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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Direct observation of molecular clusters and nucleation mode particles in the Amazon" *by* Daniela Wimmer et al.

Anonymous Referee #1

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The presented paper shows particle and ion number concentrations measured at two sites inside and close to the Amazon rainforest. The authors contrast seasonal and diurnal variations of the two different measurement stations by comparing one long-term data set to an intensive operational period during GoAmazon 2014/5. Thereby, rain-related particle and ion bursts are observed below canopy at the near-pristine forest station. New particle formation events have not been observed at the near-pristine site but at a pasture site downwind Manaus.

The manuscript contains valuable and scientifically significant information. Nevertheless, there are some aspects which need further analysis, deeper discussion and clarification. Although the paper is well written, figures and tables are lacking

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Discussion paper

significant information and need a careful revision. Some text passages, identical to existing work, are not referenced correctly. I suggest a careful major revision before considering publication in ACP.

General comments:

The authors compare two different research stations in the vicinity of Manaus. T0t - a remote, near pristine station inside the rainforest of an ecological reserve north of Manaus (e.g., Martin et al., 2016) and T3 - a pasture site (size 2.5 x 2 km) downwind of Manaus outside the rainforest. The observations in this manuscript are divided in 'inside canopy' (T0t) and 'outside canopy' (T3) which suggests a direct comparability and a close connection to forest canopies at both sites. Since T3 is not located inside the rainforest but a pasture site, I suggest to clearly discuss this in the manuscript. Instead of dividing into 'inside' and 'outside canopy', the authors should consider to rephrase the notation for T3 to 'outside forest' or 'pasture site' to avoid confusion.

Here shown particle number concentrations (in particular for the nucleation mode size range) are (at least in the wet season) significantly higher compared to former observations in the (pristine) Amazon region (e.g., Martin et al., 2010a, Martin et al., 2010b, Zhou et al., 2002). On average, the particle number concentration in the 4-20 nm size range agrees very well with the total particle concentration (> 10 nm) measured by a CPC (tables 1, 4), suggesting, that the majority of all particles is in the nucleation mode size range. These contradictory findings are not discussed by the authors. More exact information are addressed in my specific comments.

There are specific sentences and complete text passages which are identical to Martin et al., 2016. Those text passages are either not referenced or not referenced correctly. I suggest to put these sentences in quotation marks or to rephrase the respective sentences. In both cases the original source has to be cited correctly. The

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corresponding text passages are listed in my specific comments.

Specific comments:

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.

Wimmer et al. (page 3, lines 86 and following):
"The regular synoptic changes between the wet and dry seasons offered an additional important scientific contrast to study aerosol dynamics."
Martin et al. (p. 4787):
"The regular synoptic changes between the wet and dry seasons offered an additional important scientific contrast."

Wimmer et al. (page 5, lines 135 and following):
"Under the day-to-day variability in the meteorology, both clean and polluted air masses, mixed to variable degrees, arrived at T3."
Martin et al. (p. 4786):
"Under the day-to-day variability in the meteorology, both clean and polluted air masses, mixed to variable degrees, arrived at T3."

Wimmer et al. (page 7, lines 210 and following):
"The particle population is in dynamic balance with the ecosystem and anthropogenic contributions (e.g. biomass burning; which produces them directly and indirectly) and the hydrological cycle (which removes them). In the wet season (December to March), the Manaus plume aside, the Amazon basin is one of the cleanest continental regions on Earth (Andreae, 2007; Martin et al., 2010). In the dry and transition season (April to

September), biomass burning emissions are prevalent throughout the basin. The most intense biomass burning and atmospheric perturbations take place at the southern and eastern edges of the forest..."
Martin et al. (p. 4787):
"In the wet season, the Manaus plume aside, the Amazon basin is one of the cleanest continental regions on Earth (Andreae, 2007; Martin et al., 2010a). The particle population is in dynamic balance with the ecosystem (which produces them directly and indirectly) and the hydrologic cycle (which removes them). In the dry season, biomass burning is prevalent throughout the basin. The most intense burning and atmospheric perturbations take place at the southern and eastern edges of the vast forest."

Wimmer et al. (page 3, lines 114 and following):
"Manaus is situated at the confluence of the Black River (Rio Negro) with the 115 Solimões river, which together form the Amazon river. The city is an isolated urban region with a population of more than 2 million people (IBGE, 2015; Martin et al., 2017) and is surrounded by 1500 km of forests in all directions."
Martin et al. (p. 4786):
"Manaus, situated at the confluence of the Black River (Rio Negro) with the Solimões river, which together form the Amazon river, is an isolated urban region of over 2 million people (IBGE, 2015)."

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.

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?

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.

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'.

Page 6, lines 180:
A description of the applied inlet system for the PSM would be interesting for future studies under high rh conditions.

Page 6, lines 183:
"Laboratory studies have shown that the RH affects the counting efficiency of the PSM drastically" - Please provide references.

Page 7, lines 203:
"The DMPS data reported here is qualitative but not quantitative." - Please specify if there were problems with this instrument. Quantitative SMPS data are discussed in e.g., section 3.2.

Page 7, lines 220:
The planetary boundary layer development is probably different for pasture and rainforest sites. Can you please comment on that?

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?

Page 9, lines 276:
"the biomass burning during the dry season is expected to increase large ion concentrations" - Please provide a reference

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.

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."

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.

I kindly ask the authors to critically discuss these discrepancies.

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.

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.

Page 15, lines 454:
Please consider to show the results of your backward trajectory analysis in a map.

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 sub-micron aerosol population.

Page 17, lines 510:
"Similar, but weaker, rain-events were found at the site outside the rainforest canopy (T3)." - weaker in terms of what?

Technical comments:

Geolocations are indicated by e.g., -2° S, -60° W. South and west already indicate the negative latitude and longitude.

Page 2, line 72: "Martin et al., 2010" - a or b?

Page 7, line 202, 214, 225: "Martin et al., 2010" - a or b?

Page 9, lines 276: "Martin et al., 2010" - a or b?

Page 17, lines 531: "Amzon"

Technical comments related to figures:

In all boxplots, the whiskers are related to the 25th and 75th percentile. If this is true, what are the boxes referring to?

Fig 1:

- abbreviation "ZF2" used but not explained

Fig 2:

- abbreviation "ZF2" used but not explained
- lowermost 2 panels are missing
- "cm-3", "-3" has to be a superscript

Fig 3:

- "Tot part." - avoid unexplained abbreviations
- lower whiskers are all smaller than or equal zero, the authors state that the data are carefully cleaned. Please comment on the the large amount of zero and below zero particle concentrations.
- Is this the ion concentration in the upper panel and the total particle concentration in the lower panel? The caption and axis only refers to particles.

Fig 4:

- time axis of different panels not synchronized
- upper panel: tick marks invisible
- cm-3, mm h-1
- if positive and negative ions share the same axis, it should not be blue
- 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.

Fig 5:

- please check if the left axis label "%" is correct
- please consider to also show the average total rain

Fig 7:

- time axis of different panels not synchronized
- tick marks in contour plots invisible
- second panel shows additional axis on right side
- lowest panel shows more than 2 orders of magnitude with just two labels

Fig 8:

- "cm-3"

Fig 9:

- please consider to label the 2 columns with NPF no NPF
- the individual panels are irregularly distributed

Technical comments related to tables:

Tab 1:

- almost all parameters show IQR, please add missing IQR values for CS and meteorological parameters

- precipitation rate is given with 4 digits, does that represent the measurement accuracy?
- please consider to add total precipitation and wind velocity

Tab 4:

- NAIS size range indicated in caption misses units
- again, please add IQR for all parameters
- please consider to add total precipitation and wind velocity

Tab 5:

- the table does not fit the page, last column is not readable
- the size information in last columns is missing

Literature:

Manninen, H. E., Mirme, S., Mirme, A., Petäjä, T. and Kulmala, M.: How to reliably detect molecular clusters and nucleation mode particles with Neutral cluster and Air Ion Spectrometer (NAIS), Atmos. Meas. Tech., 9(8), 3577–3605, doi:10.5194/amt-9-3577-2016, 2016.

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(AMAZE-08), Atmos. Chem. Phys., 10(23), 11415–11438, doi:10.5194/acp-10-11415-2010, 2010a.

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Martin, S. T., Artaxo, P., MacHado, L. A. T., Manzi, A. O., Souza, R. A. F., Schumacher, C., Wang, J., Andreae, M. O., Barbosa, H. M. J., Fan, J., Fisch, G., Goldstein, A. H., Guenther, A., Jimenez, J. L., Pöschl, U., Silva Dias, M. A., Smith, J. N. and Wendisch, M.: Introduction: Observations and Modeling of the Green Ocean Amazon (GoAmazon2014/5), Atmos. Chem. Phys., 16(8), 4785–4797, doi:10.5194/acp-16-4785-2016, 2016.

Zhou, J., Swietlicki, E., Hansson, H. C. and Artaxo, P.: Submicrometer aerosol particle size distribution and hygroscopic growth measured in the Amazon rain forest during the wet season, J. Geophys. Res. D Atmos., 107(20), doi:10.1029/2000JD000203, 2002.

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