

## 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 thoroughly 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<sup>-3</sup>, 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<sup>-3</sup>, 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.

The authors apply a hierarchical cluster analysis to the WBS 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  $0.19\% \pm 0.07\%$ . This is well below 0.3%, which is considered to be the upper limit for background air masses not affected by biomass burning' 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  $D_p > 1 \mu\text{m}$ , 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  $0.8 \mu\text{m}$ . Please specify the 50% detection diameter of your instrument.

The instrument  $D_{50}$  is  $0.8 \mu\text{m}$ , 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.  $> 0.8 \mu\text{m}$ ).

- 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  $D > 0.8 \mu\text{m}$ , 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 they 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  $\kappa$  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<sup>-1</sup>. 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  $GF(D/D_0)$  instead of 'Growth Factor  $D/D_0$ '

We will modify the label to  $GF(D/D_0)$ .

Fig. 5:

- please specify the data gaps

As above

- The unit is probably  $\mu\text{g}/\text{m}^3$

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:

- $N_{\text{tot}}$  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:

- $N_{\text{tot}}$  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  $D_p$  instead of  $d_p$

This will be corrected for the revised manuscript

- unit of  $dN/d\log d_p$  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, 1157–11 174, 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