Response to Anonymous Referee #1

General comments:

This study describes measurements that seem to be of high quality in a very interesting region (the Amazon) during an interesting time that has previously not been characterized (the transition period between wet and dry seasons). Given that these measurements fill an important gap, I recommend them ultimately for publication. However, I have many issues with the paper in its current state, and feel that it could be much improved. While I do not think there is a fatal flaw in the manuscript, there are instances where some additional basic analysis needs to be completed and sections that need further explanation or clarification. As it currently stands, the paper lacks enough of this analysis, and is unclear enough in parts, that it should not be published.

We thank the referee for taking the time to thoroughly review our manuscript. We have addressed each of the comments and recommendations below, and will revise the manuscript accordingly.

Recommendation:

Before publication, major revisions need to be completed. I have tried to detail below those sections that either need further analysis or more detailed explanations.

Specific comments:

p3l15: The start date of the campaign is mentioned here. There should be some acknowledgement within this section that the WIBS measurements and all other measurements presented do not overlap. Perhaps this is completely unimportant insofar as the meteorology being similar over each week-long sampling period, but it should be acknowledged and discussed, if only briefly. If it is the case that these two weeks were very similar (meteorology, back-trajectories, etc.) and should be taken as descriptive of the same general time period, state so.

The referee is correct to note that the sample periods do not overlap for these instruments. In terms of meteorology, the conditions were very similar of the whole measurement period, as the referee suggests. We will include a short paragraph at the end of this section discussing this.

p3l18: Was this the same sampling location as AMAZE-08? Briefly state that this is the case if so.

It was, and we will clarify this here.

p3l30: Attention to the details of the inlet seem to have been considered, but connect the dots for the readers: are there any significant particle losses? Assuming you have done those calculations, please state any relevant conclusions. This is especially important for the coarse mode. Assuming that there are no losses that need be accounted for, please state that you have done the calculations to verify so. Do not let the reader wonder or have to do the calculations themselves.

For the range of flows rates during BUNIAACIC the transmission range has previously been calculated from 4nm to 7µm (Martin et al., 2010). As mentioned in the previous reply, the experiment was conducted at the same sampling location as AMAZE-08 and we will refer to this characterisation in the manuscript.

p6l30: You state “aerosol data were excluded if the pollution flag coincided with. . .” When I look at these plots, I see large gaps of data missing, e.g. maybe of 1/4 of the data in the Fig. 2 time series is absent. Should I conclude this is all pollution flagged? Or is some of it instrument down-time? It would be helpful if you could state in this paragraph what fraction of the data is removed due to pollution flagging. There is no data removed from the WIBS time series (fig. 6), which I assume means that there were no pollution flags during this time? This relates somewhat to my earlier comment about the WIBS and all other data not over-lapping at all in time, and the question of how

similar these two separate sampling periods actually are. It would be worth stating this explicitly, given how many gaps there seems to for the sub-micron instruments.

We will signify in the time-series figures (by shaded area, or similar) the periods removed due to pollution flags. Regarding the WIBS data, there was a mistake in that two pollution episodes were not removed from the time-series in figure 6. This makes no difference to the results or conclusions, and we will modify the figure with removed data specified in the same way as the other figures.

p5l26: You introduce the WIBS channels. Label them here as "FL1," "FL2," and "FL3."

We will add these labels in the appropriate places in the revised manuscript.

p5l27: You are using FT+3sigma to define the FL threshold. Please provide a comment here on why are you not using the ambient threshold determination used by Perring et al (2015). It should be obvious to any instrument user, but it is worth explicitly stating that because the large majority of particles you are seeing are fluorescent, the ambient thresholding approach would not be appropriate. Also, please state what the actual threshold value of FT+3sigma is.

The fluorescence threshold value of FT mean + 3 standard deviations was agreed upon by the WIBS community as the standard for determining particle fluorescence at the 2014 WIBS user group meeting (Boulder, CO, USA) and this value is used in other publications using the same instrument used here (Robinson et al, 2013, Crawford et al, 2014, Crawford et al, 2015, Crawford et al, 2016) so we use this value for consistency.

The method employed in Perring et al (2015) was used to constrain periods where the baseline was unusually variable, most likely due to the presence small fluorescent particles that were below the instrument's size detection limit or fluorescent vapours (e.g., acetone) which would increase the fluorescent background of the optical chamber. This method is unsuitable at the sampling site for the reasons suggested by the referee, and we will state this in the revised manuscript.

As requested, the FT mean +3σ thresholds were: FL1 112.4 ± 3.9, FL2 284.6 ± 7.8, FL3 164.6 ± 5.7.

p5l30: "For a particle to be considered fluorescent. . ." Why are you using 3 sigma? There are numerous examples of different thresholds being used in WIBS studies (e.g. 2.5, 3, 4 sigma). Why is 3 picked? A citation should be provided here. I also recommend stating what the actual threshold value being applied is (i.e. the actual detector counts in the PMT), and not just what FT + 3sigma is. This is very important given that you report actual fluorescence intensity values in Table 3. Additionally, do these values of FT+3sigma stay constant over the measurement campaign? How often is FT mode run? More information on the data treatment here is needed. I would recommend conducting a sensitivity analysis on how different threshold value affect the fraction of particles determined to be fluorescent and the fluorescent particle concentrations. This would lend more meaning and context to the values reported in Table 3.

The rationale for using a threshold value of FT mean + 3 standard deviations is discussed in response to the previous comment and citations to the relevant publications (Robinson et al, 2013, Crawford et al, 2014, Crawford et al, 2015, Crawford et al, 2016) will be provided in the revised manuscript.

During data processing the threshold value for each channel is subtracted from the single particle fluorescence data and the value is clipped at 0 with all values greater than 0 being considered significantly fluorescent compared to the instrument baseline. Fluorescence measurements below the threshold (i.e. less than 0 after threshold subtraction) are not considered physically meaningful and are clipped at 0. This is described in Crawford et al (2015) and we will include a short description of the processing method in this section. As such the fluorescent intensity values reported in Table 3 are relative to the applied threshold and not the absolute detector intensity.

The threshold remains consistent where 58 FT samples were made over the course of the campaign (see earlier response).

p5l34: What does it mean to "monitor instrument fluorescent channel efficiencies and baseline with time" using blue fPSLs?

This statement was misleading. In fact, the fPSLs were just used at the start of the measurements to check that the instrument was working properly. We will reword this accordingly.

p6l4: "Particles detected by this instrument" should be replaced with "Particles with fluorescent magnitudes about the threshold" or something similar (as the instrument "detects" both fluorescent and non-fluorescent particles via being an optical particle counter).

Correct. We will change the wording as the referee suggests.

p6l4: False-positive "FBAP" particles are a known issue in the WIBS. There are many WIBS studies (e.g. Toprak and Schnaiter 2013, Perring 2015 to name a few) and other single-particle fluorescence studies (e.g. Yong-Le Pan 2015) identifying nonbiological fluorescent particles as interferences. There must be an acknowledgement within this section that molecules other than tryptophan and NADH fluoresce, some of which are not biological. Please also include any thinking or analysis you have done to identify the potential presence of false-positives in the WIBS. As it stands, without any discussion of interferences within the manuscript whatsoever, the following sentence should absolutely not be used: "Particles detected by this instrument. . .represent a lower limit of PBAP. . ."

We will include a discussion of fluorescent interferents in the revised manuscript. Generally the identified interferents are smaller than the detection limit of the WIBS; polycyclic aromatic hydrocarbons (PAH) such as naphthalene have been shown to fluoresce in Fl1 (Pöhlker et al., 2012). Soot containing such interferent PAH's have also been investigated; Propane flame soot was generated at a C/O ratio of 0.5 and coagulated in a small aerosol processing chamber to detectable sizes (D_p > 0.8 µm) prior to sampling with a WIBS-4 where it was found that 0.2% of the soot population would fluoresce in Fl1 (Toprak and Schnaiter 2013). We would not expect to observe significant concentration of PAH's or soot outside of the pollution events at such a remote site so their contribution to the observed fluorescent concentration should be negligible.

Mineral dusts contain a small subset of fluorescent aerosol within their population (~10%), and given their ubiquitous nature may present a significant source of interferents to the UV-LIF method (Toprak and Schnaiter 2013), however their observed fluorescent intensity is considerably weaker than is observed for biofluorophores (Pöhlker et al., 2012) and if they were present in any significant concentration they would likely form their own cluster as was demonstrated in Crawford et al. (2016). We will add a brief discussion on this in the revised manuscript.

It is also worth adding that the technique measures "biological containing particles", which may include fluorescent material attached to non-biological particles. We will include a brief explanation of this in the revised manuscript.

p7l2: A general comment on size distributions: I would recommend adding a log-log version of Figure 1 (so have a Figure 1b perhaps) that shows size distributions over the entire size range, integrating the SMPS and WIBS data together. This would be a visual tool to very quickly convey how dominant the sub-micron mode is compared to the coarse mode in terms of particle number. Is it really true that there are no particles at e.g. 600nm (as Figure 8a indicates), or is this the WIBS detection efficiency going to zero? You state that the WIBS measures down to 500nm. Thus, the reasonable assumption from the reader is that there actually are no particles below 750nm, according to the WIBS. But how far does the accumulation mode (shown in Figure 1) tail extend to large diameters? Integrating these size distribution measurements would make all of this more clear.

We have rejected particles smaller than 800 nm from the analysis due to low collection efficiency. We will clarify this in the revised manuscript. Figure 1 shows the SMPS and WIBS size distributions together in a log-log plot. Unfortunately, there is a considerable gap between the size ranges of the two instruments, plus there are issues with trying to combine the dry mobility diameters from the SMPS with the wet optical diameters from the WIBS. The figure doesn't show how far the accumulation mode tail extends or how much it contributes to the coarse mode. We feel it therefore does not add anything to the paper, and have decided not to include it.

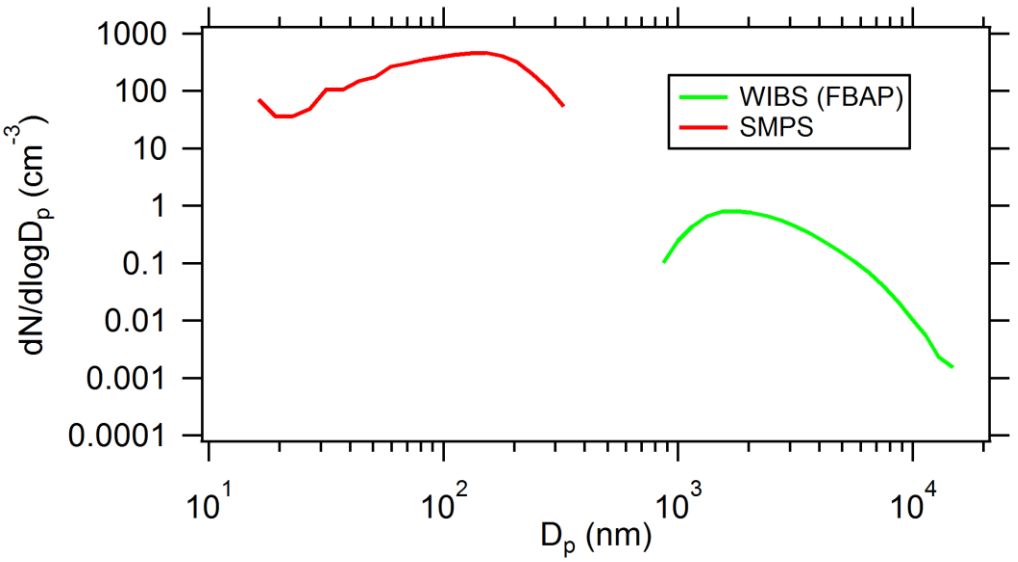

Figure 1. Combined log-log plot of total particle number size distribution (as measured with the SMPS) with FBAP number size distribution (from the WIBS).

p6l11: It seems that pollution episodes have been rigorously identified and removed. As the reader, though, I am wondering why these data were removed at all? Why not include that data, but identify it as potentially influenced by anthropogenic activities? This paragraph seems ideal to add another sentence or two as to explain further the rationale for why these episodes were removed.

The focus of this paper is on the natural (biogenic) aerosol at this time of year to compare with the wet season. We will clarify the scope of the paper in the introduction of the revised manuscript to explain why the pollution events were removed.

p719: I find this discussion of Levoglucosan-as-tracer helpful, though am confused then why f60 is not used as a direct tool in section "2.5 Removal of pollution episodes." Was f60 only considered in the context of a campaign average? Simply because the campaign average is below a reported baseline, were there not episodes of BB influence as determined by the ACSM data directly, which has the ability to directly measure this? If not a graphical presentation of these results from the ACSM, there should at least be a mention of further analysis of BBOA composition that was done beyond looking at the campaign average of this tracer.

Previous studies in the Amazon have observed that a large fraction of the biomass-burning related organic aerosols do not present a significant f60 signal, due to long-range transport (Brito et al., 2014). As such, applying a f60 threshold would remove only fresh fires and not biomass burning emissions. We accept that the text isn't at all clear on this, and we will revise it to clarify this point. The statement in the current text simply says that the relatively low f60 confirms for us that there was no sign of local BB influence during measurements.

p7l24: Please include a paragraph on the presence or absence of PBAP markers from the ACSM data. This is an obvious omission given that at least one of the co-authors on this manuscript are among the very few that have used the AMS in an attempt to identify PBAP. Refer to Schneider et al 2011

("Mass-spectrometric identification of primary biological particle markers and application to pristine submicron aerosol measurements in Amazonia").

The following text has been added to the manuscript:

"Previous studies have successfully identified FPAB markers on ambient aerosol in the Amazon using an aerosol mass spectrometer (Schneider et al., 2011), a method which relies strongly on the high-resolution capabilities of the instrument used at the time. Given the unity mass resolution of the ACSM, similar methodology has not been applied here.

p7l24: Another general comment on the Composition section: the utility of this paper, as I see it, is reporting what aerosol in the Amazon looks like during the transition between wet and dry seasons. Thus, solely reporting the organic, nitrate, and sulphate concentrations from the ACSM seems to be doing a disservice, and further analysis of the ACSM data could be included here. Was the aerosol oxidized? Were there any diurnal patterns in composition changes? How does the organic composition compare to the other studies citied? Should the conclusion drawn from the ACSM data be that there was basically no BBOA and similar organic concentrations compared to the other studies? (or, was there BBOA but it was flagged and removed?) More analysis and synthesis can be included here, given that the purpose of this paper is to give the community a baseline for this location in this season, and contrast it with the work that has been previously done. This seems to have been thoroughly done for the HTDMA data in section 3.5.1, but is absent for the aerosol composition data.

The reviewer is correct as there is a lot to explore from aerosol mass spectrometry measurements during BUNIAACIC campaign. The authors see fit that such detailed description would suit better a separated manuscript, which is currently under preparation.

p9l10: Can you verify that the size-distribution in this figure is not 'fluorescence signal limited?' It is possible, depending on the strength of the fluorescence from the material in these particles, that the signal strengths are on the same order as the threshold. If this were true and we assume an internally-mixed aerosol, there would thus be a particle size above which the average fluorescent signal would be greater than the threshold and below which the average fluorescent signal would be less than the threshold. This would make that size appear to be the true mode of the ensemble, but it would actually just be a reflection of the intrinsic fluorescent strength of the material within these particles. The 'true' diameter, so to speak, would be smaller than what it appears to be. Looking at a size-resolved average values of the FL signals for each FL channel would verify whether or not this data is in the regime. If not (and the signal strengths are sufficiently large relative to the applied threshold), this would add confidence to the reported mode diameter in Figure 8a. This is a general analysis issue for the fluorescent particle measurement community, and given that a paragraph of page 11 is devoted to comparing mode diameters between this and previous studies, I recommend this analysis.

Without knowing the identity of the particles and their resultant morphologies and whether their fluorophores are likely to be found on the surface or in the bulk of the particle, it is difficult to answer how size may influence fluorescent intensity. There is currently a lack of stable solid pure compound fluorescent calibrants to assess how particle size influences fluorescence (i.e., is there a surface area or volume dependence? Is there a maximum penetration depth?), but it not unreasonable to expect that fluorescence increases with particle size. This is an ongoing area of research, and is beyond the scope of this paper.

p9l17: "Cl2 appears to be. . .somewhat less fluorescent." What exactly do you mean by saying 'less fluorescent?' Table 3 indicates the mode diameter of the cluster is 1.9um compared to 2.5um for Cl1. Would a 1.9um Cl1 particle have the same fluorescent intensity as a 2.5 um Cl2 particle?

From table 3 of the paper, it can be seen that clusters 1 and 2 display similar characteristics, i.e., they mainly fluoresce in Fl1 with weak fluorescence in Fl2 and Fl3, however, the mean Fl1 intensity is greater for Cluster 1. This is in contrast to cluster 3, which is mainly fluorescent in Fl3 and likely of different origin. The similarities and strong correlation (p9|18) between clusters 1 and 2 suggests that they are of similar origin, with the difference in fluorescence being due to size, morphology or particle age. We will clarify this in the revised manuscript.

As stated above, it is difficult to answer how size may influence fluorescent intensity.

p9l18: "Both clusters show similar fluorescent signatures to the clusters attributed to fungal spores by Crawford." How are the fluorescent signatures similar? In absolute intensity values? If that is the case, are the two instruments using the same detector gain settings, such that it would make sense to compare the intensities on an absolute scale? Or, are they similar in the relative strengths of channels Fl1-Fl2-Fl3? Even for relative differences between the channels, differences in gain settings would still be relevant in trying to compare this instrument's response with another. Further explanation and/or analysis on the spectral information collected by this WIBS should be provided to support the conclusion that these clusters represent fungal spores. There are other WIBS studies that have identified WIBS signatures for fungal spores as well (see Healy 2012 in Atm Env; Perring, 2015 in JGR; Hernandez, 2016 in AMTD) that would be worth comparing your results to, perhaps here or in section 3.5.2.

The signatures are both referenced to the FT + 3 standard deviation threshold representing an intensity of 0 as discussed earlier. The cluster average values for this experiment and the BEACHON experiment (Crawford et al. 2014,2015), when compared, show that fluorescent signatures relative to the fluorescent detection threshold for BUNIAACIC cluster 1 and BEACHON cluster Z_1 are similar, i.e., both display strong fluorescence in Fl1 and moderately weak fluorescent in Fl2 and Fl3. Both of these clusters also display a strong diurnal cycle with a dependency on relative humidity (see figure 2, which we will include in the revised manuscript as further evidence of this dependency on RH). This behaviour is consistent with that of emission of fungal spores (Hirst, 1953; Pringle et al., 2005; Elbert et al., 2007; Jones and Harrison, 2004).

Both datasets were collected with the same WIBS-3 using identical detector gain settings. The WIBS-3 does not have a high and low gain mode as found in the WIBS-4 and WIBS-4A.

Direct comparison to other studies is not possible due to differences in detector gain (which currently cannot be calibrated) and the choice of excitation and detection wavebands. Even comparing results between the same model of instrument with identical detector/filter configurations has been difficult, as shown in Hernandez et al., (2016).

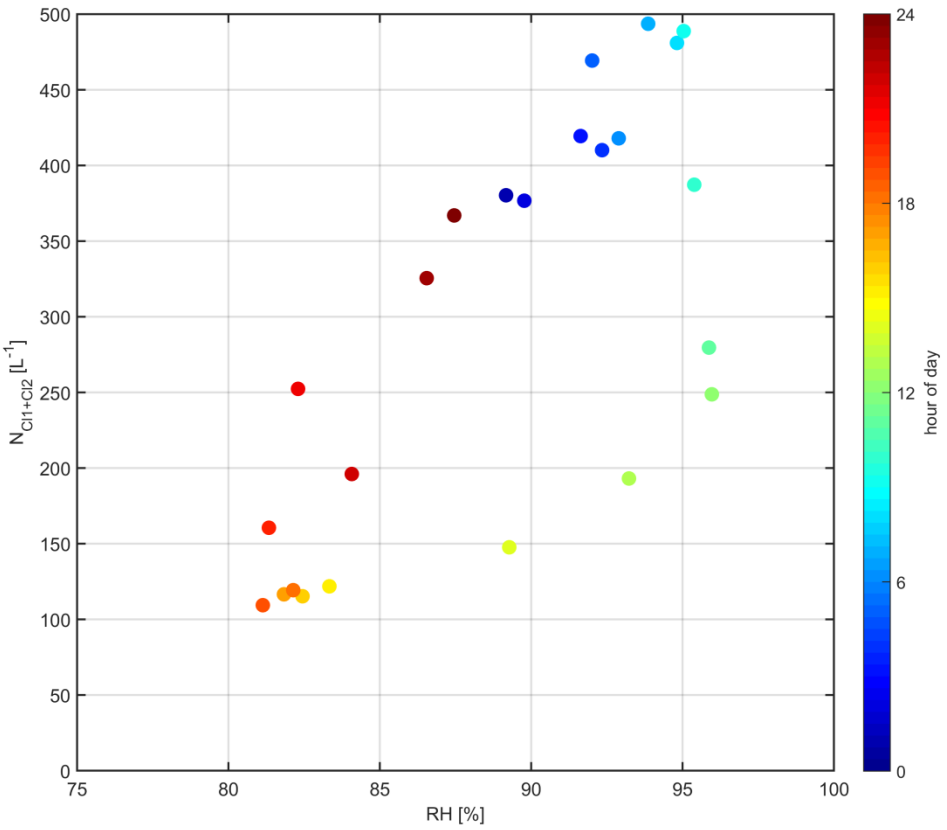

Figure 2. Total particle number in clusters 1 and 2, plotted against relative humidity.

p9l19: I find the following statement confusing: “These clusters (referring to Cl1 and Cl2) contribute approximately 70% to the total FBAP concentration, with no significant diurnal variation.” Yet there is a very strong diurnal signal in FBAP, and Cl1+Cl2 makes up 70% of FBAP. Is there a typo here, or am I misunderstanding the phrase ‘with no significant diurnal variation’ in Cl1+Cl2?

This is not a typo, just badly worded, and we apologise for the confusion. We meant to say that there was no variation in the 70% figure (i.e. there is a strong diurnal variation in Cl1+Cl2, but the make up 70% of FBAP regardless of time of day), but accept that the text is rather obscure. We will clarify this in the revised manuscript.

p9l25: A general comment on this section: the comparison of the HTDMA data made during this study with other previous work done in the same region (or similar regions) seems well done. However, there has been plenty of work done previously on submicron aerosol composition in this region, and there is very little discussion of your ACSM data within the context of this previous work. Please add some content (perhaps a paragraph) in this section comparing your ACSM results to other measurements that have been made here in the Amazon.

The following text has been added to P.9 L.25

“During BUNIAACIC, submicron non-refractory aerosol concentration shows significantly higher concentration (~2.5 µg m^{-3}) than observed at the remote sites in Central Amazonia in previous years during the wet season, ranging from 0.4 µg m^{-3} (Artaxo et al., 2013) and 0.6 µg m^{-3} (Andreae et al., 2015; Chen et al., 2009). Conversely, the concentration is significantly lower than reported during

the dry season (8.9 µg m^{-3}) (Andreae et al., 2015), as consequence of this transitional period not having extensive biomass burning activities, however with already reduced wet deposition due to reduced precipitation. Interestingly, despite the marked changes in ambient concentration, very little differences are observed in terms of relative contributions considering this and previous studies, being strongly dominated by organics (~80%), followed by sulphate and minor contribution of nitrate and ammonium (Andreae et al., 2015; Artaxo et al., 2013; Chen et al., 2009)."

p11l19: There are a number of studies not mentioned in this comparison section that the current manuscript would benefit from citing and discussing: -1. Poschl 2010: They attribute 80% of coarse-mode particles as primary biological particles. While those measurements were done with SEM, they seem to align with these results and should be mentioned. -2. Please also include in this paragraph how your results compare to PBAP modeling work that covers this region (e.g. Spracklen and Heald 2014). -3. A recent study on fungal spore measurements in the coarse mode, "Significant influence of fungi on coarse carbonaceous and potassium aerosols in a tropical rainforest." By Zhang and co-workers. They estimate fungal spore concentrations in a similar environment. There may be more studies. As this section is meant to compare your results to what has come before, a more thorough review of the literature should be done, and should not just be limited to aerosol fluorescence measurements as there are other ways of determining concentrations of airborne fungal spores.

We will include further discussion on these and other studies in the context of our work in this section.

p12l10: Similar to an earlier comment, it is not clear to me if there was no data recorded of biomass-burning influenced air, or if there was the influence of biomass burning but those data were flagged and removed. You write here ". . .the results here may reflect the transition between the two seasons, with periods consistent with each at different times (but without any influence from biomass burning)." The confusion arises because I am left wondering if the air sampled during this period is similar when you discount biomass burning influence, or if the air sampled here is similar partially because there is no biomass burning influence.

Pollution episodes, including biomass burning influences, were removed from the data prior to analysis, so that we could present the measurements of natural (biogenic) aerosol. We will clarify this in the conclusions section to avoid this confusion.

Technical corrections:

p6l27: This does not need to be a new paragraph.

We will modify the text accordingly.

Figure 1: Change "Particle number size distribution for the experiment" to "Particle number size distribution averaged over the entire measurement campaign" or something similar. Also, there are kappa and GF data here as well, which should also be mentioned in the caption.

We will change the caption in the revised manuscript.

Figure 2: Change caption to "The time-series of total particle counts (top panel) and particle number size distribution (bottom panel)." The order of what you list should go top to bottom, and with the multiple panel figures explicitly naming what is where reduces any possible confusion.

We will change the cation accordingly.

Figure 4: Can you make use of the entire range of the ROYGBIV colorscale? Almost all of the data is blue-ish/green, making use of the rest of the scale would make the data more visible here. Also given that this figure comes after a previous figure with many gaps, I would move the gaps statement ("Gaps are largely due. . .") up to Figure 2 or include this statement in each caption.

We will modify the colour-scale. In response to an earlier comment, we plan to signify gaps due to pollution events in these time-series figures with shading (or similar), and explain this in each caption.

Figure 5: I assume this is the case, but is the pie-chart for the average of all the data shown here? State this briefly in the figure caption.

This will be added to the figure caption.

Figure 9: "Mean growth factor for the dominant less hygroscopic mode" should be "Mean growth factor for the dominant, less-hygroscopic mode." Also typo with "agains."

We will correct the grammar and spelling in this cation.

Table 3: What are the units here? E.g. there should be units next to Cl1, Cl2, etc. What are the units of Asymmetry factor? (else a definition of what "Af" actually is should be provided somewhere in the text)

We will include the appropriate units in the table header. The asymmetry factor has arbitrary units, and we will define it in section 2.4.

References:

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Zhang et al., 2015: Significant influence of fungi on coarse carbonaceous and potassium aerosols in a tropical rainforest, Environ. Res. Lett., 10(3), doi:10.1088/1748-9326/10/3/034015

Response to Anonymous Referee #2

The paper 'Biogenic cloud nuclei in the Amazon' presented by Whitehead et al. contains a detailed compilation of different measurements during a 3-weeks intensive in the transition period between wet and dry season at a remote research station in the Amazon. The authors focused on different measurements of micro-physical, chemical and hygroscopic properties of the sub-micron aerosol particle population as well as the fluorescence of super-micron particles - a thoroghly interesting, comprehensive and significant data set. The collected data and shown results are relevant to the scientific community and contribute to a deeper understanding of the significance of (biogenic) aerosol particles for cloud properties and the formation of (mixed-phase) precipitation and hence the hydrological cycle in the Amazon.

The subject matter is clearly in the area of ACP. Nevertheless, I think several aspects concerning the data analysis and further technical issues need to be revisited carefully before the manuscript can be accepted for publication in ACP. Please find my major comments below.

We wish to thank the referee for taking the time for this thorough review of our manuscript. We address each of the comments below, detailing the changes made to the manuscript in response.

General Comments:

The manuscript shows an interesting but brief compilation of individual data sets, which are finally compared to previous studies. Since the whole data set comprises (as stated by the authors) a large variability e.g., for the total particle number concentration (100 - 800 cm-3, cf. Fig. 2), shape of the particle number size distribution (cf. Fig. 1), organic mass contribution measured by the ACSM (0.5 - 4 µg m-3, cf. Fig. 5), one would expect to find similar variability in GF or kappa. Nevertheless, GF and kappa are mainly discussed in terms of campaign averages and the applied color scale in Fig. 4 makes it hard to identify variability. Interestingly, the time series of GF does show clear episodes of stable conditions (cf. July 22th) versus episodes with higher variability (cf. July 23rd). Furthermore, during a short event on July 15th GF shows extraordinary high values (> 1.6), which is not discussed in the manuscript.

I suggest to carefully revisit the results section towards a more systematic and comprehensive analysis and discussion combining information from different measurements (particle number size distribution, total particle number concentration, hygroscopicity and chemical information).

In order to bring together the various measurements, we will include a greater discussion of the variability of GF and kappa (and modify the colour-scale in figure 4, also in response to a comment by referee #1), and a derivation of kappa from ACSM data to compare to those kappa from HTDMA and CCNc measurements.

We would like to add here, that although the organic mass loadings vary considerably, the mass fraction is rather stable over the course of the measurements, and this is consistent with the observed lack of variability in GF and kappa. We have expanded the discussion to take account of this, including an extra panel in Fig. 5 showing the mass fractions, and have also included black carbon measurements as they contribute to the total mass.

The authors apply a hierarchical cluster analysis to the WIBS data, which is certainly a powerful technique to identify PBAB meta-classes. However, there is significant information missing about the input to the analysis and the corresponding discussion. This paragraph is not clearly outlined making it hard to follow the argumentation.

We refer the reviewer to our responses to comments made on this subject by referee #1. We will add a short description of the method to the revised manuscript. Complete information on this analysis technique and its implementation is available from Crawford et al (2015), to which we refer in the manuscript, and it is not practical to repeat it in full in this paper.

Finally, the title is very unspecific and does not clearly reflect the content of the paper.

We will change the title to “Biogenic cloud nuclei in the Central Amazon during the transition from wet to dry season”.

I summarize more specific comments below.

Specific comments:

Section 2.1:

• first paragraph: The authors compare rainfall, temperature and humidity during their measurement period with AMAZE-08. Please specify the statement 'cooler and more humid'.

We will include numbers comparing the temperature and humidity between the two campaigns.

• second paragraph: This paragraph deals with detailed information on the location of the measurement site. Please consider to add a map. This would also be helpful for the discussion concerning the removal of pollution episodes.

We will include a map in the revised manuscript.

Section 2.5:

• The authors describe how they flag and remove pollution episodes from the entire data set. Last sentence: 'Approximately 28% of the HTDMA and CCNc data were removed in this way, with 5% of the data being flagged as possibly impacted by biomass burning and most of the rest due to the Manaus urban plume.'

• Why are only HTDMA and CCNc data removed? Additionally, data gaps in the shown figures have to be specified.

We will specify in the text how much ACSM data were removed due to flags. In addition we will include some shading (or similar) in each of the time-series figures signifying gaps due to pollution flags and add an explanation in each caption.

• I further suggest to consider to show a figure containing all geographical information including the mentioned Manaus bounding box.

We will include this in the map mentioned above.

Section 3.2:

• In section 2.5 the authors already introduce a 'cleaning procedure' to exclude pollution episodes. Does f60 show any correlation with the detected pollution events?

Previous studies in the Amazon have observed that a large fraction of the biomass-burning related organic aerosols do not present a significant f60 signal, due to long-range transport (Brito et al., 2014). As such, applying a f60 threshold would remove only fresh fires and not biomass burning emissions. We accept that the text isn't at all clear on this, and we will revise it to clarify this point. The statement in the current text simply says that the relatively low f60 confirms for us that there was no sign of local BB influence during measurements. We also refer the referee to our response to referee #1 on the same matter.

• p. 7, ll. 21: 'The mean f60 at TT34 in July 2013 was 0.19% ± 0.07%. This is well below 0.3%, which is considered to be the upper limit for background air masses not affected by biomass burning' Have the ACSM data been filtered? Is the mean value calculated after removing pollution events?

Yes, and we will add a note in the text clarifying this.

Section 3.4:

• p. 8, l. 18: 'mean total particle number concentration of FBAP ..' Do you mean the mean FBAP or the mean total particle number concentration?

We mean the mean total particle number concentration, and will correct the text in the revised manuscript.

• p. 8, l. 31: 'The observed night-time peak in FBAP number concentrations in fig. 7 is consistent with nocturnal sporulation driven by increasing RH' Where did you measure T and RH? Are the

measurements collocated (below or above canopy)or part of the regular measurements at the research tower (if so, at which height)?

The RH was from the routine measurements from the top of the tower, and so was not collocated with the WIBS. We will add a note to the revised text.

• p. 9, l. 8: '. . . FBAP clearly dominates the particle number concentrations for Dp > 1 µm, however non-FBAP concentrations are higher for submicron particles': How robust is the characterization of the WIBS instrument? I wonder if this statement might be influenced by a decrease in sensitivity of the fluorescence signal. According to Crawford et al. (2015), the WIBS-4 has a 50% detection diameter at 0.8 µm. Please specify the 50% detection diameter of your instrument.

The instrument D_{50} is 0.8 µm, we will revise §2.4 it include this information. The fluorescence response/collection efficiency is unknown for all UV-LIF instruments as there is a lack of an appropriate calibration/reference standard to perform such characterizations, as discussed in our response to referee #1. We will also modify the statement quoted here by the referee, to clarify that we mean the larger sub-micron particles (i.e. > 0.8 µm).

• p. 9, ll. 13: The authors apply a cluster analysis to the WIBS data without providing details on the data preparation and the precise input. According to the cited paper by Crawford et al. (2015), several steps are involved to filter the data before clustering. Did the authors apply exactly the same criteria? Even if so it is worth mentioning those criteria and the corresponding rejection rate in this manuscript.

The exact same method/criteria were applied in this analysis. We will revise §3.4 to clarify this. Approximately 15% of the single particle data was rejected based on this criteria, i.e., inclusion required D>0.8 µm, fluorescent in at least one channel and no detector saturation.

• p. 9, ll. 15: It is hard to follow the argumentation concerning the cluster analysis: 'Cl1 has previously been attributed to fungal spores (Crawford et al., 2014) based on comparison with other sampling techniques and the diurnal emission pattern (see fig. 7) with higher concentrations observed overnight' Was Cl1 attributed to fungal spores based on the observed diurnal cycle (in this publication) or on the mean values (of FL1-3, AF, size) of the corresponding cluster in Crawford et al., 2014?

The attribution of Cl1 to fungal spores was primarily based on the observed diurnal cycle and response to RH (see response to referee #1) and we will include a discussion of the RH dependence in the revised manuscript to clarify this. The similarity of the cluster centroids and the behaviour of the cluster to the work in Crawford et al., 2014 were presented as additional supporting information. We agree to clarify this in the revised manuscript.

• p. 9, ll. 20: 'The statistical parameters of each cluster are shown in table 3 for comparison. Together, these clusters contribute approximately 70% to the total fluorescent particle concentration, with no significant diurnal variation in this figure, suggesting that FBAP were dominated by fungal spores during this study.' Why does the hierarchical cluster analysis cluster only 70% of the data? Why is there no significant diurnal variation? And why does it in this case lead to the stated conclusion?

We agree that this section is not clear and will be revised. "Together, these clusters contribute approximately 70% to the total fluorescent particle concentration" refers to the sum of clusters 1 and 2, not the sum of all clusters, i.e., the clusters representative of fungal spores account for 70% of the fluorescent population by concentration. The HCA method used here clusters all of the input data.

We meant to say that there was no variation in the 70% figure (i.e. there is a strong diurnal variation in Cl1+Cl2, but the make up 70% of FBAP regardless of time of day), but accept that the text is rather obscure. We will clarify this in the revised manuscript.

Section 3.5.1:

• p. 10, l. 28: 'The HTDMA derived _ from the Borneo experiment shows more hygroscopic aerosol than in Amazonia, as discussed above, however the CCNc derived values are more in line with those in Amazonia. This discrepancy has been noted previously and possible reasons for it discussed by Irwin et al. (2011) and Whitehead et al. (2014).' It would be interesting to discuss the findings of the mentioned papers in the context of the here observed discrepancy.

We will add a brief summary of the discussion from those paper in the revised manuscript.

Section 3.5.2:

• p. 11, l. 7: 'The median number concentration of FPAB observed below the canopy in this study was 372 l−1'. Unprecise – which study do you mean, Gabey et al. (2010) or this study?

This study. We will clarify this in the text.

• Concerning the observed discrepancies with Huffmann et al. (2012), the authors discuss instrumental issues, mixing effects related to strong vertical gradients and pbl development. I suggest to add a discussion about possible effects of wet deposition, since the measurements of Huffmann et al. (2012) were performed during the wet season.

We will include a couple of sentences in the revised manuscript discussing the possible role of wet deposition in the differences between these measurements.

• p. 11, l. 28: 'Diurnal variations between this study and that of Huffman et al. (2012) were similar, however Gabey et al. (2010) reported an additional increase in the afternoon in Borneo'. Unprecise – which measurement parameter increases?

In this paragraph we are discussing FBAP number concentrations. We will clarify this in the text.

Technical issues:

Please reference all your physical variables in the text and/or figure captions.

We will modify the captions / text appropriately.

Please do not use abbreviations like 'don't' (e.g., p. 11, l. 32).

We will modify the text accordingly.

Figure captions miss significant information:

Fig. 1:

• information on the derived GF and kappa is missing

We will add this information to the caption

• HTDMA, CCNc data comprise different measurement periods. Please specify that in the figure. Are these data averaged over the same time period?

HTDMA and CCNc data comprise the same measurement periods. We will clarify this.

Fig. 2:

• NCN - is this measured by the CPC or integrated from the size-resolved measurements?

NCN is integrated from the size-resolved measurements, and we will clarify this in the caption.

• please specify the data gaps

We will specify the data gaps according to pollution flags and/or instrument down-time as discussed in response to a previous comment.

Fig. 4:

• please specify the data gaps#

As above

• all other figures use GF(D/D0) instead of 'Growth Factor D/D0'

We will modify the label to GF(D/D0).

Fig. 5:

• please specify the data gaps

As above

• The unit is probably µg/m3

That is the unit specified in the axis label.

• What is the collection efficiency for the ACSM data?

The following text has been added to P.7L.24:

"The instrument collection efficiency was calculated to be 1 during BUNIAACIC, through the comparison of the mass concentration of species measured by the ACSM and MAAP (black carbon) with the integrated mass of the SMPS. Further details of the method are given by Brito et al. (2014) and Stern et al. (in preparation).

Fig. 6:

• Ntot refers to the size range of the WIBS, make sure that there is no confusion with the term 'total counts' in Fig. 2

We will specify this in the figure caption

Fig. 7:

• Ntot refers to the size range of the WIBS, make sure that there is no confusion with the term 'total counts' in Fig. 2

We will specify this in the figure caption

• please add information about the sensor height and position for T and RH

We will add this information to the figure caption.

• °C

This is correct

Fig. 8 a & b:

• you use Dp instead of Dp

This will be corrected for the revised manuscript

• unit of dN/dlog dp is wrong

This will be corrected for the revised manuscript

Fig. 9: 'Irwin et al., (2011)'

References:

• page 16, line 15: lower case initials: 'Wiedensohler, Arana'

We will correct this

• page 17, line 3: full name instead of initials: 'Anna Stefaniak'

We will correct this

References:

Brito et al., 2014: Ground-based aerosol characterization during the South American Biomass Burning Analysis (SAMBBA) field experiment. Atmos. Chem. Phys. 14, 12069–12083, doi:10.5194/acp-14-12069-2014

Crawford et al., 2014: Characterisation 5 of bioaerosol emissions from a Colorado pine forest: results from the BEACHON-RoMBAS experiment, Atmospheric Chemistry and Physics, 14, 8559–8578, doi:10.5194/acp-14-8559-2014

Crawford et al., 2015: Evaluation of hierarchical agglomerative cluster analysis methods for discrimination of primary biological aerosol, Atmospheric Measurement Techniques, 8, 4979–4991, doi:10.5194/amt-8-4979-2015

Gabey et al., 2010: Measurements and comparison of primary biological aerosol above and below a tropical forest canopy using a dual channel fluorescence spectrometer, Atmospheric Chemistry and Physics, 10, 4453–4466, doi:10.5194/acp-10-4453-2010

Huffman et al., 2012: Size distributions and temporal variations of biological aerosol particles in the Amazon rainforest characterized by microscopy and realtime UV-APS fluorescence techniques during AMAZE-08, Atmospheric Chemistry and Physics, 12, 11997–12019, doi:10.5194/acp-12-11997-2012

Irwin et al., 2011: Size-resolved aerosol water uptake and cloud condensation nuclei measurements as measured above a Southeast Asian rainforest during OP3, Atmospheric Chemistry and Physics, 11, 11 157–11 174, doi:10.5194/acp-11-11157-2011

Whitehead et al., 2014: A meta-analysis of particle water uptake reconciliation studies, Atmospheric Chemistry and Physics, 14, 11 833–11 841, doi:10.5194/acp-14-11833-2014

Response to Anonymous Referee #3

This manuscript reports the results of aerosol measurements taken place in the Amazon basin during the wet-to-dry transition period. The measurements include particle size distributions, hygroscopicity, and fluorescent biological aerosol particle concentrations, and are compared to the previous measurements. The results are important and interesting, especially since there are few previous studies in that environment. However, it is not clear to me why the authors choose to remove pollution episodes from this dataset and how this "clean" dataset provides a "unique contrast (page 2, line 10) to the wet-season data?" In fact, the observed particle total number

concentrations and hygroscopicity as well as chemical composition are quite similar to those observed during the wet season. The WIBS-3 results are different but also largely because the measurements were done within the canopy. To me, the removed data are really the key feature of the transition period, meaning influences but not as strong as the dry season. It is important to add that analysis as a contrast. The authors should also pay attention to the manuscript preparation guidelines for authors provided by the journal (http://www.atmospheric-chemistry-andphysics.net/for_authors/manuscript_preparation.html). I recommend this manuscript be published after the following comments are addressed.

We wish to thank the referee for taking the time to thoroughly review our manuscript. The main focus of this paper is on the natural (biogenic) aerosol at this location during the transition from wet to dry seasons. The contrast with the wet season is due to the difference in meteorology. We will replace "contrast" with "comparison" in the quoted text to avoid confusion. We agree that it would be useful to consider biomass burning influenced air masses as well; however as we state in section 2.5, most of the removed data was due to pollution from Manaus (which is not unique to any time of year). The data flagged as *possibly* influenced by biomass burning accounts for only 5% of the data, which we did not consider sufficient to allow for a good comparison.

We address each of the referee's other comments below.

Specific comments:

(1) A 5-paragraph abstract seems unnecessary for this paper. Some of the details may be removed and the key points need to be summarized more concisely.

We will shorten the abstract as far as possible.

(2) Page 3, line 19-20: Please provide the relative humidity and temperature for both campaigns.

We will include this information in the revised manuscript.

(3) Page 4, line 30; Page 5, line 32: Do you mean "polystyrene latex spheres (PSL)" for both cases? What sizes have you used for the calibration? Do the uncertainties for growth factor derived from HTDMA vary by D0? What do you mean "blue fluorescent latex spheres"? Please clarify. Also, since different kinds of diameters are described in the paper, the authors should specify the diameter type in the text and figures.

We do mean polystyrene latex spheres (PSL), and will clarify this, as well as including the sizes used for each calibration. Sub-micron particles are measured as mobility diameter, while the WIBS measured the optical diameter; we will clarify this. We will clarify how we used the blue fluorescent latex spheres, and add that they were manufactured by Polysciences Inc., PA, USA, and Duke Scientific Corp., CA, USA.

(4) Page 6, line 4-5: Do you mean "some of the PBAP are detected by WIBS"? Please clarify and give examples.

We mean some PBAP won't necessarily be detected by the WIBS, as discussed by Gabey et al (2010) and Huffmann et al (2012). We will clarify this and expand the discussion in the revised manuscript.

(5) Section 2.5: It is not clear to me which flag was applied to which dataset and whether if the flag was properly set. The authors should provide clear information about the data processing and have consistency among datasets.

First of all, Figures 2, 4, and 5 look like having different gaps (lack of clear description in the graphs and figure captions about the gaps).

The flags were applied in the same way to the HTDMA, CCNc, ACSM and size data. We will make this clearer in section 2.5 of the revised manuscript. In addition, we will specify the periods in each of these figures where data were removed due to pollution flags (by shaded areas, or similar), and explain this more clearly in the captions. Some additional gaps were due to instrument down-time. Again, we will clarify this in the relevant captions.

Second, the back trajectories at all altitudes from 0 to 4000 m.a.s.l were used for the identification of pollution episodes (page 6, line 17). However, most sampling was taken at 39 m (10 m above canopy) and WIBS was operated on the ground level.

Issues can arise with back-trajectories initiated at ground level, due to the effects of the terrain on air flow, and the greater chance of the trajectory intersecting the ground. To overcome this, we investigated the trajectories at several heights. In terms of the pollution flags, the results were largely the same at the 0 – 2 km levels and very little influence from the upper level flow at 4 km. We will clarify this in the revised manuscript.

Third, it was said that data sampled for local wind direction of 270°-340° were flagged as potential generator contamination (line 27). But in line 31-32, the authors said that 5% of the removed data were potential biomass burning and the rest were Manaus plume. Then, which part is due to generator contamination?

In fact, there were no instances of flagged generator contamination during the measurement periods in this study. We will explain this in the revised manuscript.

Finally, in line 29-30, significant increases in black carbon concentration and particle number concentration were used as the second criteria of data removal. The question is "are there periods with such significant increases but not flagged by the back trajectories passed over Manaus, fire zone, or by wind direction for generator plumes?" If so, when and why? If not, the former (increases) is enough for identifying the pollution episodes.

There were no other increases in black carbon or particle number concentration outside the flagged periods, but we couldn't have known this when we started our analysis of pollution events. In addition, the exercise in flagging by back trajectories and wind sectors allows us to identify the nature of the pollution event (Manaus plume or biomass burning influence). We believe that this level of redundancy in flagging data for pollution events is important to ensure that we have done this as rigorously as possible, without inadvertently removing otherwise good data.

(6) Section 3.1: Both paragraphs said that the observed particle number size distributions are similar to the ones measured in the dry season (i.e., effected by biomass burning). However, the data are supposed to represent background conditions because of the removal of pollution episodes.

And they do: it is the shape of the distribution that is similar, while the number concentrations in this study are somewhat lower than during the dry season. We will add a sentence (with values) explaining this.

(7) Page 7, line 21-23: The author should clarify that the ACSM data (Fig. 5) do not cover the entire measurement period (Figs 2 and 4). "in July 2013" is inaccurate. Have the excluded periods flagged by biomass burning shown elevated f60?

Previous studies in the Amazon have observed that a large fraction of the biomass-burning related organic aerosols do not present a significant f60 signal, due to long-range transport (Brito et al., 2014). As such, applying a f60 threshold would remove only fresh fires and not biomass burning emissions. We accept that the text isn't at all clear on this, and we will revise it to clarify this point. The statement in the current text simply says that the relatively low f60 confirms for us that there was no sign of local BB influence during measurements. We also refer the referee to our response to referee #1 on the same matter.

(8) Page 7, line 28: What are the definitions of hydrophobic, less or more hygroscopic mode (page 10, line 2) in terms of growth factor? Are their definition consistent in literatures (e.g., for the comparisons done in page 10, line 1-12?

The terms in quotation marks are as defined in the cited literature, however we will define the growth factor ranges to make comparison easier.

(9) Page 7, line 29-30: What does the "local anthropogenic influence" stand for? What is "this distribution (i.e., . . .)"?

Here we are speculating that the hydrophobic mode is due to some unknown local anthropogenic source, and will insert the word "unknown" to make it clear. Then we refer to the growth factor distribution, and will insert the term "growth factor".

(10) Page 8, line 1-5: Increased growth factor with particle dry diameter can be explained by many possibilities (it doesn't have to be greater sulfate contribution at larger diameter; organic material at different diameter may different as well). Without careful analysis, I think it is hard to demonstrate that the observations here reflect similar size-resolved chemical information to the previous studies. And the particle number size distributions observed in this study are indeed different from what was observed in previous wet-season studies as described in Sect. 3.1.

We agree with the referee that careful analysis is needed before we can draw this conclusion. We will modify this paragraph to say that higher sulphate concentration is a possible explanation.

(11) Page 9, line 15-24: The analysis here is confusing and needs clarifications. It was said first that C11 is attributed to fungal spores and C12 remain unclassified. Then why "both clusters show similar fluorescent signatures to the clusters attributed to fungal spores"? Aren't all the three classes distinct in fluorescent signatures (line 15)?

We accept that this isn't clear and we agree to revise this section to the following:

"Cl1 like particles have previously been attributed to fungal spores (Crawford et al., 2014) based on comparison with other sampling techniques and the diurnal emission pattern (see fig. 7) with higher concentrations observed overnight. Cl2 appears to be a distinct sub-class of Cl1 which is less fluorescent in FL1. Cl2 shows similar behaviour to, and correlates strongly (r2 = 0.86) with Cl1, hence both have been combined in fig. 7. Both clusters show similar fluorescent signatures to the clusters attributed to fungal spores by Crawford et al. (2014, 2015)."

Second, in line 21, it was said that "these clusters . . ., with no significant diurnal variation in this figure, suggesting that FBAP were dominated by fungal spore during this study." Does "these" mean C11+C12 or C11+C12+C13? Don't C11 and C12 show nighttime increase in Fig. 7? Finally, if C13's concentration is low, what about the residuals in the cluster analysis (meaning Fig. 7 showed a difference of hundreds in number concentration between FBAP and C11+C12)? What does the "insufficient data" mean in line 24?

We are referring to Cl1+Cl2, and we will clarify this in the revised manuscript. Cl1 and Cl2 do show a nighttime increase; we meant to say that there was no variation in the 70% figure, but accept that the text is rather obscure. We will clarify this in the revised manuscript.

The residual difference between N_{FBAP} and $N_{Cl1}+N_{Cl2}$ is a result of rejecting saturated particles from the cluster analysis input.

Insufficient data refers to a lack of additional supporting data which could be used to infer the origin of Cl3, e.g., response to rainfall may infer that the particles are bacterial. We will clarify this in the revised manuscript.

(12) Page 10, line 9 and line 12: What does “strong diurnal cycles” mean? Daytime peak? Please clarify.

The “strong diurnal cycles” refers to an increase in the fraction of moderately hygroscopic particles, and we will clarify this in the revised manuscript.

(13) Page 10, line 31-32: What about the removed data? Do those data show very different results compared to the “clean” conditions? Also it is important to explain why the particle concentrations and hygroscopic properties are similar to those during the wet season but the particle size distributions are similar to those observed in the dry season (my comment #6, Sect. 3.1).

The removed data was mostly flagged as pollution from Manaus, which is of no interest to this study, while the data flagged as possibly influenced by biomass burning made up less than 5% of the total, which we considered insufficient to provide a significant result. We will expand the discussion on the differences / similarities with the wet season.

(14) Page 11, line 11-12: What kind of meteorological conditions? Need a reference or example to support this hypothesis. Also, what are “other locations”? Please specify.

We refer to the wetter, more humid conditions of the wet season favouring sporulation, and we will include references in support. We will specify in the revised manuscript the locations for each citation here.

Technical remarks:

Page 3, line 18-19: Revise “the AMAZE-08 campaign saw 370 mm fall” and move the reference to the end.

We will revise as necessary

Page 3, line 27: Revise “local time was UTC – 4 hours”.

We will revise as necessary

Page 3, line 31: “RH” has not been defined yet.

We will define RH here

Page 4, line 21 and 25: Properly revise “dry sizes” since the DMA selects a band of the electric mobility not just one size.

We will use appropriate terminology in the revised manuscript

Page 4, line 28-29: “a bubble flowmeter” is an improper description. Also, shouldn’t be “Gillibrator-2”?

We will change this to “air flow calibrator”, and “Gillibrator-2”.

Page 5, line 23-26: What is NADH? What do you mean “3 fluorescence channels”?

We will define NADH here. The 3 fluorescence channels are already defined in the preceding text.

Page 6, line 4: Add “as” after “termed” and revise the later part of the sentence.

We believe “termed” is used correctly here, but will revise the end of the sentence.

Page 7, line 3 and later text: “fig. ” should be “Fig. ”.

We will revise accordingly

Page 7, line 8: “particle counts” should be “particle number concentrations”.

We will revise accordingly

Figure 5. Remove frame. Figure 5 appeared earlier than Fig. 4.

We will remove the frame, and change the order of the figures.

Page 7, line 27: Should be “in the range of 1.2 to 1.4” (the word “of” is missing).

We will revise accordingly

Page 8, line 8-9: Check the grammar for “ at larger diameters _ _ . . . and _ _ 0.18 around the accumulation mode. ” SI units should be used, and units in the denominator should be formatted with negative exponents.

We will revise the grammar as necessary. Kappa has no units.

References:

Brito et al., 2014: Ground-based aerosol characterization during the South American Biomass Burning Analysis (SAMBBA) field experiment. Atmos. Chem. Phys. 14, 12069–12083, doi:10.5194/acp-14-12069-2014

Crawford et al., 2014: Characterisation 5 of bioaerosol emissions from a Colorado pine forest: results from the BEACHON-RoMBAS experiment, Atmospheric Chemistry and Physics, 14, 8559–8578, doi:10.5194/acp-14-8559-2014

Crawford et al., 2015: Evaluation of hierarchical agglomerative cluster analysis methods for discrimination of primary biological aerosol, Atmospheric Measurement Techniques, 8, 4979–4991, doi:10.5194/amt-8-4979-2015

Gabey et al., 2010: Measurements and comparison of primary biological aerosol above and below a tropical forest canopy using a dual channel fluorescence spectrometer, Atmospheric Chemistry and Physics, 10, 4453–4466, doi:10.5194/acp-10-4453-2010

Huffman et al., 2012: Size distributions and temporal variations of biological aerosol particles in the Amazon rainforest characterized by microscopy and realtime UV-APS fluorescence techniques during AMAZE-08, Atmospheric Chemistry and Physics, 12, 11997–12019, doi:10.5194/acp-12-11997-2012

Biogenic cloud nuclei in the Central Amazon during the transition from wet to dry season

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Abstract. The Amazon basin is a vast continental area in which atmospheric composition is relatively unaffected by anthropogenic aerosol particles. Understanding the properties of the natural biogenic aerosol particles over the Amazon rainforest is key to understanding their influence on regional and global climate. While there have been a number of studies during the wet season, and of biomass burning particles in the dry season, there has been relatively little work on the transition period - the start of the dry season in the absence of biomass burning. As part of the Brazil-UK Network for Investigation of Amazonian Atmospheric Composition and Impacts on Climate (BUNIAACIC) project, aerosol measurements, focussing on unpolluted biogenic air masses, were conducted ~~above the canopy~~ at a remote rainforest site in the Central Amazon, during the transition from wet to dry seasons, in July, 2013. This period marks the start of the dry season, but before significant biomass burning occurs in the region.

Median particle number concentrations were 266 cm^{-3}, with size distributions dominated by an accumulation mode of 130 - 150 nm. During periods of low particle counts, a smaller Aitken mode could also be seen around 80 nm. While the concentrations were similar in magnitude to those seen during the wet season, the size distributions suggest an enhancement in the accumulation mode compared to the wet season, but not yet to the extent seen later in the dry season, when significant biomass burning takes place. Submicron non-refractory aerosol composition, as measured by an Aerosol Chemical Speciation Monitor (ACSM), was dominated by organic material (~~86~~around 81%).

Aerosol hygroscopicity was probed using measurements from a Hygroscopicity Tandem Differential Mobility Analyser (HTDMA), and a quasi-monodisperse Cloud Condensation Nuclei counter (CCNc). The hygroscopicity parameter, κ, was found to be low, ranging from 0.12 for Aitken mode particles to 0.18 for accumulation mode particles. This was consistent with previous studies in the region, but lower than similar measurements conducted in Borneo, where κ ranged 0.17 - 0.37~~, possibly due to a stronger marine influence at that location, bringing higher sulphate loadings than are typically seen in the Amazon~~.

A Wide Issue Bioaerosol Sensor (WIBS-3M) was deployed at ground level to probe the coarse mode, detecting primary biological aerosol by fluorescence (Fluorescent Biological Aerosol Particles, or FBAP). The mean FBAP number concentration

was ~~404~~ 400 ± ~~237~~ 242 l^{-1}, however this ~~was subject to a strong diurnal cycle, and~~ ranged from around 200 l^{-1} during the day to as much as 1200 l^{-1} at night. FBAP dominated the coarse mode particles, comprising ~~more than 90% of particles detected by the WIBS-3 during the night. This proportion was also subject to a diurnal cycle, dropping to~~ between 55% and 75% of particles during the day ~~, since non-FBAP~~ to more than 90% at night. Non-FBAP did not show a strong diurnal pattern. Comparison with previous FBAP measurements above canopy at the same location suggests there is a strong vertical gradient in FBAP concentrations through the canopy. ~~Application of Ward linkage cluster analysis using the z-score normalisation to the~~ Cluster analysis of the data suggests that FBAP were dominated (around 70%) by fungal spores. Further, long-term measurements will be required in order to fully examine the seasonal variability, and distribution through the canopy of primary biological aerosol particles.

This is the first time that such a suite of measurements has been deployed at this site to investigate the chemical composition and properties of the biogenic contributions to Amazonian aerosol during the transition period from the wet to dry seasons, and thus provides a unique ~~contrast~~ comparison to the aerosol properties observed during the wet season in previous, similar campaigns. This was also the first deployment of a WIBS in the Amazon rainforest to study coarse mode particles, particularly primary biological aerosol particles, which is likely to play an important role as ice nuclei in the region.

1 Introduction

The Amazon Basin consists of the world's largest rainforest, covering an area of 5.5 million square kilometres. The Amazon rainforest is one of the few continental regions where atmospheric processes are minimally influenced by anthropogenic emissions, particularly during the wet season, and ambient conditions can represent, to some extent, those of the pristine pre-industrial era (Pöschl et al., 2010). Concentrations and properties of aerosol particles are largely governed by biogenic emissions, of both primary biological aerosol particles (PBAP) and biogenic volatile organic compounds (BVOC), which contribute to secondary organic aerosol (SOA). On a regional scale, in the wet season, the hydrological cycle is strongly influenced by these biogenic aerosol emissions, which provide most of the cloud condensation nuclei and thereby influence the radiation balance and cloud lifetime (Pöschl et al., 2010). In the dry season, by contrast, widespread biomass burning can result in a substantially increased aerosol optical depth over large areas of Amazonia, as well as modified cloud properties and suppressed precipitation (Andreae et al., 2004).

Previous studies in the pristine Amazon rainforest showed that fine particles (which account for most of the cloud condensation nuclei), consist mostly of secondary organic material derived from oxidised biogenic gases (Pöschl et al., 2010; Martin et al., 2010a; Allan et al., 2014; Chen et al., 2015). A lack of evidence for new particle formation during ground-based measurements (Zhou et al., 2002; Rissler et al., 2004; Martin et al., 2010a) implies that nucleation processes occur at higher altitudes, and new particles are entrained into the boundary layer from aloft (Krejci et al., 2005; Martin et al., 2010b). Larger super-micron particles are dominated by primary biological aerosol particles (PBAP) released from rainforest biota (Elbert et al., 2007; Pöschl et al., 2010; Huffman et al., 2012), which can play a significant role as ice nuclei (Prenni et al., 2009). These PBAP consist of wind-driven particles, such as pollen, bacteria, and plant debris, as well as actively ejected material,

such as fungal and plant spores. Non-biological particles observed in the Amazon in the super-micron size range largely consist of advected Saharan dust and sea-salt from the Atlantic (Formenti et al., 2001; Worobiec et al., 2007; Martin et al., 2010b).

The low aerosol number concentrations in the pristine Amazon rainforest (typically a few hundred cm^{-3}) mean that CCN activation in convective clouds is often aerosol limited (Pöschl et al., 2010). It is clear that there is a strong coupling between the rainforest biosphere and the hydrological cycle in the Amazon Basin, with biogenic aerosol particles providing the nuclei for clouds, which in turn sustain the rainforest through precipitation (Pöschl et al., 2010).

Improving our knowledge of these processes is necessary to understanding the influence the Amazon rainforest has on regional and global climate and atmospheric composition, and how changing land use and climate in Amazonia will impact on this (Artaxo et al., 2013). To this end, the Brazil-UK Network for Investigation of Amazonian Atmospheric Composition and Impacts on Climate (BUNIAACIC) was established to define and nurture a framework within which future UK contributions to studies in these areas may be coordinated. As part of the BUNIAACIC project, a short-term intensive measurement campaign was undertaken at a pristine rainforest site in July, ~~2013, focusing on aerosol particle concentrations and properties during clean, pollution-free conditions. The timing permitted a study of the natural aerosol at the start~~ 2013. The main focus of this study was to look at natural (biogenic) aerosol at this site at the beginning of the dry season ~~, which could be compared to~~ (also referred to as the transition fron wet to dry season), and to compare to previous measurements made during the wet season at the same location (Martin et al., 2010a). Here we present the results of this study.

2 Methodology

2.1 Measurement Site and Sampling

The measurements were conducted at a remote site in pristine Amazonian rainforest between the 4th and 28th July 2013, during the transition from the wet to dry seasons. This is around the start of the dry season, but before significant biomass burning takes place. In July 2013, the total rainfall measured was 153 mm, mostly concentrated at the start and end of the month (during the measurement period itself, the rainfall was 77 mm). For the purposes of comparison, the AMAZE-08 campaign~~(Martin et al., 2010a) saw~~ , which was conducted at the same site, had 370 mm ~~fall~~ of rainfall over the course of 5 weeks during the wet season ~~. Conditions were also~~ (Martin et al., 2010a). In this study, the quartile ranges in temperature were 24°C - 29°C during the daytime, and 23°C - 25°C at night; RH was 72% - 92% by day and 85% - 96% at night. By contrast, the conditions during AMAZE-08 were cooler and more humid~~than during the current study~~, with temperature ranging 23°C - 27°C during the day and 22°C - 24°C at night; RH ranging 88% - 99% by day and 96% - 100% at night (Martin et al., 2010a).

Sampling was done at the TT34 tower (2°35'40"S 60°12'33"W, elevation 110 m), in the Reserva Biológica do Cuieiras, approximately 60 km NNW of the city of Manaus in Brazil ~~.~~(see Fig. 1). The site is representative of near-pristine conditions, and no biomass burning takes place within the reservation, however the site can be affected by regional transport of pollutants including emissions from Manaus and biomass burning (Artaxo et al., 2013; Rizzo et al., 2013). Locally, accommodation for researchers and a 60 kW diesel generator were situated 0.33 km and 0.72 km, respectively, in a WNW direction from the tower. Intensive measurement campaigns have taken place at this site in the past (e.g. Martin et al., 2010a), and long term

measurements have been conducted since 2008 (Artaxo et al., 2013; Rizzo et al., 2013). During this experiment, local time was ~~UTC -~~ 4 hours behind UTC.

A laminar sample flow of about 17 lpm was drawn through a 3/4" OD stainless steel line from a height of 39 m (about 10 m above canopy height) down to a ground level air conditioned container, in which the instruments were housed. Before entering the container, the sample was passed through an automatic regenerating adsorption aerosol dryer (Tuch et al., 2009). This kept the ~~RH~~ relative humidity (RH) in the sample flow to between 20% and 40%. For the range of flows rates during this campaign the transmission range has previously been calculated from 4 nm to 7 µm (Martin et al., 2010a). Instruments drawing off this dried sample flow included a Hygroscopicity Tandem Differential Mobility Analyser (HTDMA; University of Manchester), and a Cloud Condensation Nuclei counter (CCNc; CCN-100, Droplet Measurement Technologies). Upstream of these instruments, the sample flow (2 lpm) was further dried to an RH of between 15% and 25% with a nafion dryer operating with a counterflow of dry compressed air. The flow then passed through an electrical ionizer (model 1090, MSP Corporation), providing a charge-neutralised aerosol sample to the instruments. These same instruments were deployed in Borneo during the OP3 project (Irwin et al., 2011). Further details of the HTDMA and CCNc are given below.

Core instruments running at the site, on the same inlet, included a Multi Angle Absorption Photometer (MAAP; model 5012, Thermo-Scientific), a Condensation Particle Counter (CPC; model 3772, TSI), and an Aerosol Chemical Speciation Monitor (ACSM; Aerodyne Research Inc.). The ACSM was used to measure mass concentrations of particulate ammonium, nitrate, sulphate, chloride, and organic species in the submicron size range. Mass calibration was obtained by sampling mono disperse ammonium nitrate and ammonium sulphate. The instrument collection efficiency was calculated to be 1 during BUNIAACIC, through the comparison of the mass concentration of species measured by the ACSM and MAAP (black carbon equivalent; BC_e) with the integrated mass of the SMPS. Further instrumental details and data post-processing is given by Brito et al. (2014) ~~.~~ and Stern et al. (in preparation). A weather station (Davis, USA) at the top of the tower provided meteorological data (wind speed and direction, temperature, RH, etc.).

As well as the instruments in the container, a Wide Issue Bioaerosol Sensor (WIBS; model 3M, University of Hertfordshire) was operated in a weatherproof box on the ground, a short distance from the base of the tower, with a short (1 m) 1/4" OD stainless steel inlet (more details are provided below). Other core instruments running at the site, but not used in this study, are detailed by Artaxo et al. (2013).

2.2 HTDMA measurements

In the HTDMA (Cubison et al., 2005; Good et al., 2010), a dry aerosol sample is mobility size-selected with the first DMA and then humidified to a set RH. The second DMA is then used to measure the size distribution of the humidified aerosol, to give the distribution of Growth Factor (defined as the ratio of humidified to dry aerosol diameter: D/D_0) as a function of RH and dry diameter (GF_{RH,D_0}). Quality assurance and inversion of the data was performed using the TDMAinv toolkit of Gysel et al. (2009). During normal operation, the first DMA cycled through 5 ~~dry~~ mobility sizes (45 nm, 69 nm, 102 nm, 154 nm and 269 nm; calibrated values), and the monodisperse flow after the first DMA was humidified to a target RH of 90%. The RH measured in DMA2 remained fairly stable ($\pm$2%) for most of the measurement period, and the variation was accounted

for by correcting the data to the target RH within the inversion toolkit (Gysel et al., 2009). In addition to this normal mode of operation, humidograms were run on the 21st and 23rd July. In this mode, cycling through 3 dry sizes (45 nm, 102 nm and 269 nm), the RH in the second DMA was gradually varied between 45% and 95% in order to determine how the GF of ambient aerosol varies with RH.

In both DMAs, a ratio of 10:1 was maintained between the sheath and sample flows, and these were calibrated using ~~a bubble flowmeter (Gillibrator~~an airflow calibrator (Gillibrator-2, Sensidyne). The first DMA was size calibrated at the start of measurements using NIST-traceable polystyrene latex spheres (PSL; Fisher Scientific), sizes 100, 150, 200 and 300 nm, nebulised with an aerosol generator (model ATM 226; TOPAS). Dry scans (in which the sample is not humidified between the DMAs) were run on an approximately weekly basis in order to monitor the size offset between the two DMAs and to define the width of the DMA transfer functions (Gysel et al., 2009). The HTDMA was further verified by sampling nebulised ammonium sulphate, monitoring the growth factors for a range of RH (68% to 92%) at a given size (140 nm), and comparing to modelled values (ADDEM; Topping et al., 2005). More details of the calibration procedures for this instrument are given by Good et al. (2010).

2.3 CCNc measurements

The CCNc (Roberts and Nenes, 2005) operated downstream of a DMA (model 3081, TSI), the voltage of which was controlled with a classifier (TSI, model 3080) stepping discretely through a mobility size range 16 nm to 325 nm. This quasi-monodisperse aerosol sample flow was then split isokinetically between the CCNc and a CPC (TSI, model 3010). The flow into the CPC was further diluted with filtered air by a factor of 2 in order to match the flow into the CCNc. Inside the CCNc, the aerosol flowed through a wetted column with a temperature gradient, providing supersaturated conditions in which a proportion of the particles activated and were detected by an Optical Particle Counter (OPC) at the bottom of the column. Throughout the deployment, the CCNc cycled through 5 calibrated supersaturation setpoints: 0.15%, 0.26%, 0.47%, 0.80% and 1.13%. The ratio of activated particles to total particles (measured by the CPC), can be determined as a function of dry particle diameter and supersaturation (the activated fraction: AF). By fitting a sigmoid curve function to this activation spectrum, the dry diameter at which 50% of particles activate (D_{50}) was derived. The hygroscopicity parameter, κ (Petters and Kreidenweis, 2007), was then derived from D_{50} and supersaturation using the κ-Köhler model. In addition, the total number of CCN (N_{CCN}) was calculated by integrating the number size distribution above D_{50}.

~~As with the HTDMA, the~~ The DMA was calibrated using ~~nebulised latex spheres~~PSLs of the same sizes as with the HTDMA. The CCNc was calibrated by flowing nebulised ammonium sulphate into the system and determining the supersaturation at which 50% of the particles of a given dry size activate. This critical supersaturation is then compared to modelled values (ADDEM; Topping et al., 2005) to determine the slope and offset.

2.4 Bio-aerosol measurements

Fluorescent Biological Aerosol Particles (FBAP) in the optical size range $0.5 \leq D_p \leq 20$ µm were detected using the WIBS-3M (Kaye et al., 2005; Foot et al., 2008; Stanley et al., 2011), which operates on the principle of ultraviolet light induced

fluorescence of molecules common to most biological material, specifically Tryptophan and the co-enzyme NADH. Two sequential pulses of UV light are provided by filtered Xenon lamps at 280 nm and 370 nm to excite Tryptophan and NADH, respectively. Fluorescence is then detected in the ranges 310–400 nm and 400–600 nm following the Tryptophan excitation, and 400–600 nm following the NADH excitation (i.e. 3 fluorescence channels~~).~~ ; FL1, FL2, and FL3, respectively). In addition, the WIBS-3M provides a dimensionless particle assymmetry factor (A_f) as a proxy for particle morphology, as detailed by Crawford et al. (2015). Particles smaller than 0.8µm were rejected from analysis due to low counting efficiency.

The baseline fluorescence of the instrument is measured during so-called forced trigger (FT) sampling periods, where the instrument triggers the flash lamps and records the resultant fluorescence in the absence of aerosol in the sample volume. The mean fluorescence in a FT period is treated as the baseline fluorescence of the optical chamber during the sample period. For a particle to be considered fluorescent (FBAP) it must exhibit a fluorescence greater than a threshold value, defined as the baseline fluorescence plus 3 standard deviations, in any channel. During data processing the threshold value for each channel is subtracted from the single particle fluorescence data and the value is clipped at zero with all values greater than zero being considered significantly fluorescent compared to the instrument baseline. All reported fluorescence measurements are relative to the applied threshold and not the absolute detector intensities. This is consistent with previous studies using this instrument (Robinson et al., 2013; Crawford et al., 2014, 2015, 2016), and a detailed description of this data processing method is provided by Crawford et al. (2015). The thresholds remained consistent over 58 FT periods throughout the measurements at: 112.4 ± 3.9 for channel FL1, 284.6 ± 7.8 for FL2, and 164.6 ± 5.7 for FL3. The ambient threshold determination method (Perring et al., 2015) was not used here due to the majority of particles being fluorescent in nature.

Size calibration of the WIBS-3M consisted of using ~~latex spheres~~ PSLs with a physical diameter of 1.0 µm. Blue fluorescent latex spheres (1.0 µm diameter; Thermo Scientific) were also used to ~~monitor the instrument fluorescent channel efficiencies and baseline with time~~ensure that the excitation and fluorescence channels were operating correctly. The WIBS-3M inlet was operated at a total flow rate of 2.3 lpm (±5%). 90% of this was directed through a HEPA filter and used as a sheath flow, constraining the remaining 0.23 lpm for the scattering chamber sample flow from which particle concentrations were derived.

Particles ~~detected by this instrument~~ with fluorescent magnitudes above the threshold are termed FBAP, as they represent a lower limit of PBAP, some of which ~~will not necessarily~~ may not be detected by this method ~~(Gabey et al., 2010; Huffman et al., 2012).~~ if their fluorescence goes undetected, or they simply don't fluoresce (Gabey et al., 2010; Huffman et al., 2012). Non-biological fluorescent material can also be detected by the WIBS should its excitation and emission profile match that of the instrument. Generally the identified interferents are smaller than the detection limit of the instrument. Polycyclic aromatic hydrocarbons (PAH) such as naphthalene, and soot containing PAHs have been shown to fluoresce in FL1 (Pöhlker et al., 2012; Toprak and Schnaiter, 201 however they would not be expected to be seen in significant concentrations outside of the pollution events at such a remote site. Mineral dusts contain a small subset of fluorescent aerosol within their population (≈10%), and given their ubiquitous nature may present a significant source of interferents to the UV-LIF method (Toprak and Schnaiter, 2013). Their observed fluorescent intensity, however, is considerably weaker than is observed for biofluorophores (Pöhlker et al., 2012), and if they were present in any significant concentrations they would likely form their own cluster in the cluster analysis discussed in

section 3.4 (Crawford et al., 2016). It should also be noted that the technique does not distinguish between biological particles and fluorescent material attached to non-biological particles (e.g. dust).

This instrument has previously been deployed in Borneo, and further details of its operation are given by Gabey et al. (2010). In this experiment, the instrument was positioned in a small clearing, a few metres away from the rainforest understorey. It should be noted that the WIBS-3M measurements only ran until the 10th July, and so did not overlap with the other principle measurements (HTDMA, CCNc, ACSM), which began on the 10th July. Meteorological conditions were fairly consistent over the whole measurement period, and so all measurements discussed here are considered representative of the same general period (i.e. the transition from wet to dry season).

2.5 Removal of pollution episodes

In order to focus on the natural (biogenic) aerosol, for comparison with the wet season, it was necessary to exclude periods affected by pollution. While the site is described as pristine, it can nevertheless be affected by local emissions and regional transport of pollutants: biomass burning emissions from outside the reserve; the urban plume from Manaus; and pollution from the nearby diesel generator.

For each day of the campaign 7-day back trajectories were calculated using the HYSPLIT model (Draxler and Hess, 1998) at 30 minute intervals and 6 altitudes above TT34 (0, 250, 500, 1000, 2000 and 4000 m.a.s.l). The horizontal and vertical wind fields employed here were from the NCEP/NOAA 1°x 1°Global Data Assimilation System (GDAS) reanalysis product. These back trajectories were used to identify air masses arriving at TT34 which had either passed over Manaus or passed nearby active fire zones. The results were largely the same between 0 and 2000 m, with very little influence from the upper level flow at 4000 m.

A bounding box was drawn between -3.16°to -2.88°~~longitude~~ latitude and -60.12°to -59.81°longitude to define the Manaus influence zone (see Fig. 1), and any back trajectory passing over this box at any altitude up to 2000 m was flagged.

Air masses potentially impacted by biomass burning were identified by coupling the back trajectory measurements to satellite detected fires as measured by the MODIS instrument. This operates on the Aqua and Terra satellites, which have local overpass times in the morning and afternoon respectively. The fire detection data (specific product: MCD14ML) was produced by the University of Maryland and acquired from the online Fire Information for Resource Management System (FIRMS; https://earthdata.nasa.gov/data/near-real-time-data/firms/about). At each location along the back trajectories the surrounding 1°box was interrogated for any fire counts at the nearest terra/aqua overpass. If any were present this trajectory was flagged as potentially influenced by biomass burning. This technique is subject to uncertainties associated with trajectory errors (e.g. Fleming et al., 2012) and the detection of fires in cloudy scenes, or false detection of fires (Schroeder et al., 2008), and therefore can only be considered qualitative.

Finally, data were ~~flagged~~ investigated for possible contamination from the generator if the local wind direction was in the range 270°- 340°, however there were no instances of generator contamination during the measurement periods of this study.

In the event of any flag, the black carbon data (from the MAAP) were checked along with the particle counts (where available), and aerosol data were excluded if the pollution flag coincided with a significant increase in these concentrations.

No other increases in black carbon concentrations were seen outside the flagged periods. Approximately 28% of the HTDMA and CCNc data were removed in this way, with 5% of the data being flagged as possibly impacted by biomass burning and most of the rest due to the Manaus urban plume. The ACSM, which was not necessarily operating at the same times as the HTDMA and CCNc, had approximately 9% of its data removed due to pollution flags (almost entirely due to the urban plume from Manaus).

3 Results and Discussion

3.1 Size distributions

The particle number size distribution recorded over the measurement period of this study can be seen in ~~fig~~Fig. 2. This shows a broad accumulation mode peak at 130 - 150 nm with a median number concentration of 266 cm^{-3} (calculated from the integral of the size distribution curve). Despite observing aerosol number concentrations comparable to previous observations during the wet season, the shape of the distribution resembles those measured in the dry season, although the concentrations during the latter are considerably higher at 2200 cm^{-3} Artaxo et al. (2013).

The size distribution, however, was quite variable over the period of the measurements, as can be seen in the time-series in ~~fig~~Fig. 3, and varied with total particle ~~counts~~number concentrations. Median size distributions observed when the total number concentration was above or below 200 cm^{-3} are shown in ~~fig~~Fig. 4. During periods of low particle counts, an Aitken mode is also seen, with a mode around 80 nm, while the size distribution during episodes of higher concentrations is dominated by the accumulation mode, possibly masking the smaller mode. Such a size distribution profile, dominated by accumulation mode aerosols, has also been reported during the dry season in western Amazonia, in the deforestation arc, during biomass burning events (Brito et al., 2014), albeit with substantially higher concentrations.

3.2 Composition

Submicron non-refractory aerosol composition, as measured by the ACSM during the period of this study, is illustrated in ~~fig~~Fig. 5. The mean mass loadings for organic material, sulphate and nitrate were 2.13 $\pm$ 0.75 µg m^{-3}, 0.11 $\pm$ 0.04 µg m^{-3}, 0.08 $\pm$ 0.03 µg m^{-3}, respectively ($\pm$ 1 standard deviation). Organic material dominated the submicron aerosol, comprising around ~~86~~81% of the total mass (86% of non-refractory material), on average. Such a high fraction of organics compares well with previous observations in the Amazon basin (Artaxo et al., 2013; Brito et al., 2014; Andreae et al., 2015). BC_e concentrations are also shown, with a mean mass loading of 0.25 $\pm$ 0.01 µg m^{-3}. This is consistent with previous wet season measurements in the Amazon (Artaxo et al., 2013; Andreae et al., 2015).

The mass fractions of non-refractory aerosol and BC_e are shown in the bottom panel of Fig. 5. Due to the noise in the ammonium signal (see Fig. 5), resulting from concentrations below the limit of detection of 0.3 µg m^{-3} for the ACSM, it was necessary to estimate the ammonium from the nitrate and sulphate mass loadings for the purpose of mass fraction calculations. The time-series of mass fractions show that, while the mass loadings vary considerably, particularly organics, the composition

is relatively consistent as a proportion of the aerosol mass. Organic mass fractions remain steady around 81% of the total mass, until the 22nd and 23rd July, when a slight increase in BC_e is seen.

Levoglucosan, a major constituent of biomass burning aerosol, fragments in AMS and ACSM instruments at a mass-to-charge ratio (m/z) of 60 (Alfarra et al., 2007), and so the fraction f_{60} is frequently used as a marker for biomass burning (Artaxo et al., 2013; Chen et al., 2009). The mean f_{60} ~~at TT34 in July 2013~~ from the ACSM data in this study, after removal of pollution episodes, was 0.19% $\pm$ 0.07%. This is well below 0.3%, which is considered to be the upper limit for background air masses not affected by biomass burning (Cubison et al., 2011). It should be noted that previous studies in the Amazon have observed that a large fraction of the biomass-burning related organic aerosols do not present a significant f60 signal, due to long-range transport (Brito et al., 2014). It can be said, however that the relatively low f_{60} observed here suggests that, ~~indicating that ,~~ on average, these measurements were not strongly impacted by local biomass burning emissions.

Previous studies have successfully identified FPAB markers on ambient aerosol in the Amazon using an aerosol mass spectrometer (Schneider et al., 2011), a method which relies strongly on the high-resolution capabilities of the instrument used at the time. Given the unity mass resolution of the ACSM, similar methodology has not been applied here.

3.3 Aerosol water uptake

The HTDMA ran from the 13th to the 28th July. Figure 6 shows the time-series of RH-corrected GF distributions for all dry sizes, as derived from the HTDMA data using the TDMAinv toolkit. These largely exhibit a single mode at each size, ~~which varied little over the period of measurements,~~ roughly in the range of 1.2 ~~–~~ to 1.4. Some variability can be seen, for example on the 21st and 23rd July, but for the most part, peak growth factors remained relatively stable over the measurement period. This is consistent with the stable mass fractions seen in the composition data from the ACSM (see Fig. 5, bottom panel). The variability and slight decrease in GF at some sizes on the 22nd and 23rd July may also be attributed to the slight increase in the mass fraction of BC_e (Fig. 5). High peak growth factors (> 1.6) can briefly be seen on the night of the 15th July, shortly before a pollution event, however without composition data available on that day (Fig. 5), it is difficult to speculate as to the nature of this.

Smaller, more hygroscopic (GF > 1.5) modes can be seen at the lower dry diameters, while the larger particles also show a hydrophobic mode in the growth factor distribution. The contribution of the hydrophobic mode to the larger particles is small (~~<~~ < 10% in number) and may be due to ~~a~~ some unknown local anthropogenic influence that was not accounted for. The averages of the growth factor at the peak of ~~this~~ the growth factor distribution (i.e. the dominant mode) are shown in table 1 and ~~fig~~Fig. 2. They show an increase with dry diameter, reflecting the difference between Aitken and accumulation mode aerosol: organic mass fractions are highest in the Aitken mode, while elevated sulphate mass fractions have been previously seen in the accumulation mode (Gunthe et al., 2009; Pöschl et al., 2010). It should be noted, however, that the elevated sulphate events observed by Gunthe et al. (2009) were likely linked to long-range transport of biomass burning aerosol from Africa, which, due to a combination the African burning season and large scale circulation, tends to impact the Amazon forest more often during the wet season (Ben-Ami et al., 2010).

The campaign averages of the CCNc derived parameters, D_{50}, κ and N_{CCN} are given for each set supersaturation in table 2. The κ values are also plotted against D_{50} in ~~fig~~Fig. 2. Consistent with the growth factor data, and with previous measurements at this site (Gunthe et al., 2009), they show more hygroscopic particles at larger diameters ($\kappa \approx 0.12$ below 100 nm, and $\kappa \approx 0.18$ around the accumulation mode).

Reconciliation between sub- and super-saturated particle water uptake for these measurements has already been investigated by Whitehead et al. (2014). They showed that there was agreement within the variability of the data, with a slightly underestimated hygroscopicity from the HTDMA data compared to the CCNc at lower supersaturations (larger dry diameters). The analysis of Whitehead et al. (2014) considered the full dataset without separating out the pollution events, however performing the same analysis on the 'clean' data did not result in any significant difference.

3.4 FBAP measurements

Measurements of biological particles in the Amazon are important as they are considered to have a strong influence on clouds as ice nuclei (Pöschl et al., 2010). The ~~WIBS-3~~ WIBS-3M operated uninterrupted from the morning of the 3rd July until 10th July. The mean ~~total~~ particle number concentration ~~of FBAP~~ measured by the ~~WIBS-3~~ WIBS-3M during this period was ~~475~~ 464 $\pm$ ~~244~~ 250 l^{-1} (1 standard deviation), while the mean FBAP number concentration was ~~404~~ 400 $\pm$ ~~237~~ 242 l^{-1} (i.e. accounting for ~~85~~86% of the particles in the size range of the instrument). The time-series of number concentrations for the duration of this period is shown in ~~fig~~Fig. 7. This shows coarse mode particles were dominated by FBAP number concentrations, which exhibited a strong diurnal cycle with concentrations varying from around 200 l^{-1} during the daytime up to as much as 1200 l^{-1} at night. The diurnal variation (~~fig~~Fig. 8) shows that FBAP number concentrations plateaued from around 21:00 through the night, began to drop from 05:00, reached a minimum by 11:00 and started increasing again from 15:00. The FBAP fraction was highest (more than 90%) at night, and remained high until around 08:00 - even after FBAP number concentrations began decreasing. This dropped to between 55% and 75% during the day, helped in part by an apparent increase in non-FBAP concentrations, before steadily increasing in line with the FBAP concentrations through the late afternoon / early evening.

There are a number of factors driving the diurnal cycle in coarse mode particles, as discussed by Huffman et al. (2012). Previous studies at this and a nearby site, utilizing electron and light microscopy, have identified the FBAP as predominantly fungal spores (Graham, 2003; Huffman et al., 2012). Similar diurnal cycles have been seen in airborne fungal spore densities at other tropical rainforest locations (Gilbert and Reynolds, 2005; Elbert et al., 2007). The observed night-time peak in FBAP number concentrations in ~~fig~~Fig. 8 is consistent with nocturnal sporulation driven by increasing RH (see bottom panel; note that RH is measured above the canopy). The dependence of fungal spore release on meteorological conditions, however, varies greatly according to species, and any relationship is non-trivial (Jones and Harrison, 2004). FBAP number concentrations begin dropping several hours before any decrease in RH, and the FBAP fraction also remains high (~~fig~~Fig. 8). This suggests that the morning decrease in FBAP is not necessarily due to a cessation of emission processes, but may also be the result of a break-up of the nocturnal boundary layer around sunrise (Whitehead et al., 2010; Huffman et al., 2012). Graham (2003) and Huffman et al. (2012) suggest that the night-time increase in coarse mode particles is due, at least in part, to the shallow nocturnal boundary layer. The slight increase in non-FBAP concentrations during the day may be a result of enhanced particle exchange

through the canopy, facilitated by sporadic turbulent events, as described by Whitehead et al. (2010), bringing non-FBAP that had originated elsewhere into the space below canopy.

Figure 9 shows the number size distributions reported by the ~~WIBS-3~~ WIBS-3M during the measurement period. Again, FBAP clearly dominates the particle number concentrations for $D_p > 1$ µm, however non-FBAP concentrations are higher for ~~submicron particles .~~ particles smaller than 1 µm measured by the WIBS-3M (i.e. down to the instruments 50% detection diameter of 0.8 µm). The FBAP number size distribution shows a peak at around 1.8 µm, while the non-FBAP distribution is characterized by a flatter, broader peak between 0.8 and 1.3 µm. Non-fluorescent particles at this site have previously been identified as mineral dust, non-fluorescent biological aerosol, and inorganic salts (Huffman et al., 2012). Caution must be applied when interpreting the sub-micron fluorescent aerosol fraction due to the reduced fluorescent counting efficiency for particles $D_p < 0.8$ µm (Gabey et al., 2011), which may lead to an underestimation of the fluorescent aerosol fraction at small sizes.

A Ward linkage cluster analysis using the z-score normalisation was applied to the data, ~~as described by Crawford et al. (2015),~~ where the optimum number of retained distinct clusters was determined using the Calinski–Harabasz criterion. Prior to analysis, all non-fluorescent and saturated particles, and particles smaller than 0.8 µm in diameter were excluded, resulting in approximately 15% of the single particle data being rejected. The asymmetry factor and size inputs were converted to log space prior to normalisation and clustering. Complete information on this technique is given by Crawford et al. (2015). This analysis revealed three distinct fluorescent classes of particles (Cl1-3). ~~Cl1 has previously been attributed to fungal spores (Crawford et al., 2014) based on comparison with other sampling techniques and the diurnal emission pattern (see fig. 8) with higher concentrations observed overnight.~~ The statistical parameters of each cluster are shown in table 3. It can be seen that CL1 and Cl2 ~~appears to be a distinct sub-class of somewhat less fluorescent particles which remain unclassified, but shows similar behaviour to, and correlates~~ display similar characteristics; specifically, they mainly fluoresce in FL1 with weak fluorescence in the other channels, although the intensities are greater for Cl1, suggesting they are distict sub-classes. The two clusters correlate strongly ($r^2 = 0.86$) with ~~Cl1~~each other, hence both have been combined in ~~fig~~Fig. 8. ~~Both clusters~~ They show similar fluorescent signatures to the clusters attributed to fungal spores by Crawford et al. (2014, 2015) ~~. The statistical parameters of each cluster are shown in table 3 for comparison . Together, these clusters~~ based on comparison with other sampling techniques and the diurnal emission pattern. In this study, they show higher concentration overnight (Fig. 8), and a strong correlation to RH (Fig. 10. Together, clusters Cl1 and Cl2 contribute approximately 70% to the total fluorescent particle concentration, ~~with no significant diurnal variation in this figure~~regardless of time of day, suggesting that FBAP were dominated by fungal spores during this study. A third cluster, Cl3, shows very low concentrations (around 20 l^{-1}), with no strong diurnal trend, however there is insufficient data to speculate upon the nature of this cluster (such as response to rainfall).

PBAP classification via the comparison of single particle fluorescent signatures to laboratory samples is an ongoing area of research (e.g. Hernandez et al., 2016). Such direct comparison for this purpose is not possible here due to differences in the instruments used (i.e., different excitation/detection wavebands and optical chamber design). Even comparing results between the same model of instrument with identical detector/filter configurations has been difficult (Hernandez et al., 2016) due to the current lack of a robust fluorescence calibration method.

3.5 Comparison with previous studies

3.5.1 Submicron aerosol

Aerosol water-uptake studies have previously been conducted at the TT34 site by Gunthe et al. (2009) using size-selected CCNc measurements, and at Balbina (110 km NE of TT34) by Zhou et al. (2002) using a HTDMA, both during the wet season. HTDMA and CCNc measurements were also made at Balbina during the transition from wet to dry season by Rissler et al. (2004). In addition, HTDMA measurements from pasture-land in SW Amazonia at the end of the dry season / beginning of wet season are presented by Rissler et al. (2006) and Vestin et al. (2007). This study represents the first measurements with HTDMA and monodisperse CCN instruments at TT34 during the transition from wet to dry seasons. Concurrent CCNc and HTDMA measurements have also been conducted in Borneo, SE Asia, by Irwin et al. (2011), providing a useful comparison with a different tropical rainforest region.

The HTDMA growth factor measurements of Zhou et al. (2002) showed a similar pattern to this study: a dominant mode of "less hygroscopic" particles (GF $\approx$ 1.16 - 1.32), accompanied at times by a hydrophobic mode (GF < 1.06; particularly at the larger particle sizes), and a more hygroscopic mode (GF $\approx$ 1.38 - 1.54). The growth factors of the less hygroscopic particles are compared in ~~fig~~Fig. 11, along with the other studies (note that Rissler et al. (2004) define "less hygroscopic" as GF $\approx$ 1.17 - 1.5). All the measurements showed a similar increase in growth factor with dry diameter. The growth factor values from this study were slightly higher than those of Zhou et al. (2002) and Rissler et al. (2004), but the difference is within the variability of the measurements, and probably within the variability that has been seen between different HTDMA instruments (Duplissy et al., 2009; Massling et al., 2011). The "moderately hygroscopic" particles (GF = 1.26) observed by Rissler et al. (2006) exhibited growth factors in the same range as the other studies in Amazonia, however in this case, the hydrophobic mode (GF $\approx$ 1.05 - 1.13) was dominant for all but the larger particles ($>$ 135 nm). Furthermore, strong diurnal cycles (daytime increases in the fraction of moderately hygroscopic particles) were observed (Rissler et al., 2006; Vestin et al., 2007), which were not seen during the current study. In contrast to the current study, the measurements of Rissler et al. (2006) and Vestin et al. (2007) were conducted in a region that has undergone heavy land use change and is strongly influenced by anthropogenic sources (Andreae et al., 2002), which may contribute to the observed diurnal pattern.

In contrast to the studies from Amazonia, aerosol growth factors measured in Borneo (Irwin et al., 2011) were somewhat higher: in the range 1.3 - 1.7 (~~fig~~Fig. 11). This can be explained by the fact that, while the site in Amazon benefited from a fetch of hundreds of kilometres of undisturbed rainforest, the site in Borneo was heavily influenced by marine air masses (Robinson et al., 2011). As discussed by Robinson et al. (2011), the sulphate loadings in Borneo were substantially higher than in Amazonia, even in air masses from across the island, which, with sulphate being more hygroscopic than organic aerosol, ~~would explain~~ is a possible explanation for the higher growth factors.

The results of the humidogram are shown in ~~fig~~Fig. 12, and compared to the humidogram data from Borneo (Irwin et al., 2011) and the humidogram fit for the wet season data of Rissler et al. (2006). Growth factors in Borneo were higher across the RH range than in Amazonia. As with previous measurements, no deliquesence behaviour was seen in this study.

Values of κ derived from the HTDMA and CCNc measurements during these studies are compared in ~~fig~~Fig. 13, as a function of dry diameter. Here, the κ from HTDMA measurements is derived using the average growth factor, rather than the peak of the less hygroscopic mode, for direct comparison with the CCNc derived values. The various measurements in Amazonia showed very similar κ, largely agreeing within the variability of the individual measurements. It can be said that water uptake measurements in Amazonia are consistent, and, as noted by Gunthe et al. (2009), show κ to be around half that typically seen in other continental regions (Andreae and Rosenfeld, 2008).

The HTDMA derived κ from the Borneo experiment shows more hygroscopic aerosol than in Amazonia, as discussed above, however the CCNc derived values are more in line with those in Amazonia. This discrepancy has been noted previously and possible reasons for it discussed by Irwin et al. (2011) and Whitehead et al. (2014). These were mainly related to differences in the instrument setups and how they treat the aerosol. It should be noted that the discrepancy in the data from the Borneo experiment was the largest amongst a number of datasets studied by Whitehead et al. (2014), but the reason for this is not clearly understood.

In general, the particle concentrations and hygroscopic properties observed during this study were similar to those seen during wet season measurements in the Amazon rainforest. The main difference seen was that size distributions in this study were more strongly dominated by the accumulation mode: similar to those seen in the dry season (Artaxo et al., 2013), but in clean conditions with significantly lower number concentrations. Under these conditions, cloud droplet formation in convective clouds in this region is likely to be aerosol- limited (Reutter et al., 2009). Previous modelling studies have suggested this is the case during the wet season (Pöschl et al., 2010), in contrast to the dry season during periods of intense biomass burning when droplet number is largely controlled by the updraft velocity (Reutter et al., 2009).

In terms of composition, submicron non-refractory aerosol concentration during this experiment showed significantly higher concentration (≈2.5 µg m^{-3}) than observed at the remote sites in Central Amazonia in previous years during the wet season, ranging from 0.4 µg m^{-3} (Artaxo et al., 2013) and 0.6 µg m^{-3} (Chen et al., 2009; Andreae et al., 2015). Conversely, the concentration is significantly lower than reported during the dry season (8.9 µg m^{-3}) (Andreae et al., 2015), due to this transitional period having no extensive biomass burning activities, albeit with already reduced wet deposition due to reduced precipitation. Interestingly, despite the marked changes in ambient concentration, very little difference is observed in terms of relative contributions considering this and previous studies, being strongly dominated by organics (≈80%), followed by sulphate and minor contribution of nitrate and ammonium (Chen et al., 2009; Artaxo et al., 2013; Andreae et al., 2015).

3.5.2 Coarse mode aerosol

Huffman et al. (2012) conducted measurements of FBAP at the TT34 tower using an ultraviolet aerodynamic particle sizer (UV-APS) during the AMAZE-08 campaign. In contrast to this study, the AMAZE-08 measurements were taken during the wet season (February to March), from the top of the tower (i.e. above canopy). It is also worth comparing with the measurements of Gabey et al. (2010), who used the same WIBS-3 instrument to sample the aerosol above and below canopy in the rainforest of north-east Borneo.

The median number concentration of FPAB observed below the canopy in ~~this study was 372~~ the current study was 363 l^{-1}, while the UV-APS measurements at the top of the tower by Huffman et al. (2012) were around a fifth of this, at 73 l^{-1} (also median). In an intercomparison between the two different measurement techniques, Healy et al. (2014) found that, while there was agreement in total number concentrations, the counts in the fluorescence channels of the WIBS (particularly FL1) were substantially higher than the UV-APS fluoresence counts, which would at least partly explain the difference here. ~~Meteorological conditions~~ The wetter, more humid conditions during the wet season measurement period of Huffman et al. (2012) would be expected to favour emission (Jones and Harrison, 2004; Zhang et al., 2015). On the other hand, the higher rainfall would also result in enhanced wet deposition during the wet season~~measurement period of Huffman et al. (2012)~~, especially above canopy. At other locations, Gabey et al. (2010) saw concentrations in Borneo often in excess of 1500 l^{-1} below canopy, and around 200 l^{-1} above, using the same instrument at each site, while Gilbert and Reynolds (2005) observed substantially higher concentrations of fungal spores in the understorey than in the canopy during measurements in Queensland, Australia. Strong vertical gradients in biological particles are therefore regularly seen in rainforest environments, and would be an additional factor in the differences observed between the measurements at TT34. In a remote tropical rainforest in China, Zhang et al. (2015) estimated fungal spore concentrations to be around 50 l^{-1} based on chemical analysis of filters, and found higher concentrations associated with rainfall events. A global modelling study by Spracklen and Heald (2014) found simulated surface annual mean concetrations of fungal spores to be around 100 l^{-1} over tropical forests (including Central Amazonia), which is consistent with this and other measurements at this site.

The fraction of FBAP in this study was, on average 85% of total coarse mode particles (and as much as 90%) whereas it was 24% in the AMAZE-08 campaign (41% in unpolluted conditions). The higher fraction at ground level would be expected, being closer to the source, whereas above canopy, there is a stronger influence from non-fluorescent particles from external sources. Elbert et al. (2007) found fungal spores accounted for 35% of coarse mode particles, also in Central Amazonia, but their filter samples were taken at a pasture site adjacent to the rainforest. In Borneo, as in the Amazon, there was a higher fraction below the canopy (55%) than above (28%), however not as high as the ~~85~~86% observed in this study. Reasons for this difference are unclear, but may include a stronger influence in Borneo of non-fluorescent particles from external sources, such as the nearby coast. More consistent with the current study were the scanning electron microscopy (SEM) measurements reported by Pöschl et al. (2010) and Huffman et al. (2012), which attributed 80% of coarse mode particles to primary biological aerosol during the AMAZE-08 campaign, and also identified particles likely to be fungal spores.

One difference between the measurements of this study and others is the position of the mode in the FBAP number size distribution. Gabey et al. (2010) report the peak at 2.5 µm, while Huffman et al. (2012) observe the peak around 2.3 µm. By contrast, the peak in this study was 1.8 µm. The difference between the two measurements at TT34 is likely due to the different measurement techniques, with the UV-APS found to be less sensitive to smaller fluorescent particles (Healy et al., 2014).

Diurnal variations between this study and that of Huffman et al. (2012) were similar, however Gabey et al. (2010) reported an additional increase in FBAP number concentrations in the afternoon in Borneo. This increase coincided with a peak in RH, and it is believed that this is linked (Gabey et al., 2010). In this study, the RH increased more gradually through the afternoon

and evening (see ~~fig~~Fig. 8, bottom panel), which may explain the lack of afternoon peak in FBAP compared to the Borneo results. Huffman et al. (2012) also ~~don't~~ do not observe a mid-afternoon peak in FBAP.

4 Conclusions

Measurements of aerosol concentrations and water uptake properties were conducted at a remote site in pristine Amazonian rainforest in July, 2013, during the transition from the wet to dry seasons. Back trajectories and wind sectors were examined in conjunction with black carbon concentrations in order to exclude any pollution episodes and ensure the aerosol measured were representative of background aerosol over the rainforest.

~~In the absence of polluted periods~~With any pollution episodes removed from the data, particle concentrations were low, with a median of 266 cm^{-3}. The particle size distributions were largely dominated by an accumulation mode around 130 - 150 nm, with a smaller Aitken mode apparent during periods of lower particle counts. Based on previous measurements contrasting wet and dry seasons (Artaxo et al., 2013), the results here may reflect the transition between the two seasons, with periods consistent with each at different times (but without considering any influence from biomass burning).

Aerosol chemical composition, as measured with an ACSM, was dominated by organic material, comprising around 81% of the total mass of non-refractory aerosol and BC_e. The mass fraction of organics was relatively consistent over the measurement period.

Aerosol water uptake and hygroscopicity was measured using an HTDMA and a CCNc. Good agreement was found between the measurements of both instruments. Particle growth factors from the HTDMA varied little over most of the measurement period and were typically between 1.2 and 1.4 (low hygroscopicity mode). Aerosol hygroscopicity was found to be low (κ = 0.12) for Aitken mode particles, and increased slightly to κ = 0.18 for accumulation mode particles. This is consistent with previous measurements at, or near this site, and with the observation that Aitken mode particle composition is dominated by organic material, while accumulation mode particles exhibited higher sulphate mass fractions (Pöschl et al., 2010).

Particles in the size range $0.5 \leq D_p \leq 20$ µm were measured using the WIBS-3M, which distinguishes fluorescent (representing a subset of primary biological aerosols, or FBAP) and non-fluorescent. FBAP dominated the coarse mode aerosol, accounting for as much as 90%. Concentrations of FBAP followed a strong diurnal cycle, with maximum concentrations during the night. This is likely driven by a combination of the dependence of emission processes on meteorological conditions and the diurnal cycle of the boundary layer.

The results from this study were also compared to measurements conducted in Borneo in 2008 (Irwin et al., 2011; Gabey et al., 2010; Robinson et al., 2011), contrasting the vast 'Green Ocean' of the Amazon rainforest to the island rainforest geography of SE Asia. In the submicron range, aerosol hygroscopicity was greater in Borneo, possibly due to the stronger marine influence of that region (Irwin et al., 2011). Coarse mode particles at both locations were dominated by FBAP (probably mostly fungal spores). Below canopy, the Amazon exhibited a higher fraction of FBAP than Borneo, though higher FBAP concentrations were seen at the latter.

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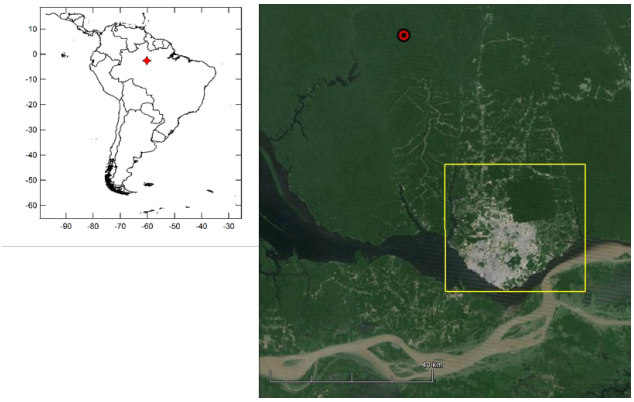

Figure 1. Location of the sampling site, shown as the red markers. The yellow rectangle represents the bounding box around Manaus used to flag air masses influenced by pollution from the city.

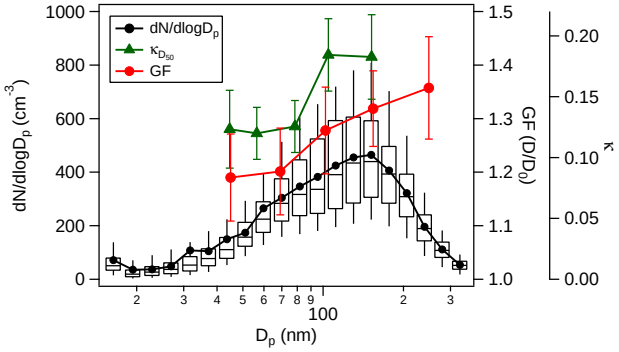

Figure 2. Particle number size distribution ~~for~~ averaged over the ~~experiment~~entire measurement campaign. Box-and-whisker plots ~~showing~~ show the median, interquartile ranges and 5th and 95th percentiles, and lines and markers ~~showing~~ show mean $dN/dlogD_P$. Also shown are κ derived from the D_{50} from the CCNc, and growth factor from the HTDMA, both as a function of particle diameter. Error bars represent $\pm$ 1 standard deviation. Note that the HTDMA and CCNc / particle size data have been averaged over slightly different measurement periods, as shown in Figs. 3 and 6.

~~The time-series of normalised RH-corrected (to 90%) growth factor distributions derived from HTDMA measurements, for all 5 dry diameters. Gaps are largely due to removal of data from pollution episodes and humidograms.~~

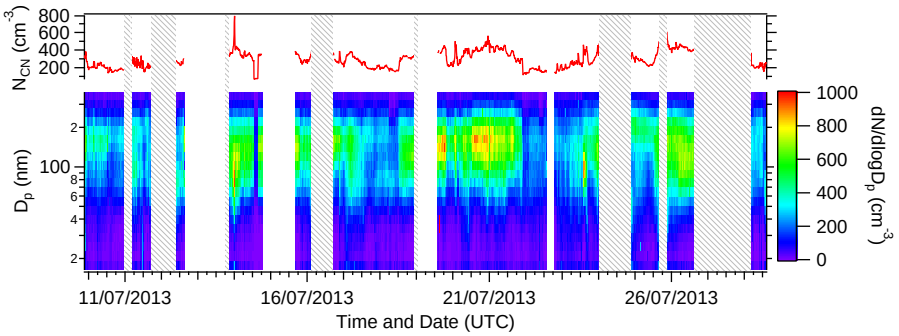

Figure 3. The time-series of total particle counts (integrated from size distributions; top panel) and particle number size distribution ~~and total counts~~(bottom panel). Shaded areas represent pollution episodes removed from the data. Any other gaps are due to instrument down-time.

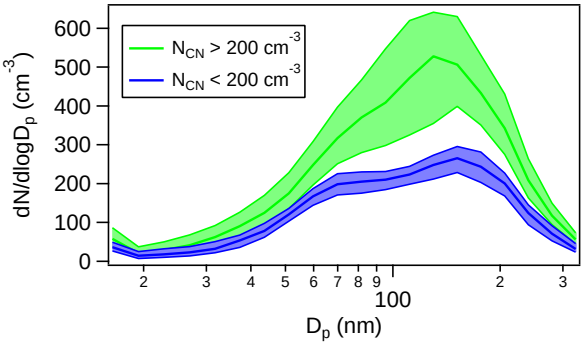

Figure 4. Median and interquartile ranges of particle number size distributions observed during high ($>$ 200 cm^{-3}) and low ($<$ 200 cm^{-3}) total particle number concentrations.

Table 1. Mean peak growth factors and derived κ from HTDMA measurements for each dry diameter, along with $\pm$ standard deviation.

D0 (nm)	GF	κ
45	1.19 ± 0.08	0.09 ± 0.10
69	1.20 ± 0.08	0.09 ± 0.09
102	1.28 ± 0.08	0.12 ± 0.10
154	1.32 ± 0.07	0.15 ± 0.09
249	1.36 ± 0.10	0.17 ± 0.09

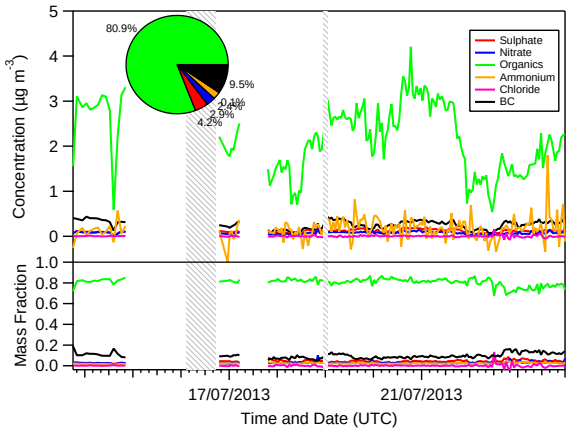

Figure 5. Submicron non-refractory aerosol composition from the ACSM measurements along with equivalent black carbon from the MAAP measurements: Concentration (top panel) and mass fraction (bottom panel). The pie chart shows the average proportions over the measurements. Shaded areas represent pollution episodes removed from the data. Any other gaps are due to instrument down-time.

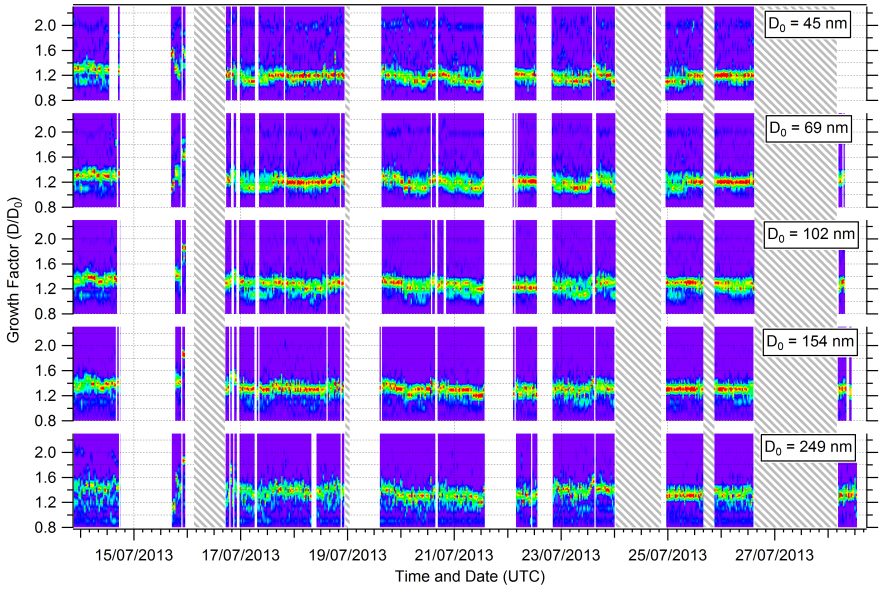

Figure 6. The time-series of normalised RH-corrected (to 90%) growth factor distributions derived from HTDMA measurements, for all 5 dry diameters. Shaded areas represent pollution episodes removed from the data. Any other gaps are due to instrument down-time and humidograms.

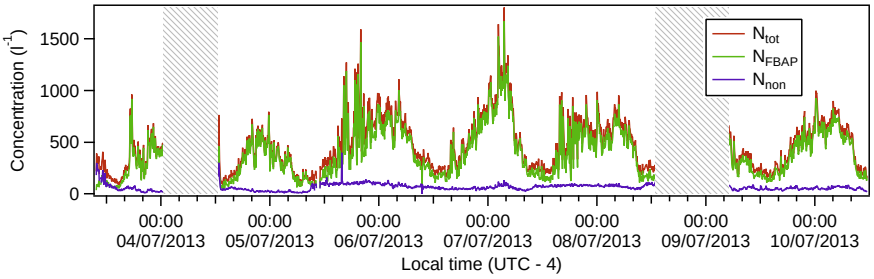

Figure 7. The time-series of total, FBAP and non-FBAP number concentrations as measured by the WIBS-3M. Shaded areas represent pollution episodes removed from the data. Any other gaps are due to instrument down-time.

Table 2. Mean derived parameters from CCNc measurements for each set supersaturation, along with ± standard deviation.

SS (%)	D_{50} (nm)	κ	N_{CCN} (cm^{-3})
0.15	152 ± 9.5	0.18 ± 0.03	87 ± 35
0.26	105 ± 5.5	0.18 ± 0.03	161 ± 60
0.47	78 ± 4.2	0.13 ± 0.02	212 ± 74
0.80	56 ± 3.0	0.12 ± 0.02	248 ± 82
1.13	45 ± 3.4	0.12 ± 0.03	268 ± 86

Table 3. Solutions to the Ward linkage cluster analysis, showing mean (± 1 standard deviation) intensity in each fluorescence channel (FL1 - 3), optical particle diameter (D_p) and assymmetry factor (A_f). The intensities are referenced to the FT + 3 standard deviation threshold representing an intensity of zero, as discussed is section 2.5. Fluorescent intensities and assymmetry factor are in arbitrary units.

	Cl1	Cl2	Cl3
FL1 (280 nm)	1400 ± 302	478 ± 386	386 ± 533
FL2 (280 nm)	120 ± 96	33 ± 47	351 ± 212
FL3 (370 nm)	94 ± 106	47 ± 73	721 ± 379
D_p(µm)	2.5 ± 1.3	1.9 ± 1.0	2.3 ± 1.1
A_f	30.9 ± 15.0	30.2 ± 15.7	29.0 ± 15.1

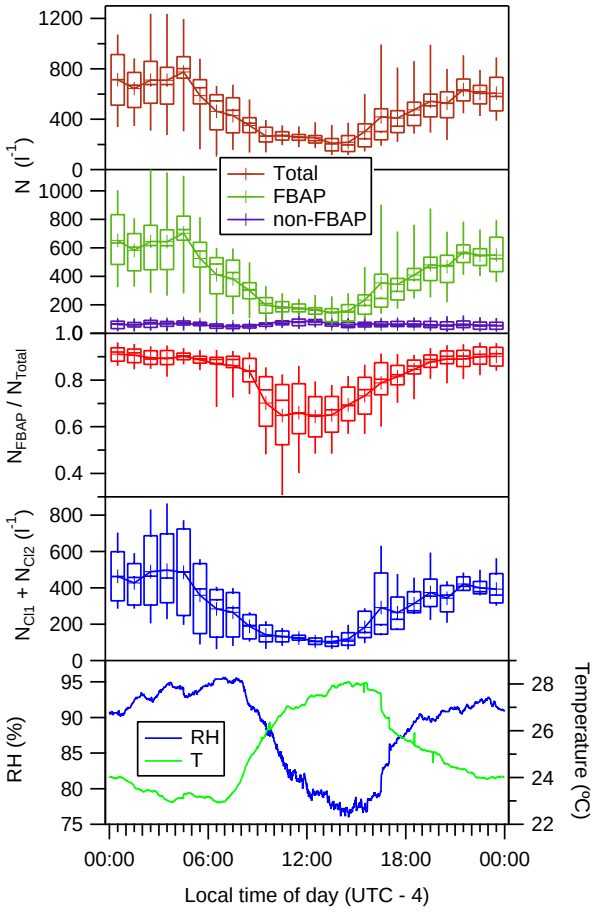

Figure 8. Diurnal variations in total, FBAP and non-FBAP number concentrations, as measured by the WIBS-3M, as well as the fraction of FBAP, and the combined number concentrations of clusters Cl1 and Cl2. Shown are the means (lines and markers), medians and inter-quartile ranges (boxes) and 5th and 95th percentiles (whiskers). Also shown at the bottom are the mean diurnal variations in temperature and RH, measured on the tower above the canopy, for the same period.

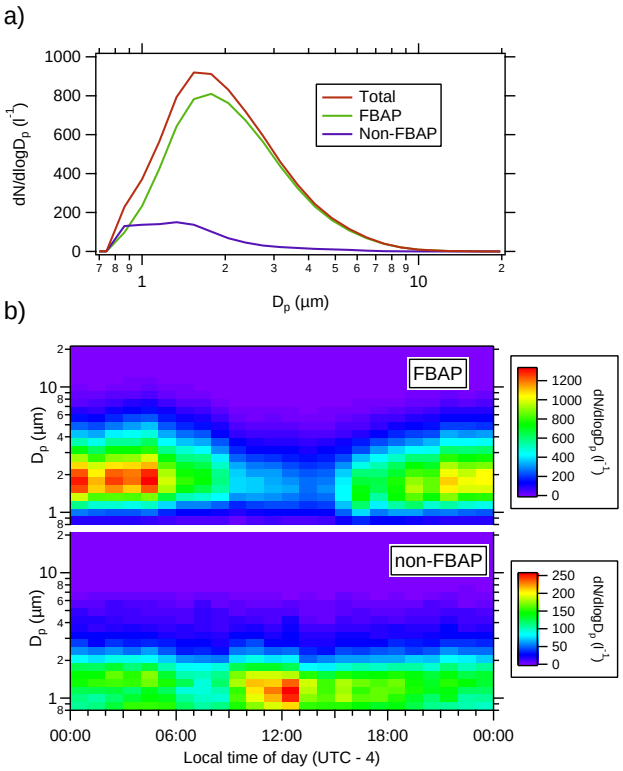

Figure 9. Particle number size distributions measured with the WIBS-3: a) mean size distributions for Total, FBAP and non-FBAP; and b) diurnal variation of size distribution for FBAP and non-FBAP (note that the colour scales are not the same).

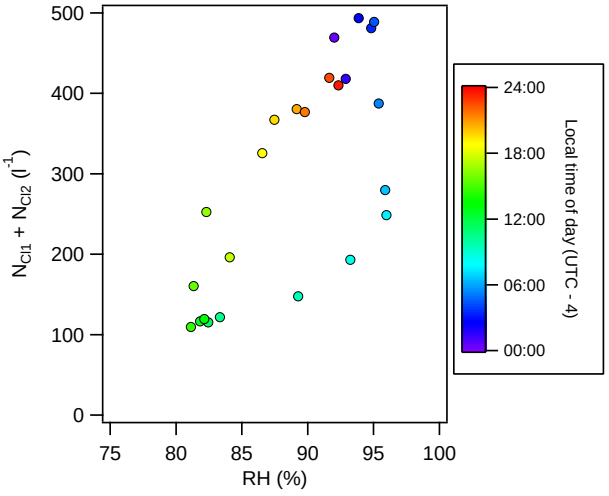

Figure 10. Diurnal means of total particle number concentrations in clusters 1 and 2, plotted against RH.

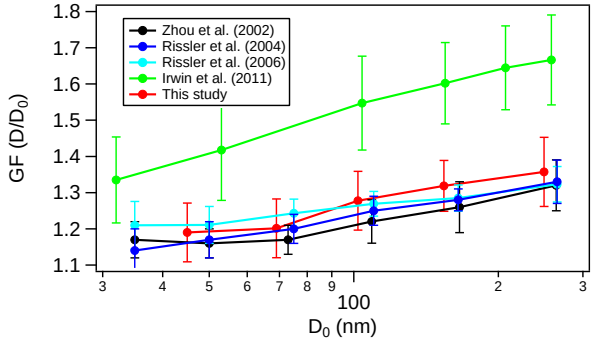

Figure 11. Mean growth factor for the dominant, less hygroscopic mode plotted ~~agains~~ against dry diameter, comparing this to previous studies in Amazonia and Borneo. The data from Rissler et al. (2004) and Rissler et al. (2006) represent "less" and "moderately hygroscopic" particles (respectively) during the wet season. The definitions differ slightly between the studies in terms of GF range, but the modes represented here broadly fit into the "less hygroscopic" classification of Swietlicki et al. (2008). Error bars represent ± 1 standard deviation.

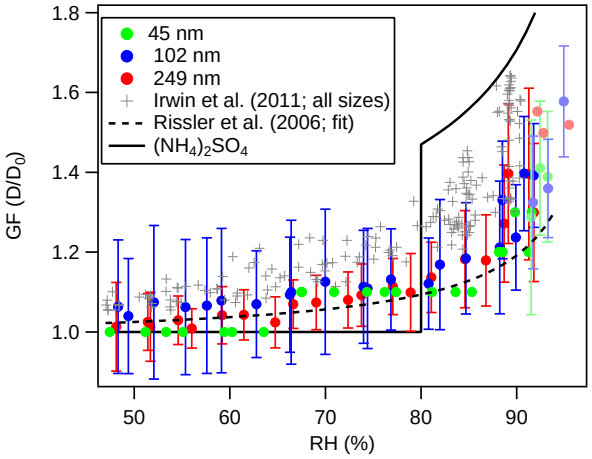

Figure 12. Humidogram (dependency of growth factor on RH), taken between 14:00 and 20:30 UTC on the 21st July. The fainter points at higher RH were taken between 13:30 and 14:30 UTC on the 23rd July. The humidogram data from Irwin et al. (2011), and the humidogram fit from Rissler et al. (2006) are also shown, for comparison. The black line shows the modelled humidogram for ammonium sulphate (Topping et al., 2005) for reference.

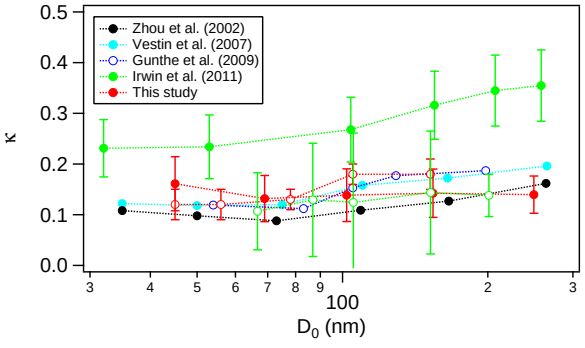

Figure 13. Comparing κ as a function of diameter for this and previous studies in Amazonia and Borneo. Filled circles represent HTDMA derived values, while empty circles are CCNc derived values. Error bars represent $\pm$ 1 standard deviation, where this data is available. The values for Zhou et al. (2002) and Vestin et al. (2007) were calculated by Gunthe et al. (2009).