

Interactive comment on “Biogenic cloud nuclei in the Amazon” by J. D. Whitehead et al.

Anonymous Referee #1

Received and published: 15 February 2016

Biogenic cloud nuclei in the Amazon

J.D. Whitehead, E. Darbyshire, J. Brito, H. M. J. Barbosa, I. Crawford, R. Stern, M. W. Gallagher, P. H. Kaye, J. D. Allan, H. Coe, P. Artaxo, and G. McFiggans

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

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this analysis, and is unclear enough in parts, that it should not be published.

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.

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

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 to 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.

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

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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 be for the sub-micron instruments.

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

p5l27: You are using $FT+3\sigma$ 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 $FT+3\sigma$ is.

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 $FT + 3\sigma$ is. This is very important given that you report actual fluorescence intensity values in Table 3. Additionally, do these values of $FT+3\sigma$ 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 values 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.

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

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

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counter).

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 non-biological 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. . .”

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.

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.

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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.

p7124: 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”).

p7124: 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 cited? 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.

p9110: Can you verify that the size-distribution in this figure is not ‘fluorescence signal

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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.

p9117: “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?

p9118: “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 FI1-FI2-FI3? 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 WIFS should be provided to support the conclusion that these clusters represent fungal spores. There

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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.

p9l19: I find the following statement confusing: “These clusters (referring to C11 and C12) contribute approximately 70% to the total FBAP concentration, with no significant diurnal variation.” Yet there is a very strong diurnal signal in FBAP, and C11+C12 makes up 70% of FBAP. Is there a typo here, or am I misunderstanding the phrase ‘with no significant diurnal variation’ in C11+C12?

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 sub-micron 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.

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.

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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.

Technical corrections:

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

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.

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.

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.

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.

Figure 9: “Mean growth factor for the dominant less hygroscopic mode” should be

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“Mean growth factor for the dominant, less-hygroscopic mode.” Also typo with “agains.”

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)

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2015-1020, 2016.