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Replies to referee #1

We thank the referee for the careful revision of the manuscript 'Direct observations of molecular clusters and nucleation mode particles in the Amazon'.

The comments improve the current manuscript. We will address all the comments and concerns in detail as shown below/ as in the following paragraphs.

General comments.
We thank the reviewer for suggesting the comparison of the two research sites. This issue has been addressed carefully in the revised manuscript.

The identity of specific sentences in the current manuscript were a mistake. We have re-phrased the identical sentences from previous publications in the revised manuscript.

We address the specific comments of the referee here below.

Referee comment:
There are specific sentences and complete text passages which are identical to Martin et al., 2016. The following list is not necessary complete. The authors should make sure that further text passages similar to other work are referenced correctly. I encourage to use the similarity report provided by the iThenticate plagiarism screening service.

Reply: All the identical text passages to previous publications have been re-phrased in the revised manuscript.

Specific comments:
Page 4, lines 134:
The authors state that T3 is located in a pristine environment. According to e.g., Martin et al., 2016 T3 (time points three) is located downwind of the pollution in a pasture area. I suggest to not use 'pristine' in this context.
Reply: We agree with the referee. The term pristine has been removed from the revised manuscript

Page 4, lines 118:
"T0t is mostly unaffected by the Manaus pollution and is surrounded by dense rainforest. It allows the characterization of an almost completely undisturbed natural environment"
- Did the authors filter for pollution affected periods? If so, what are the filter criteria?
Reply: In the general data analysis, we did not filter for pollution affected periods, since we report average values for the whole measurement period and wet/dry season specifically.
However, for the analysis of the NPF events, pollution events would appear in the NAIS/SMPS data as elevated aerosol concentrations in the accumulation mode. Also, the calculation of the condensation sink gives a good criterion for polluted days, which is clearly higher on non NPF days. Since we observed two nucleation events, with GR of approximately 10-20 nmh^{-1} and about 1 nmh^{-1}, it might be that the days with the higher GR are days which are more influenced by the Manaus pollution plume. Since the sulfuric acid concentrations seemed to be about the same on days with

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high and low GR, we may assume that the Manaus pollution is not the main factor influencing the air masses.

Page 4, lines 124:

The introduced DMPS measurements are performed using an inlet line above canopy. Nevertheless, the section is called 'inside canopy measurements' which is confusing. I further wonder if there are any comparisons of the DMPS and NAIS during the 3-year period to confirm the quality of measurements.

Reply: The section has been re-named to 'Measurements inside the rainforest' to avoid confusion. We changed the classification of the two sites in the whole manuscript accordingly. T0t is called inside rainforest site and T3 pasture or outside rainforest site.

The instrumentation was calibrated before shipping to the campaign and regular maintenance including flow adjustments and zero checks were performed.

Page 5, lines 136:

"The site is located in a clearing of the rainforest." According to Martin et al., 2016 the site is located in a pasture area (2.5 x 2 km) outside the rainforest. I suggest to rephrase the text accordingly from 'outside canopy' to 'outside forest' or 'pasture site'.

Reply: We thank the referee for the suggestion. The text has been rephrased accordingly, ***line 177-180****:*

'The site is an open pasture site, where the Manaus pollution plume regularly intersects and the rainforest canopy did not hinder mixing. Due to the site location, T3 is either a pristine environment or highly influenced by the Manaus pollution plume, mainly depending on the wind direction.'

Page 6, lines 180:

A description of the applied inlet system for the PSM would be interesting for future studies under high rh conditions.

Reply: We agree with the referee. A description has been added to the revised manuscript, ***line 236-243:***

The inlet system consists of a core sampling probe combined with a sintered tube. The core sampling probe consists of two cylindrical tubes with different outer diameters (10 mm and 6 mm). The larger diameter of the outer tube allows up to 10 Lpm total laminar flowrate, to minimize diffusional losses. The inner tube is directly attached to the PSM with an airflow of 2.5 Lpm. The excess airflow is discarded into an exhaust line (Kangasluoma et al, 2016). Downstream of the core sampling line is a sintered tube where dry pressurized air is introduced. The water molecules in the sample flow are pushed towards the outer walls of the sinter material by diffusion, drying the airflow.

Page 6, lines 183:

"Laboratory studies have shown that the RH affects the counting efficiency of the PSM drastically" - Please provide references.

Reply: the sentence has been rephrased as follows:

Line 244-246: Laboratory studies have shown that the RH affects the counting efficiency of the PSM drastically (higher sensitivity at smaller sizes at higher RH; Kangasluoma et al. 2013, Iida et al, 2009).

Page 7, lines 203:

"The DMPS data reported here is qualitative but not quantitative." - Please specify if

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there were problems with this instrument. Quantitative SMPS data are discussed in e.g., section 3.2.

Reply: the issue is addressed more precisely in the revised manuscript. Since the particle losses in the sampling line due to diffusion are not precisely known, the SMPS data has not been corrected for those losses. Hence, for the data shown in Figure 7, where the concentrations of 6-10 nm and 10-20 nm are shown, we feel comfortable only at making assumptions based on the trend of the data but not absolute numbers.

We added the following sentence to the revised manuscript:

Page 7, lines 264-267: 'The DMPS data reported here are qualitative, not quantitative, as the losses due to diffusion in the sampling line are not precisely known and therefore not taken into account in the data presented later in this manuscript.'

Page 7, lines 220:

The planetary boundary layer development is probably different for pasture and rainforest sites. Can you please comment on that?

Reply: The local features and land-use affect the development of convective boundary layer as well as their emission spectra in terms of volatile organic compounds are different. In the morning, the boundary layer develops more rapidly in the pasture area due to lower evapotranspiration and the sensitive heat flux is dominating. This induces a more rapidly growing mixed layer, causing more efficient vertical mixing of precursors and aerosols. Also, photochemistry is more pronounced in (semi) open area than under the canopy. However, during the daytime the small-scale variability in boundary layer dynamics and in VOC concentrations tends to even out. The rapid oxidation chemistry remains characteristic for each site.

We added a sentence in the revised manuscript. ***Page 8, line 293-297****: '*The boundary layer development is also different at the two different measurement sites. It develops more rapidly in the pasture area, causing a more efficient vertical mixing compared to the site enclosed by rainforest. From our observations, we conclude that the main differences in the dynamics of the aerosol particle population at the two measurement sites is due to the 'umbrella effect' of the rainforest canopy.

Page 8, lines 234:

"We observed an unexplained increase in the concentrations of the cluster ions in the NAIS towards the end of October 2013 to January" - Can you please comment on possible reasons for that drift? Is it possible that this drift continued after moving to T3?

Reply: we carefully looked at the flow rates and other NAIS technical data that could give some input, but we could not find any clear indicator of an instrumental drift. The drift continued after moving to T3, which is why we corrected all the data after we observed the drift for the first time accordingly. We attribute the drift is caused by a slow change in the differential mobility analyzer flow rates and charger ion filtration that cause erroneously some of the corona charger generated ions to penetrate into the detectors.

We explain this with the following sentences on ***page 8, lines 306-323*** *in the revised manuscript:*

'We observed an unexplained increase in the concentrations of the cluster ions in the NAIS towards the end of October 2013 to January 2014 at the T0t site. This increased level continued when the NAIS was taken to the T3 site. We consider this drift instrumental. By comparing the 2014 concentrations of the NAIS channels to those prior to the increase (January 2012 and 2013), a correction factor of 1.8 was applied to the 4 smallest size channels of the NAIS (0.8-1.25 nm) to account for the drift for the subsequent data.'

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Page 9, lines 276:

"the biomass burning during the dry season is expected to increase large ion concentrations"
- Please provide a reference

Reply: we rephrased the sentence as follows:

***Page 9, lines 361-365**:*

'Additionally, the wet and dry seasonality characteristic for the Amazon (Rissler et al. 2006, Martin et al. 2010a) can be observed in the concentration of the large ions (4-20nm): the biomass burning during the dry season seems to increase large ion concentrations, whereas during the wet season their concentrations decreased, most likely due to wet deposition and reduced source strengths.

Page 10, lines 287:

"Figure 2 shows the seasonal variability of ions and particles in the three size ranges (0.8-2nm, 2-4 nm and 4-20 nm)" - the lowermost panel in Fig. 2 is missing.

Reply: This is a mistake. During the writing process, we decided not to show the 4-20 nm size range as it does not add any additional valuable information. The sentence was changed in the revised manuscript as follows:

Page 10, lines 373-374:

'Figure 2 shows the monthly variability of ions and particles in two size ranges (0.8-2nm, 2-4 nm) for the 2011-2014 period.'

Page 10, lines 305:

In this paragraph it is not clear to which figure or table the authors refer to. Some examples:
"Positive and negative cluster ion concentrations were, on average, higher during the wet season compared to the dry season."
"Additionally, cluster ions (0.8-2 nm) showed slightly higher concentrations in the morning and evening, compared to other times of the day"
"A dip in the median ion concentration after midday coincides with a higher median concentration of large ions, which is a sign of a larger sink for cluster ions."
"Lastly, 4-20 nm ions peaked at around midday during the wet season, while their diel pattern was more irregular during the dry season."

Reply: this paragraph has been deleted from the revised manuscript. The numbers refer to a Figure that has been removed from the final manuscript, as we decided to only show the particle concentrations, as the data shows a very similar behavior as the ion data. The ion data does not add any additional information to the manuscript.

Page 11, lines 343:

"The median total particle concentrations were about a factor of two higher during dry season (about 1500 cm-3) compared with the wet season (about 700 cm-3)." - In table 1 different values are shown. Furthermore, large particle (4-20 nm) concentrations are very similar to CPC measurements (> 10 nm), implying that on average all particles are in the size range between 10 and 20 nm.
Also, the average particle concentrations (4-20 nm) at T0t (250-800, for the wet season) compares well to total particle concentrations (e.g., in 10-500 nm size range) reported in earlier studies (e.g., Martin et al., 2010a, Martin et al., 2010b, Zhou et al., 2002).
This again implies that the size distribution is dominated by nucleation mode particles, which is in contrast to the same mentioned references.

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*Reply: The numbers reported in the text are a mistake. The sentence has been re-phrased as follows (**page 11, lines 426-428** in the revised manuscript)*: The median total particle concentrations were about a factor of 1.5 higher during wet season (about 1000 cm^{-3}) compared with the dry season (about 700 cm^{-3}).

The presented manuscript is (to our knowledge) the first comprehensive study of small ions and particles in the Amazon basin. We agree with the referee that from looking at those numbers, we could conclude that the aerosol particle population in the Amazon is dominated by the nucleation mode.

Nevertheless, we should be careful since previous studies have not been focusing on nucleation mode particles. All the numbers presented in the current manuscript for the T0t site are directly from the measurements with the NAIS. Whereas the previous results have been using different instrumentation and the measurement locations have been different. We think that from our current knowledge we cannot conclude that the aerosol particle population in the Amazon is dominated by the nucleation mode.

Page 12, lines 361:

"The rain events were more common during the wet season (Fig. 5) when also the median rain intensity was higher." According to Fig 5, the median rain intensity is highest in August.

*Reply: The sentence has been rephrased as follows: **p. 12, lines 482-483** in the revised manuscript:* 'The rain events were more common during the wet season, peaking in August which can be considered as transition season (Fig. 5; Martin et al, 2010) when also the median rain intensity was higher.'

Page 11, lines 377 and following:

In section 3.2 the authors describe a very interesting and scientifically significant phenomenom of increased particle and ion concentrations during rain. Concentrations increase by 2 orders of magnitute towards more than 10000 particles/ions per cubic centimeter. In the following discussion, the authors mention that the particle concentration (nucleation mode size) above canopy (SMPS) does not increase accordingly.

Instead, particle concentration increases only by 20 particles per cubic centimeter (6-20 nm size range), strongly contrasting the conditions below. They conclude that the high particle/ion concentration is a below canopy phenomenom. Furthermore, these nucleation mode particles are not able to leave the canopy which is acting as an umbrella preventing mixing.

In contrast, the presented diurnal variation suggests that mixing and planetary boundary layer development is efficient (although less efficient as compared to the pasture site). Also, the authors argue that they are able to measure ions and particles related to transported biomass burning plumes (page 9, lines 275). Why are those particles able to be mixed into the canopy. It is hard to believe that the forest canopy can maintain such a strong gradient of particle number concentration.

Please justify your statement.

Reply: Earlier studies have shown that rain and particularly shattering of water droplets will result in high concentration of ions (e.g. Tammet et al., 2009). Typically, these effects are not seen with aerosol instruments as the ions are neutralized in the measurement process. Our main point here is that this increase in ion concentrations is mainly an effect that can be observed inside the canopy as the ions that we observe are produced by splashing of the water droplets on the tree leaves. Those ions will not survive until the measurements by the DMPS as it is sampling from above the

canopy and they ions are filtered out by the leaves before reaching the inlet of the DMPS. From the current measurements, we cannot make any statement of the source of the larger neutral particles that are seen by the DMPS above the canopy. It is likely that they are produced in cloud outflow regions and due to strong downdrafts entrained back into the mixing layer (Wang et al, 2016). Most likely the increase of 4-20nm ions during the dry season is a combination of local biomass burning sources and a decrease in wet deposition.
The sentence has been re-phrased in the revised manuscript:
***Page 9, lines 361-365**:*
'Additionally, the wet and dry seasonality characteristic for the Amazon (Rissler et al. 2006, Martin et al. 2010a) can be observed in the concentration of the large ions (4-20nm): the biomass burning during the dry season seems to increase large ion concentrations, whereas during the wet season their concentrations decreased, most likely due to wet deposition and reduced source strengths.

Page 15, lines 45

Please consider to show the results of your backward trajectory analysis in a map.

Reply: We thank the referee for the suggestion, as it improves the manuscript. We have added a Figure showing the map to clarify the back trajectory calculations

We also rephrased the sentence as follows, ***line 594-598: '***These air masses all originate from upstream of the Amazon river, where the NPF day air mass originate from further north, which is an area with dense rainforest. The results of the back trajectory calculations are shown in Figure 10. The red line shows the median of an ensemble or the non event days and the blue line for NPF days.

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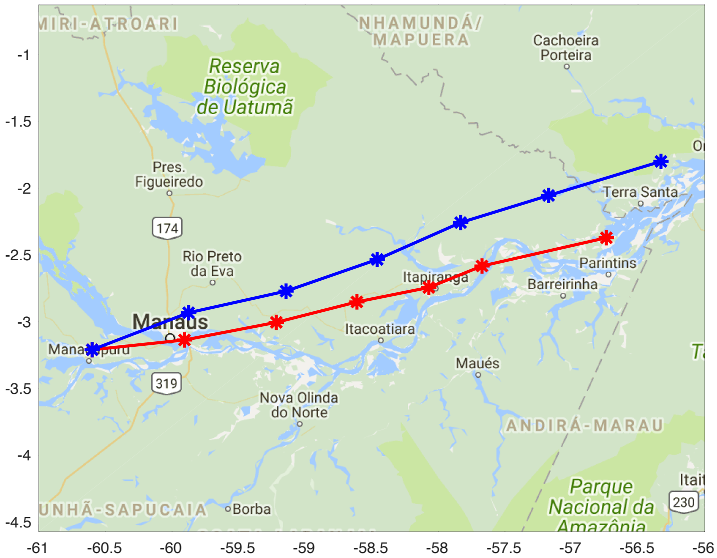

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Figure 10: median back trajectories for NPF (blue) and non NPF (red) days. The trajectories were calculated 24hours backwards arriving at 09:00 local time at 500m a.s.l. at the measurement site.

Page 15, lines 459:
In Fig. 10 a new particle formation event is shown: Please consider to add SMPS contour plots and SMPS particle number concentrations in the nucleation mode size range. Statistical information of SMPS nucleation mode particle number concentration will add further valuable information to Figure 9 and Tables 1 and 4.
The absence of the forest canopy at T3 gives the opportunity to combine NAIS and SMPS measurements, which allows to investigate the entire evolution of the submicron aerosol population.
Reply: we thank the referee for this suggestion. The SMPS Figure has been added to the Figure.

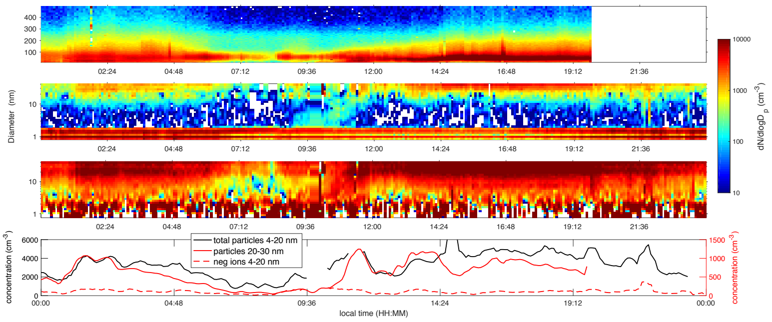

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Figure 11 One example NPF day as observed at the outside canopy (T3) site. (a) shows the surface Figure from the SMPS, (b) and (c) show the surface Figures from the NAIS, (b) for negative ions, (c) for total particles. The color code indicates the measured concentrations. Panel (d) shows concentrations for the 20-30nm size range from the SMPS (black line) and from the NAIS the negative ion (dashed red line) and total particle concentrations (solid red line) in the 4-20 nm size range.

Page 17, lines 510:
"Similar, but weaker, rain-events were found at the site outside the rainforest canopy (T3)." - weaker in terms of what?
Reply: We have re-phrased the sentence as follows in the revised manuscript:
Page 17, lines 679-683: 'Similar rain-events were found at the pasture site (T3). The production of small (0.8-2 nm) and intermediate ions (2-4 nm) during rain events reached a maximum of 10^4 cm^{-3} at the pasture site, where it was one order of magnitude higher at the T0t site. Large ion concentrations reached similar concentrations during rain events at both measurement sites.'

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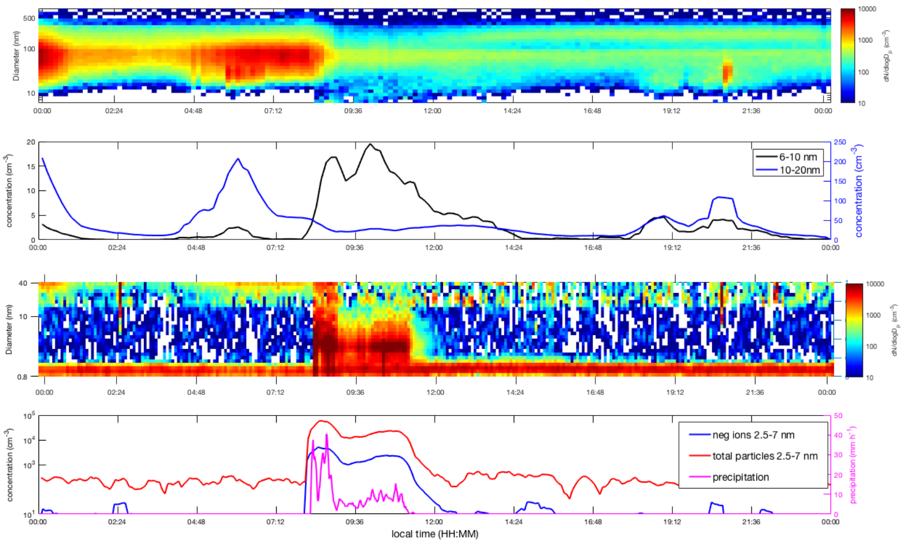
Technical comments related to Figures
The boxes refer to the 25th-75th percentile.
Reply: The whiskers show the extreme values of the data set which are not considered outliers.

The tables and Figures have been changed according to the suggestions of the referee in the revised manuscript.

Fig 4:
number concentration of small positive and negative ions disagrees by a factor of 2. According to Manninen et al., 2016 there should be an agreement within 20%. Please comment on the instrument performance and data quality.

Reply: Table 1 shows a very good agreement between the positive and negative ion concentrations. We believe that the difference seen in Figure 4 is due to a problem with the instrument performance, which might be different on certain days, but which does not affect the overall good instrument performance and data quality.



updated Figure 7

Figure 7. Example for a rain-induced event for total particles (DMPS). The DMPS measurements are taken above the canopy (60 m height), NAIS measurements are inside the canopy. Panel (a) shows the DMPS surface Figure. Panel (b) shows the particles measured by the DMPS for 6-10 nm (black line, left hand axis) and 10-20 nm (blue line, right hand axis). Panel (c) shows the surface Figure for the negative ions, measured by the NAIS. Panel (d) shows the negative ion concentrations for 2.5-7

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nm in blue and the total particle concentration in the same size range from the NAIS in red with the scale on the left axis. The pink trace shows the precipitation in mm h^{-1} on the right axis.

References

Iida, K., Stolzenburg, M. R., and McMurry, P. H.: Effect of Working Fluid on Sub-2 nm Particle Detection with a Laminar Flow Ultrafine Condensation Particle Counter, Aerosol Sci. Tech., 43, 81–96, 2009.

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Wang J., Krejci R., Giangrande S., Kuang, C., Barbosa H. M. J., Brito J., Carbone S., Chi X., ComstockJ., Ditas F., Lavric J., Manninen H. E., Mei F., Moran-Zuloaga D., Pöhlker C., Pöhlker M. L., Saturno J., Schmid B., Souza R. A. F., Springston S. R., Tomlinson J. M., Toto T., Walter D., Wimmer D., Smith J. N., Kulmala M., Machado L.A. T., Artaxo P., Andreae M. O., Petäjä T. & Martin S. T., Amazon boundary layer aerosol concentration sustained by vertical transport during rainfall, *Nature*, doi:10.1038/nature19819, 2016.

Referee comment: 1) Title: “Direct” seems unnecessary. Perhaps better replaced by “Ground-based”? Also, “molecular clusters” seems inappropriate. Perhaps “particle ions”?

Reply: we thank the referee for the valuable suggestion for the title. We agree that ‘’Ground-based’’ suits the content of the paper better than ‘’direct’’. We would like to keep ‘’molecular clusters though, since with the instrumentation used in our work, we are able to measure down to the molecular cluster level.

Referee comment 2) Line 38 – “Pristine” is used here and in a few other places. It needs to be defined.

Reply: we agree with the reviewer that an explanation of pristine in our manuscript is missing. We added the following sentences in the revised manuscript:

Line 38 and following; ‘The occurrence of NPF on ground level in the Amazon region has not been observed previously in pristine conditions. In this work, pristine refers to CCN concentrations of a few hundred cm^{-3}.’

Referee comment 3) Lines 40-42 - Define the sites as locations relative to Manaus, much as you did on lines 80-83. You can’t expect all readers to identify with T0t and T3.

Reply: We agree with the referee that a description of the measurement locations is missing at this point of the manuscript.

*We changed the sentence in the revised manuscript as follows, **line 40-45**:* We measured the variability of air ion concentrations (0.8–20 nm) with an ion spectrometer between 2011 and 2014 at the T0t site and between February and October 2014 at the GoAmazon 2014/5 T3 site. The T0t site is surrounded by dense rainforest, mostly unaffected by the Manaus pollution plume. The T3 site, instead is an open pasture site, 70km downwind of Manaus.

Referee comment 4) Lines 43-44 – “T0t is reached by the pollution about 1 day in 7, where the T3 site is about 15% of the time affected by Manaus.” The statement implies a difference between T0t and T3, but 1 in 7 is 14%, which is not different from 15%. What are you trying to say here?

*Reply: We agree with the referee that here the numbers are similar and the sentence is confusing. We were trying to point out the differences between the two measurement sites. The T0t site is parallel wind to the Manaus pollution plume, where the T3 site is downwind of the Manaus pollution plume. We discussed the numbers again. Based on AMAZE-08, we concluded T0t is affected about once a week (Martin et al. 2010 (in Supp Material) T3 gets influenced between once every day and once every two days for a few hours, especially in the afternoon (de Sa et al. 2017, Thalman et al. 2017). We rephrased the sentence to, **line 47-50**: ‘*T0t is influenced by pollution about once per week, where T3 on the other hand is reached once per day/once per every second day, especially in the afternoon (Martin et al., 2010b supplementary material, Thalmann et al, 2017, de Sa et al, 2017).’

Referee comment 5) Lines 59-60 – This sentence is not useful. Also, you state in the paper that the back trajectories in both cases pass over Manaus. Does not the source strength of Manaus even out other differences in the trajectories? Your last sentence of the conclusions is that “Most likely, during the dry season the condensation sink is too high for new particle formation.” That appears to be the main factor that differentiates between the NPF and non-NPF days. Why is that not mentioned in the abstract?

Reply: We agree with the referee that the difference in the condensation sink between NPF and non-NPF is a major finding in our manuscript and should be in the abstract. We also think that the difference in the back trajectories is a relevant difference; therefore, we want to keep the statement in the abstract.

*We changed the last sentence of the abstract in the revised manuscript**, line 65-68**:*

The two major differences between NPF days and non event days are two. A factor of 2 lower condensation sink on NPF days and different air mass origins for the NPF days compared to non event days.

We followed the suggestion by Referee 1 and included a new Figure (Fig. 10) in the revised manuscript, which shows the calculated back trajectories on a map. The map shows actually that the trajectories on do not pass over Manaus on NPF days.

*The sentence '*Nevertheless, all air masses pass over Manaus before reaching the measurement site.' *has been deleted from the revised manuscript. The first set of back trajectory calculations was made without looking at the map, which lead to the wrong conclusion.*

New Figure 10

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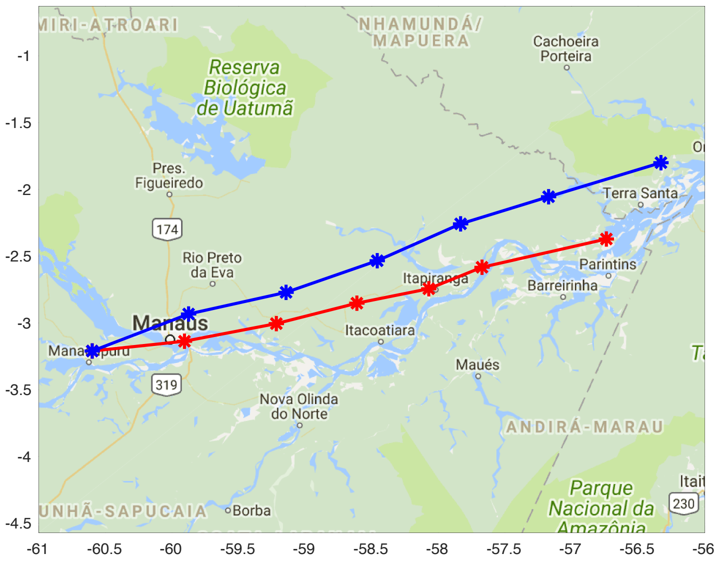

Figure 10: median back trajectories for NPF (blue) and non NPF (red) days. The trajectories were calculated 24hours backwards arriving at 09:00 local time at 500m a.s.l. at the measurement site.

Referee comment 6) Lines 221-222 – You say "The vertical mixing can be enhanced during the wet season due to convective clouds." Are you saying that convective clouds lift the mixed layer or that convective clouds lift particles out of the mixed layer or something else? Clouds formed at the top of a mixed layer will tend to cool below, which does not help the development of a mixed layer.

Reply: We agree with the referee that this sentence slightly confusing. We were trying to say that convective clouds lift particles out of the mixed layer.
We re-phrased the sentence in the revised manuscript, ***line 289-290:*** 'The vertical mixing can be enhanced during the wet season as particles are lifted out of the mixed layer due to convective clouds.'

Referee comment 7) Lines 275-279 – This may be true for inside the canopy, but not for outside the canopy. Please clarify. Also, why would the pattern outside of the canopy not reflect biomass burning and wet deposition more than that inside the canopy?
Reply: We do believe that the statement is true for both outside and inside the canopy. The chapter title in this paragraph. 'Number concentrations of ions and particles at

*the two sites' includes both measurement sites. It seems like this statement is not fully clear, so we rephrased the sentence in the revised manuscript as follows, **line 361-365:*** 'Additionally, the wet and dry seasonality characteristic for the Amazon (Rissler et al. 2006, Martin et al. 2010a) can be observed in the concentration of the large ions (4-20nm): the local biomass burning during the dry season seems to increase large ion concentrations, whereas during the wet season their concentrations are decreased most likely due to wet deposition and reduced source strengths.'

Referee comment 8) Lines 281-282 - That appears to be true for the wet season, but the factor is less than 2 during the dry season. Did you mean "up to a factor of 3"?

Reply: we agree with the referee that the statement should be **'**The average concentrations of 4 – 20 nm particles were up to a factor of 3 higher in comparison to the less polluted site (T0t).**'**

Referee comment 9) Lines 287-288 – The 4-20 nm ions are not shown in the Figure 2 I have.

*Reply: We apologize for the mistake in the manuscript. The Figure has been changed during the writing process of the manuscript. The sentence has been re-phrased as follows in the revised manuscript, **line 367-368**:* **'**Figure 2 shows the monthly variability of particles in two size ranges (0.8-2nm, 2-4 nm) for the 2011-2014 period. The cluster ions had a median concentration of 814 cm^{-3} and 968 cm^{-3} (wet) and 605 cm^{-3} and 765cm^{-3} (dry) for negative and positive ions, respectively.'

Referee comment 10) Line 301 – "Oct-Dec for both seasons"? Oct-Dec is a season (fall). Specify wet and dry seasons.

*Reply: we agree with the referee that this sentence is confusing. We removed the sentence completely from the revised manuscript. We agree with the referee that the seasons should be specified in our manuscript. We added a sentence in the Methods section, **line 139-142**:* **'**Wet and dry season in the Amazon are Dec-March and June-September respectively (Martin et al, 2010a). Due to the measurement periods available for our dataset, we define the dry season as dry and transition season Apr-Oct.'

Referee comment 11) Line 305 – On line 214 the dry and transition season is April to September, whereas here it is Apr-Oct. Please correct.

Reply: this sentence was removed in the revised manuscript. The whole paragraph was changed following the suggestions of Referee 1.

***Line 401-405**:* 'These values are comparable, for example, to intermediate and large ion concentrations found in coastal Mace Head (Vana et al. 2008) outside the periods of rain or active NPF. In general, the positive cluster ion concentrations are higher in all the cluster ion and intermediate ion size classes for all the months. Table 2 summarizes the annual concentrations of ions and total particles for the three size bins.'

Referee comment: 12) Line 311-312 – Cluster ions are not shown in Figure 3. Where are we supposed to view this?

*Reply: this has been removed in the revised manuscript, as we do not present the Figure in the final manuscript. The paragraph in the revised manuscript was re-phrased as follows, **line 406-409**: '*Differences between the wet (Dec-Mar) and dry and transition season (Apr-Oct) were also observed in the diel cycle of the ion and particle concentration. Positive and negative cluster ion concentrations were, on average, higher during the wet season compared to the dry season as shown in Table 1.'

Referee comment 13) Lines 321-323 – Again, 4-20 nm ions are not shown in Figure 2

Reply: this has changed in the revised manuscript. The paragraph was re-phrased as stated in the reply to the previous Referee comment 12.

Referee comment 14) Line 358 – What do you mean when you say that "negative ions are smaller than positive ions"? Do you mean fewer in number?

*Reply: We mean that the negative ions are smaller in size compared to positive ions. The sentence has been re-phrased in the revised manuscript as follows, **line 478-484**:* 'Rain-induced bursts are likely a result of a balloelectric effect, in which splashing water produces intermediate ions such that the negative ions are smaller in size than the positive ions (Horrak et al., 2005, Hirsikko et al., 2007, Tammet et al., 2009). The duration of the 579 rain events varied from a couple of minutes to 22 hours, with over half the rain events lasting for two hours or less. The rain events were more common during the wet season, peaking in August which can be considered as transition season (Fig. 5) when also the median rain intensity was higher.'

Referee comment: 15) Lines 374-376 and figure 6 – For the ions in the 0.8-2 nm particles, it looks like they simply turn on at rain intensities above 1.

Reply: We made Figure 6 in order to show the relation between rain intensity and ion concentrations. At rain intensities below 1 mm/h the ion concentration especially in the cluster ion size range only contains the natural in background as they are produced via radon decay or galactic cosmic rays. The background cluster ion band can be observed worldwide, yet the concentrations depend on the location as it depends on the sources and sinks for the ions.

Referee comment 16) Figure 7 – Indicate which axis corresponds with which particle size class in Panel B; presumably, the LH axis is 6-10 nm.

Reply: we agree with the referee that the Figure is confusing. The Figure was changed to improve the clarity in the revised manuscript.

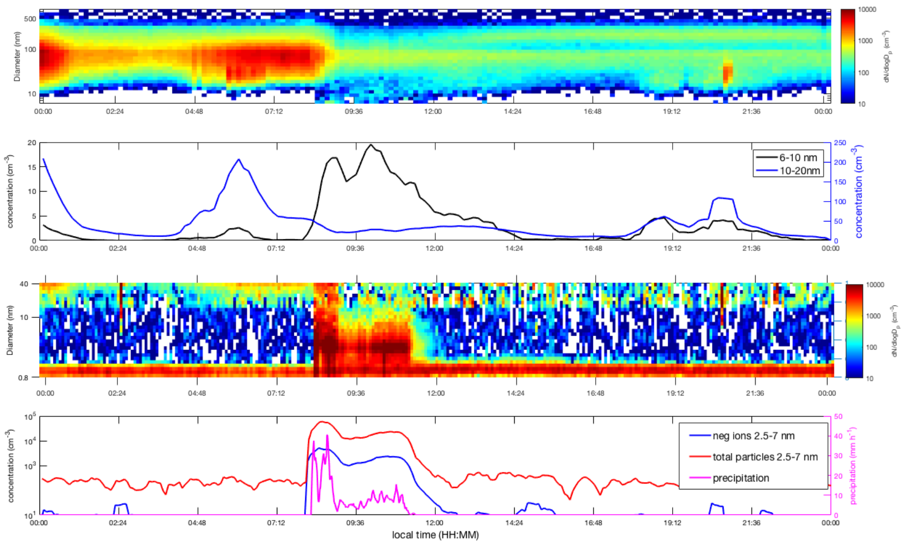

Figure 7. Example for a rain-induced event for total particles (DMPS). The DMPS measurements are taken above the canopy (60 m height), NAIS measurements are inside the canopy. Panel (a) shows the DMPS surface Figure. Panel (b) shows the particles measured by the DMPS for 6-10 nm (black line, left hand axis) and 10-20 nm (blue line, right hand axis). Panel (c) shows the surface Figure for the negative ions, measured by the NAIS. Panel (d) shows the negative ion concentrations for 2.5-7 nm in blue and the total particle concentration in the same size range from the NAIS in red with the scale on the left axis. The pink trace shows the precipitation in mm h^{-1} on the right axis.

Referee comment 17) Line 379 – "followed by a second one at about 11:00". Here, indicate the relative difference in rain intensity.

Reply: The second rain intensity peak was lower than the first one in this example. The first one was about 40 mm/h and the second one about 10 mm/h. So, the difference was about 30 mm/h. We added the relative difference in the revised manuscript, ***line 502-503****.* 'Rain events were evident also when looking at the total particle concentrations measured by the NAIS, as depicted in Figure 7. The first rain event showed a maximum of about 40 mm h^{-1} and the second one about 10 mm h^{-1}.'

Referee comment 18) Figure 7 and lines 385-395 – This is a very interesting set of observations. If particles descending with the rain were responsible for the increase in 6-10 nm particles above the canopy, how do you explain the apparent evolution of 6-10 nm particles to 10-20 nm over a few hours? Given the roughly 3 orders of magnitude difference in particle number concentrations from ground to above canopy and the potential canopy filtering you mention, why instead is it not possible that the few 6-20 nm particles above the canopy were due to the rain-induced particles mixing and filtering upwards?

Reply: There are two main points about Figure 7. The first one should show a clear correlation between the rain intensity and increase in ion concentrations inside the canopy. This effect is due to the splashing of the water droplets on the leaves mainly of the trees. The water droplets explode and release high amounts of small ions. This phenomenon has been observed and explained by Tammet et al, 2009. The second effect is that these ions seemingly do not contribute to the total particle population as the DMPS which is measuring from above the canopy does not show an according increase in particle number concentration. We assume that this effect is due to the filtering effect by the rainforest canopy. We cannot say very much about the source of the particle that are seen by the DMPS, but from our current knowledge it is likely that they are transported via downdraft from production at convective cloud outflow regions (Wang et al, 2016).

*We rephrased the paragraph in the revised manuscript, **line 509-518**:* The 10-20 nm particle concentration showed first a decrease followed by a slight increase up to ~35 cm^{-3}, peaking later than the 6-10 nm particles. However, it is unlikely that these 10- 20 nm particles originate from the same rain-induced burst as seen inside the canopy, as there is no apparent particle growth from the NAIS measurements. It is unlikely that those particles survive until the top of the canopy, as the tree leaves would filter them out. Wang et al. (2016) reported that nucleation mode particles produced in cloud outflows will be transported down with the rain, such that they can be observed at the ground level as an increase in nucleation and Aitken mode concentrations (Dp <50 nm). The appearance of 6-10 nm particles with its peak concentration, could present a similar scenario of small particles brought down from the free troposphere.'

Referee comment 19) Table 3 and lines 404-406 - Table 3 shows 65 and 49 for a total of 114, while you state 64 and 46 and 113. Please correct.

Reply: the numbers have been corrected in the table.

Updated Table 3:

	NPF days	Undefined	Non-events	Rain events	No-rain events
Wet season (Jan-Mar)	8/64 (12.5%)	0/64 (0%)	57/64 (89%)	61/64 (95%)	04/64 (6%)
Dry season (Aug-Oct)	0/46 (0%)	0/46 (0%)	46/46 (100%)	15/46 (32.6%)	34/46 (74%)

Referee comment 20) Figure 9 – On either side, you show four panels. The top two are labelled ions and the bottom two are labelled total particles, which is consistent with the text. In the caption, we are led to believe that the top three are ions. Please correct.

Reply: we agree the Figure and description was unclear. The Figure and caption were updated in the revised manuscript.

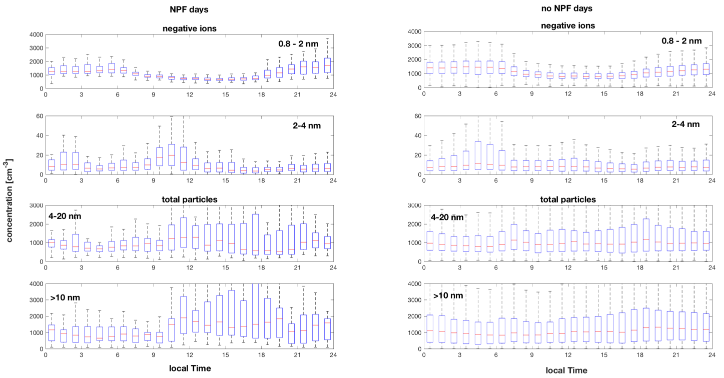

Figure 9. Diel cycle of ions measured outside the canopy by the NAIS (small: 0.8–2 nm; intermediate: 2–4 nm; The lowest two panels show the total particles (large: 4–20 nm) from the NAIS and total particles >10 nm as measured by the MAOS CPC. The left column shows the NPF event days and the right column the non NPF days. The markers are hourly median number concentrations and the whiskers 25th and 75th percentiles.

Referee comment 21) The RH side of Table 5 is cut off in my copy.

Reply: we are sorry about that. Table 4 is better readable in the revised manuscript.

Updated Table 4

Particle and ion concentrations- 09:00 – 12:00 LT		
	NPF day	**Non NPF day**
Cluster ions (0.8-2 nm) [cm^{-3}]	800 (-) (692-905)	870 (-) (687-1000)
Intermediate ions (2-4 nm) [cm^{-3}]	13 (-) (6-23)	8 (-) (4-15)

Large ions (4-20 nm) [cm^{-3}]	83 (-) (44-137)	62 (-) (25-119)
Intermediate particles (2-4 nm) [cm^{-3}]	606 (303-969)	547 (522-1600)
Large particles (4-20 nm) [cm^{-3}]	1000 (604-1600)	970 (238-1000)
Full day data		
SMPS Condensation sink [s^{-1}]	1.6e-3 (8.4e-4-2.6e-3)	3.3e-3 (1.7e-3-5.5e-3)
CPC total particles (>10 nm) [cm^{-3}]	1100 (579-1860)	1000 (404-2000)
Environmental parameters-full day		
	NPF day	**Non NPF day**
Temp [°C]	25.6 (23.8 – 28.9)	26 (24.5 – 29.3)
RH [%]	94.2 (78.8 – 98.1)	93.5 (78.9 – 97.6)
Precipitation rate [mm hr^{-1}]	0 (0 - 0)	0 (0 – 0.16)
Total average precipitation [mm day^{-1}]	6.9 (5.8-8.2)	5.6 (0.9-15.3)
Wind direction [°; relative to north]	83 (56.95 – 120.8)	105.5 (38.8 – 217)
Wind speed [m s^{-1}]	1.85 (0.96 – 3.04)	1.2 (0.6 – 2.3)

Referee comment 22) Line 520 – Should be Jan-March for wet season?
Reply: We agree with the referee. The statement has been corrected in the revised manuscript, ***line 691-692***. 'We observed eight NPF events showing particle growth at site T3 outside the canopy during Jan-March 2014, which is during the wet season.'

Referee comment 23) A couple of more general comments: Is there some sort of summary connecting the ion concentrations with NPF that can be drawn? The rain-induced events are prominent, but we are not given any sense of how important these might be. For example, is there any evidence that a significant number of rain-induced particles survive to become CCN size, or is Figure 7 the best example of their potential longevity?

Reply: Our analysis has clearly shown that the connection between rain and NPF events is not clear. There was no rain observed during any of the NPF events, yet sometimes there was rain in the evening after or shortly before the start of an NPF event. That indicates that the rain clears the air of pre-existing particles and therefore the conditions for NPF events to happen are favorable.
To our current understanding, the increase in ion concentrations due to rain events that were mainly observed inside the canopy do not significantly contribute to the production of bigger neutral particles as there is no concomitant increase in neutral particle concentrations as measured by the DMPS, which is sampling from above the canopy (see Figure 7). We believe that the ion production due to rain is mainly an inside canopy effect and the ions are filtered out by the canopy and therefore do not survive until they would be able to reach bigger sizes. We did observe some rain events that lasted up to 20 hours but still we did not observe any increase in neutral particle concentrations above the canopy.

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