General comments from Referee 1
I thank the authors for addressing my concerns. Nevertheless, there are still open questions, incorrect statements, internal inconsistencies and technical mistakes.

The high amount of mistakes in form and content as well as the incorrect citations leave me with serious concerns about the data quality, the analysis and manuscript preparation. The updated manuscript is not improved significantly and does not meet the standards of ACP.

A thorough and careful major revision of the data analysis and the entire manuscript is necessary before considering publication.

General author comments
We thank the referee for the careful revision and concerns about the manuscript. We have carefully revised the manuscript and re-analyzed the dataset we present.

Changes in the current data analysis:

- All the precipitation days have been excluded in the revised manuscript for the results shown in Tables 2 and 3 and Figures 2 and 3.

- The NAIS data have been carefully quality-checked. The NAIS data, especially in the neutral mode, are unreliable due to the multiple charging effect at sizes above 20 nm (e.g. Manninen et al., 2016). In our presented dataset, we observed the most intense noise levels at sizes above 15 nm, so we decided to restrict our data analysis up to 12 nm in the revised manuscript.

- The T3 meteorological data are also derived from a Vaisala system provided by ARM, to make the data more comparable from both sites.

- The definition of the wet and dry season follows now Artaxo et al. (2013) who defined the wet season in the Amazon from January to June and the dry season from July to December. This definition has been applied throughout the whole analysis in the revised manuscript.

- We removed pristine from the revised manuscript, as our focus for the dataset presented in our manuscript is not on a pristine environment. Nucleation mode particles have been observed in the Amazon region in the vicinity of Sao Paulo (Backman et al., 2012). The dataset presented shows aerosol and ion characteristics at measurement sites 40 km and 70 km away from any major anthropogenic pollution sources.

- To address the concerns about the data availability and statistics, we included Table 1, summarizing the NAIS data availability and rain data availability for the whole measurement period.

In order to make it easier to follow the discussion, we address the concerns of the referee that are still open questions from the previous version of the manuscript.

Regarding the comments by the referee concerning the Figures and Tables, we have re-analyzed the whole dataset as described above. Almost all Figures and Tables have changed in the revised manuscript and all the mistakes in the Figures and Figure captions have been thoroughly taken care of. We thank the referee for pointing these mistakes out.

Referee comment
Rephrasing the original sentences does not solve the citation issues. If you rephrase a sentence from a different source, you still have to cite the original source. I am concerned the authors do not take reasonable care to check their citations.
Make sure all the identical sentences and text passages mentioned in my first comments are now correctly referenced - even if rephrased.

Author comment
We take the concern about the referencing and similarities to previous publications very seriously and carefully re-formulated the revised manuscript, including cross-checking all our references.

Referee comment
You mention the term pristine already in the abstract. Your answer 'The term pristine has been removed from the revised manuscript' is incorrect and misleading. Be precise what parts of the manuscript are changed.
As referee 2 already pointed out, you have to clearly define 'pristine'.

Author comment
We removed all the phrases that included pristine in the revised manuscript. This study does not focus on pristine conditions. The comparison of the seasonal characteristics of ions and neutral particles in the Amazonian atmosphere is the emphasis of the presented manuscript.

The effect of the rainforest canopy on the characteristics of ion and neutral particle size distributions in the sub-3 nm sizes have not been studied before. We put our results into this context and highlight the effect of the rainforest canopy on these quantities in our manuscript.

Referee comment
The authors argue that '.. pollution events would appear in the NAIS/SMPS data as elevated aerosol concentrations in the accumulation mode.'.
According to Kuhn et al., 2010, the Manaus pollution plume consists to a significant degree of fuel combustion. Kuhn et al., 2010 found CN concentrations up to several 10000 particles per cubic centimeter - the majority of these particles were likely smaller than 40 nm. Hence, pollution events can influence NAIS and SMPS data to highly variable degrees in a broad size range not only in the accumulation mode. The author's argumentation that filter criteria are not necessary is not convincing.
Furthermore, in your abstract you insist to present measurements under pristine conditions. You further state, that even the parallel-wind station T0t site is affected by the Manaus pollution plume about once per week. To my understanding, pristine refers to undisturbed, clean or natural conditions.
Without a proper filter to exclude pollution sources, your results can not be considered as pristine. Again, please provide a clear definition for 'pristine'.
Presenting average values does not help in this case since these average values will be affected by pollution episodes to variable degrees.¨

Author reply
We agree with the referee in that pollution can influence NAIS and SMPS data in smaller sizes as well. However, we did not filter the results in the revised manuscript for pollution, since our aim in this paper was not to investigate pristine conditions. We present our results at two measurement

sites that are both (60 and 70 km) far away from major pollution sources, which is Manaus in this case. Nevertheless, we are not measuring in pristine conditions, as both sites are to various extents influenced by the Manaus pollution plume.

Referee comment
Even a rough estimate would already help to put your integral DMPS data into context. Are these concentrations underestimated by a factor of 2 or an order of magnitude?

Author comment
My educated guess is that the DMPS diffusion losses are about 70% transmission at 6 nm and close to 100% at 10 nm for the specific inlet used in this experiment. In the AMAZE-08 experiment where the same aerosol inlet was used, 50% transmission of aerosol particles was achieved at 4 nm.

Reply to author comment:
I do not understand what the authors are trying to say with: 'However, during the daytime the small-scale variability in boundary layer dynamics and in VOC concentrations tends to even out.' Can you please provide references?

Author comment
The message here is that our analysis does not rely e.g. on eddy covariance techniques that enable identification of flux of VOCs or aerosols in the footprint of the measurements, which depends e.g. on the measurement height and atmospheric lifetime of the compound (e.g. Rinne et al. 2012, ACP). Instead we look into typical concentrations with averaging time of 1 hour. Such averaging masks the small-scale variability in the boundary layer dynamics in the vicinity of the observation site. A similar analysis is done for the VOCs in Wei et al. 2018 (Agric Forest Met).

Referee comment
I have a few questions on this. Does the drift affect positive and negative cluster ions? Furthermore, you relate the drift in cluster ions to ions generated by the corona charger. According to Manninen et al., 2016, all parts of the preconditioning unit are switched off while naturally charged ions are measured. How can then natural ions be affected?

Author comment
We agree with the referee that this drift in concentrations raises some questions. We looked at the raw NAIS data files to investigate in detail the instrumental performance. The reason for the drift was found to be due to too low sheath filter currents. Not all the ions will be filtered out of the re-circulating sheath air flow, leading to an over-estimation of the ion concentrations.
At T0t site, the sheath filter currents were too high in both polarities after October 7, 2013. The data has been corrected for a factor of 1.8 for the 4 smallest size channels in the NAIS for the time period October 7 to January 7, 2014.
The sheath filter performance was reasonable, after the NAIS was moved to the T3 site, but only in the negative channel. The positive channel is considered unreliable for the whole time period of the measurements at the T3 site and hence no data from the positive ion channel is shown in the revised manuscript. Additionally, the NAIS data from Sept 09, 2014 to Sept 26, 2014 is considered unreliable and has therefore been removed from the analysis.
We included a section describing the phenomenon in the revised manuscript as follows:

We observed an increase in the concentrations of the cluster ions in the NAIS starting from October 7, 2013 to January 21, 2014. By investigating the raw data files, this drift was observed to be due to too low currents in the sheath air filters. The sheath air filters are electrical filters, using corona

needles to neutralize all the remaining ions. The sheath air is re-circulating in the NAIS, hence an inefficient filtering leads to an over-estimation of ion concentrations. The increased ion concentration was due to too low sheath air filter currents in both polarities after October 6, 2013. A correction factor of 1.8 was applied for both polarities in the 4 smallest size channels of the NAIS (0.8-1.25 nm) for the data taken at the T0t site after that.

This increased level in the positive polarity of the natural ions continued when the NAIS was redeployed at the T3 site. The cause was the same (too low a current in the sheath air filters). The negative polarity was performing well at the T3 site. We consider the positive polarity of the natural charged ions in the NAIS at the T3 site unreliable, therefore no data from the positive channel for the T3 site is shown in this study. Additionally, the ion data from 9-26 September 2014 at the T3 site was considered unreliable and also excluded from our analysis.

Referee comment
The authors agree, that from their presented particle number concentrations from NAIS and CPC measurements one could conclude that their findings indicate a dominating nucleation mode in the Amazon aerosol particle number size distribution but at the same time they do not. This is confusing.
I would like to outline my concerns based on the results shown in Table 1:
1. At T3, the measured particle number concentrations from NAIS (4-20 nm) and CPC (> 10 nm) are on average very similar. If both instruments are comparable this means, almost all or at least a very large amount of the measured particles must be in the size range 10 - 20 nm. Hence, it follows from these measurements, that the aerosol population is dominated by nucleation mode particles. If true, this has to be supported by further size resolved measurements, since it is a stark contrast to the mentioned existing literature (see my first review, e.g., Martin et al., 2010a, Martin et al., 2010b, Zhou et al., 2002).
If one cannot draw this conclusion, NAIS and CPC measurements are not consistent. In any case, a detailed paragraph with a thorough discussion on this discrepancy (if it is one) has to be added to this manuscript.

2. At T0t this comparison (CPC, NAIS) is not possible. Nevertheless, comparing the particle number concentrations shown in this manuscript (NAIS, 4-20 nm) with total particle number concentrations in the mentioned references, one can again conclude, that the majority of particles is in the nucleation mode size range. The authors argue, that the mentioned references did not focus on new particle formation. But clearly, these authors used instrumentation sensitive to the nucleation mode size range.
The authors have to put their findings (at least for the particle concentration data) into context of existing results and have to discuss their high concentration of nucleation mode size particles.

Author comment
We agree with the referee. We carefully investigated the data quality of the NAIS measurements in our revised analysis. The NAIS is over-estimating neutral particle concentrations, mainly at sizes >20 nm due to the multiple charging effect (Manninen et al., 2016).
To address these issues, we included two new analysis methods in the revised manuscript.

First: we observed most noise in the NAIS in our data at size above 15 nm, for both neutral and ion measurements. Therefore, we decide to restrict our current analysis to the size of 12 nm for both neutral and ion measurements.

Second: For the comparison of the seasonal characteristics at both measurement sites, we excluded all the days with occurring precipitation for the median diel cycles and median numbers shown in Table 2 and 3 and Figures 2 and 3. The concentrations of NAIS neutral 4-12 nm size channel at the T3 site are about a factor of 2.5 (wet) to 4 (dry) lower compared to the CPC concentrations. At T0t the intermediate size range neutral concentrations are even lower (80-90cm^{-3}) after filtering out the precipitation days and restricting the upper size limit to 12 nm.
We can conclude from these results, that neutral particles are also produced during precipitation (as also shown in Figure 4) and that with our current analysis, the NAIS neutral particle mode is most reliable up to 12 nm.

Referee comment
The rephrased sentence is confusing. How can the rain events be more common in the wet season, when they 'peak' in the transitional season? Also, the reference is unclear. Do you reference Fig. 5 in Martin et al., 2010?

Author comment
We have also re-analyzed the rain statistics at the T0t site, as shown in Figure 5. The Figure shows the total monthly average precipitation on the right hand-axis and the monthly median number of rain days and no rain days as green and blue bars with the scale on the left-hand axis. The Figure shows a minimum in rain days from July to November and a maximum number of rain days from December to June. This confirms the definition of wet and dry season as presented in Artaxo et al. (2013), as used in the revised manuscript. Also, the total average precipitation shows a minimum between June and November.

Reply to author comment:
The rephrased sentence does not address my main criticism. My main concern is that the 2 orders of magnitude increase for the particle number concentration is not visible at all above the canopy.
I agree that the increase in ion concentration will not be detected by the DMPS, but according to you methods section, the DMPS should be able to detect particles larger than 6 nm (taking into account the CPC cutoff and the inlet losses as stated by you).
The forest canopy is certainly hindering mixing. Nevertheless, it is likely that a certain amount of these small particles (if generated by droplet splashing) is already produced at the top of the canopy. At least a fraction of these small particles does not have to go through the canopy and should therefore appear in the DMPS measurements.
I put up the comparison with the ions produced by biomass burning for another reason. You argue that those ions are able to pass the canopy, but the others are not - why?

Author comment
We agree with the referee that at least a fraction of those small particles should be produced also above the canopy. Based on our dataset, we cannot make any firm conclusion on the source of the neutral particles above as observed by the DMPS. In the revised manuscript in Figure 2 all the precipitation days are excluded. We can exclude the precipitation as a source of cluster and intermediate ion concentrations at the T0t site.

References
Artaxo, P., Rizzo, L. V., Brito, J. F., Barbosa, H. M. J., Arana, A., Sena, E. T., Cirino, G. G., Bastos, W., Martin, S. T., and Andreae, M. O.: Atmospheric aerosols in Amazonia and land use change: from natural biogenic to biomass burning conditions, Faraday Discuss., 165, 203–235, 2013.

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General author comments

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- The NAIS data have been carefully quality-checked. The NAIS data, especially in the neutral mode, is unreliable due to the multiple charging effect at sizes above 20 nm (e.g. Manninen et al., 2016). In our presented dataset, we observed the most intense noise levels at sizes above 15 nm, so we decided to restrict our data analysis to up 12 nm in the revised manuscript.

- The T3 meteorological data are also derived from a Vaisala system provided by ARM, to make the data more comparable from both sites.

- The definition of the wet and dry season follows now Artaxo et al. (2013) who defined the wet season in the Amazon from January to June and the dry season from July to December. This definition was applied throughout the whole analysis in the revised manuscript.

- We removed pristine from the revised manuscript, as our focus for the dataset presented in our manuscript is not on a pristine environment. Nucleation mode particles have been observed in the Amazon region in the vicinity of Sao Paulo (Backman et al., 2012). The

dataset presented shows aerosol and ion characteristics at measurement sites 40 km and 70 km away from any major anthropogenic pollution sources.

- To address the concerns about the data availability and statistics, we included Table 1, summarizing the NAIS data availability and rain data availability for the whole measurement period.

Referee comment

The authors defined pristine by saying "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." That definition is a little careless. It is quite possible to measure CCN at 0.15% supersaturation and find number concentrations of a 'few hundred cm-3'. At that supersaturation, the particles would be about 100 nm diameter or larger, which for a number concentration of a few hundred/cc is unlikely to be a pristine situation. Also, the authors never define CCN in the manuscript. If you truly mean pristine, then you should rephrase the above sentences as follows: "The occurrence of NPF on ground level in the Amazon region has not been observed previously in pristine conditions, in which the aerosol has not been influenced by anthropogenic pollution." If that is inappropriate, then find a word other than pristine to use.

Author reply

We completely removed the word pristine from the revised manuscript. This particular study does not focus on pristine conditions. The focus of the presented manuscript is to compare the seasonal characteristics of ions and neutral particles in the Amazonian atmosphere. Pristine was mentioned in the previous manuscript as we wanted to point out that nucleation mode particles have only been observed in the Amazon region in the vicinity of Sao Paulo (Backman et al., 2012). We present our results at two measurement sites that are both (40 and 70 km) far away from major pollution sources, which is Manaus in this case. Nevertheless, we are not measuring in pristine conditions, as both sites are to various extents influenced by the Manaus pollution plume.
The second major results in our manuscript focus on the ion and neutral aerosol characterization of the two different measurement locations. The effect of the rainforest canopy on the characteristics of ion and neutral particle size distributions in the very small sizes have not been studied previously, so the manuscript focuses on this characterization.

Referee comment

2) There is a problem with the revised sentence on line 66. Perhaps "…event days are a factor of two lower…"

Author reply

We thank the referee for pointing out the mistake in the manuscript. Since we followed the suggestions by referee1, and thoroughly re-analyzed our dataset, also the text in the revised manuscript has changed significantly.

Referee comment

3) In response to another comment, the authors wrote the following modification: '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).' Instead of 'where T3 on the other hand is reached once…', I assume you meant to say something like "whereas T3 is impacted about once…"

Author reply

We thank the referee for pointing out this mistake. As we re-wrote the manuscript based on the suggestions of referee 1, also the text in the revised manuscript has changed significantly.

Referee comment

4) The authors missed my point regarding my previous comment "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." You

responded "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." My point, which I should have made clearer, was that your statement that "some log-linear relation between the ion concentration and rain intensity could be observed for rain intensities >1 mm h-1 for all the three size bins" is incorrect for Fig 6a. In that case, the log-linear relationship for the T0t ion concentrations is not evident: their variation with rainfall appears to turn on about a rainfall intensity of about 10 mm/hr, and it does not exhibit the clear increase with increasing rainfall intensity as it does in the other five plots. Perhaps that is connected to a higher background concentration of smaller ions, but the exception needs to be mentioned. Also, please correct the legend in Figure 6 that refers to ZF2 rather than T0t.

Author comment
We re-analyzed our dataset based on the suggestions of referee 1. Therefore, most of the Figures were changed in the revised manuscript. Figure 6 now shows the maximum negative ion concentrations during precipitation events for both measurement sites as a function of the rain intensity. We added horizontal lines in Figure 6 to indicate the background ion concentrations at the different sites and different cluster sizes that we studied. We removed the sentence about the log-linear relationship as we do not want to make any parameterization of the ion enhancement due to rain.

Referee comment
5) Concerning my comment 18) Figure 7 and lines 385-395, you "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.'" Why is it that 6-10 nm particles going up will be filtered out by the canopy, but 6-10 nm particles going down will make it to the ground: are the downward particles carried in the wake of the rain drops, leaving less time and for diffusion to the vegetation compared with the upward particles? Please elaborate a little on the mechanisms that differentiate the upward- versus downward-moving particles.

Author comment
Based on our dataset, we cannot make any conclusion on the source of the neutral particles above as observed by the DMPS. The ions inside the rainforest canopy produced by the precipitation are very short-lived. Ion concentrations are only increased during the precipitation events and drop to background levels as soon as the precipitation stops.
Figure 2 shows enhanced ion concentrations in the months October to January in the absence of precipitation (all days with precipitation were excluded from the current analysis in Figures 2 and 3 and Tables 2 and 3). Since those concentrations are increased during the dry season months, when local biomass burning is most frequent in the Amazon region, we thought that the source of those could be due to anthropogenic influence. We agree with the referee that based on our dataset, we cannot make a firm conclusion on the source of those ions.

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