## Response to reviews

We thank all reviewers for the constructive suggestions and comments. Reviewer comments are listed in red. Author responses are in plain text labeled with [R]. Specific modifications to the manuscript are in blue.

### Reviewer #3 (published 19 September 2014)

The paper of Chen et al. describes…

In my opinion the significance of this paper is that different SOM production pathways can be found by using the PMF. This paper addresses relevant scientific questions, however, the main issue is that a substantial portion of the results (and conclusions) presented in this paper have already been published in Chen et al. (2009) even though the data analysis was different. Therefore my main concern regards the novelty of the paper. Overall, this paper is well written and the structure of paper is clear and easy to follow. I think this paper merits publication after addressing the issues listed below.

General comments

As mentioned above, a large part of the results and following conclusions have been published earlier. Therefore I'd suggest making a clear difference which results are novel in this paper and which ones have already been published. That would be fair to the readers.

[R0] We thank the reviewer for the valuable feedback. This paper presents a follow-up research of Chen et al. (2009), in which in-Basin sources that have been suggested as major contributors of submicron OA are segmented quantitatively. For clarification, the manuscript is carefully revised. New results are highlighted. Statements related to the results that have been published in Chen et al. (2009) are either removed or revised with proper citations. Specifically, the Introduction section (p16155, line 13-28) is revised to clarify the goal of this research and the difference between results published in Chen et al. (2009) and this study. Experimental details and results that have been provided in Chen et al. (2009) are deleted (including p16157, line 1-24; p16158, line 13-17, 21-25; p16159, line 1-2; p16164, line 4-6; p16167, line 11-25; and lines in Sect. A and Sect. B of the Supplement). Moreover, modifications are made in the Results and Discussion section (p16163, line 16-22; p.16164, line 10 to p1616, line 22) to clarify the new results vs. published ones.

Specific comments

1. Abstract: page 16153, line 7; "Ammonium was present in sufficient quantities to partially neutralize sulfate". Later it was said that: "there was insufficient ammonium to neutralize sulfate" (page 16161). I understand that the meaning is same but I prefer using the latter for clarity.

[R1] This sentence is revised as suggested.

2. Abstract and throughout the manuscript; make sure that when the abbreviations are used for the first time, you also write the whole definition e.g. HOA and OOA in abstract, IEPOX, MVK and MACR in results and discussion.

[R2] Changes are made to make sure that abbreviations are defined in parentheses the first time they are used.

3. Site and instrument description: page 16157, lines 25- ; PMF was conducted on medium-resolution V-mode data but reported in unit mass resolution. Why?

[R3] The resolution of V-mode AMS data is about 2000. The data are typically analyzed in two ways. In one way, the signals are summed for each m/z window. Although there might be multiple peaks in one window, data are reported in unit mass resolution (UMR). Alternatively, the ion signals can be determined by multi-peak fitting (i.e., called high-resolution data). However, such non-linear fitting process may increase the noise level for low signals. For cases like Amazon having very low OA concentrations, the PMF analysis on the high-resolution data show similar four factors compared to the analysis on the UMR data, but worse separation for HOA. That's because although more ions are quantified in the high-resolution data, many of them have signal-to-noise ratios lower than the corresponding UMR fragments. We therefore decided to only report in UMR.

4. Results and discussion; I suggest keeping the discussion on inorganic species as short as possible as the title of the paper is "Fine mode organic mass concentrations. . .". Only if they are relevant to organics they should be discussed.

[R4] We think the information about inorganic species are important because this is the first on-line measurements of the chemical composition of submicron particles in Amazon and measurements are challenging, and the majority of this part has not been published in Chen et al. (2009). We have revised the title to match with the content. The new title is "Submicron Particle Mass Concentrations and Sources in the Amazonian Wet Season". Redundant discussions that have been covered in Chen et al. (2009) are deleted in the revised manuscript.

5. Results and discussion: page 16163, lines 26-28; you said that species correlated but R2 values were 0.35-0.52. To me these were only moderate correlations.

[R5] We agree with the reviewer that the $R^2$ values are not high. The purpose was to show the correlations of OOA-1 with those tracers are better than other factors with them. We have revised this part as "The correlations of the statistical loadings of the OOA-1 factor with the measured mass concentrations of biomass burning tracers, such as chloride ($R^2 = 0.52$), potassium ($R^2 = 0.35$), and black carbon ($R^2 = 0.43$) in the submicron particle population, were not high, possibly because of the mixing of sources to these tracers such as primary biological particles (i.e., contributing chloride and potassium) and regional pollution from Manaus to (i.e., black carbon). These correlation value were, however, significantly greater than those of the other three factors (HOA, OOA2, and OOA-3) with the tracers (i.e., $R^2 < 0.10$ for chloride, $R^2 < 0.02$ for potassium, and $R^2 < 0.20$ for black carbon) (Fig. 6b)".

6. Results and discussion, page 16166, lines 27- ; you talk about the different fractional contributions of OOA-2 and OOA-3 during different periods. Have you found any reasons why the fractions differed so much?

[R6] The weather conditions were different for the two periods. It was sunny and warmer with occasional clouds during the first period but rainy and cooler during the second period, which possibly drove the difference in the mass fraction of PMF factors. We have added the discussion of the meteorological conditions as follows: "Figure 9 shows that the relative importance of each process as a contributor differed with time and highlights two focus periods. Precipitation and temperature were the major meteorological factors that differed between the two periods. The first period was sunny, warmer with occasional clouds, and the second period had frequent heavy rainfall events. Long-range back-trajectory analyses presented in Martin et al. (2010b) showed that the air masses consistently arrived from the equatorial Atlantic Ocean passing as northeasterlies through the Amazon basin. Local measurements showed that the daytime winds mainly came from the north and northeast (Fig. 9, top)… Figure 9 shows that the loading fraction of the OOA-2 factor consistently dropped following heavy rainfall events, suggesting more efficient in-cloud or below-cloud scavenging for the types of material represented by OOA-2 than for those types represented by OOA-3. This finding further supports the interpretation that the OOA-2 factor represents, at least in part, aqueous-phase production pathways because SOM produced in this way has greater water solubility and hence greater wet deposition rates than SOM produced freshly by gas-to-particle condensation, as interpreted for the OOA-3 factor. Figure 9 also shows that the mode diameter of organic material in period 1, which has a higher OOA-2 loading fraction, is significantly larger than that in period 2, which has a higher OOA-3 loading fraction. Aqueous-phase processing is anticipated to add additional organic material that results in larger mode diameters after dehydration."

7. Results and discussion; I guess you also measured particle size distributions with the AMS. Could you get any support for the PMF factors from the pToF data?

[R7] Yes, we measured particle size distributions. The data are however very noisy because of the low concentrations. We are unable to derive the statistic size distributions for the PMF factors. When comparing the two case periods (i.e., the period having dominant contribution of OOA-2 and the period mainly contributed by OOA-3, we found the mode-diameter for OOA-2 was greater than that for OOA-3, which is consistent with our interpretation of OOA-2 from sustained particle-phase pathways although not conclusive. We have modified Fig. 9 with this addition and modified the related discussions (please see R6 for more details).

8. Conclusions; This section is mostly summarizing the results. Only the last paragraph concludes. Maybe the title should be summary and conclusions?

[R8] We have revised this section with the focus of the last paragraph.

9. Figure 5, page 16182; Figure is a bit unclear. I suggest using dots only to PMF factors and lines to all the other components, or something similar.

[R9] We thank the reviewer for the suggestion and have updated the figure.

## Reviewer #4 (published 6 November 2014)

The study presents new information of chemical composition of organic aerosols in the Amazon. The results are important and interesting, especially since there are few previous studies in this environment.

My main concern is that the results in some parts seem to be over-interpreted, and some conclusions are not well-supported by the data analysis. This regards the interpretation of the PMF factors OOA-2 and OOA-3, which only differ slightly in average mass spectrum and time variation. The associated uncertainty should be better reflected in the abstract, discussion and conclusions. OOA-2 is "implicated as associated with reactive uptake of isoprene products... to haze, fog or cloud droplets", while OOA-3 is "consistent with fresh production SOM by a mechanism of gas-phase oxidation of BVOC followed by gas-to-particle conversion of the oxidation products" (from abstract). These strong statements should be modified to better reflect the data, uncertainty and analysis.

[R10] We thank reviewer #4 for the valuable feedback. We have carefully revised the results and discussion section, particularly on the interpretation of OOA-2 and OOA-3. Detailed comparisons between the mass spectra of the PMF factors and the spectra of chamber SOM are made. Additional information from literature and the analysis of meteorological conditions is added to support conclusions. Statements are rephrased based on the associated uncertainties. Detailed responses to the individual comments are given below.

The supplemental material is quite extensive. 34 pages supplement to an 18 page manuscript seem like too much for a journal like ACP. I suggest moving some of the most important sections and figures to the main text.

[R11] We have moved section D and Figure S9 to the main text. Also redundant details in Sect. A of the Supplement that have been provided in Chen et al. (2009) are deleted.

The results and discussion section should include at least some information on meteorology (boundary layer heights and wind patterns) at the site, especially in the discussion of time series and daily patterns.

[R12] We have added some information on the boundary layer, local wind, back trajectories, and weather conditions to support our discussion of PMF factors. Please see R6 and Fig. 9 for more details. More detailed information on meteorology is provided in the overview paper by Martin et al. (2010).

Previous studies in the Amazon have observed primary biological aerosols – were there any indications of primary biological aerosols in the present data?

[R13] The PMF analysis conducted herein does not suggest a factor for primary biological aerosol particles (PBAP). Possible reasons include (1) the marker ions are not unique and have contributions from other sources (e.g., secondary organic material), (2) the concentration level is low so that the PMF analysis only extracted information from the UMR data (see R3), and (3) the contribution of PBAP to the submicron organic aerosol mass is small. Schneider et al. (2011)

semi-quantified the primary biological aerosols during AMAZE-08 by using mass spectral markers and placed an upper limit of 20% to their mass contributions to submicron organic aerosols. This information is now provided in Introduction.

Specific comments:
Page 16154 line 15: Oxidation of BVOC correlates with other factors beside sunlight – please be more specific.

[R14] This sentence is revised.

Page 16157 line 5-8: The statement on density seems misplaced here.

[R15] Since the calculation of density is discussed in the Supplement, we have deleted these lines in the main text.

p. 16158 and supplement: The authors spend quite some time to discuss that they do not observe organosulfates in their data. The sulfate levels are quite low, and since organosulfates are only expected to constitute a fraction of this, their concentrations should be very low. What is the detection limit for organosulfates using AMS?

[R16] The molecular weight of organosulfates is much greater than sulfate. If a substantial fraction of the detected sulfate is from organosulfates, the concentration of organosulfates might not be low. Some studies suggest a large contribution of organosulfate to the organic mass concentration, especially for acidic particles (Surratt et al., 2008; Iinuma et al., 2009). During AMAZE-08, submicron particles are acidic. Therefore, it is important to evaluate the potential contribution of organosulfates. The AMS detection limit for organosulfates is estimated to be ~0.06 μg $m^{-3}$ given a detection limit of sulfate of 0.02 μg $m^{-3}$ and an assumed molecular weight of organosulfates of 300 g $mol^{-1}$.

P. 16160 L4: Would biomass burning in Africa give a covariance of BC with sulfate at your site?

[R17] Studies have shown that sulfate and BC in the fine mode were both elevated during long-range transport events in the central Amazon (Martin et al., 2010b and references therein). Based on satellite, lidar, and back-trajectory analysis, we concluded that the particles during AMAZE-08 were affected at times by dust transport from northern Africa and biomass burning in equatorial Africa.

P16161 L17-22: The argument is not clear here. First you say that the charges nearly balanced and then you state the relative proportions within each group, which does not provide information on the degree of charge balance. In several places you mention that the aerosols were acidic. How did you arrive at that conclusion based on your data?

[R18] The charges were nearly balanced when looking at the fine mode ($PM_2$) data obtained by IC. For the submicron domain, we integrated information of the concentrations of non-refractory species detected by AMS, concentrations of other cations obtained from $PM_2$-IC data (for the entire campaign) and the size distributions of these cations obtained from MOUDI-IC data (one sample for AMAZE-08 as well as Fig. 9c in Fuzzi et al. (2007)). The results indicated that the particles were acidic and probably presented as bisulfate. For clarification, we have deleted p16161, L16-22 and added the following statement in L12: "The AMS is unable to quantify refractory components such as K+, Na+, Ca2+, and Mg2+. Mass-diameter distributions of these ions obtained by IC analysis of samples collected by a Multi-Orifice Uniform Deposit Impactor (MOUDI) on 22 March 2008 suggest that 40% of the mass of these ions was distributed to the submicron particle fraction. For comparison, Fuzzi et al. (2007) for the wet season reported 50-60% of K+ and Ca2+ in the submicron fraction, compared to the predominance of Na+ and Mg2+ in the supermicron fraction. The campaign-average fine-mode mass concentrations of K+, Ca2+, Na+, and Mg2+ measured by IC were 0.03, 0.01, 0.02, and 0.01 μg m 3, respectively. The implication of the relative concentrations (i.e., sulfate concentration of 0.19 ±0.06 μg m-3) is that the submicron inorganic ion composition is reasonably approximated as ammonium bisulfate during AMAZE-08.".

P16161 L. 8-11: statements 1) and 2) seem somewhat redundant.

[R19] The statement of "(i.e., requiring a molar ratio of 2)" is removed.

P. 16163 L26: The correlations are not very strong, probably due to a mix of sources for the tracers. A term like "correlated somewhat" would describe this better.

[R20] We agree with the reviewer. This statement is revised. Please see R5 for more details.

P. 16164 L. 11: The peak at m/z 82 is not prominent in Fig. 4c. I agree that is present and visible, but it is far from being prominent. Furthermore the relative intensities of m/z 53 and 55 in OOA-2 and OOA-3 are too similar to state that 53 is "elevated". The text should better reflect the data here.

[R21] We have gone through the manuscript. Similar descriptions are rephrased (e.g., "characteristic peak".

P. 16164 L25: What is number of observations included in calculation of the correlation coefficient? How many days did the correlation include?

[R22] About seven days including ~1000 observations.

P. 16164 L27-29: Should be rewritten to reflect that your OOA-2 factor somewhat resemble the spectra of Budisulistiorini et al, not the other way around.

[R23] We meant the IEPOX-OA factor reported in Budisulistiorini et al. (2013) not our OOA-2 factor in this sentence. For clarification, we have revised this paragraph by specifying the factor name used in the references such as "… Borneo (named as the "82Fac" factor) …Ontario, Canada (named as the "UNKN" factor) … Atlanta, Georgia, USA (named as the "IEPOX-OA" factor) … "OOA-2" of this study and "82Fac", "UNKN", and "IEPOX-OA" of earlier studies".

P16165 L1: Unless the reference Kuwata et al., has been updated, the statement should be removed, since it does not add to the readers understanding of the present work.

[R24] We have replaced Kuwata et al. by Liu et al. (2014) which is from the same study and is already published.

P16165 L5: How did you measure/calculate that the particles were acidic? How acidic were they?

[R25] Please see R18 for the answers.

P16165 L7-22: The paragraph is quite speculative and ends with a conclusion that OOA-2 represents both degree of emission and oxidation of isoprene, as well as uptake of these products in particles, in addition to uptake and aqueous phase processes involving other precursors. It would be nice if the latter part could be supported by data or more precise references. I agree that these are possible explanations for the factor, but it is important that the text (here, as well as in the conclusion and abstract) reflects the associated uncertainty.

[R26] We have carefully revised the discussion in p16164-16165. Results from a recent publication (Liu et al., 2014) are cited to support the discussion. Detailed comparisons between the mass spectra of the OOA-2 factor and the spectra of chamber IEPOX or isoprene SOA are made. Possible explanations (not just aqueous-phase processing) are listed.

P16165 L23-: The figure (4d) does not support the statement that m/z 55 and 91 are “distinct”.

[R27] The statement is changed to be “The OOA-3 factor had a prominent peak at *m*/*z* 43. For *m*/*z* > 80, the most intense peak occurred at *m*/*z* 91”.

P16166 L1-2: “a linear combination of the three chamber spectra largely reproduced the OOA-3 factor”. The ion of highest intensity m/z 29 is not well modeled with the chamber spectra – why?

[R28] One reason is that our chamber isoprene SOM likely formed from both gas-to-particle condensation and reactive-uptake of IEPOX pathways. The spectra of laboratory IEPOX SOM also show high intensity at *m*/*z* 29 (Budisulistiorini et al., 2013; Liu et al., 2014). We added this discussion in the revised manuscript.

P16166 L 10-11: For how many days did OOA-3 track the BVOC concentrations?

[R29] About 6 days.

P16167 L18: “the OOA-2 factor was consistent with the reactive uptake of isoprene oxidation products”. Consistent is a very strong word here. It is more fair to say that it showed similarities or it was interpreted as uptake isoprene oxidation products on aerosol particles.

[R30] We have gone through the manuscript and rephrased such statements.

P16166 L22-24. Was there changes in the boundary layer height during the day?

[R31] The boundary layer height was 50-100 m at night and increased to 700-800 m by local noon time. This information is added to the main text.

Figure 6: The legends should be more clearly described in the figure text. What is "the OH family"? It is very interesting that these experiments have been performed at different concentrations. The results show the variation in the relative intensities, and thus give information on the uncertainty in these studies.

[R32] This figure has been updated. The legend information is added to the figure caption. We also changed "OH family" to be "$H_yO_1^+$ family" for clarification.

Supplement
Please check that the references to figures and tables are in chronological order.

[R33] We checked the order and changes are made.

P5: Please update and add the reference Canagaratna et al. 2013. "The contributions of organonitrates and organosulfates, detected as inorganic nitrate or sulfate ions, to the elemental ratios were negligible because their low mass concentrations." The statement should have a reference to the discussion later in the Appendix.

[R34] The reference is updated to "Canagaratna et al. (2015)". The statement that the reviewer pointed out has been revised as a separate paragraph that provides quantitative discussion on the concentrations of organosulfate and organonitrate species. Previous description about organosulfate (also in sect. A of the supplement) and organonitrate (in main text, p16159 Line 14-20) are combined to this new paragraph.

P6: "The two types of filters show reasonable agreement". The statement should be further elaborated or at least give a reference to data in the manuscript or published papers.

[R35] The statement is changed to be "The two types of filters show reasonable agreement on the particle mass concentration (Table S2)."

P12: "The prominent C7H7+ companied with a negligible signal of C6H13+ at m/z 85 is also unique for the three biogenic SOA studied in the chamber". The reader cannot judge the statement since only data for BSOA is given. Give at least a reference here. The last sentence of "prominent" m/z 82 should be corrected as discussed above.
I suggest to move section D to the main manuscript. Furthermore, I suggest that Figure S9 is included in the main manuscript.

[R36] We have moved Sect. D and Fig. S9 to the main text. The descriptions of the mass spectra are carefully revised. Comparisons to the spectra of other types of organic aerosol from the literature with references are made.

Please extend the figure texts for figures S12-S14, so the reader is able to understand them without reading the full text.

[R37] This change is made.

**Reviewer #5 (published 10 November 2014)**

General comments
This paper presents the results of on-line measurements of sub-micrometer organic aerosol (OA) obtained in a tropical rainforest in the Amazon during the wet season. The study examines in detail the formation pathways for OA, by mainly focusing on statistical analysis (i.e., PMF) of the HR-ToF-AMS data sets. Based on these analysis, the authors suggest the comparable importance of particle-phase and gas-phase pathways for the secondary production of OA during the study period.

The present work may provide valuable data in our understanding on formation processes of OAs particularly associated with the oxidation of biogenic VOCs. The manuscript fits with the scientific scope of ACP. In my opinion, however, new scientific finding does not seem to be emphasized in comparison with some previous works. Although a data set presented is valuable, there are some important issues that need to be worked out. I recommend its publication in ACP after some revisions raised below.

[R38] We thank the reviewer for the valuable feedback. We have carefully revised the manuscript to highlight our new findings. Please see R0 for more details.

Specific comments
(1) My major concern is about the discussion on Figure 8: What is the major difference between the Period 1 and Period 2 from viewpoints of meteorological conditions, photochemical field, etc.? What is the major factor controlling the fractional contributions by each factor, particularly OOA-2 and OOA-3? None of the explanation has been made in the text. The authors should discuss these points because this is one of the most important part of this paper.

[R39] Yes, we have added the discussion on the meteorological conditions for the two focus periods. We think that the precipitation is one driven factor of different fractional contributions of OOA-2 and OOA-3. Please see R6 and Fig. 9 for more details.

(2) Figure 9 summarizes the processes for BSOA in the Amazonian wet season. However, most of the processes shown in the figure have been already reported or suggested in previous studies and seem to be somewhat “general” picture which one can find in a textbook. The authors should

emphasize in the figure what the new findings are in this study with making a quantitative statement.

[R40] The caveat is that multivariate statistical factors do not correspond to segregated individual chemical components (e.g., unlike molecules or families of molecules), although the factors can be indicative of the relative importance of different atmospheric emissions and pathways. Given this caveat in mind and the fact that the main text is already quite extensive after taking other reviewer's suggestions, we decided to remove Fig. 9 in the main text.

(3) If the OOA-2 factor represents the particle-phase reactions, then what is the time scale for this factor? The reaction is expected to be much faster than that represented by the OOA-3 factor which is associated with gas-to-particle partitioning of the BVOC oxidation products on a timescale of several hours (P. 16166, L7-9). Are these timescales supported by the time lag between OOA and BVOC? It is not clear from the diurnal profiles in Figure 7.

[R41] We agree with the reviewer that the reactive uptake of IEPOX isomers is probably rapid. The gas-to-particle condensation is also considered to be spontaneous. The formation of OOA-2 and OOA-3 is therefore regulated by the timescale of the production of gas-phase products by BVOC photochemistry. The diel profiles of OOA-2 and OOA-3 both peak in the earlier afternoon as is consistent with photochemically driven production. It is however difficult to quantify the time lag (if any) between OOA and BVOC based on the diel profiles because these profiles were influenced by local precipitation (e.g., Fig. S7). By looking at the time series (Fig. 6), there may be 1-2 h lag between OOA and BVOC (e.g., on 13 Feb and 17 Feb). The data coverage is however limited. We are unable to draw a quantitative conclusion on these timescales.

(4) P.16163, L.16-18: "The OOA-1 factor… African biomass burning." I cannot logically understand this sentence. Do the sentences below in this paragraph (P.16163, L.18-P.16164, L.9) support this sentence?

[R42] Yes, later sentences provide support. We have removed revised this sentence as "The OOA-1 factor had the feature of a singularly dominant peak at *m*/*z* 44 (Fig. 5b), which has been linked to organic material that has undergone extensive oxidation during a prolonged atmospheric residence time (~10 days)". The statement of "was believed to be mainly associated with long-range transport of African biomass burning" is removed since it has been address in the later text.

(5) P.16163, L.19: "a prolonged atmospheric residence time" What is the exact time scale for this? Please provide more quantitative statement.

[R43] We meant a time scale of "~10 days", which has been added to this statement.

(6) P.16163, L.27: Chloride is also a tracer for sea salt, and black carbon can be attributed to not only the biomass burning emissions, but also other burning sources, such as fossil fuel burning. The authors should rephrase the sentence or change the logic of it.

[R44] We have rephrased the sentence. Please see R5 for more details.

(7) P.16163, L.23: “Africa” should be “African”.

[R45] This change is made in the revised manuscript.

## References

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