

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 DO ? 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 differ 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:

“C11 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. C12 appears to be a distinct sub-class of C11 which is less fluorescent in FL1. C12 shows similar behaviour to, and correlates strongly ($r^2 = 0.86$) with C11, 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 CI1+CI2, and we will clarify this in the revised manuscript. CI1 and CI2 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 $N_{\text{CI1}}+N_{\text{CI2}}$ 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 CI3, 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