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 As it currently stands, the paper lacks enough of this analysis, and is unclear enough in parts, that it should not be published.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.

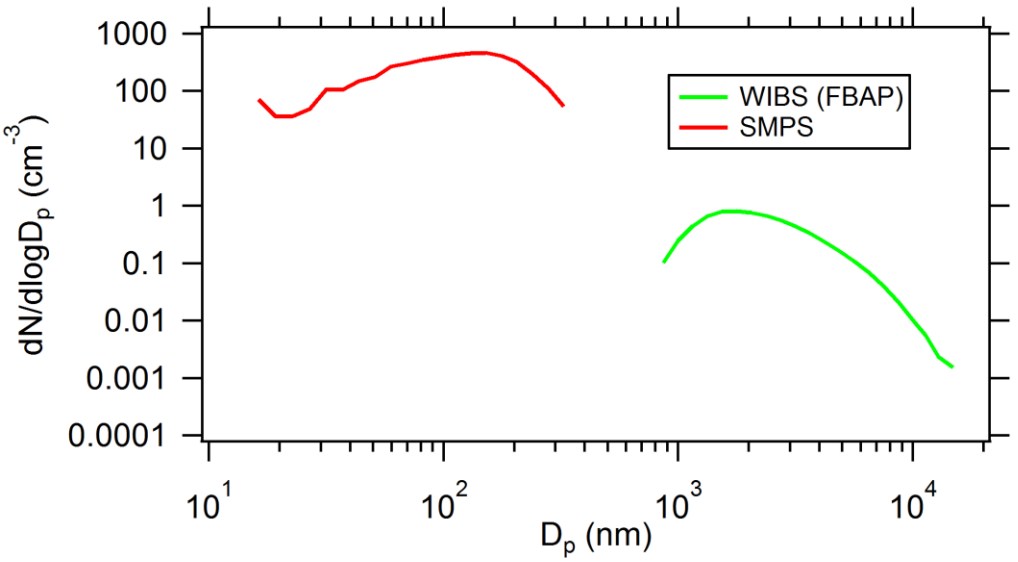

Figure 1. Combined log-log plot of total particle number size distribution (as measured with the SMPS) with FBAP number size distribution (from the WIBS).

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).

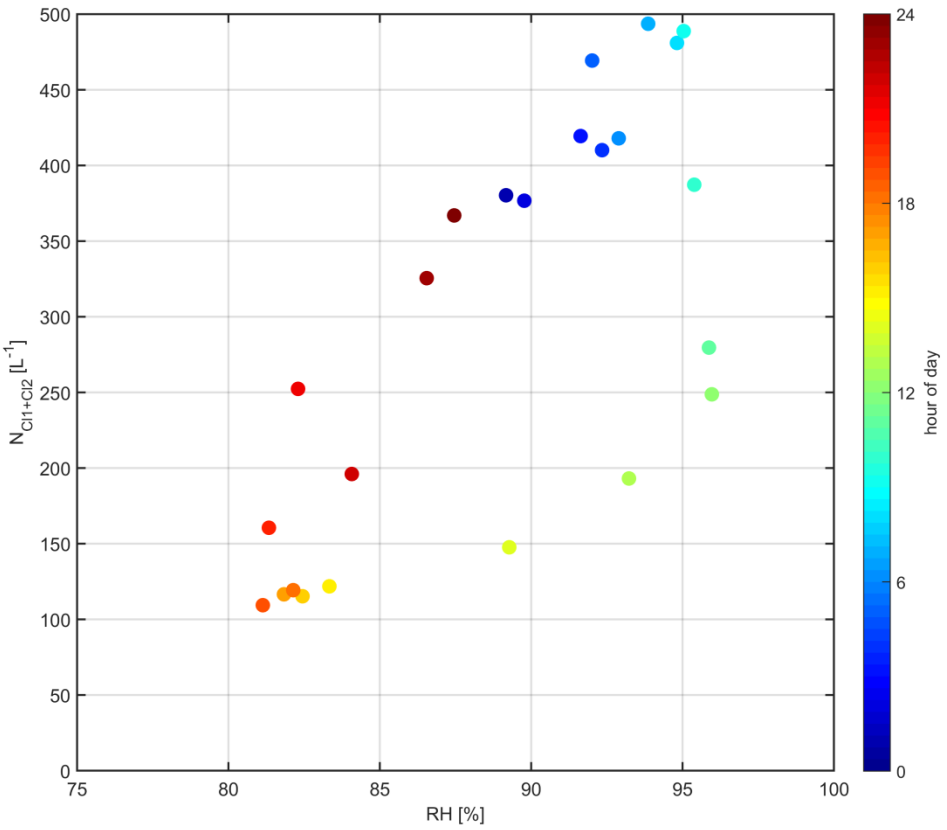

Figure 2. Total particle number in clusters 1 and 2, plotted against relative humidity.

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.

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