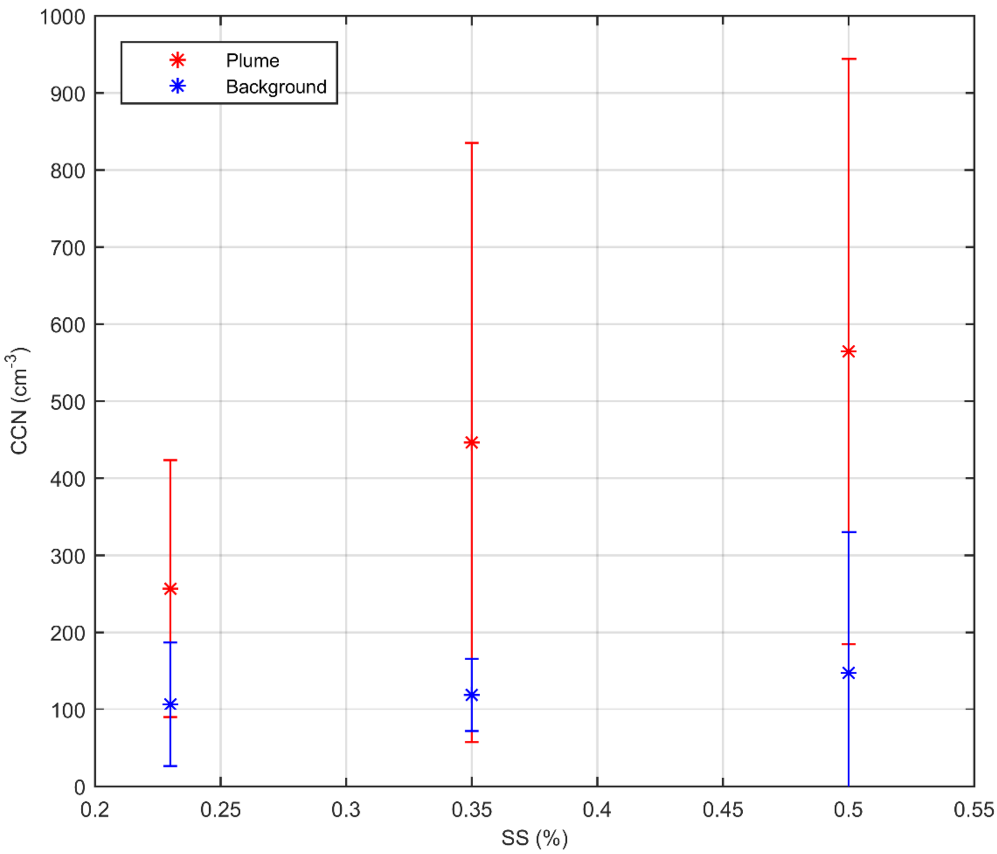

Figure R.1: CCN concentrations as function of SS. Measurements classified as plume are shown in red, while background is in blue.

4.

a. **(Question)** For selecting the in-plume events, the authors defined a cone where the plume was most likely to be found. This cone was centered at the airport as if the pollution plume were being dispersed from that single position. While this approach might work for larger distances, for the short distances from Manaus (closer legs) the airport-angle will not confine the plume.

b. **(Answer)** Yes, this is true and we failed to make it clearer in the text the intention of centering the origin on Manaus airport. By keeping the origin of the coordinate system over the airport, we avoid including measurements over the city on the plume classification because the airport is located on the far west corner of the city. In this way, the heat island effect is avoided, which could introduce different thermodynamic conditions to be considered. See question 46 in the next section for more details on how we addressed this issue on the manuscript text.

5.

a. **(Question)** The authors based their whole analyses on a bold hypothesis:

Given the nature of the meteorology in the Amazonian wet season, i.e. (...) horizontal homogeneity, there is no significant difference between the thermodynamic conditions inside and outside the plume region (...). This would be true only for Amazon regions with an uniform vegetation cover, which is not the case at all for the region of Manaus. The Manaus plume goes towards T3, which is the direction of the Solimoes river. Hence, the in-plume cases studied are mostly over or close to the river. On the other hand, the out-plume clouds are far away from the plume and hence from the river. Therefore, as we know from previous studies that the river breeze is significant, one cannot assume that the thermodynamics are the same (over the river and far away)!

To assess the validity of their hypothesis, the authors could, for instance:

- use radiosondes close and away from the river

- look at the specific humidity around the clouds (polluted vs pristine)

- verify the average time of day when polluted vs pristine clouds were sampled

- verify the location (lat/lon) where the polluted vs pristine clouds were sampled

- etc...

b. **(Answer)** This is a valid concern you expressed and we worked hard to study it and prove that our results really reflect the plume effects and not the river breeze. We found it difficult to prove our point based on the radiosondes, so we will focus on the cloud observations over the rivers or land.

First, it should be noted that most of the flights started at around 10 am local time. At this time the river breeze is not fully developed but is present nonetheless. In this way, there is suppression of the convection over the rivers (Solimões and Negro) and the clouds are predominantly located over land. To check the effects on the DSD measurements, we produced figures similar to

Figure 7 and 8 of the paper substituting the plume-background classification by “over river” or “over land” conditions. Figures R.2 and R.3 shows the averaged DSDs for the vertical layers and w conditions mentioned in the paper. It is quite clear from this that the convection suppression reflects on the growth of droplets. In other words, the droplets tend to be smaller in the clouds over the rivers, with less growth with altitude. This is somewhat expected and the question to address now is how the plume and background classifications relate to the positioning of the rivers.

From the 350 seconds of plume DSDs, 161 are over the rivers and 189 over land. For the background classification, 115 are over rivers and 456 over land. There is a higher contribution of river-DSDs for the plume classification than for the background one. However, the next figures will show that the effects of the pollution on clouds is consistent even considering the land-river differences. Figures R.4 and R.5 shows the averaged DSDs in the same way as in Figure R.2, but restricting for the background and plume classifications, respectively. No DSDs were observed on background conditions over the rivers for the mid and top layers, either by physical or sampling reasons. Focusing on the DSDs over land, it is possible to observe that the plume has a suppression effect on droplet growth, as is seen in Figure 7 of the paper. This shows that the plume effects on the Amazonian clouds is the one noted in the paper even though the clouds over land are more vigorous. We decided to leave the figures unchanged as we feel they consistently represent the effect of Manaus plume on the DSD properties. The manuscript text was updated with comments to this feature in the last paragraph of the methodology section.

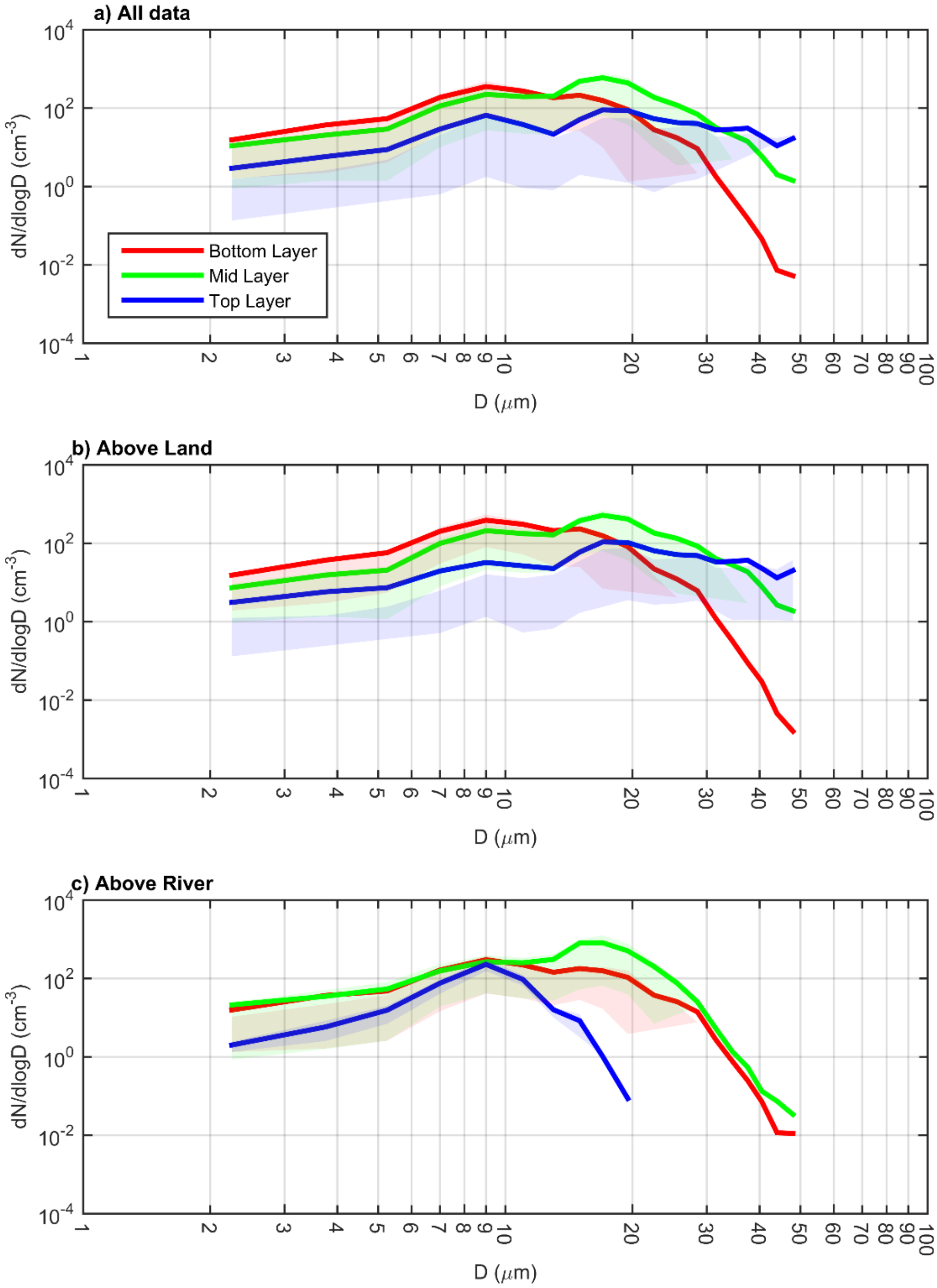

Figure R.2: averaged DSDs for the bottom, mid and top layers defined in the paper. a) all data, b) only DSDs measured above land and c) only DSDs observed in clouds over the rivers.

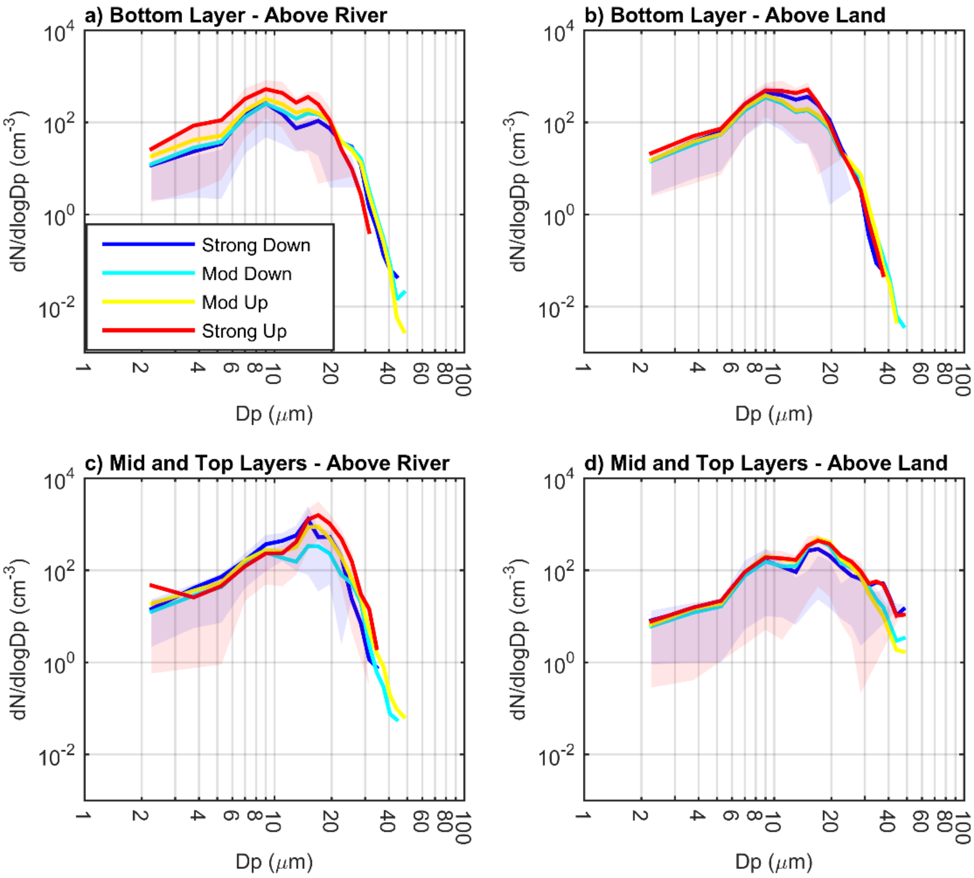

Figure R.3: the same as Figure 8 in the paper, but the plume DSDs are substituted by those measured over the rivers. The background DSDs are substituted by those observed over land.

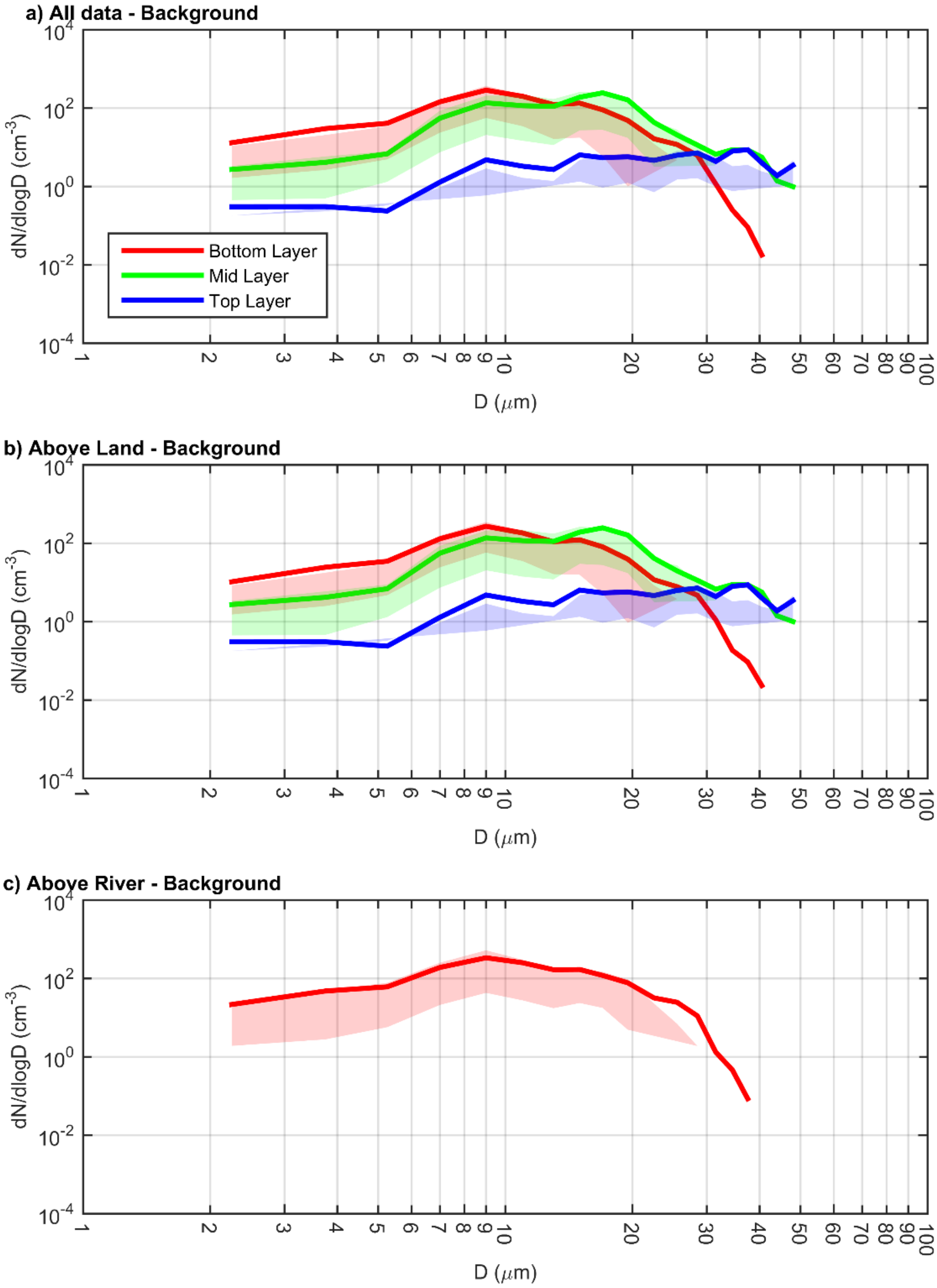

Figure R.4: the same as Figure R.2, but only considering the points classified as background.

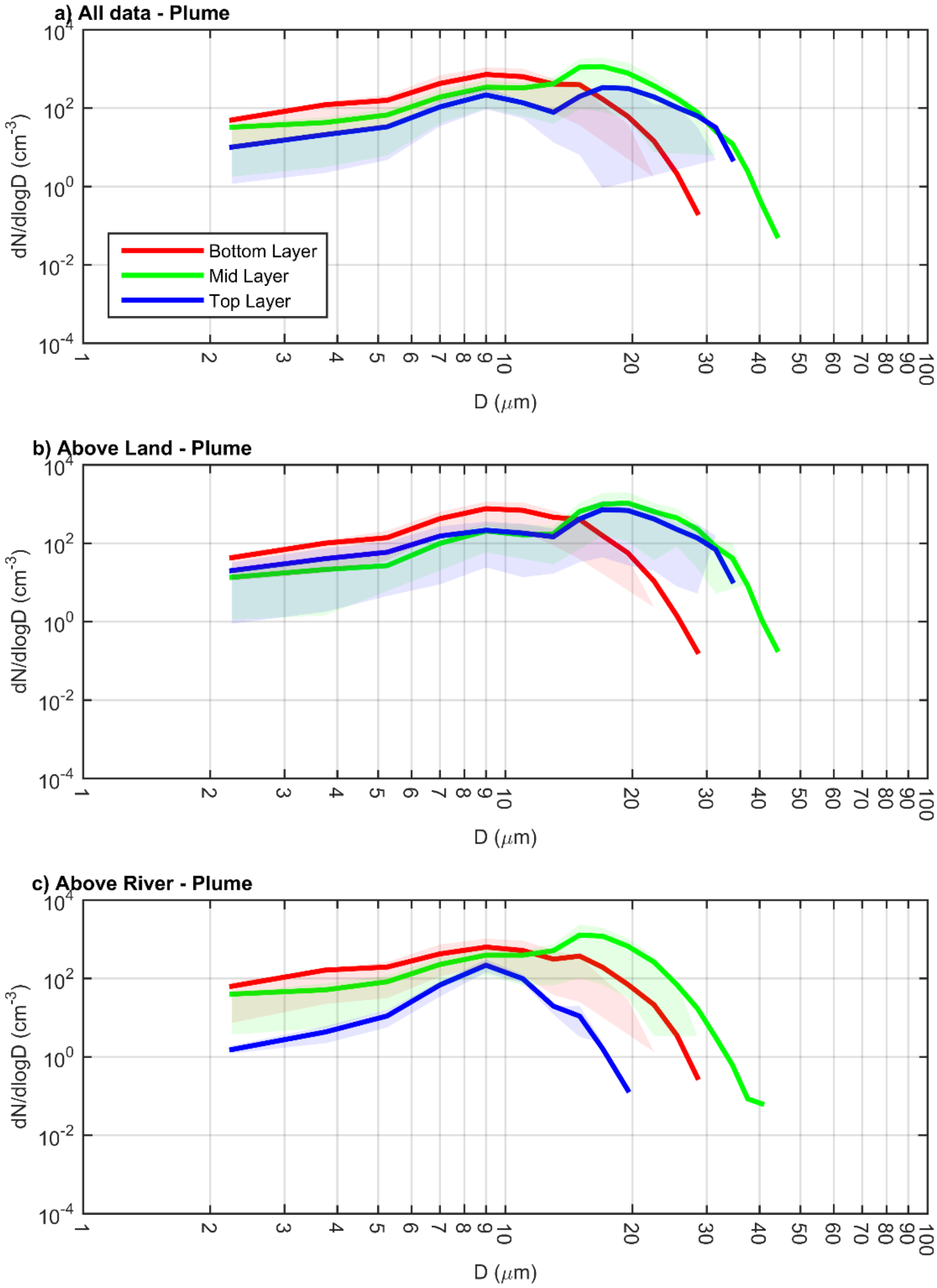

Figure R.5: the same as Figure R.2, but only for the points classified as plume.

6.

a. **(Question)** When authors look at DSD from different altitudes, they divide the vertical from LCL (0%) to freezing level (100%). Then they made averages for relative altitude ranges of 0-20, 20-50 and >50%. There are two things going on. Firstly, the G1 samples are not well distributed in the vertical, hence the authors had to choose uneven limits to get the same number of samples in each. However, not all shallow clouds will develop as high as the freezing level. Therefore, the average for the bottom layer includes some clouds that did not extend at altitudes >20% and more clouds that did not develop > 50%. On the contrary, samples for the top layer are, by definition, all from clouds that extended from the LCL up to > 50%. Hence, this introduces a large bias. It is mixing clouds of different total vertical development, in different amounts, in each of the three categories. Hence one could not compare DSD from different altitudes, just DSDs from the same altitude for polluted/pristine cases.

Please also note the supplement to this comment:

http://www.atmos-chem-phys-discuss.net/acp-2015-1049/acp-2015-1049-RC2-supplement.pdf

b. **(Answer)** It is indeed possible that there is a bias regarding cloud top altitude representation in each of our vertical layers. However, we do not believe this should be a concern to our conclusions. Firstly, the main reason to even separate the clouds into the same vertical intervals is to be able to compare different clouds under the same benchmark. We are mainly focusing on comparing plume-affected and background DSDs for the same vertical levels. Otherwise, those layers wouldn't even be needed.

Additionally, there is a common practice by the cloud physicist's community in which measurements made in different clouds in a region can be combined and interpreted as if they were made in a single cloud. For instance, Rosenfeld and Lensky (1998) were able to calculate vertical profiles of effective droplet sizes from satellite based on this assumption. They select a region with clouds with different top heights and use this assumption to produce the profiles as if they were measuring a single well-developed cloud. Freud et al. (2008), using in-situ measurements, found that this assumption is reasonable for the Amazon. We added a comment on the end of the first paragraph of the section "Vertical DSD development and the role of the vertical wind speed":

"Despite probing individual clouds, the DSD measurements can be combined into the three layers defined and interpreted as representative of a single system. It is conceptually similar to satellite retrievals of vertical profiles of droplets effective radii (e.g. Rosenfeld and Lensky, 1998), where the cloud top radius is measured for different clouds with distinct depths and combined into one profile. This approach was validated with in-situ measurements for the Amazon region by Freud et al (2008)".

SPECIFIC COMMENTS FROM THE SUPPLEMENT

The specific comments from the supplement are listed and numbered below. The "q" after the numbers refer to "question" (i.e., the comment from the reviewer), while "a" stands for the answer given by the authors.

1q) p.1, l.11: …terms of aerosol concentrations…

1a) Done.

2q) p.1, l.15: … between the Manaus plume ??

2a) Yes, we changed for Manaus plume. The intended meaning is that the city has an effect on the local atmosphere through the production of the plume.

3q) p.1, l.17: review the English. You probably want to start a new sentence here. It is also not clear if this range is a result of your observations or an arbitrary cut you choose to use, or a cut imposed by the instruments you had at disposal.

3a) We changed the sentence to: "The droplet size distributions reported are in the range 1 μm ≤ D ≤ 50 μm in order to capture the processes leading up to the precipitation formation". In this way the goal is clearer.

4q) p.1, l.21: aerodynamic or physical or other? Or doesn't matter for your case?

4a) The effective diameter stands for the ratio between the third and second moments of the DSD. In this way, it is a physical diameter and the name of the variable should be self-explanatory.

5q) p.1, l.22: the average value has an exact value, hence it cannot range from 10 to 40%. Do you mean "The differences range from 10% to 50%"?

5a) We calculated mean effective diameters and droplet number concentrations in each 400 m vertical bins. The "average" was meant to address the vertical averaging process. However, we left the sentence as you suggested because we feel it sufficiently explains the results without complicating for the reader.

6q) p.1, l.22: This is confusing. Of course droplet concentration varies a lot across different vertical levels! But you were talking about differences between polluted and pristine clouds. Please rephrase.

6a) We left the final sentence as: "The differences range from 10% to 40% for the effective diameter and are as high as 1000% for droplet concentration for the same vertical levels". We are comparing diameters and concentrations between polluted and background clouds for the same altitudes. Because, as you pointed out, they vary greatly with altitude.

7q) p.1, l.30: droplets sizes.

7a) Corrected.

8q) p.2, l.3: This is not true! The water vapor comes from the ocean!!!

8a) Indeed, we removed the "self-contained" to avoid confusion.

9q) p.2, l.8: by.

9a) Corrected.

10q) p.2, l.8: by.

10a) Corrected.

11q) p.2, l.11: review. You probably want to say "the atmospheric…".

11a) We changed "its" for "the", it should be clearer now.

12q) p.2, l.12: ,

12a) Corrected.

13q) p.2,l.12: "around" or "of about".

13a) Corrected.

14q) p.2, l.13: with

14a) Corrected.

15q) p.2, l.22: in the case of Manaus.

15a) Added.

16q) p.2, l.22: focused

16a) Corrected.

17q) p.2, l.24: study evaluated.

17a) Corrected.

18q) p.2, l.25: Review the text. Disconnected meanings. You probably want to say something like: “This is important because the city plume is always there, all year long, and polluted clouds…”

18a) We changed the sentence to: “However, no study evaluated the urban aerosol interaction with clouds over the rain forest during the wet season, when biomass-burning is strongly reduced given the frequent rain showers that leave the forest wet and more difficult to burn. In this case, the effects of the Manaus plume can be studied separately and in detail. Polluted clouds over the Amazon usually present more numerous but smaller droplets that grow inefficiently by collision-coalescence and therefore delay the onset of precipitation to higher altitudes within clouds (Rosenfeld et al., 2008)”.

19q) p.2, l.29: Please review. Previous studies showed that precipitation during the monsoon seasons is more stratiform while that during the dry season is more showering. More over, the reason why the wet season is more clean is because the vegetation is wet (not mattering the type of rain) and hence cannot be burned.

19a) Regarding the cleaner atmosphere for the period, we rephrased in order to make clearer that the air is clean due to the reduction in biomass burning. The sentence is now: “The period is in the wet season, which presents a clean atmosphere due to the reduction in biomass burning”. In the previous question, we included one comment saying that there is less biomass burning because the forest is wet.

The clouds are surely more convective during the dry season. However, cumulus fields are very common during the wet season, being frequent during the campaign days. We observed stratiform rain during the campaign, but we can confirm that most of the clouds during the flights were cumulus. We are not sure what to review in this sentence related to this discussion given that we don’t mention the convective x stratiform regimes here.

20q) p.2, l.31: It is not the background air who provides the opportunity. Please rephrase.

20a) The sentence is now: “The pristine characteristic of the background air provides the opportunity for contrasting the microphysics of natural and urban pollution-affected clouds”.

21q) p.2, l.31: Unless you are going to state that this is the focus of GoAmazon, I don't see why you should talk about chemistry if you will focus on clouds.

21a) See previous question.

22q) p.3, l.2: you probably mean something else.

22a) The sentence is now: "This scenario allows for the first time the direct comparison between clouds formed under background conditions and those affected by pollution in the wet season".

23q) p.3, l.3: you probably mean clouds formed under background conditions.

23a) see previous question.

24q) p.3, l.4: you will only analyze data from the wet season. It is not clear then why you are discussing difference between the dry/wet seasons. You didn't even mention what was found by Machado (2004) in terms of cloud microphysics.

24a) The intent of this paragraph is to show that there is a large scale forcing for the clouds during the wet season. This forcing is related to the monsoon system. We feel that it is important to give a general picture of the large scale features in place during the campaign. The paragraph mentions that the large scale is related to the monsoon system and further details can be seen in the citations given.

25q) p.3, l.18: give the number or rephrase.

25a) Gave the number – 16.

26q) p.3, l.26: see my comment on figure 1. It is very confusing!

26a)

27q) p.3, l.29: past tense.

27a) Corrected.

28q) p.3, l.31: past tense.

28a) Corrected.

29q) p.3, l.31: past tense.

29a) Corrected.

30q) p.4, l.3: sections.

30a) Corrected.

31q) p.4, l.4: past tense.

31a) Corrected.

32q) p.4, l.4: past tense.

32a) No correction needed as far as we know.

33q) p.4, l.7: this should be at the beginning of the section.

33a) Relocated the sentence to the beginning of the section. It is now the second sentence of the Methodology section.

34q) p.4, l.11: Typical CPC that use butanol measure only > 10nm. Did the G1 had an ultra-fine CPC? If that is the case, you should say. Moreover, what are the losses on the G1 inlet? Does it allow particles of 3nm or 3 microns to reach the CPC?

34a) The characteristics of the CPC used are the ones given in the text. The cut-off diameter is 3 nm, making it sensible to small particles and, therefore, able to readily detect urban pollution. The intent of using this CPC instead of the model 3010 (also available on the plane) is to better locate the plume, even though particles as small as 3 nm are not able to activate droplets. As mentioned in the text, the intent of using CN concentrations instead of CCN is to produce a qualitative classification of the plume. Note that the quantitative CN values are not used in any part of the study. We consider that there is not enough statistics to analyze DSD characteristics for several levels of pollution. That is one of the reasons we only produce a binary classification, it is a way to characterize clouds inside and outside the plume regardless of the exact amount of CCN produced by the urban pollution.

As mentioned in question 35 (below), inlet losses are lower than 4% (penetration higher than 96%), with an up limit of 5 µm in diameter.

35q) p.4, l.13: Please give a reference. TSI instruments, for instance, measure only up to 10000. After that point, the chance of coincidence is non-negligible.

35a) We updated the manuscript text with more information on the CPC instrument used (model 3025). The first paragraph of Section 2.1 is now:

"The two main instruments used for this study were the Condensational Particle Counter (CPC, TSI model 3025), and the Fast Cloud Droplet Probe (SPEC Inc., FCDP). The CPC instrument measures number concentration of aerosols between 3 nm and 3 µm using an optical detector after a supersaturated vapor condenses onto the particles, growing them into larger droplets. Particle concentrations can be detected between 0 and 105 cm-3, with an accuracy of ±10%. Coincidence is less than 2% at 104 cm-3 concentration, and corrections are automatically applied for concentrations between 104 cm-3 and 105 cm-3. The CPC was mounted in a rack inside the cabin and connected to an isokinetic inlet and an aerosol flow diluter and was operated using an external pump. The isokinetic inlet has an up limit of 5 µm for particle diameter, with penetration efficiency higher than 96%. A 1.5 LPM flow rate was maintained using a critical orifice. The dilution factor varied between 1 and 5".

36q) p.4, l.19: aircraft measurements are tough! You should be clear about which corrections were applied or not to the data, how it was cleaned, etc…

36a) We added sentences explaining the filter applied to the FCDP-measured DSDs. The end of the second paragraph in Section 2.1 now is as follows: "Shattering effects were filtered from the FCDP-measured DSDs (Droplet Size Distributions), which is a built-in feature of the provider software. Additionally, measurements with low number concentrations (< 0.3 cm^{-3}) and low water contents (< 0.02 gm^{-1}) were excluded".

For the CPC measurements, we changed the first paragraph of Section 2.1 – see question 35 above.

37q) p.4, l.21: how many % of the dataset had to be interpolated? You should also mention how you average the data, as I don't think you used 1Hz… If you average over 30s or more, I don't see why you would need this interpolation.

37a) Around 16% of the CPC data was flagged as "bad" and was interpolated, while 0.02% was excluded because no good measurement was close enough. Additionally, we performed tests with moving averages on the CN data and noted no significant impact on the results with periods of up to 10 s. Higher averaging periods seem too large, given that the airplane flew at around 100 ms^{-1} and 10 s represents roughly 1 km. In this case, we chose to continue using 1 Hz measurements. The second paragraph of Section 2.1 was changed in order to reflect those comments:

"The quality flag of the CPC instrument was used to correct the concentration measurements. Whenever an observation was flagged as "bad", it was substituted by an interpolation between the closest measurements before and after it that were either "questionable" or "good". For "good" measurements, which represents 59% of all the measurements, the uncertainty is less than 10%. The interpolation weights decayed exponentially with the time difference between the current observation and the reference ones. If the reference observations were more than 10 s apart, these data were excluded. 16% of the data was interpolated in that manner, while only 0.02% had to be excluded. This process was required not only to smooth out the bad measurements but also was important to maintain significant sample sizes (instead of simply excluding “bad” measurements). No averaging was applied to the 1 Hz CPC data. However, tests were made in order to check the impact that the sample frequency had on the results. The results were not sensible to moving averages of up to 10 seconds, which corresponds to roughly 1 km displacement given that the G-1 flew around 100 ms^{-1} in speed. Given this observation, the analyzes are based on the 1 Hz CPC measurements”.

38q) p.4, l.29: do you mean accuracy (distance from the true value) or precision distance from average value)? If you mean accuracy, how was it even calculated? Which other instrument has been used for giving the true vertical wind speed?

38a) We mean precision, the text is corrected.

39q) p.4, l.29: was 75m/s the approximate G1 speed? The accuracy of 0.75m/s is too large when compared to the typical vertical wind speeds (1-3m/s). You should try to estimate how much miss-classifications you might have.

39a) The updated text after question 37 notes that the G-1 speed is around 100 ms^{-1}. Therefore, the vertical wind speed precision should be close to 0.75 ms^{-1}. This is one of the reasons we use relatively wide w bins in Figure 8. We believe there is not much impact of miss-classifications in that case.

40q) p.5, l.2: these are too small to become CCN.

40a) They may be right after emission, but they become more efficient CCN as they age. We updated the sentence to reflect this: “Urban activities such as traffic emit large quantities of particles to the atmosphere, which are then transported by atmospheric motions and can participate in cloud formation, especially when they grow, age and become more effective droplet activators”.

41q) p.5, l.4: it will only affect if they have the size (and chemistry) to compete for the available water vapor.

41a) The urban aerosol are indeed smaller than the background ones and are less effective to become CCN. However, their high concentrations are able to produce more CCN even so. See, for instance, Fig. 4 in Kuhn et al (2010) – the study you mention in the next question.

42q) P.5, l.6: why not using CCN? Was it not measured by G1? As the plume is chemically and physically transformed downwind of Manaus, the extra aerosol loading will interfere more and more with the CCN population. See for instance Kuhn et al (2010). Hence, as the G1 flight legs are at different distances from Manaus, CCN would be a better indicator than CN.

42a) See major question 3 in the previous section.

43q) p.5, l.19: it is not clear what you mean.

43a) We believe this is explained in the next sentence on the manuscript (i.e. a CN measurement inside the cloud is substituted by the closest cloud-free measurement).

44q) p.5, l.20: You are using CN just to build a plume/background mask… so why making it so complicated? For instance, what happens if CN before the cloud says "plume" while CN after the cloud says "background"? By your criteria of time-distance, half of the plume will be polluted and half will be clean. Does it make sense? Why don't you do the mask on a cloud basis instead of 1Hz basis?

44a) As mentioned before in this document and also on the manuscript, most of the clouds probed were part of the Cu fields usually observed during the wet season. This makes it almost impossible to have a "background" classification in one side and a "plume" classification on the other side of the cloud because of the size of the systems. Additionally, the 90% percentile requirement on the CN concentrations for the plume classification results in measurements closer to the center of the plume. The "background" classification also requires the measurements to be outside the plume angular section in order to avoid this issue.

45q) p.5, l.26: so you are throwing out 65% of the data?

45a) Not necessarily, and the percentage is actually 74%. There are CPC measurements not only during the cloud penetrations but also in clear sky. The 90% and 25% percentiles refer only to the CPC measurements. Only a portion of the CPC data points have a corresponding FCDP one. Even though most of the data remains unclassified, we believe it is necessary in order to differentiate as much as possible both populations.

You may have noted that the number of measurements changed slightly in this new version of the manuscript (see page 6, line 12). We changed the DSD filtering slightly, eliminating the cases where DNC < 0.3 cm^{-3} and LWC < 0.02 gm^{-3}. Before we only eliminated the DSDs where LWC < 0.02 gm^{-3}. The change is to make it more consistent with the cloud flag we use – described in the methodology. Minimal impacts on the dataset resulted from this and no impact whatsoever on the results.

46q) p.6, l.3: This is not true. The plume does not originate on the airport!! Hence, calculating theta from there is misleading, particularly for the short distances from Manaus. See, for example, the dark blue lines on Fig. 3. The plume is much wider than what is indicated by the vertical lines, exactly because of this reason!

46a) Yes, this is true and we failed to make it clearer in the text the intention of centering the origin on Manaus airport. By keeping the origin of the coordinate system over the airport, we avoid including measurements over the city on the plume classification because the airport is located on the far west corner of the city. In this way, the heat island effect is avoided, which could introduce different thermodynamic conditions to be considered. The manuscript text was updated in the following manner, starting from the line of the comment:

"Note that there is an angular section where the concentrations are high not only close to the city but also as far as 70 km. This section is defined to be affected by Manaus pollution plume (delimited by grey dashed lines in Figure 3). Note that the coordinate system is centered on Manaus' airport, where the G-1 took off, and not on the center of the city or other point of interest. For this reason, it is also possible to observe relatively high CN concentrations close to the origin and to the northeast and southeast directions. This corresponds to high CN concentrations over the city. By keeping those directions outside the plume angular section, this data is not considered as plume. This is intentional because other aspects occur over the city that may contribute to the cloud formation. For instance, the heat island effect may contribute to the convection, changing the thermodynamic conditions compared to those over the forest. By keeping the origin point as the airport, which is located on the west section of the city, this problem is avoided."

47q) p.6, l.5: where is this plot? And why didn't you look at the radio sonde from the ponta-pelada airport? It makes much more sense, as it will travel between Manaus and T3.

47a) We did not feel the need to show this plot, we are just confirming that we looked into it. We feel that it is quite reasonable that the plume and the radiosonde would have a similar trajectory in lower level given that they are subject to the same wind field. The mentioned radiosondes were not released from T3, it was a mistake on the text. They were indeed released from Ponta Pelada airport, thanks for pointing that out.

48q) p.6, l.8: this would be true only if considering an uniform vegetation at the surface. Which is not the case at all for the region of Manaus. The Manaus plume goes towards T3, which is the direction of the Solimões river. Hence, the in-plume cases you selected are, I guess, mostly over or close to the river. On the other hand, the out-plume clouds will be far away from the plume and hence from the river. Therefore, you cannot assume waving hands that the thermodynamics are the same! But you have the radiosondes from T0z and from T3. You should compare them to prove.

48a) This question is addressed in the item 5 of the major concerns (previous section in this document). Please refer to it.

49q) p.6, l.11: this is not the first place CCN is defined.

49a) Corrected.

50q) p.6, l.19: what about the environmental specific humidity outside the clouds? As I said, probably the environmental conditions are not the same for the clean and plume samples.

50a) The wet season in Manaus and its surroundings is characterized by very high relative humidity values (e.g. 90%) given the constant inflow of humidity from the trade winds. We believe the humidity outside the clouds should be similar for all cases.

51q) p.6, l.25: that only means that the difference in LWC is smaller than the difference in DNC.

51a) Changed the sentence to: "This factor shows that, despite condensing lesser amounts of total liquid water, the background clouds are able to produce bigger droplets than their polluted counterparts". The issue should be clearer now.

52q) p.6, l.28: Please note that availability of water is different than LWC. Hence you should look at the specific humidity around the polluted and pristine clouds (or below their cloud bases) to be able to say that the water availability is different… or that aerosols have an effect on that.

52a) The idea is to separate the bulk condensation efficiency of the clouds (that affects LWC values) and the water vapor competition scenario (which is usually analyzed in fixed LWC). The sentence is now: "The other factor is how much bulk water the systems are able to condense while the vapor competition is ongoing".

53q) p.7, l.4: under polluted conditions?

53a) Yes, corrected.

54q) p.7, l.4: slower? If droplets are smaller, the condensation ratio is lower, and hence they grow slower then initially larger droplets, isn't it? You even say that later on…

54a) The smaller droplets in polluted conditions grow faster by the condensation process only – the condensation rate is inversely proportional to size. The droplets under background conditions grow faster overall, because they anticipate the start of the collision-coalescence process.

55q) p.7, l.13: But that would change the LWC near the top of the cloud, where droplets could be large enough to precipitate. This, by the way, rings a beçç: it does not make sense to include in Fig. 5 data from all altitudes. At this point you are discussing the impact of aerosols on the droplet formation at cloud base… Hence the analysis would be more coherent if Fig. 5 showed on near-base data. If you do for both (base x top) you may be even able to disentangle the two mechanisms you identified.

55a) By comparing the histograms in the different layers of the clouds, the same observations are possible (i.e. higher LWC and NDC and lower De for polluted clouds). Se Figure R.6 and R.7 below. In this way, it is possible to observe that the aerosols affected the whole warm layer structure of the clouds. We chose to leave the manuscript unchanged in that regards as it already illustrates the issue adequately.

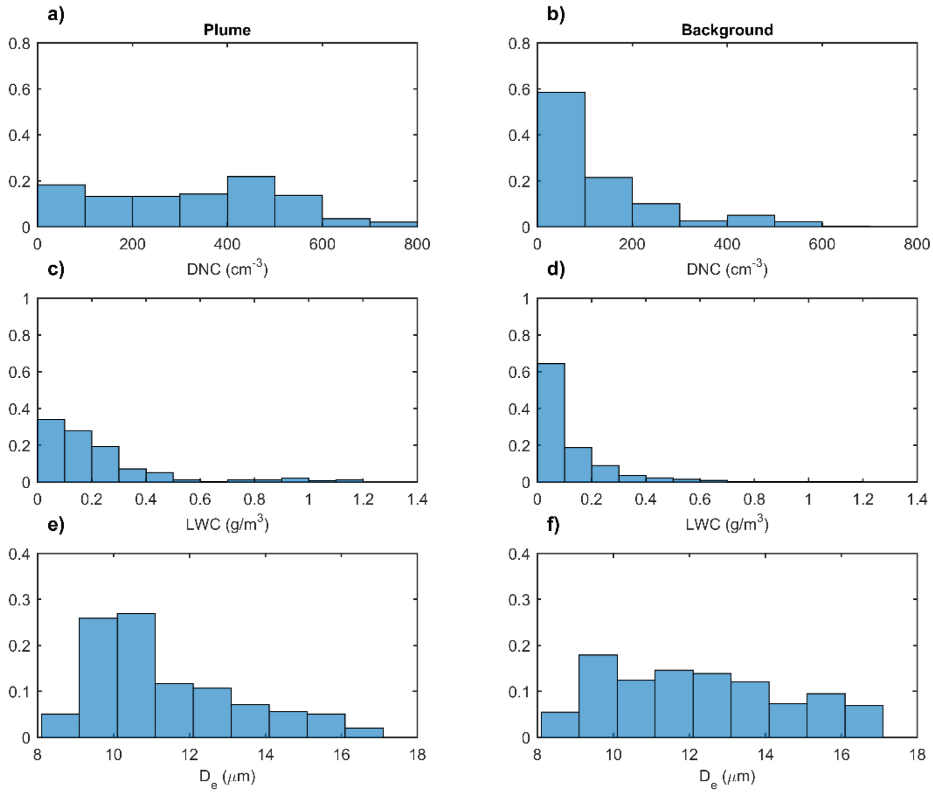

Figure R.6: the same as Figure 5 in the paper, but only for the bottom layer.

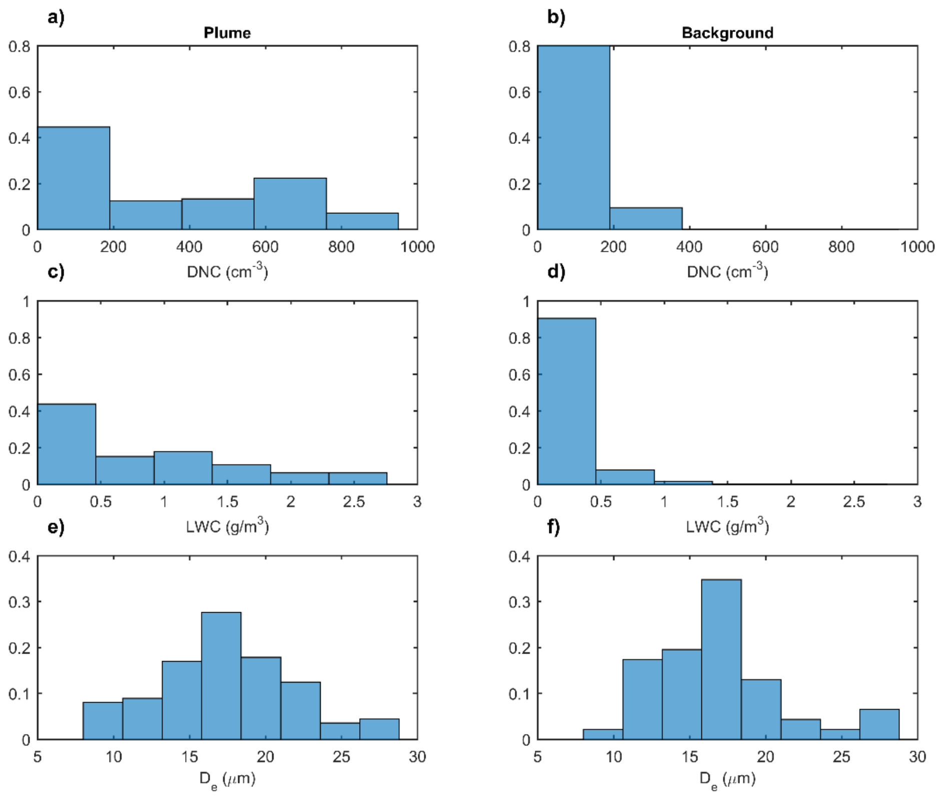

Figure R.7: the same as Figure 5 in the paper, but only for the mid and top layers.

56q) p.7, l.17: hence hypothesis 2 is bad.

56a) We believe hypothesis 1 should be the most significant, therefore we added a sentence right after: "The second process identified (i.e. suppressed precipitation staying longer inside the clouds) probably has a lesser impact".

57q) p.7, l.19: why would that be? You are talking about the area of the droplets… and then go back to a previous step to consider that what more CCN could do? Isn't there a confusion between the aerosol area and the droplet area? I mean, when you first discussed hypothesis #1 you are talking about the larger area of aerosol surface under polluted conditions… But then you calculated the area for your DSD.

57a) Although initially the vapor condenses onto the aerosol particles, when the droplets form the vapor continues to condense onto them. By calculating the average second moment of the polluted and background DSDs, it is possible to calculate the overall area available for condensation onto the droplets. In this way, we do not need to look into aerosol size distributions and can focus solely on the cloud-DSDs.

58q) p.7, l.20: We have already enough evidence (at least published on conferences) to show that under polluted conditions (urban or biomass burning) the organic (75%)/inorganic (25%) fractions are the same. Besides, we also have hygroscopicity measurements showing lower values that under polluted conditions because of the much lower hygroscopicity of the organic component.

58a) Indeed, we removed the hygroscopicity mention.

59q) p.7, l.27: why would it not be?

59a) Correct, we changed "considering that…" to "given that…".

60q) p.7, l.31: (hopefully) the non-precipitating shallow cumulus. But why < 1km? You should restrict yourself to the cloud base, and I find 1km too deep for shallow clouds. You should justify why 1km and not 300m or 2km.

60a) There are two reasons for choosing 1km instead of 500m or 300m. Firstly, the amount of data more than doubles from 500m to 1km. Additionally, in polluted clouds the aerosol activation process may last longer and higher in the cloud when compared to the background clouds. For that reason, the higher LWC bins can be underrepresented with a 500m limit. We added a sentence following your comment: "The 1000m limit is chosen for both maximizing statistics and also capturing the layer in which the aerosol activation takes place. That layer is possibly thicker under polluted conditions, given the higher availability of nuclei".

61q) p.7, l.34: it is influenced by the aerosol population with sizes allowing it to be activated. But, since you have aerosol size distribution and CCN measured on the G1… Why don't you estimate LWC for each updraft (ie. For each max SS)?

61a) It is not the intent of this paper to model the expected LWC from the aerosol size distribution. We defined a strategy to locate the plume and the background regions and accumulate statistics for each case. What this affirmation means is that the clouds affected by the plume presented higher LWC, which can only be associated to aerosols given we eliminated thermodynamic conditions. In Kuhn et al. (2010) paper it is possible to see that CCN is enhanced in Manaus plume, showing that the pollution increases the number of aerosol particles that have sizes above the critical diameter to activate. As we mentioned before, the intent here is not to analyze the DSD properties for specific quantitative CCN values. Instead, we report on the mean microphysical properties of clouds formed in and out the plume.

62q) p.8, l.1: Come on, you cannot say that by just visual inspection of the red points in the figure 6a! You have to make a line fit (considering the error bars!!) and make a null hypothesis test. Hence setting a confidence level for your statement.

62a) We agree that a more robust statistical analysis would be required to estimate the confidence level. However, the physical processes identified are consistent to the average results we obtained. As such, we left the confidence level question open (a bigger dataset is desirable), but discussed the possible physical mechanisms. The paragraph is now (starting from the sentence of this question):

"This figure shows that, on average, not only are the polluted clouds more efficient at the bulk water condensation but also the resulting LWC scales with updraft speed (linear coefficients, considering the error bars, are 0.13 g s m^{-2} for plume measurements and 0.033 g s m^{-2} for background clouds). In a background atmosphere, most of the aerosols readily activate, and increases in updraft strength does not result in further condensation. On the other hand, the higher availability of aerosols inside the plume allows for more condensational growth as long as enough supersaturation is generated, especially considering that the critical dry diameter for activation is inversely proportional to supersaturation and, consequently, to the updraft speed. However, a deeper analysis in a bigger dataset would be required to assess the statistical significance. The enhanced condensation efficiency and the possible LWC scaling with updraft strength at least partly explain the higher liquid water contents in the plume-affected clouds. The standard deviation bars in Figure 6a indicate that while there is high variability for the LWC in polluted clouds, the clean ones are rather consistent regarding the condensation efficiency".

63q) p.8, l.12: again, you have to do an statistical test.

63a) Changed the sentence to: "It is clear that, even with the dispersion observed, the two DSD populations present consistently different average behaviors for all LWC intervals".

64q) p.8, l.19: why almost? Please make the line fit and statistical tests, so that you can state that with statistical confidence.

64a) Removed the "almost" and included R^2 information to show that we calculated the linear fits.

65q) p.8, l.22: by your own argument, this might not be the case. You can have a low LWC content at higher altitude inside the cloud that did not develop high LWC. I understand that you are onlu showing data < 1000m from cloud base, but this might be too deep. It is not obvious that low LWC means cloud base high LWC means near 1km.

65a) It is true that LWC usually increases with altitude both for polluted or background clouds. It even has a more pronounced increase in clouds under the effect of the plume. Given that LWC increases with altitude, this factor is implicit in our affirmation. We said that for low LWC (or closer to cloud base), increases in DNC have a higher impact on the LWC value. However, for higher LWC (or higher in the cloud), new droplet formation won't affect as much LWC because the cloud droplet are bigger – LWC depends cubically on the diameter.

66q) p.8, l.22: at this stage of the cloud life you are discussing, the droplets present are those activated from aerosols at cloud base. The number of droplets activated on the cloud base depend (among other things) on the maximum SS achieved (i.e updraft). Hence, it might make sense to plot DNC x W near cloud base…

66a) It is indeed an interesting analysis. Se Figure R.8 below with this calculation (graph b). It shows that, on average, DNC is always higher for plume-affected clouds and it tends to grow with w. However, we think that the discussion presented on the paper already covers the main physical mechanisms at play and this graph is not actually needed, despite being interesting.

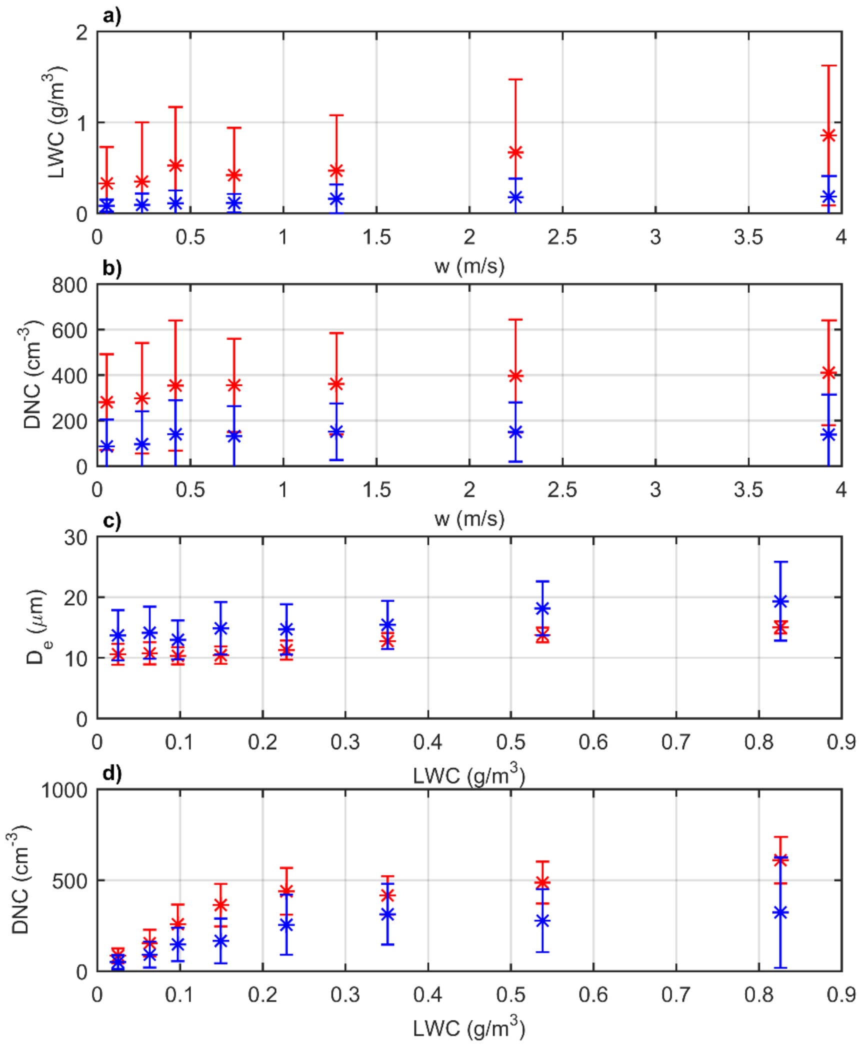

Figure R.8: the same as Figure 5 in the paper, but adding averaged DNC according to w intervals (b).

67q) p.9, l.2: There are two things at the same time going on. Firstly, the G1 samples are not well distributed in the vertical, hence you had to choose uneven limits to get the same number of samples. However, another point is that not all shallow clouds develop as high as the freezing level (yours 100%). Hence, your average for the bottom layer includes some clouds that did not extend at altitudes > 20% and more clouds that did not develop > 50%. On the contrary, your samples for the top layer are, by definition, all from clouds that extended from the LCL up to > 50%. You have, thus, and important bias! You are mixing clouds of different total vertical development, in different amount, in each of the three categories.

67a) It is indeed possible that there is a bias regarding cloud top altitude representation in each of our vertical layers. However, we do not believe this should be a concern to our conclusions. Firstly, the main reason to even separate the clouds into the same vertical intervals is to be able to compare different clouds under the same benchmark. We are mainly focusing on comparing plume-affected and background DSDs for the same vertical levels. Otherwise those layers wouldn't even be needed.

Additionally, there is a common practice by the cloud physicist's community in which measurements made in different clouds in a region can be combined and interpreted as if they were made in a single cloud. For instance, Rosenfeld and Lensky (1998) were able to calculate vertical profiles of effective droplet sizes from satellite based on this assumption. They select a region with clouds with different top heights and use this assumption to produce the profiles as if they were measuring a single well-developed cloud. Freud et al. (2008), using in-situ measurements, found that this assumption is reasonable for the Amazon.

68q) p.9, l.14: check the quartiles. There seems to be an error as they, sometime, go to zero at the begin/end of each curve. They also, in some case, do not contain the average value.

68a) When the quartiles go to zero it only mean that 25% or 75% of the respective bins concentrations are 0. For instance, the polluted DSDs on the bottom layer present, in most cases (higher than 75%), bin concentrations equal to 0 for D>20 µm. In that case, the quartiles will go to zero in that size range.

The average is not required to be inside the interquartile range, do not take it by the median. When the average is outside the interquartile range, it means that a few measurements presented high enough concentrations to bring the mean value even higher than the 75% quartile. When the average is not null and the quartiles are zero, it means that there are only a few DSDs (frequency lower than 25%) contributing to the respective size range.

69q) p.10, l.8: please check the quartiles and averages, as in the last figure.

69a) See previous question.

70q) p.10, l.17: the size of the activated droplet depend mostly on the super saturation and not on the aerosol.

70a) The size of the aerosol where the water is condensing defines the initial size of the droplet. As such, bigger aerosols would favor the formation of bigger droplets. If this was not the case, giant CCNs would have a similar impacts on the clouds as a smaller CCNs.

71q) p.12, l.22: Please review all references. This one, for example, was not cited in the text.

71a) We cited Kuhn et al. (2010) while answering question 41. Checked all other citations to make sure everything is correct.

72q) p.19, l.2: what is theta?

72a) Changed the sentence to: “θ is the azimuth angle and is zero for East direction and grows counterclockwise”. Theta is the azimuth angle.

73q) p.20, l.1: these fluctuations at higher altitudes doesn’t make sense… it clearly shows that you have lower statistics and hence large standard deviation. You should decrease the vertical resolution.

73a) Changed the resolution to 800m, no significant impacts on the results.

74q) p.22, l.4: please make it centered. You choose a log-x scale, hence it is not possible to infer the limits (and hence the center) of each horizontal bin.

74a) It is centered now.

75q) p.23, l.1: It seems there is something wrong. The average value (dark blue) is not within the 25-75% quantile (light blue). Interquartile range for red and green goes to zero at larger sizes.

75a) See question 68.

MARKED-UP MANUSCRIPT STARTS ON NEXT PAGE

Impacts of the Manaus pollution plume on the microphysical properties of Amazonian warm-phase clouds in the wet season

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Abstract. The remote atmosphere over the Amazon can be similar to oceanic regions in terms of aerosol conditions and cloud type formations. This is especially true during the wet season. The main aerosol-related disturbances over the Amazon have both natural sources, such as dust transport from Africa, and anthropogenic sources, such as biomass burning or urban pollution. The present work considers the impacts of the latter on the microphysical properties of warm-phase clouds by analyzing observations of the interactions between Manaus ~~city~~pollution plume and its surroundings, as part of the GoAmazon2014/5 Experiment. The analyzed period corresponds to the wet season ~~over a tropical rain forest (i.e.,~~(specifically from Feb to Mar 2014 and corresponding to the first Intensive Operating Period (IOP1) of GoAmazon2014/5~~), and the droplets observed~~). The droplet size distributions reported are in the range 1 μm ≤ D ≤ 50 μm in order to capture the processes leading up to the precipitation formation. The wet season largely presents a clean background atmosphere characterized by frequent rain showers. As such, the contrast between background clouds compared to those affected by the Manaus pollution can be observed and detailed. The focus is on the characteristics of the initial microphysical properties in cumulus clouds predominantly at their early stages. The pollution-affected clouds are found to have ~~lower~~smaller effective diameters and higher droplet number concentrations. The~~ average~~ differences range from 10% to 40% for the effective diameter and are as high as 1000% for droplet concentration ~~across different~~for the same vertical levels~~ (0 to 3200 m).~~. The growth rates of droplets with altitude are slower for pollution-affected clouds (2.90 compared to 5.59 μm km^{-1}), as explained by the absence of bigger droplets at the onset of cloud development. Clouds under background conditions have higher concentrations of larger droplets ~~(e.g., >~~(≥ 20 μm) ~~close to~~near the cloud base, which would contribute significantly to the growth rates through the collision-coalescence process. The overall shape of the droplet size distribution (DSD) ~~overall shape do~~does not appear to be predominantly determined by updraft strength, especially beyond the 20 μm range. The aerosol conditions play a major role in that case. However, the updrafts modulate the DSD concentrations and are responsible for the vertical transport of water in the cloud. The larger droplets found in background clouds are associated with weak water vapour competition and a bimodal distribution of droplets sizes in the lower levels of the cloud, ~~that~~which enables an earlier initiation of the collision-coalescence process. This study shows that the pollution produced by Manaus

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affects significantly warm-phase microphysical properties of the surrounding clouds by changing the initial DSD formation. The corresponding effects on ice-phase processes and precipitation formation shouldwill be the focus of future endeavors.

1 Introduction

The natural atmosphere of the Amazon is a self-contained system where the forest itself provides the nuclei for clouds, which in turn activate the hydrological cycle and help distribute the water that maintains the local flora. Under undisturbed conditions the aerosol particles that serve as cloud condensation nuclei (CCN) are mainly secondarily generated from the oxidation of biogenic gases (Pöschl et al., 2010). Primary aerosols emitted directly from the forest may also contribute to the overall CCN population and are especially active as ice nuclei (IN). A review of the cloud-active aerosols' properties and sources in general is provided inby Andreae and Rosenfeld (2008) and specifically for the Amazon refer toby Martin et al. (2010). The results presented herein relate to the local wet season, which presents a relatively clean atmosphere compared to the local dry season when biomass burning is more frequent (Artaxo et al., 2002).

Given such an environment it is interesting to study the impacts that a city like Manaus haves on itsthe atmospheric conditions. Manaus is located in the Brazilian Amazonas state, in the middle of the forest, and has a population of arboundt 2 million people. The human activities associated towith the city produce air pollution, which interacts with the natural background gases and particles. Several studies found that city pollution enhanced atmospheric oxidation (Logan et al., 1981; Thompson, 1992; Kanakidou et al., 2000; Lelieveld et al., 2008), which not only impacts human health but also may interact with biogenic gases to increase secondary aerosol formation. Another example is the interaction between volatile organic compounds (VOCs) with the urban NO_x, which leads to enhanced ozone concentrations through a photochemical process (Trainer et al 1987, Chameides et al., 1992; Biesenthal et al. 1997; Starn et al. 1998; Roberts et al. 1998; Wiedinmyer et al., 2001).

The effects that the Manaus city has on the chemical properties of the local atmosphere potentially alter the way in which clouds are formed. Not only can the human activities change particles chemical properties, they can also increase the number concentration available for droplet formation. Most of this additional particulate matter is tied to emissions from traffic and power plants. in the case of Manaus. Previous studies regarding the effects of anthropogenic aerosols on Amazonian clouds propertiescloud generally focused on biomass-burning related occasions (e.g. Roberts et al., 2003; Andreae et al., 2004; Freud et al., 2008, Martins and Silva Dias, 2009) in the dry season.or transition seasons. However, no study evaluatesd the urban aerosol interaction with clouds over the rain forest during the wet season, when biomass-burning is strongly reduced and the background is very clean. Polluted cloudsgiven the frequent rain showers that leave the forest wet and more difficult to burn. In this case, the effects of the Manaus plume can be studied separately and in detail. Polluted clouds over the Amazon usually present more numerous but smaller droplets that grow inefficiently by collision-coalescence and therefore delay the onset of precipitation to higher altitudes within clouds (Rosenfeld et al., 2008).

The results presented herein are based on data sets collected between February and March 2014 during the first Intensive Operati~~o~~n~~gs~~ Period (IOP1) of The Observations and Modeling of the Green Ocean Amazon (GoAmazon2014/5) experiment (Martin et al., submitted). The period is in the wet season, which presents a clean atmosphere due to the ~~frequent rain showers.~~reduction in biomass burning. The pristine characteristic of the background air provides the opportunity for ~~studying~~contrasting the ~~impact~~microphysics of ~~the Manaus~~natural and urban pollution~~ plume on the chemistry and concentrations of biogenic particles released by the Amazon forest~~-affected clouds. Due to the proximity to the Intertropical Convergence Zone (ITCZ) and the trade winds, the large-scale motions are rather stable over the region for the campaign period. Most of the time, trade winds from the northeast prevail, advecting the pollution plume southwestward. This ~~scope~~scenario allows for the first time the direct comparison between clouds formed under background ~~clouds~~conditions and those affected by pollution in the wet season.

Clouds in the wet season differ from those in the dry and transition periods both because of aerosol conditions and large-scale meteorology (Machado et al, 2004). Although there is not a complete reversal of the mean wind directions intra-annually, the wet season clouds can be related to a monsoon system, usually referred as South American Monsoon System (SAMS). Zhou and Lau (1998) suggest~~s~~ that the monsoon-like flow can be understood when analyzing monthly anomalies on the wind fields. During the austral summer months, the winds tend to have a stronger northeastern component over Manaus area, while at austral winter time the stronger wind component is from the southeast. More details on the SAMS, including comparisons with other monsoon systems, can be found in Vera et al. (2006).

The main objective of this work is to understand the effects that anthropogenic urban pollution have on cloud droplets properties and development in the Amazon during the wet season. Specifically, the focus is on the comparison between warm-phase properties of clouds affected and not affected by the pollution emitted from Manaus city. The urban aerosol effect will be analyzed as function of height above the cloud base and vertical velocity. Section 2 describes the instrumental setup and the methods used for the analysis. The main findings are detailed in Section 3, while the summary and discussion are presented in Section 4.

2 Methodology

~~Several~~Sixteen research flights took place near Manaus in the Amazon forest between February and March 2014. Manaus coordinates are 3°06'S, 60°01'W and the dates and time periods of the flights are listed in Table 1, with times in UTC (local time is UTC-4). The U.S. Department of Energy Atmospheric Radiation Measurement program Gulfstream-1 (G-1) airplane (Schmid et al., 2014) performed 16 flights while measuring aerosol concentrations and composition, radiation quantities, gas-phase chemistry and cloud~~s~~ microphysical properties. The G-1 aircraft performed mostly short-ranged flights from Manaus, with most of the observations being held closer than 100 km from Manaus. The flight patterns were mainly focused on measuring properties in and around the city pollution plume. A schematic for the concepts of the flight planning is shown in Figure 1. The actual patterns varied daily depending on the weather forecast and plume dispersion prediction (Figure 2).

Additionally, other patterns were performed such as a run upwind from Manaus in order to probe a background air reference, or cloud profiling missions (vertical slices of the cloud field). However, the kind of pattern shown in Figure 1 was the most used and is the determinant to assess the interaction between the urban plume with the background atmosphere.

During the wet season it is very common to observe ~~Cu fields~~cumulus clouds as exemplified in Figure 1 and the G-1 cloud measurements consist~~s~~ed mostly of quick penetrations in those types of systems. From Manaus airport, the aircraft performed several legs perpendicular (or as close to as possible) to the plume direction while moving away from the city. At the end of the pattern, the aircraft start~~s~~ed over in a different altitude and perform~~s~~ed the same flight legs. In this way, it was possible to collect not only data regarding the plume but also on the surrounding background air. During the local wet season, the background atmosphere is rather clean and the effects of the plume can be readily observed. The pollution-aerosols in this situation are almost only urban and biomass-burning contribution is very exceptional. The main idea to compare the background and polluted clouds is to accumulate statistics inside and outside the plume sections as shown in Figure 1. By concatenating the observations for the different flights, it ~~is~~was possible to obtain a dataset of background and polluted droplet size distributions (DSDs), which can then be used to look at aerosol impacts in different ways. All G-1 flights were used in order to obtain the highest sample size possible. Figure 2 shows the trajectories for all flights, where the ~~color represents the flight number, numbered chronologically from 1 to 16. Manaus' coordinates are 3°06'S, 60°01'W. The dates and time periods of the flights are listed in Table 1, with times in UTC (local time is UTC-4).~~dashed grey lines represent the plume angular section considered from the airplane data. Note that the plume usually disperses from Manaus to the T3 site, with relatively small variations on the direction based on the wind field. Two flights (4 and 6) had low sampling on the plume given the trajectories and the grey lines may not represent the overall region of the plume. However, the directions identified presented higher CN concentrations than the other ones.

2.1 Instrumentation

The two main instruments used for this study were the Condensational Particle Counter (CPC, TSI model 3025), and the Fast Cloud Droplet Probe (SPEC Inc., FCDP). The CPC instrument measures number concentration of aerosols between 3 nm and 3 μm using an optical detector after a supersaturated vapor condenses onto the particles, growing them into larger droplets. Particle concentrations can be detected between 0 and 10^5 cm^{-3}~~.~~, with an accuracy of ±10%. Coincidence is less than 2% at 10^4 cm^{-3} concentration, and corrections are automatically applied for concentrations between 10^4 cm^{-3} and 10^5 cm^{-3}. The CPC was mounted in a rack inside the cabin and connected to an isokinetic inlet and an aerosol flow diluter and was operated using an external pump. The isokinetic inlet has an up limit of 5 μm for particle diameter, with penetration efficiency higher than 96%. A 1.5 LPM flow rate was maintained using a critical orifice. The dilution factor varied between 1 and 5.

The FCDP measures particle size and concentration by using focused laser light that scatters off particles into collection lens optics and is split and redirected toward 2 detectors. The FCDP bins particles into twenty bins ranging between 1 and 50 μm~~.~~, with an accuracy of approximately 3 μm in the diameters. Bin sizes were calibrated using glass beads at several sizes in the total range. The FCDP was mounted on the right wing of the G-1 aircraft. Shattering effects were filtered from the FCDP-measured DSDs (Droplet Size Distributions), which is a built-in feature of the provider software. Additionally, measurements with low number concentrations (< 0.3 cm^{-3}) and low water contents (< 0.02 gm^{-1}) were excluded.

The quality flag of the CPC instrument was used to correct the concentration measurements. Whenever an observation was flagged as "bad", it was substituted by an interpolation between the closest measurements before and after it that were either "questionable" or "good". For "good" measurements, which represents 59% of all the measurements, the uncertainty is less than 10%. The interpolation weights decayed exponentially with the time difference between the current observation and the reference ones. If the reference observations were more than 10 s apart, these data were excluded. 16% of the data was interpolated in that manner, while only 0.02% had to be excluded. This process was required not only to smooth out the bad measurements but also was important to maintain significant sample sizes (instead of simply excluding "bad" measurements). No averaging was applied to the 1 Hz CPC data. However, tests were made in order to check the impact that the sample frequency had on the results. The results were not sensible to moving averages of up to 10 seconds, which corresponds to roughly 1 km displacement given that the G-1 flew around 100 ms^{-1} in speed. Given this observation, the analyzes are based on the 1 Hz CPC measurements.

Complementary measurements of meteorological conditions were obtained from the Aventech Research Inc. AIMMS-20 instrument (Aircraft-Integrated Meteorological Measurement System). – Beswick et al., 2008). This instrument combines temperature, humidity, pressure, and aircraft-relative flow sensors in order to provide the atmospheric conditions during the measurements. From the aircraft measurements of relative flow, the vertical wind speed was obtained and was used herein to compare cloud properties in the up and downdraft regions. The accuracyprecision of vertical wind speeds is 0.75 m s^{-1} at 75 m s^{-1} true airspeed.

2.2 Plume classification

In order to compare two different populations of clouds, namely those formed under background conditions compared to those affected by pollution, a classification scheme was developed. The most discernible and readily observable difference between a polluted and background atmosphere is the number concentration of aerosol particles per unit volume. Urban activities such as traffic emit large quantities of particles to the atmosphere, which are then transported by atmospheric motions and can participate in cloud formation., especially when they grow, age and become more effective droplet activators. Their number concentration and sizes primarily determine their role on the initial condensational growth of cloud droplets through the aerosol activation mechanism. Even though the urban aerosols have a lower efficiency to become CCN (cloud condensation nuclei), their number concentrations are high enough to potentially produce a higher number of cloud droplets (see, for example, Kuhn et al., 2010). By affecting the initial formation of the droplets, increased aerosol concentrations due to urban activities can alter the cloud microphysical properties throughout its whole life cycle. It will be considered here that a simple, yet effective, classification scheme should consider primarily aerosol number concentrations to discriminate polluted and background conditions with respect to cloud formation environments. The intent of the classification scheme is not to quantify specifically the aerosols concentrations available for cloud formation under background and polluted conditions. Rather, it is a way to identify atmospheric sections that presented urban or natural aerosol characteristics.

Aerosol particle number concentrations (CN) measured by the CPC-3025 instrument were used to identify the plume location. The first procedure required is the elimination of possible artifacts related to measurements while the aircraft was inside a cloud. For that purpose, a cloud mask must be considered. The data are considered to be in-cloud by examining particle concentrations detected by several aircraft probes. The aircraft probes used to determine the presence of cloud are the Passive Cavity Aerosol Spectrometer (PCASP, SPEC Inc.), the 2D-Stero Probe (2D-S), and the Cloud Droplet Probe (CDP, Droplet Measurement Technologies). The thresholds for detection of cloud are when either the PCASP bins larger than 2.8 μm have a total concentration larger than 80 cm^{-3}, the 2D-S total concentration is larger than 0.05 cm^{-3}, or the CDP total concentration is larger than 0.3 cm^{-3}. Thresholds were determined by examining the sensitivity of each instrument. Assuming that the presence of clouds can affect the CN measurements, the concentrations inside clouds were related to those in clear air. Whenever an in-cloud observation is detected, the CN concentration is substituted by the closest cloud-free measurement (given that they are not more than 15 s apart, in which case the data are excluded from the analysis). In this way, possible cloud and rain effects on aerosols concentrations, such as rainout or washout, can be mitigated on the analysis.

A simple and fixed threshold to separate the background and polluted observations is not enough because the altitude of the measurements should also be taken into account. For that purpose, all CPC data were used to compute vertical profiles of particle number concentrations in ~~4~~800-m altitude bins. This resolution was chosen in order to result in significant amounts of data in each vertical bin. A background volume is identified whenever the measured particle concentration is below the 25% quartile profile. The polluted ones are considered to be the ones above the 90% profile. Additionally, it is required that the measurement is located in the general direction of the urban pollution dispersion in order to be considered a plume volume. Similarly, the background measurements are limited to the section outside the plume location only. It is important to note that, while the CPC data are available for the whole duration of the flights, in-cloud observations are limited to the times of actual penetrations. The choice of asymmetric 25% and 90% profiles result in similar sample sizes for the classified polluted and background in-clouds measurements (~~319~~305 and ~~431~~424 s, respectively), while maximizing the differences between the populations.

Given the daily variations of meteorological characteristics, the plume direction, width, and overall particle concentrations may vary. For that reason, the plume angular section must be obtained for each day individually. Figure 3 shows an example of plume classification for the flight on 10 March 2014. The CN concentrations are shown as a function of the azimuth angle with respect to Manaus airport (0° is east, grows counterclockwise), irrespective of altitude. The color represents the horizontal distance (km) from the airport. Note that there is an angular section where the concentrations are high not only close to the city but also as far as 70 km. This section is defined to be affected by Manaus pollution plume (delimited by grey dashed lines in Figure 3). Note that the coordinate system is centered on Manaus' airport, where the G-1 took off, and not on the center of the city or other point of interest. For this reason, it is also possible to observe relatively high CN concentrations close to the origin and to the northeast and southeast directions. This corresponds to high CN concentrations over the city. By keeping those directions outside the plume angular section, this data is not considered as plume. This is intentional because other aspects occur over the city that may contribute to the cloud formation. For instance, the heat island effect may contribute to the convection, changing the thermodynamic conditions compared to those over the forest. By keeping the origin point as the airport, which is located on the west section of the city, this problem is avoided.

The final result of the classification scheme for March 10 is shown in Figure 4. A visual inspection of radiosonde (released from the T3 sitePonta Pelada airport located on southern Manaus) trajectory plots confirmed the overall direction of the plume for each flight. Given the nature of the meteorology in the Amazonian wet season, i.e. its similarities with oceanic conditions concerning horizontal homogeneity, there isshould be no significant difference between the thermodynamic conditions inside and outside the plume region for the G-1 flights. In this way, differences observed in pollution-affected clouds are primarily due to the urban aerosol effects. It should be noted that even though the plume classification is defined from the CN measurements, there are also observable differences regarding CCN concentrations. The in-plume CCN concentrations (for altitudes lower than 1000 m) averages at 257 cm^{-3} for a 0.23% supersaturation, while the respective background concentration is 107 cm^{-3} (Figure 5). Note the overall low concentrations representative of the wet season. In that case, the plume increases the CCN concentrations by more than a factor of 2. For higher supersaturation conditions (which can be achieved in strong updrafts), the differences are even more pronounced. At 0.5% supersaturation, the average CCN concentration inside the plume is 564 cm^{-3}, while outside it is 148 cm^{-3}. This shows that the plume increases the concentration of aerosol particles that are able to form cloud droplets under reasonable supersaturation conditions, even though they are less efficient than the particles in the background air.

In addition to the plume, the river breeze also plays a role on the convection characteristics over the region and the respective microphysics. The clouds directly above the rivers are usually suppressed given the subsidence from the breeze circulation. This was addressed by comparing the DSDs under plume and background conditions only for measurements over land and it showed a similar picture to what will be shown in the next section. In this way it is possible to confirm that the results presented here reflects the effect of Manaus pollution plume and not the river breeze, even though the clouds over land were indeed more vigorous. The results shown on the next section consists of the data probed both above rivers and above land.

3 Results

Bulk DSD properties for polluted and background clouds

Given that the aerosol population directly affects cloud formation during the CCN (Cloud Condensation Nuclei) activation process, bulk DSD properties under polluted and background conditions may differ. Figure 56 shows the frequency distribution of the droplet number concentrations (DNC), liquid water content (LWC), and effective diameter (D_e) for all measurements inside the plume and under background conditions, irrespective of altitude. Those bulk properties were obtained from the FCDP-measured DSDs. The background clouds presented droplet number concentrations below 200 cm^{-3}

for most cases, while being more dispersed for the polluted DSDs. It shows that it is much more likely to find higher DNC under polluted conditions than on background air. This observation may be tentatively justified as an increase in the water vapor competition, which leads to the formation of a higher number of droplets with ~~lower~~smaller diameters. However, the water vapor competition is usually discussed for a fixed LWC, which is not the case for the statistics shown here. The background clouds measured presented lower water contents overall, which could also partly justify the lower concentrations observed.

The effective diameter histograms show distinct droplet sizes distributions for both populations. While around 50% of droplets in the polluted clouds have ~~effective sizes~~D_e between 8 and 12 μm, the frequency distribution for the background clouds shows more frequent occurrence of D_e > 12 μm~~. The distributions~~, even though they peak at ~~different~~similar diameters~~, with the modal D_e being larger in background conditions.~~. This factor shows that, ~~even with~~despite condensing lesser amounts of total liquid water, the background clouds are able to produce bigger droplets than their polluted counterparts. Overall, Figure ~~5~~6 shows a picture consistent with the water vapor competition concept. However, the DSD formation under a water vapor competition scenario depends on two factors. One is commonly cited on the literature (e.g. Albrecht, 1989) and is related to the impacts on effective droplet sizes as function of aerosol number concentrations. The other factor is how much bulk water ~~there is for~~ the ~~aerosol population~~systems are able to ~~compete on.~~condense while the vapor competition is ongoing. Figure ~~5~~6 suggests that the Manaus pollution plume affects both mechanisms, ~~and the analysis is~~which are more complex than the water vapor competition process.

An interesting question to address is why LWC is lower for background clouds, i.e., why this type of cloud is relatively inefficient to convert water vapor to liquid droplets. One possible answer is related to total particle surface area in a given volume. Considering a constant aerosol size distribution, when their total number concentration is increased, the total particle surface area per unit volume also increases. In this way there is a wider area for the condensation to occur, leading to higher liquid water contents. Additionally, if there is higher competition for the water vapor, the more numerous and smaller droplets formed under polluted conditions will grow faster by condensation than their background counterparts (because the condensation rate is inversely proportional to droplet size) and will readily reach the threshold for detection by the FCDP (around 1 μm). One point to remember is the high amount of water vapor available during the wet season. Those differences in the bulk condensational growth under polluted or background conditions may explain in part the differences observed in Figures ~~5c~~6c-d, even if the aerosol size distribution changes from the background to the polluted sections. If the bulk condensation is more effective in a polluted environment, it should also lead to increased latent heat release and stronger updrafts. In a stronger updraft the supersaturations tend to be higher, which feeds back into an even higher condensation rate. Other possible physical explanations for the higher LWC in polluted clouds include processes associated with precipitation-sized droplets (i.e., outside the FCDP size range) and aerosol characteristics. If the aerosol-rich plume is able to reduce the effective sizes of the liquid droplets, it will also be able to delay the drizzle formation. In this way, the liquid water would remain inside the cloud instead of precipitating. On the other hand, the fast-growing droplets in the background clouds may grow past the FCDP upper threshold, effectively removing water from the instrument size range. However, the clouds penetrated were predominantly non-precipitating cumulus at early stages of their life cycle. Therefore, the warm-phase was not completely developed and the condensational growth plays a major role in determining the overall DSD properties. Calculations show thatThe second process identified (i.e. suppressed precipitation staying longer inside the clouds) probably has a lesser impact. The averaged ratio between second moment of the polluted DSDsand background DSDs is around 2, which shows that the former have around twice of the total area for condensation (in average) than their background counterparts, which suggests that. In this way, the increase in the bulk condensation efficiencty is probably significant. Given a higher area for condensation, the type of aerosol can play a significant role. Urban emissions may contribute to higher inorganic fractions, increasing the aerosol hygroscopicity and contributing for enhanced condensation. Further studies are encouraged in order to detail and quantify the processes that lead to the observed LWC amount. However, based on Koren et al. (2014), the most determinant factor contributing for the high amount of cloud water under polluted conditions seems to be related to the condensation process. In the referred paper, it is shown that the amount of total condensed water tends to grow with aerosol concentration in a pristine atmosphere.

In order to detail the pollution effects on the total condensation rate and on the DSD properties, averaged properties for different water content and updraft speeds are analyzed. Firstly, consideringgiven that the LWC is a measure of the total amount of water condensed onto the aerosol population, its correlation with the updrafts should be assessed. The updraft speed at cloud base can be understood as a proxy for the thermodynamic conditions, as it is a result of the meteorological properties profiles in lower levels. In this way, it is possible to disentangle the aerosol and thermodynamic effects by averaging the LWC data at different updraft speeds levels. Figure 6a7a shows the result of this calculation for only the lower 1000 m of the clouds, while also differentiating between polluted and background clouds. The 1000m limit is chosen for both maximizing statistics and also capturing the layer in which the aerosol activation takes place. That layer is possibly thicker under polluted conditions, given the higher availability of nuclei. For similar updraft conditions, i.e., similar thermodynamics, the averaged total liquid water is always higher for polluted clouds. By eliminating the dependence on the thermodynamic conditions, it is possible to conclude that the LWC values are significantly influenced by the aerosol population. This figure shows that, on average, not only are the polluted clouds are more efficient at the bulk water condensation but also the resulting LWC also scales with updraft speed. (linear coefficients, considering the error bars, are 0.13 g s m^{-2} for plume measurements and 0.033 g s m^{-2} for background clouds). In a background atmosphere, most of the aerosols readily activatehave been activated, and increases inincreasing updraft strength does not result in further condensation. On the other hand, the higher availability of aerosols inside the plume allows for more condensational growth as long as enough supersaturation is generated, especially considering that the critical dry diameter for activation is inversely proportional to supersaturation and, consequently, to the updraft speed. However, a deeper analysis in a bigger dataset would be required to assess the statistical significance. The enhanced condensation efficiency and the possible LWC scaling with updraft strength at least partly explain the higher liquid water contents in the plume-affected clouds. The standard deviation bars in Figure 6a7a indicate that while there is high variability for the LWC in polluted clouds, the clean ones are rather consistent regarding the condensation efficiency.

The water vapor competition effect can be observed by examining droplet effective diameter and number concentrations at a certain LWC interval, as shown in Figures 6b7b and 6c7c. In this way, the polluted and background DSD properties can be evaluated irrespective of the bulk efficiency of the cloud to convert water vapor into liquid water. It is clear that, even with the dispersion observed, the two DSD populations arepresent consistently different at anyaverage behaviors for all LWC intervals. For similar LWC, the averaged effective diameter is always higherlarger on background clouds, with lower droplet number concentrations on average. Those results show a picture clearly consistent with enhanced water vapor competition in polluted clouds. It shows that, given a bulk water content value, droplet growth is more efficient in background clouds. In other words, the liquid water is transported quicker into higher diameter ranges. This process should make background clouds more efficient to produce rain from the warm-phase mechanisms because of the early acinitivation of the collision-coalescence growth.

Another noteworthy point shown in Figure 67 is the difference between the relationships of D_e x LWC and LWC, and of DNC xand LWC. While the average effective diameter varies almost linearly with LWC, (R^2=0.95 for plume and R^2=0.92 for background DSDs), there seems to be a capping on DNC. This means that for low LWC (e.g., <(< 0.4 g m^{-3}), increases in the total water content are reflected in increased droplet concentrations. For higher LWC values, the averaged DNC remains relatively constant while the effective diameter grows with the water content. This suggests that at low water content levels, i.e., at the early stages of cloud formation, the formation of new droplets has a relatively higher impact on the overall LWC. As the cloud develops, the LWC is tied to the effective diameter of the droplets, as the impact of new droplet formation is weaker at this point. This effect is clearer in background clouds given the limited aerosol availability.

Vertical DSD development and the role of the vertical wind speed

The analysis of bulk DSD properties indicates a clear difference between the polluted and background cloud microphysics. However, it is desirable now to further detail those differences. As most of the aerosol activation takes place close to cloud base (Hoffmann et al., 2015), the direct effects of enhancements in particle concentrations should be limited to this region. However, the aerosol effect can carry over to later stages of the cloud life cycle given that it will develop under perturbed initial conditions. One proxy for the cloud DSD evolution in time is to analyze its vertical distribution. For a statistical comparison, a relative altitude for all flights is defined. This relative altitude is calculated as follows: firstly, the cloud base altitude is computed from the closest radiosonde is used in order to obtain the cloud base altitude (as the lifting condensation level, the 0°C isotherm as) and the freezing level (unless. In case the airplane did reach suchreached high enough altitudes, in which case its data is used instead). In general, most used to obtain the altitude of the clouds probed by G1 were cumulus clouds at their early stages0ºC isotherm. From those two levels, the relative altitude is calculated as percentages where 0% represents the cloud base and 100% is the freezing level. The altitudes of the cloud base and freezing levels range, respectively, from 100 m to 1200 m and from 4670 m to 5300 m approximately. Three layers are then defined: 1) bottom layer in which relative altitudes vary between 0% and 20%; 2) mid layer for 20% to 50%; and 3) top layer, where the altitude is above 50%. Those specific relative altitude intervals were chosen in order to capture the physics of the cloud vertical structure and to minimize the differences in sample sizes for each layer~~ (there are more measurements for lower levels~~, as there are more measurements for lower levels. Despite probing individual clouds, the DSD measurements can be combined into the three layers defined and interpreted as representative of a single system. It is conceptually similar to satellite retrievals of vertical profiles of droplets effective radii (e.g. Rosenfeld and Lensky, 1998), where the cloud top radius is measured for different clouds with distinct depths and combined into one profile. This approach was validated with in-situ measurements for the Amazon region by Freud et al (2008)).

Figure ~~7~~8 shows statistical results for the DSDs in the three warm layers defined, while Table 2 shows the respective mean bulk properties. The altitude-averaged values show that the polluted clouds present higher number concentrations and water contents and lower diameters for all layers. Additionally, DNC decays much slower with altitude and droplet growth is significantly suppressed. Those observations point to enhanced collisional growth in the background clouds.

The overall picture of cloud DSD vertical evolution can be seen in Figure ~~7a~~8a. The most discerning feature between the DSDs at different altitudes is related to the concentrations of droplets greater than 25 μm. The concentrations in this size range grow with altitude on average. On the other hand, the concentrations of droplets smaller than 15 μm tend to diminish from the bottom to the top layer. Considering that the vertical dispersion of the DSDs represents at least in part its temporal evolution, this feature is associated with droplet growth where the bigger droplets grow in detriment of the smaller ones. This growth mechanism is the collision-coalescence process, where the bigger droplets collect the smaller ones and acquire ~~its~~their mass. The shaded areas on the figure show that this is not only an average feature, but is also visible in the quantiles.

The statistical results of the vertical evolution of the DSDs are discriminated for the measurements inside the plume and in background regions in Figures ~~7b~~8b-c. At first glance, it is quite clear that the two DSD populations present different behaviors with altitude, meaning that the droplets grow differently depending on the aerosol loading. The plume DSDs present a high concentration on the bottom layer and shows weak growth with altitude. The concentration of small droplets ~~(e.g., <~~(< 15 μm) does not change much with altitude and the top layer DSD is relatively similar to the middle one. On the other hand, the DSDs in the background clouds show a stronger growth with altitude (Figure ~~7c~~8c). The bottom layer DSD presents lower concentrations of small droplets but higher concentrations of bigger droplets than its polluted counterpart does. This coexistence of relatively big and small droplets readily activates the collision-coalescence process, accelerating droplet growth. Comparing both polluted and background DSDs with the overall averages (Figure ~~7a~~8a), it is clear that enhanced aerosol loading leads to less-than-average growth rates and the opposite is true for background clouds. The average growth rate for D_e is 2.90 μm km^{-1} and 5.59 μm km^{-1} for polluted and background clouds, respectively.

The vertical speed inside the cloud is a critical factor as it helps determine the supersaturation and, consequently, the condensation rates in the updrafts. The interaction~~s~~ between the updraft speeds and aerosol loadings ultimately determines the initial DSD formations at cloud base. As mentioned before, the characteristics of the initial DSD may have impacts on the whole cloud life cycle, making the study of the vertical velocities critical for understanding the system development. Figure ~~8~~9 shows averaged DSDs for different cloud layers and vertical velocities conditions, discriminating between the p~~o~~l~~l~~u~~t~~med~~d~~ and background cases. The first row shows results for the bottom layer under a) p~~o~~l~~l~~u~~t~~med~~d~~ and b) background conditions. The mid and top layer results are shown together in the second row, for c) plume and d) background conditions. “Strong” and “Mod” are references to the up- or downdraft speed (strong or moderate). The mid and top layers are considered in conjunction in order to increase the sample size.

For the bottom layer, the vertical velocity has an impact mainly on the concentration of small droplets on polluted DSDs in the range $D < 5$ µm. The regions that presented updrafts are associated with higher concentrations of such droplets~~, as a result~~ because of new droplets nucleated under supersaturation. The downdraft regions mainly contain droplets that already suffered some processing in the cloud system and have relatively lower concentrations of small droplets that were probably collected by bigger ones. Additionally, small droplets ascend readily with the updrafts given their low mass, which is also a factor that can contribute to the differences between up- and downdraft~~s~~ DSDs. However, the dispersion shown in the shaded areas shows that the populations of DSDs in up- and downdrafts are relatively similar, suggesting a homogeneous layer with respect to DSD types. The DSDs shown on Figure ~~8a~~9a indicate single-mode distributions, which hampers collection processes and ~~justifies~~explains the similarities between the different vertical velocit~~ies~~y regions. On the other hand, the background clouds have a second mode, especially in the downdrafts given the additional cloud processing, which favor the collision-coalescence process. The particles associated with background air in the Amazon are not only less numerous but also bigger overall compared to the urban pollution, and both of those features favor faster growth by condensation because of less vapor competition and larger initial sizes. It is interesting to note that the background DSDs in the strong updraft regions are narrower when compared to their polluted counterpart. In a polluted environment, there is not only the natural background aerosol population but also the urban particles emitted from Manaus. The mixture of the two, with the consequent physicochemical interactions, permits the formation of droplets over a wider size range, with a prolonged tail towards the lower diameters. The shaded areas show that the differences between the DSDs in the up- and downdraft regions are statistically relevant for the background clouds and are not a mere averaging feature.

Cloud droplets keep growing as they move to higher altitudes, but the way in which it occurs is rather different in a background or plume-affected environment. For polluted DSDs, there are two modes at the higher altitudes: one reminiscent of the lower levels and the other is probably mainly a result of additional condensational growth. In those systems, the additional processing does not seem to be effective to produce bigger droplets, as shown by the blue line and shaded area in Figure ~~8c~~9c. For the background clouds, DSDs in the updraft regions ~~DSDs~~ show similar modes to their polluted counterparts, one close to 10 µm and the other at around 18 µm. However, there are ~~rare~~ appearances of droplets bigger than 30 µm that contribute to the formation of a third mode~~.~~ in the mid and top layers. This mode ~~is further highlighted~~appears on the strong downdraft regions, which suggests it appears after in-cloud processing.

4 Summary and conclusions

This study focused on the analysis of ~~cloud~~ microphysics of warm-phase clouds in Amazonia~~n~~ during the wet season, with a specific emphasis on interactions with the pollution emitted by Manaus city. A statistical approach was used to compare several clouds probed in different flights on different days. Concerning the effects of the pollution plume on the cloud DSDs bulk properties, there are two processes to consider. A polluted environment with high particle count presents a high total area for the condensation, favoring higher bulk liquid water on the DSDs. Additionally, the total amount of condensed water scales with updraft speed in the plume-affected clouds, which is not the case for background clouds. ~~On the other hand, the~~The growth processes under background aerosol levels are much more effective even with lower bulk liquid water contents. ~~The~~Despite the lower amount of water condensed in background DSDs~~ is readily transported to~~, bigger droplets readily form given the early start of the collision-coalescence process (which does not increase LWC). Polluted clouds ~~presented~~had droplets 10%-40% smaller on average and more numerous droplets (as high as 1000% difference) in ~~different~~the same vertical layers~~ from 0 m to 3200 m~~ inside the cloud.

The averaged DSDs in different layers of warm clouds show droplets grow with altitude overall, with bigger droplets acquiring mass from the smaller ones. However, the growth rates with altitude are much slower for plume-affected clouds (almost half of the clean growth rate) due to the enhanced water vapor competition and the lack of bigger droplets at the onset of the systems. Background clouds present~~ed~~ relatively high concentrations of droplets greater than 20 μm near cloud base that contributed to the growth rates, especially taking into account the non-linear nature of the collection process. With respect to warm-phase cloud DSDs, the updraft strength does not seem to be the major driving force for effective droplet growth, especially beyond the 20 μm range. The most important features to produce such big droplets are weak water vapor competition (usually observed in background clouds) and the existence of bi-modality at the lower levels of the cloud. The weak water vapor competition favors the formation of big droplets ~~(e.g., >~~(> 20 μm) required for the collision-coalescence process, while the bi-modality favors the efficiency of the collision-coalescence process due to the large terminal velocity differences between the modes. However, the thermodynamic role of the updraft speeds should not be underestimated. It is responsible for transporting hydrometeors beyond the freezing level, activating the cold processes. Those processes are known to be associated to thunderstorms and intense precipitation. ~~But~~Nevertheless, the main feature that determines warm-phase DSD shapes seems to be the aerosol~~s~~ conditions, with the vertical velocities playing a role in the modulation of the distributions.

While the effects of aerosol particles in the warm layer of the clouds are relatively straightforward, this may not be the case for the mixed and frozen portions. An aspect that was not directly addressed in this work is the impacts that warm layer characteristics have on the formation of the mixed phase (above the 0°C isotherm). Given that aerosols alter the properties of the whole warm phase, it is reasonable to assume that this would have an impact on the initial formation of the mixed layer. Such impacts can be in the form of the timing and physical characteristics of the first ice particles and the maximum altitude with supercooled droplets above the freezing level. This issue will be addressed in future studies, taking advantage of data provided by the HALO (High Altitude and Long Range Aircraft) airplane that operated in the second GoAmazon2014/5 IOP between September and October, 2014.

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Figure captions

Figure 1: Conceptual schematic for the flight patterns planning. It shows Manaus city and its pollution plume dispersing over the surrounding Amazon forest. The Cu field shown is very common during the wet season and is representative for most of the cloud conditions during the flights. The yellow circles indicate a 100 km radius from Manaus airport, although the figure is not meant to be quantitatively accurate. The lines with arrow heads show the most common flight plan used, where blue regions are possible locations for the background air measurements and the red ones indicate measurements inside the plume section (dashed white lines). T3 is a GoAmazon site to the north of Manacapuru.

Figure 2: Trajectories for all G-1 flights during GoAmazon2014/5 IOP1. Colors represent a numbering of the flights. Manaus is located close to the {-60, -3} point, marked with an "X", while the T3 site is marked with the black circle.

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Figure 3: CN concentrations around Manaus for 10 March 10, 2014. θ is the azimuth angle and is zero for East direction and grows counterclockwise. Colors are proportional to the horizontal distance (km) between Manaus airport and the aircraft. The black dots represent the angular mean CN concentration for each one of the 60 bins (azimuth). The vertical dashed lines represents the limits of the plume location.

Figure 4: The same as Figure 2, with the coloring representing the plume classification for 10 March 2014. The green-colored dots represent unclassified points, red is for plume, and cyan is for background conditions. The inset shows the median (cyan) and the 25% (Blue) and 90% (red) percentiles profiles of CN concentrations.

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Figure 6: Normalized histograms of cloud droplets properties affected or notand unaffected by the Manaus plume. (a-b) Total droplet number concentrations (cm^{-3}), (c-d) liquid water content (g cmgm^{-3}), and (e-f) effective diameter (µm).

Figure 6: (a)7: Mean (a) LWC values for different log-spaced w intervals and mean D_e, (b) and DNC, (c) for log-spaced LWC intervals. Error bars are the standard deviation for each interval. Blue points indicate background measurements, while red ones are relative to the polluted ones. The points are located at the upper limitmiddle of the respective bin intervals. Those results are limited to the first 1000 m of the clouds.

Figure 78: Averaged DSDs for three different cloud layers - bottom, mid and top of the warm layer. Graph (a) shows the results for all DSDs irrespective of classification, while (b) is for polluted DSDs only, and (c) for background. Lines represent averages, while the shaded areas represent the dispersion between the 25% and 75% quantiles.

Figure 89: Averaged DSDs as function of altitude, presence of up/downdrafts, and aerosol conditions. The first row shows results for the bottom layer under (a) polluted and (b) background conditions. The mid and top layers results are shown together in the second row for (c) plume and (d) background conditions. "Strong Down" means the presence of strong downdrafts, with velocities lower than -2 m s^{-1}. "Mod Down" is moderate downdrafts, with -2 m s^{-1} < $w \leq 0$. "Mod Up" and "Strong Up" are the equivalents for updrafts. Their velocities ranges are, respectively, $0 < w \leq 2$ m s^{-1} and $w > 2$ m s^{-1}. The shaded areas represent the dispersion between the 25% and 75% for the strong downdrafts (in blue) and updrafts (in red).

Table captions

Table 1: Dates and times for all G-1 flights during GoAmazon2014/5 IOP1. Local time for Manaus is UTC-4. All flights were carried out in the year 2014.

Table 2: Averaged bulk DSD properties for the three warm-phase layers and the respective standard deviations. The bottom layer is defined by relative altitudes between 0% and 20%, the mid layer between 20% and 50% and the top between 50% and 100%.

Tables

Table 1: Dates and times for all G-1 flights during GoAmazon2014/5 IOP1. Local time for Manaus is UTC-4. All flights were carried out in the year 2014.

Flight Number	Date	Start Time (UTC)	End Time (UTC)
1	February 22	14:38:27	17:25:26
2	February 25	16:32:06	18:40:07
3	March 1	13:35:37	15:27:35
4	March 1	17:18:48	18:47:07
5	March 3	17:46:34	19:11:57
6	March 7	13:09:51	15:35:25
7	March 10	14:26:37	17:09:35
8	March 11	14:42:23	17:51:08
9	March 12	17:21:25	19:29:42
10	March 13	14:16:09	17:21:27
11	March 14	14:18:54	16:48:23
12	March 16	14:40:17	17:26:32
13	March 17	16:24:40	19:26:36
14	March 19	14:26:38	17:17:48
15	March 21	16:33:47	18:56:07

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16	March 23	14:59:05	17:43:34

Table 2: Averaged bulk DSD properties for the three warm-phase layers and the respective standard deviations. The bottom layer is defined by relative altitudes between 0% and 20%, the mid layer between 20% and 50% and the top between 50% and 100%.

Layer	**DNC (cm^{-3})**		**D_e (µm)**		**LWC ($g\ m^{-3}$)**	
	Plume	Background	Plume	Background	Plume	Background
Bottom	317 ± 190	127 ± 131	11.3 ± 2.00	14.2 ± 4.19	0.206 ± 0.216	0.114 ± 0.122
Mid	360 ± 276	81.6 ± 77.4	17.7 ± 4.12	18.4 ± 6.18	0.848 ± 0.788	0.183 ± 0.218
Top	191 ± 203	7.64 ± 14.9	15.5 ± 5.28	31.7 ± 4.12	0.522 ± 0.703	0.0766 ± 0.151

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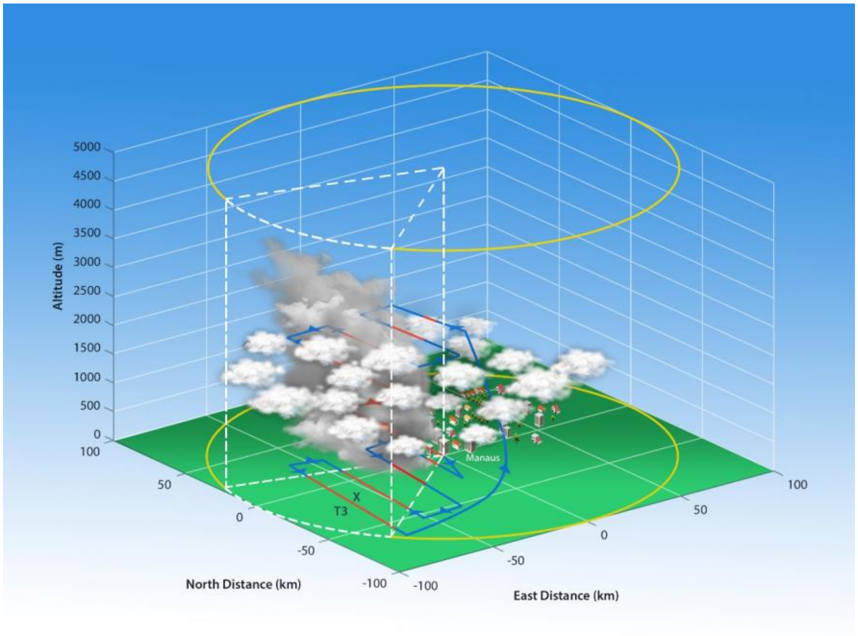
Figures



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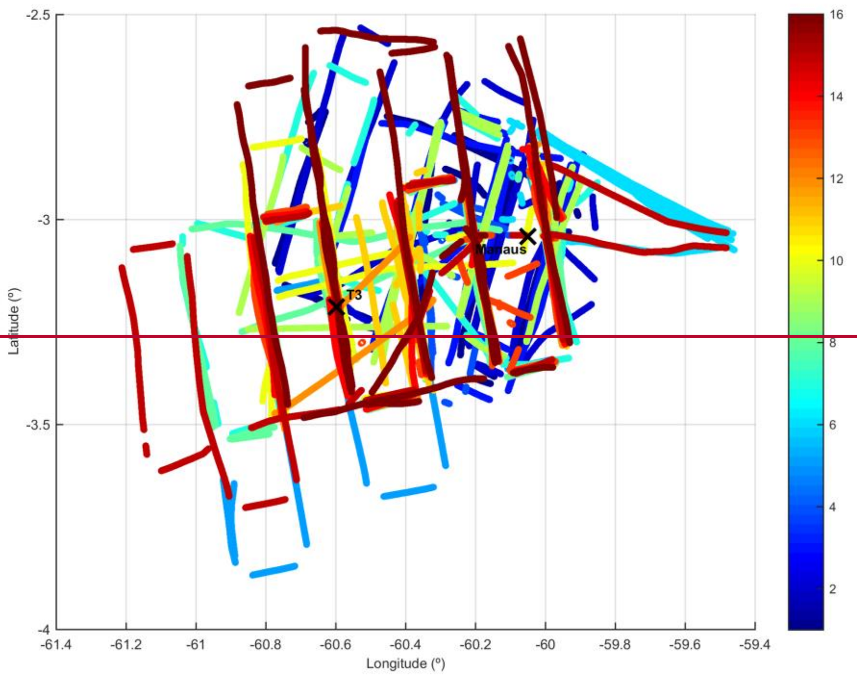
Latitude (º)
Longitude (º)
Manaus
T3
-2.5
-3
-3.5
-4
-61.4
-61.2
-61
-60.8
-60.6
-60.4
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-60
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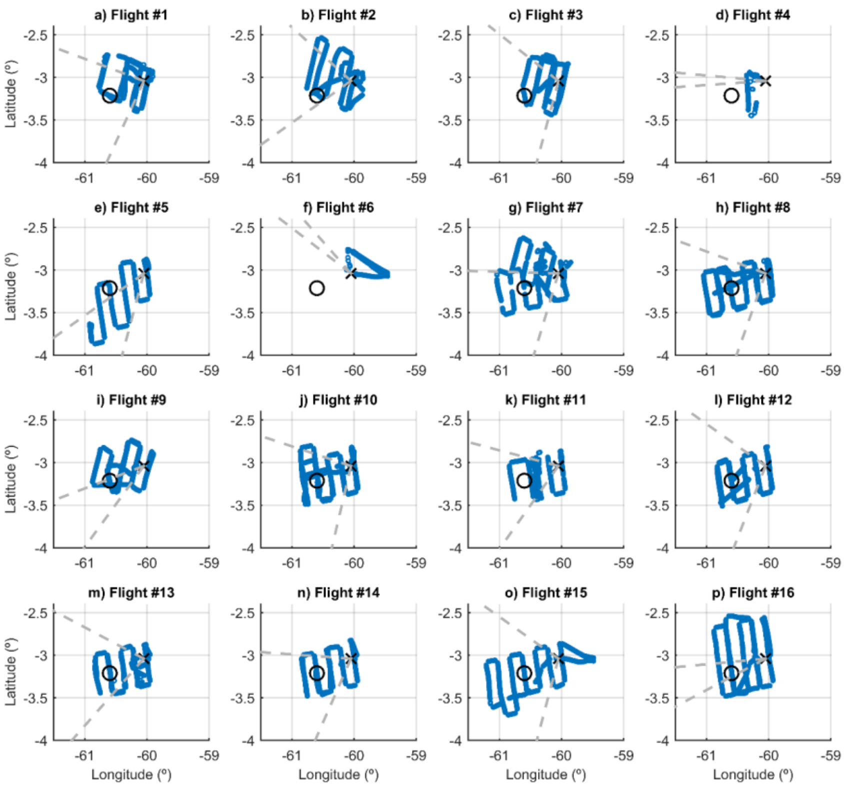
10
8
6
4
2



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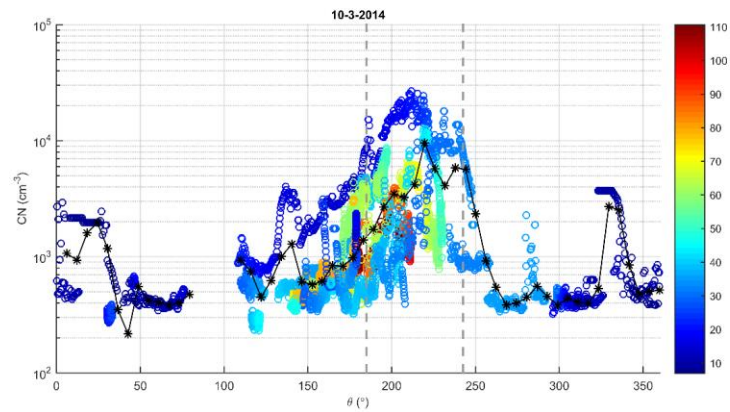

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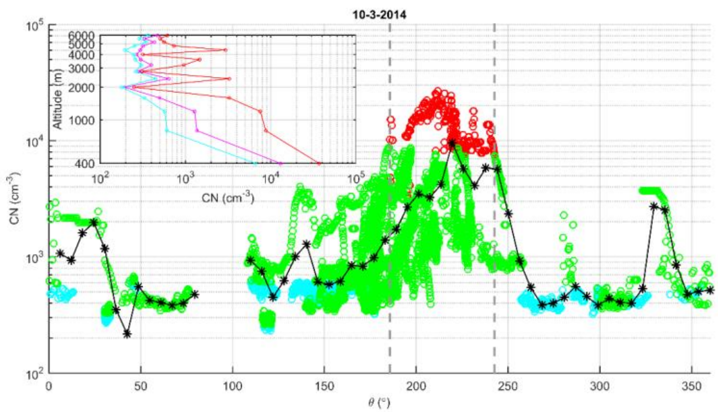
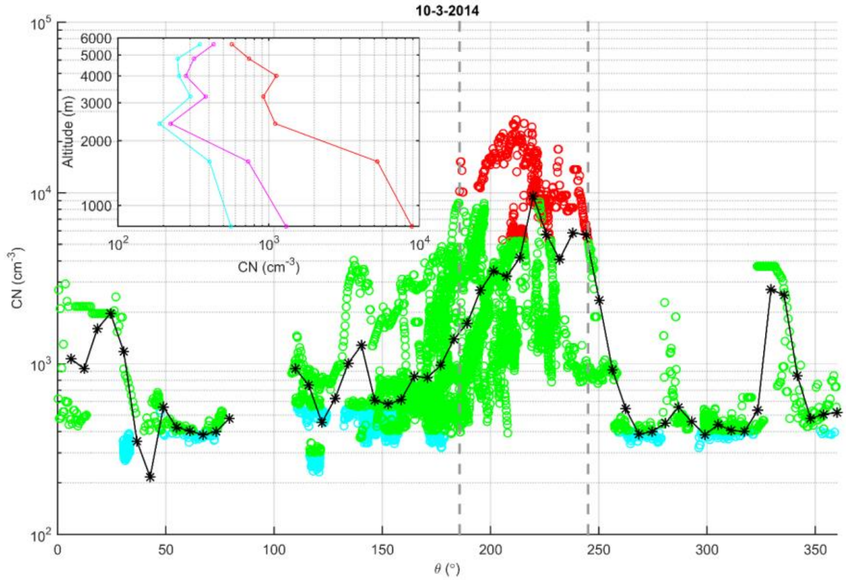

10-3-2014
CN (cm^{-3})
θ (°)
Altitude (m)
CN (cm^{-3})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CN (cm^{-3})
θ (°)
Altitude (m)
CN (cm^{-3})

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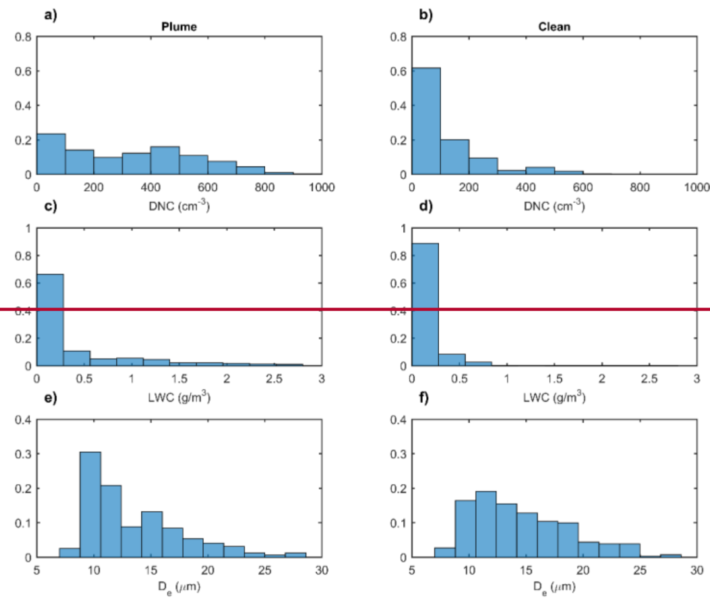
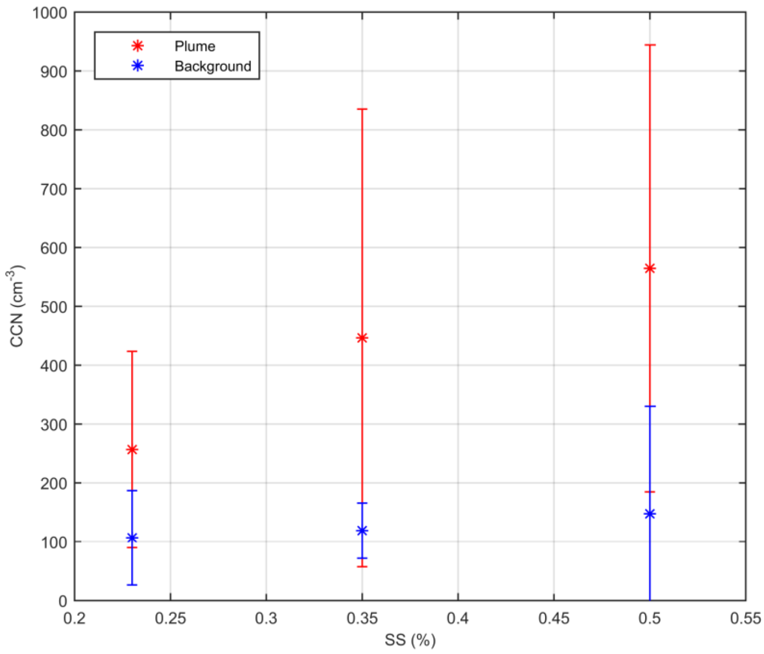

a)
Plume
b)
Clean
DNC (cm^{-3})
c)
d)
LWC (g/m^3)
e)
f)
D_e (μm)



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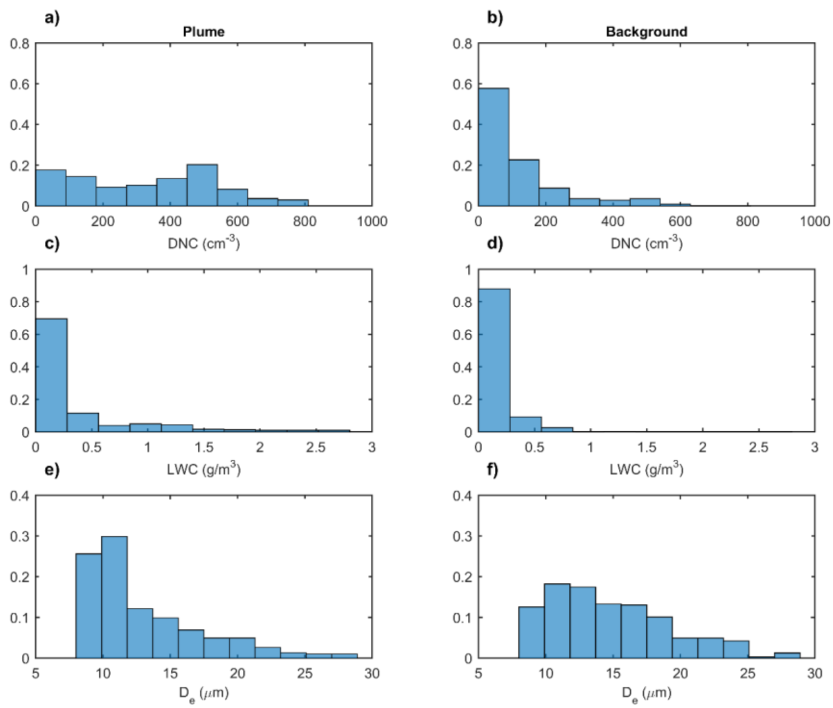

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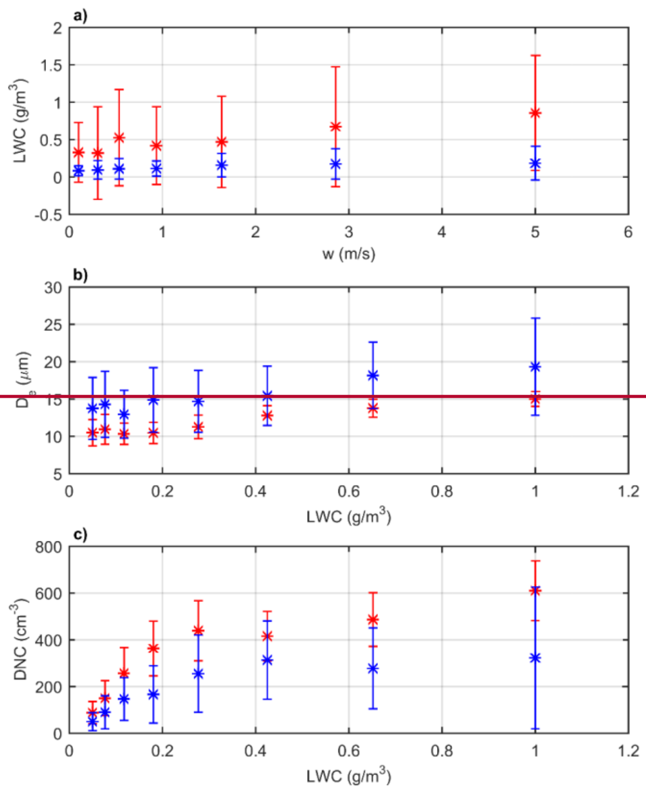

a)
LWC (g/m^3)
w (m/s)
b)
D_e (μm)
LWC (g/m^3)
c)
DNC (cm^{-3})
LWC (g/m^3)

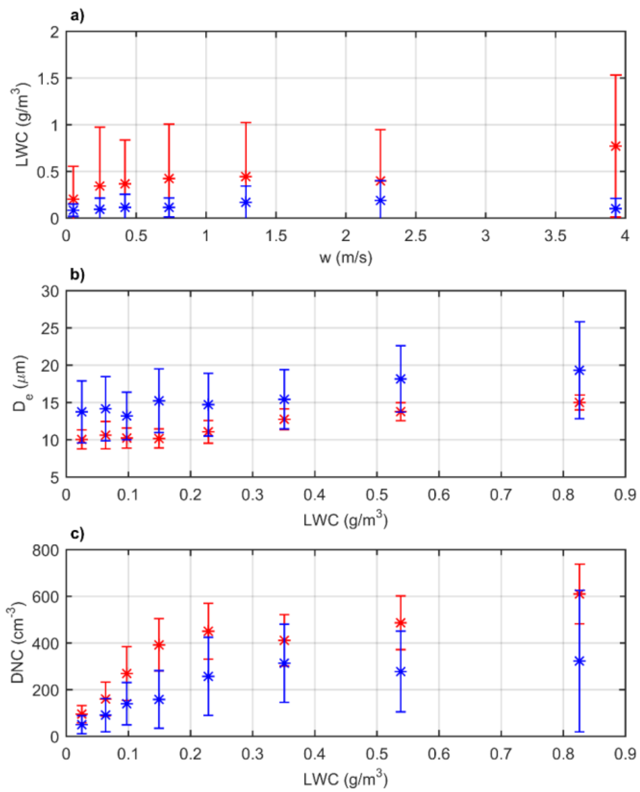
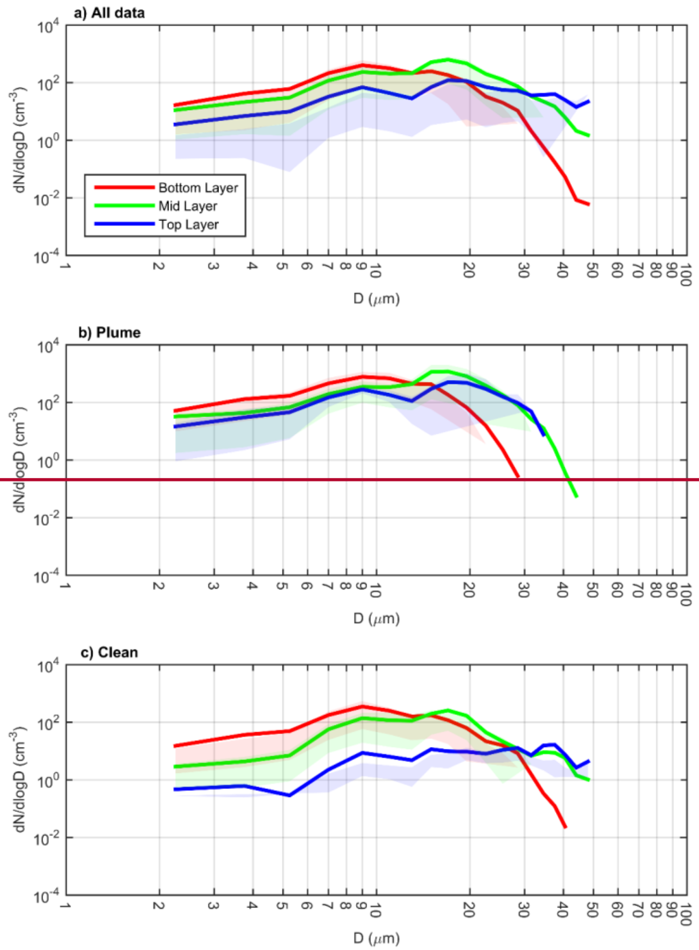


Figure ~~6:~~7: Mean (a) ~~mean~~ LWC values for different log-spaced w intervals and mean D_e (b) and DNC (c) for log-spaced LWC intervals. Error bars are the standard deviation for each interval. Blue points indicate background measurements, while red ones are relative to the polluted ones. The points are located at the ~~upper limit~~middle of the respective bin intervals. Those results are limited to the first 1000 m of the clouds.


a) All data
dN/dlogD (cm-3)
Bottom Layer
Mid Layer
Top Layer
D (μm)
b) Plume
dN/dlogD (cm-3)
D (μm)
c) Clean
dN/dlogD (cm-3)
D (μm)

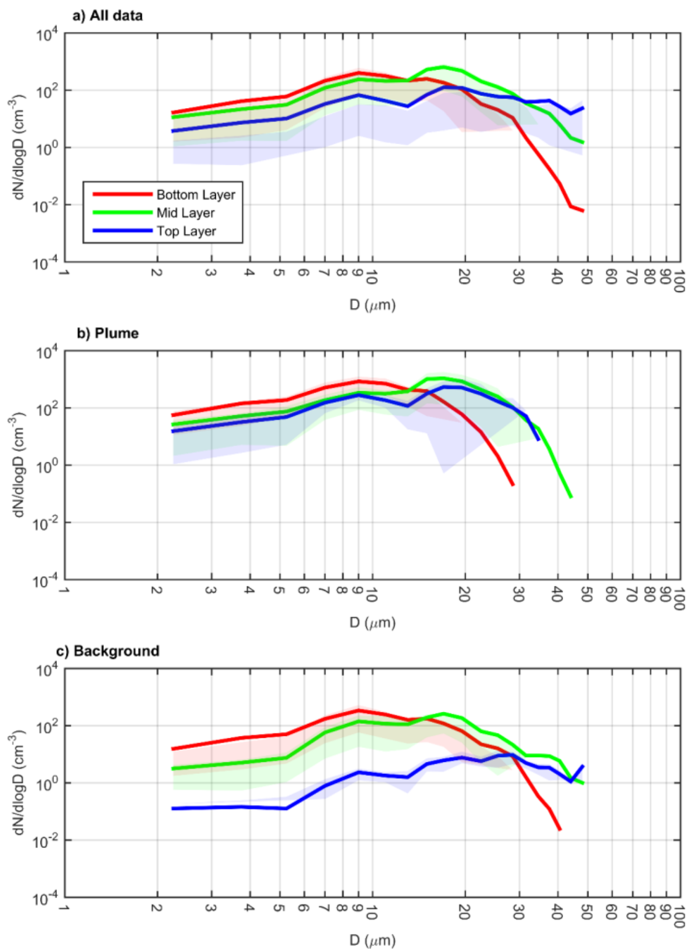
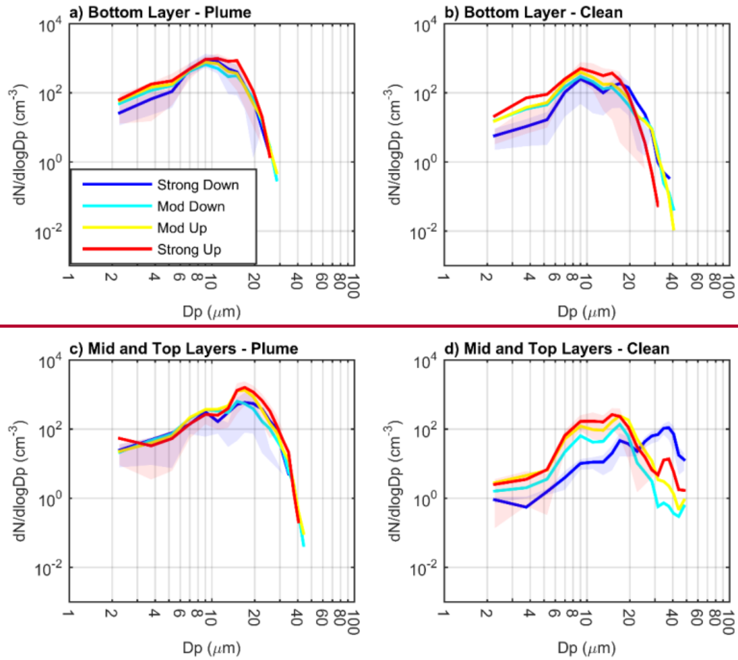


Figure ~~7~~8: Averaged DSDs for three different cloud layers of bottom, mid and top of the warm layer. Graph (a) shows the results for all DSDs irrespective of classification, while (b) is for polluted DSDs only and (c) for background. Lines represent averages, while the shaded areas represent the dispersion between the 25% and 75% quantiles.


a) Bottom Layer - Plume
b) Bottom Layer - Clean
c) Mid and Top Layers - Plume
d) Mid and Top Layers - Clean
Strong Down
Mod Down
Mod Up
Strong Up
dN/dlogDp (cm-3)
Dp (μm)

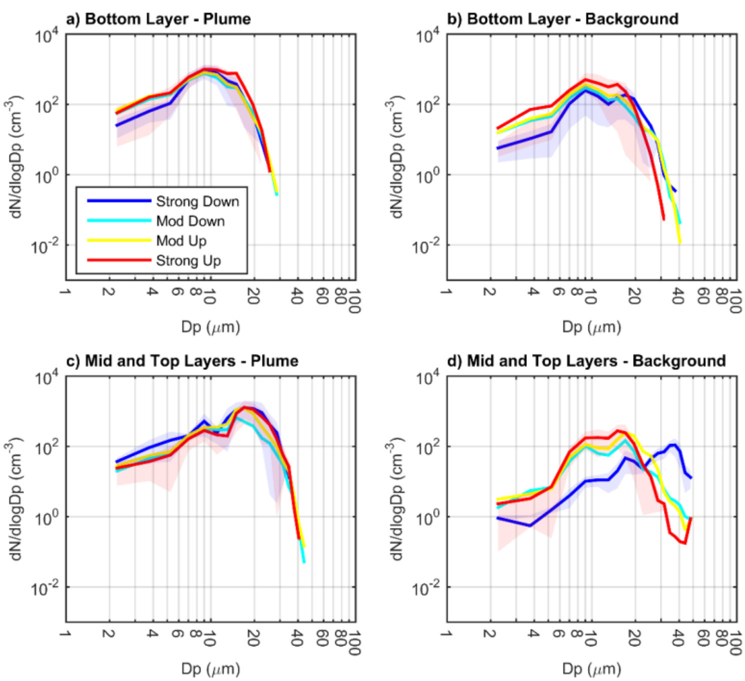


Figure 89: Averaged DSDs as function of altitude, presence of up/downdrafts and aerosol conditions. The first row shows results for the bottom layer under (a) polluted and (b) background conditions. The mid and top layers results are shown together in the second row for (c) plume and (d) background conditions. “Strong Down” means the presence of strong downdrafts, with velocities lower than -2 m s^{-1}. “Mod Down” is moderate downdrafts, with -2 m s^{-1} < $w \leq 0$. “Mod Up” and “Strong Up” are the equivalents for updrafts. Their velocities ranges are, respectively, $0 < w \leq 2$ m s^{-1} and w > 2 m s^{-1}. The shaded areas represent the dispersion between the 25% and 75% for the strong downdrafts (in blue) and updrafts (in red).