

We thank the anonymous reviewers for providing helpful comments and suggestions on this manuscript. The responses to the Referees comments are found below.

Referee #1

Major comments:

1. Indicator for the chemical aging

Authors used oxygen to carbon (C:O) ratio as an indicator for the chemical aging of submicron aerosols. In most cases, this can be useful. Other indicators such as hydrocarbon ratio, NO_x/NO_y (or NO_z/NO_y) ratio, and photochemical age have been used in a number of previous studies. These parameters are significantly affected by not only aging and but also mixing processes with background air (e.g., Mckeen and Liu, 1993). We need to discuss the aging process with the careful consideration of the effect of air mass mixing. “Abstract” and “Conclusions” did not include the connection of C:O ratios with the actual time scale of aging. Negligible OA production (to carbon monoxide, CO) itself should be of importance, but the quantitative information on time scale is also crucial for discussing the OA aging and comparing with previous studies such as DeCarlo et al. (2010). My question to this point is whether you have considered the effect of air mass mixing to the C:O ratios and have evaluated the time scale of chemical aging found in this study. When you have done, please include this point in the manuscript. If not, please consider this point for the better description of the observed changes in the elemental compositions and formation processes of organic aerosol.

The reviewer raises an important point and the authors agree that this topic must be more clearly addressed in the manuscript. During the measurement campaign, the sampling site was continuously impacted by biomass burning emissions, both close and far. Figure S3, depicting identified fire spots for a single day, illustrates the large number of biomass burning sources found in a 100 km radius. Therefore, unlike the case of atmospheric processing of a unique source, where chemical processing can be linked to a given time scale, mixing processes of many BB sources at varying distances allows us to examine $\Delta\text{OA}/\Delta\text{CO}$ mass ratios on a broader scale. Fig. 08 now spans the entire O:C range, and conclusions are based on a “big picture”, which still depicts little $\Delta\text{OA}/\Delta\text{CO}$ mass ratio change with varying O:C. The text has been altered as follows:

The text on Page 12290, L.25 has been replaced by:

“The sampling site being located in a region with intense fire activity, fire spots were distributed both near and far from the site. In this context, an analysis of changes in chemical composition throughout the campaign has been carried out. An important method...”

Furthermore, text on Page 12292, L.21-L.29 has been replaced by:

“Figure 8 depicts the $\Delta\text{OA}/\Delta\text{CO}$ mass ratio for a given O:C ratio. For the calculation of ΔCO , we chose as background the lowest CO mixing ratio observed throughout the campaign, 240 ppb. High values of $\Delta\text{OA}/\Delta\text{CO}$, characteristic of biogenic air masses, are observed for $f_{60} < 0.08$. The values

in this range should be viewed with caution as they correspond to data with low ΔCO , and are thus subject to large uncertainty related to the CO background subtraction. Excepting those, $\Delta\text{OA}/\Delta\text{CO}$ is fairly constant around 0.03, indicating no net OA production tendency from biomass burning emissions. “

2. *Background concentrations of CO (CO_{bg})*

Authors assumed the constant value of CO_{bg} for the whole period of the campaign. During the phase 1, especially before 17 Sep. 2012, CO_{bg} should be higher than the assumed value of CO_{bg}. And this point is also significant to the calculation of ΔOA . How does the constant CO_{bg} affect the calculation of the $\Delta\text{OA}/\Delta\text{CO}$ ratio? As the evolution of $\Delta\text{OA}/\Delta\text{CO}$ ratio is one of major results in this study, authors should clarify and/or validate the effect of the assumption.

The CO_{bg} value before 17 Sep. 2012 is 290 ppb, 50 ppb higher than previously assumed for this period of time. Given that peak and average CO concentrations before 17 September were 2.2 ppm and 605 ppb, respectively, no large effect has been observed on the $\Delta\text{OA}/\Delta\text{CO}$ mass ratios.

3. *Impacts of brickyard*

Authors suggested the influence of the brickyard upon the observed data sets as discussed in section 4.2 and 4.3. In my view, this study aims to discuss the biomass burning impacts on the submicron aerosols. Therefore, authors need to eliminate in advance the data sets significantly affected by the brickyard emissions for the better presentation of the results.

The authors agree with the reviewer. The data from 24 September 15:00 up to 25 September 06:00 was removed from the analysis of the data throughout the manuscript.

4. *Evolution of number size distributions*

Authors summarized the tri-modal fitting to the averaged number size distributions (NSD) in Table 2. This data is very useful. Therefore, I strongly recommend providing the fitting results for the NSD classified by C/O ratios and including some of the highlight results in sections “Abstract” and “Conclusion”. Normalization approach as given in Takegawa et al. (2006) should be useful to interpret the evolution of NSD. They used the observed acetylene concentrations ($[\text{C}_2\text{H}_2]$) and their average ($[\text{C}_2\text{H}_2]_{\text{avg}}$), namely $[\text{C}_2\text{H}_2]_{\text{avg}}/[\text{C}_2\text{H}_2]$, for minimizing the effect of the air mass dilutions. In this study, ΔCO can be used as a surrogate. Please consider the reanalysis of the evolution of NSD normalized by the $[\Delta\text{CO}]_{\text{avg}}/[\Delta\text{CO}]$. Furthermore, how about the case of volume size distributions (VSD)? VSD normalized by $[\Delta\text{CO}]_{\text{avg}}/[\Delta\text{CO}]$ should be useful for discussing the evolution of total aerosol mass with air mass aging and be a compatible parameter to $\Delta\text{OA}/\Delta\text{CO}$ ratio.

Following the reviewer suggestion, fitting results were added to the text. Volume size distribution for a given O:C ratio has been included as well (Fig. 09), indicating little variation in volume distribution, following number size distribution.

Minor comments:

5. *P12284, L16-18. What does this sentence actually mean?*

The sentence has been rewritten as:

“However, Amazonian biomass burning aerosol were only recently, during SAMBBA, characterized using aerosol mass spectrometry.”

6. *P12285, L24 - P12286, L2. Aethalometer was used for measuring black carbon aerosols (BCA). Some studies suggest the possibility of the condensed materials on BCA to affect the measurement of absorption coefficient of BCA. Heater or thermodenuder have been used for reducing the effect of the condensed materials on the absorption measurements (e.g., Miyakawa et al., 2008; Kondo et al., 2011). How does this point affect the observed BC concentrations?*

While the data discussed in this manuscript were measured during SAMBBA, several instruments were deployed at the site for a longer period of time, including a MAAP (Thermo) and filter samplers (Artaxo et al., 2013). Comparison between Elemental Carbon (EC) concentrations, retrieved from thermo-optical analysis of filters, and aerosol light absorption from MAAP during the dry season of 2011 allowed to retrieve a Mass Absorption Coefficient (MAC) of $8.4 \text{ m}^2 \text{ g}^{-1}$ (unpublished data), not much higher than the standard value of $6.6 \text{ m}^2 \text{ g}^{-1}$.

7. *P12286, L25. Authors assumed a constant value of CE of 0.5. This assumed value was validated by the comparison with other measurements (SMPS+OPC). My concerns on this point are (1) the assumed density to calculate the mass concentrations of total aerosols and (2) absolute value of the CE which assumed as being constant.*

(1) The assumed density of 1.2 g m^{-3} is very low, 1.2 g cm^{-3} is correct? If so, what is the basis of this assumption? To the best of my knowledge, inorganic and carbonaceous aerosols typically have larger densities (for example, $\sim 1.7 \text{ g cm}^{-3}$ for major inorganics and BCA and $1.0\text{-}1.9 \text{ g cm}^{-3}$ for organics, Pan et al., 2006). The typical values of the measured C:O ratios of $0.4\text{-}0.6$ suggest that the larger value of the particle density should be better. This should be significant for the data sets obtained, especially, in Phase II, because the relative contribution of organics to total particle mass was smaller during the period.

(2) Is the assumed constant value of CE reasonable? This question is closely related to (1). The good correlation of reconstructed and measured total aerosol mass concentrations only supports the assumption of the constant value of CE. For validating the assumed value of CE, AMS users have conducted the intercomparison experiments using a Particle-into-liquid-sampler coupled with ion chromatography (Drewnick et al., 2003; Takegawa et al., 2005). Recently, Middlebrook et al. (2012) showed the way to estimate the composition-dependent CE. I recommend the comparison of the assumed CE of 0.5 with the composition-dependent one by using the algorithm given by Middlebrook et al. (2012).

Following the suggestion of both reviewers, a composition-dependent density has been calculated for the entire dataset. Furthermore, the average upper limit of the aerosol aerodynamic diameter measured by the SMPS is found to be roughly 620 nm, compatible with ACSM size range. Therefore, the OPC data has not been considered for validating the applied CE in the revised calculation. Figure 2, now including a larger dataset due to removal of the OPC contribution, has been updated accordingly. Besides the analysis undertaken according to the reviewers' suggestions, a data handling mistake has been found as well. Due to changes in recent versions of the Igor routines for treating ACSM data, all ACSM data described in the manuscript had effectively a CE of 1, instead of 0.5, which used to be the ACSM data analysis CE standard value. The text on Page 12286, L.24 has been replaced by:

“The collection efficiency (CE) has been previously reported to range from 0.5 to 1, depending on system setup and aerosol physical-chemical characteristics (Huffman et al., 2005; Canagaratna et al., 2007; Matthew et al., 2008; Middlebrook et al., 2012). Here, the CE was evaluated through the comparison of the mass concentration of species measured by the ACSM and Aethalometer (BC_e, measured at 880 nm) with the integrated mass of the SMPS (aerodynamic diameter upper limit ~620 nm). The density used for each species was 1.78, 1.72, 1.72, 1.52, and 1.77 g cm⁻³ for sulfate, nitrate, ammonium, chloride, and BC, respectively (Lide, 1991; Park et al., 2004). The density of organics was estimated based on the oxygen to carbon (O:C) and hydrogen to carbon (H:C) ratios (Kuwata et al., 2012). The median aerosol density was calculated to be 1.48 g cm⁻³, being 1.21 g cm⁻³ during the biomass burning peak. Results of intercomparison suggest that a CE of 1 provides the best fit for the dataset presented here (Fig. 2). Such a CE is in good agreement with the results of Middlebrook et al., (2012), who have suggested an increase in CE in environments dominated by organic aerosols.”

8. *P12289, Fig 5. Time series in Fig 5 should include the full of the observation period (6 - 30 Sep., 2012). Even though authors mentioned that the reported data focus on the period of 13 – 30 Sep. 2012 in section 2, the data analysis in Phase I was applied to the data obtained before 13 Sep. 2012 (according to the definition of the period given in section 4.2).*

The text was corrected following the reviewer's suggestion.

9. *P12290, Fig 6. I recommend modifying Fig 6 by coloring the data points based on the observed concentrations of OA. This is because the low concentrations comparable to the lower limit of detection or quantification (LOD or LOQ) tend to make the values of f60 scattered in Fig 6.*

Figure 6 has been changed accordingly.

10. *P12291, L2 – L11. These sentences should be moved to section 3, because they are technical descriptions and not actually result. This will help the improvement of the readability.*

The text was changed accordingly.

11. P12291, Fig 7. Please include markers or area of data points corresponding to biogenic OA suggested by previous studies (AMAZE-08, Heald et al., 2008) as describe in the manuscript. Furthermore, a guideline for slope of -1 (corresponding to carboxylation) should be useful for connecting the descriptions with the figure.

Figure 7 was changed accordingly.

12. P12295, L15 – L21. These sentences are speculative, because there are no data of elemental compositions of OA directly connected to the hygroscopicity in this study. Such descriptions should not be included in the section of the result. Please consider to make a new subsection “Discussion” or “Implications” in “Results and discussion” for clearly separating the facts and speculations. Other parts of the speculative discussion also should be moved into the new subsection.

The text was changed accordingly.

Referee #2

Major comments:

1. The authors report on measurements made during the dry-to-wet transition period (Sept.) in the Brazilian Amazon in 2012. A suite of instrumentation was deployed to characterize particle composition in real time (equivalent BC + non-refractory sub-micron mass), as well as size distributions and CO. They find that organic aerosol (OA) dominates. They find that the OA can be decomposed into fresh biomass burning-derived OA (BBOA) and aged BBOA, along with an oxygenated OA component. The observations themselves are certainly unique, as detailed characterization of particles in this area has been extremely limited until recently. I have some concerns that the authors are over-interpreting their OA composition data in terms of atomic ratios. I suggest that the authors focus on what was actually measured (e.g. f43, f44 from the ACSM) rather than the derived properties (H:C and O:C), and carefully consider how strongly their conclusions can actually be made regarding variations in particle composition, and also how good a metric O:C (or f44) is for atmospheric processing in this region.

The authors acknowledge the concerns raised by the reviewer. The replies are found along the specific comments, below.

Minor comments:

- 2. Aethelometer: It is entirely unclear how the multiple scattering effects correction was performed given that Aethelometer correction algorithms require knowledge of the particle scattering. It is not stated that scattering was measured. Was scattering calculated from the size distributions? More information is required. Without further information, I have concerns over aethelometer accuracy.*

Aerosol light scattering was measured using an Ecotech Aurora 3000 Nephelometer, which allowed to perform multiple-scattering corrections of the Aethalometer. The text on P.12285 L.24 has been replaced as:

“An Aethalometer (Magee Scientific, model AE30) provided Equivalent Black Carbon (BC_e) mass concentrations throughout the campaign, which were corrected for filter loading and multiple scattering effects according to Arnott et al. (2005) and Schmid et al. (2006) using aerosol light scattering measured using an Ecotech Aurora 3000 Nephelometer.”

- 3. ACSM calibration: Calibration was performed using size-selected ammonium nitrate particles. Undoubtedly, there were some fraction of multiply charged particles present following size selection. The much larger size of the multiply charged particles means that they have much greater mass per particle than the singly charged particles. In the AMS, this can be accounted for in part because the size distribution can be measured as there is a chopper wheel within the AMS. No such component exists in the ACSM. It is thus unclear how multiply charged particles are accounted for in the calibration. This must be addressed.*

During ammonium nitrate calibration, we always observed a very good linearity between number concentration and detector signal, which hardly would have been obtained if multiple-charged particles had played an important role. Furthermore, a very good mass concentration comparison between different instruments, e.g., ACSM and AMS, has consistently been observed, following the same calibration procedure as reported here. As such, the authors of this work believe that under normal conditions, multiple-charged particles do not contribute strongly to the instrument mass calibration.

- 4. ACSM+BC vs. SMPS+OPC comparison: It is not clear that differences in definition of diameter are taken into account when making this comparison. The ACSM measures by aerodynamic diameter. But the SMPS measures by mobility diameter and the OPC by an optically-defined diameter. These are not all identical, and thus the upper-limit for integration of the SMPS+OPC distributions is not clear in relation to the 650 nm upper limit for the AMS. Perhaps this is why the SMPS+OPC distribution is integrated to 750 nm, but this is not stated. This must be addressed. The authors argue for good closure between the ACSM+BC vs. SMPS+OPC, but this has a few assumptions built in, such as the ACSM CE and the particle density. The CE is assumed to be 0.5. Such a value has been shown to be appropriate for ammonium nitrate, but it is not clear to me that this same value is appropriate for a system that is strongly dominated by biomass burning organic particles. Although Middlebrook et al. argue for no strong deviation from a CE = 0.5 for organics, it is clear in their ARCPAC data, which has the largest BBOA contribution from any of the studies*

considered, there is a noticeable upturn in the CE at the highest organic mass fractions (see their Fig. 4). Here, the BBOA mass fractions were very large, and thus it is possible that the ACSM CE in this study is too small (leading to an overestimate of the Organic concentration).

Please refer to comment #7 from Reviewer 1.

5. *Atomic ratios: The authors convert their observed fraction of signal at $m/z = 43$ to H:C ratios using the relationship in Ng et al. (2011). One only need look at the relationship in Ng et al. (2011) (their Fig. 2) to realize that it is of limited utility, as it is exceptionally flat over a large range of f_{43} and within this flat region substantial differences are observed between different studies. In other words, the uncertainties are substantial because f_{43} is not a particularly sensitive metric of H:C, except when f_{43} is small. Now, it is possible that for a given instrument/region, there is a stronger relationship between H:C and f_{43} than is indicated when the various campaigns are combined (c.f. Fig. 2 in Ng et al.). But since the authors adopt the multi-campaign fit from Ng et al., there is an inherent weak relationship between f_{43} and H:C.*

If the H:C ratio had been the variable measured, then the rationale developed by the reviewer, from such flat relationship, would indeed imply in a large uncertainty in estimating f_{43} . However, since f_{43} was the variable measured, the fact that H:C doesn't depict large changes for a significant range of f_{43} does not make H:C estimation less precise. The standard deviation for the entire dataset is 10%, including BB emissions.

Given that the measured spectra presented here correlate well with the AMS database (Table 2), with airborne C-TOF AMS during SAMBBA (Table 2), as well as with other parameters (e.g. f_{60} /CO/aerosol number concentrations, in Fig. 5, or Fig. 9), the authors find no evidence to believe that the H:C ratio has not been estimated within the uncertainties described by Ng et al., (2011). This topic is further discussed after the following comment.

6. *Related, the authors compare their derived H:C to H:C values from Heald et al. (2010). But Heald et al. determined H:C from consideration of high-resolution AMS data, not from the Ng et al. f_{43} vs H:C relationship. The difference in behavior could easily arise because equivalent things are not being compared. Again, if one looks at Fig. 2 in Ng et al. there does seem to be some indication of a more linear relationship between H:C and f_{43} for individual studies than there is for the overall combined datasets. I am not convinced that these are comparable. It is my opinion that the data are being overinterpreted here with respect to what can actually be concluded regarding the behavior of H:C, and the relationship between H:C and O:C. I am not convinced by the composition arguments that are put forward on p. 12292. The authors are working hard to provide chemical explanations for observations that may simply be wrong. I realize that the authors are trying to make the most out of the data that they have available, and that measurements in the Amazonian rainforest are no easy task, but I have concerns that they are pushing the interpretation of their data beyond what is justifiable.*

Fig. 2 from Ng et al., (2011) has been put together using a suite of ambient and laboratory measurements. For laboratory measurements, the majority of the data (alpha-pinene, toluene, m-xylene, acrolein and so forth) depict very little changes in the H:C ratios and indeed appear as a cluster around a

midpoint, as described by the reviewer. For ambient data, a single point is usually shown, the OOA profile resolved from factor analysis, and it thus is not possible to observe f43-H:C changes for single ambient studies. The ambient data presented here, unlike the results discussed above, spans over a large range of f43 values (0.03 – 0.09), roughly one third of the full range of Fig. 2 from Ng et al., (2011). This is comparable with two studies shown in Fig. 2 from Ng et al., (2011): Riverside OOA time-dependent data (yellow markers) and laboratory studies of isoprene SOA (dark green markers). These studies do show a very good agreement over a large range of f43 values and allow O:C to H:C changes to be derived, within their uncertainties.

The authors of this study agree that the uncertainties associated with the V-K diagram and its interpretation were not described in the required detail. Furthermore, recent results improving O:C and H:C ratios from f44 and f43 were published in ACPD. Appropriate changes were inserted in the text. The text on P. 12291, L. 29 has been changed to:

“The biomass burning aerosol in Rondônia, however, shows a different behavior, with the majority of the oxygen-to-carbon ratio change (from O:C \sim 0.25 to O:C \sim 0.6) accompanied by only a small change in the H:C ratio. Such processing is consistent with the replacement of a hydrogen with an alcohol group (-OH), for example. For O:C > 0.6, the average slope changes to -0.5. Such a slope could arise from an increase in the importance of biogenic aerosol processing (slope = -1) relative to BBOA processing (slope = 0) and/or a change in the reaction tendency.

Being derived from an UMR instrument, the atomic ratios presented here were not directly measured and thus contain substantial uncertainties compared to the references above. Considering a 10% standard deviation from the f43 to H:C relationship as described by Ng et al. (2011a) alone, the BBOA slope in the Van Krevelen diagram, identified to be 0, would range from -0.5 to 0.5, considerably affecting the conclusions relative to BBOA chemical processing. Furthermore, recent results, currently under validation, suggest a significant underestimation of O:C and H:C ratios from HR-AMS measurements (Canagaratna et al., 2014). For the dataset presented here, O:C and H:C would be underestimated by 11 and 24%, respectively. Such change, however, does not impact substantially the Van Krevelen diagram, shown in supplemental material (Fig. S8). Even with these large uncertainties, the results shown here suggest a different processing path in the Van Krevelen diagram than previously reported, and certainly motivate a better characterization of fast changes in the chemical processing of BBOA in the Amazon region.”

7. P.12292: I do not see how limiting O:C to <0.6 “avoid[s] influence from biogenic aerosols.” This is not a binary system, where one does/does not have biogenic aerosols. It is a binary system with mixing. Variations in O:C can come about due to chemical processing of the BBOA, or can come about as BBOA mixes with biogenic OA (that presumably has a higher O:C). There is therefore no threshold below which contributions from biogenic OA can be ignored. .the contributions simply become of lessening importance.

The authors agree with the reviewer that the limitation of O:C<0.6 in this analysis must be more thoroughly addressed. Therefore, Fig 6 has been reformulated to include the entire dataset. The fact that it was originally limited to O:C<0.6 follows from the observation that the OA/CO mass ratio strongly

increases at this range, as expected for biogenically dominated air masses. The text has been revised and is further discussed with comment #1 from Reviewer 1.

8. *The authors have in no way convinced me that, for this environment, O:C is really a marker of atmospheric processing (i.e. ageing). They simply state that O:C is a surrogate for atmospheric processing, but provide no justification that this is appropriate for this location. Importantly, variations in O:C can easily come about from mixing between BBOA and biogenic (secondary) OA. The observed three factors: two BBOA factors and one OOA factor. Do they find, for example, that O:C varies with the ratio between OOA/(BBOA1+BBOA2)? Or some other combination? This is not discussed, but I think critical to any advancement of an argument that O:C is a good metric for ageing.*

Please refer to comment #1 from Reviewer 1.

9. *P.12293: It is not important that that size distributions were fit using Matlab or that this fitting was automated, unless the authors mean to provide the specific code.*

The text was changed accordingly.

10. *P.12295, L19: I think that this mention of hygroscopicity should be removed, as the authors seem to be basing their argument on an entirely different study. I do not see how mention of this point strengthens the story here.*

The text was changed accordingly.

11. *P12295/12296: I am not a PMF expert. But I must ask the question: if two PMF factors have a good temporal correlation, then are they really different factors? Isn't the whole point of PMF to separate components based on their unique temporal behavior?*

The choice of splitting the BBOA factor in two relies on the considerable reduction in the residuals of m/z 60, the mass-to-charge value relevant for biomass burning organic aerosols, as shown in the supplemental material. The fact that it separates a fresh and aged BBOA factors arises indeed from the fact that there is not always a temporal correlation between these two factors, e.g., during periods of time when no fire spots are found near the sampling site.

12. *Fig. 8 and P.12292: The authors state specifically how they determined deltaCO (difference between observed value and 240 ppb). It is never stated how deltaOA is calculated. Presumably this is the difference between the observed OA and some lower value. But what is the lower value used? Zero?*

Please refer to comment #2 from Reviewer 1.

13. *Fig. 2: Is the fit constrained to go through zero? Does it go through zero if unconstrained? What type of fit was done? Both axes have uncertainty, and thus a standard linear fit is not appropriate.*

Figure 2 has been reformulated with a linear fit not constrained to pass through zero.

14. *Fig. 5: For panels b and c, the differences between the axes are subtle, and thus the different lines are not easily distinguished. I suggest making the entire right-hand axis and axis label the red color.*

The figure was changed accordingly.

15. *Fig. 5: For panel a, the different axis for the organics is exceptionally subtle, i.e. the green is only transferred as a thin line. The green color must be applied to the entire axis. I had initially missed this, which led me to question the relationship between Fig. 5 and Fig. 6. The authors should aim to avoid creating such (potential) confusion for their readers by being more explicit in their representation of the data.*

The figure was changed accordingly.

16. *Fig. 5: “Average diameter” is insufficient. This should state what type of diameter (e.g. number weighted, surface area weighted)*

The caption of figure 5 now reads:

“Concentration time series of: organics, SO₄, NO₃ and NH₄ (a), B_{Ce} and f₆₀ (b), particle number concentration and number-weighted average diameter (c) and CO mixing ratio (d).”

17. *Fig. 6: This figure could probably benefit from adding a few lines that indicate values from previous studies, so as to place this in context. If this figure is not placed in a broader context, then it loses some of its relevance.*

The figure was changed accordingly.

18. *Fig. 7: The authors should make the x-axis minimum 0.*

The figure was changed accordingly.

19. *Fig. 10: It is not clear why the caption contains mention of Phase I and Phase II. . .these are not evident in the actual figure.*

The reference to Phase I and Phase II were removed from the caption.

References

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- Ng, N. L., Canagaratna, M. R., Jimenez, J. L., Chhabra, P. S., Seinfeld, J. H. and Worsnop, D. R.: Changes in organic aerosol composition with aging inferred from aerosol mass spectra, *Atmos. Chem. Phys.*, 11(13), 6465–6474, doi:10.5194/acp-11-6465-2011, 2011.

Figures

Figure 2

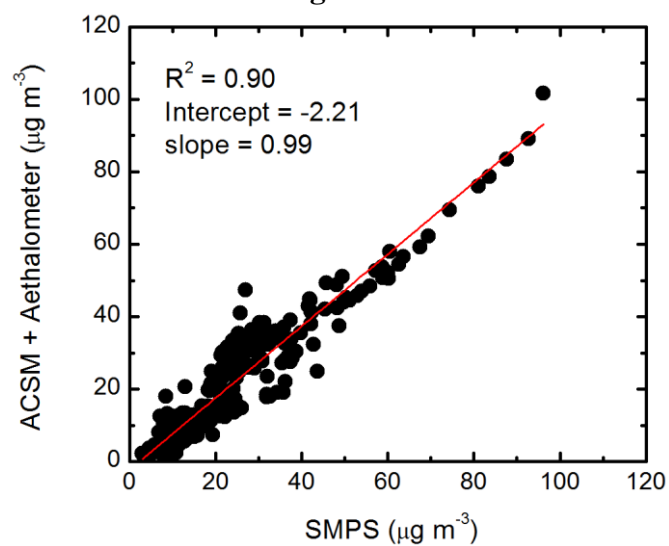


Fig. 2. Comparison between the sum of the mass concentrations of species measured by the ACSM and the Aethalometer (at 880 nm) vs. integrated mass concentration obtained from the SMPS.

Figure 5

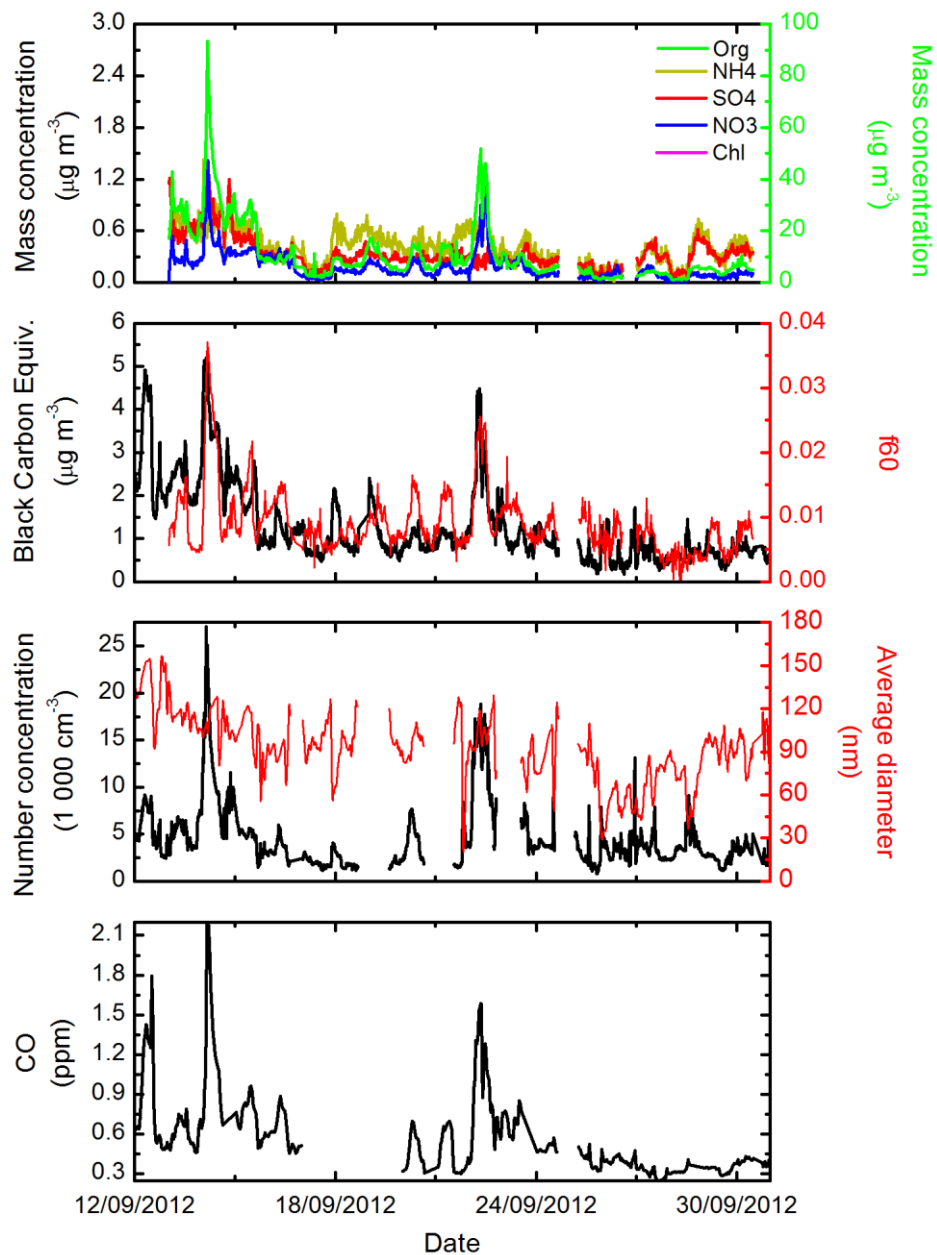


Fig. 5. Concentration time series of: organics, SO₄, NO₃ and NH₄(a), BCe and f60(b), particle number concentration and average diameter (c) and CO mixing ratio (d).

Figure 6

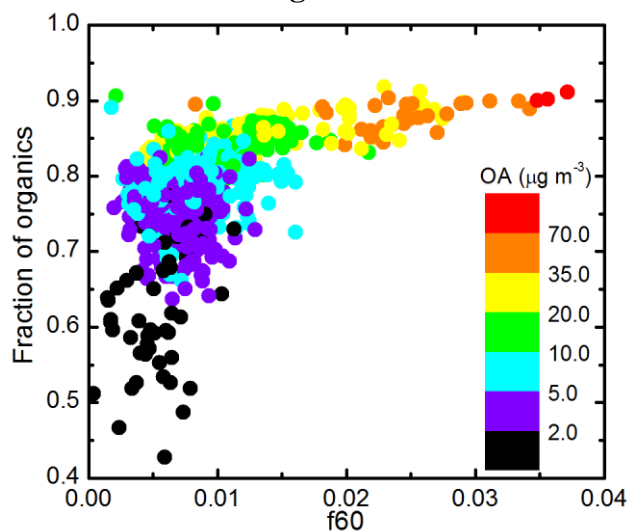


Fig. 6. The fraction of organic aerosol relative to total mass (non-refractory and BCe) vs. f_{60} , a marker for biomass burning impacted air masses.

Figure 7

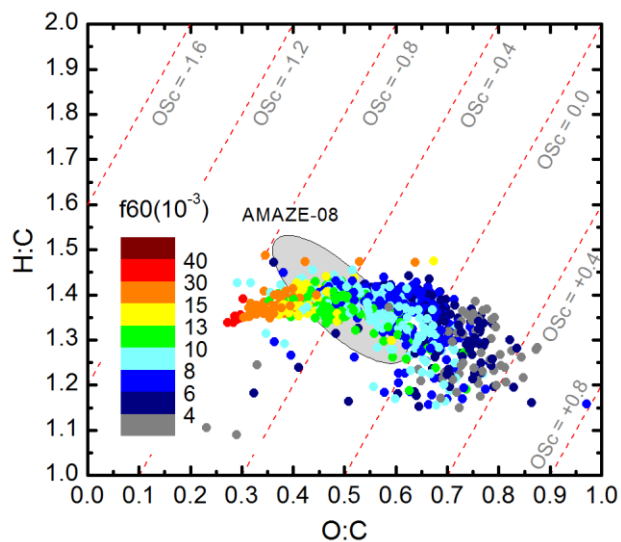


Fig. 7. Representation of the OA components into the VK-triangle diagram. The estimated carbon oxidation states ($OSC \approx 2 \times O/C - H/C$) are shown as red dotted lines. Gray area indicates the V-K space from AMAZE-08 as described by Heald et al., (2010).

Figure 8

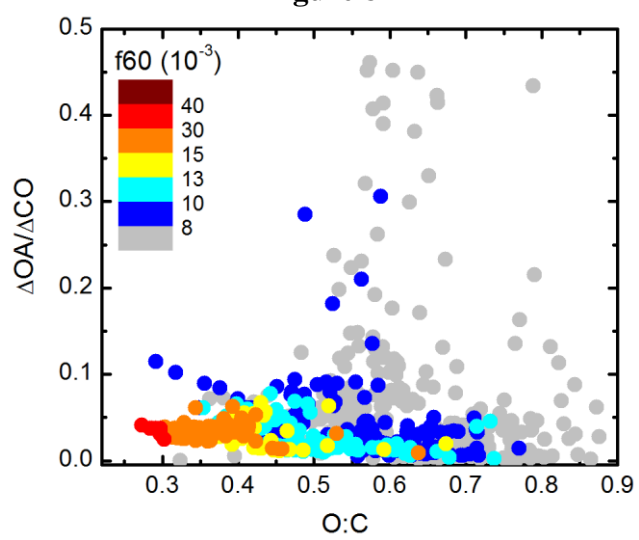


Fig. 8. $\Delta\text{OA}/\Delta\text{CO}$ mass ratios vs. O : C ratio.

Figure 9

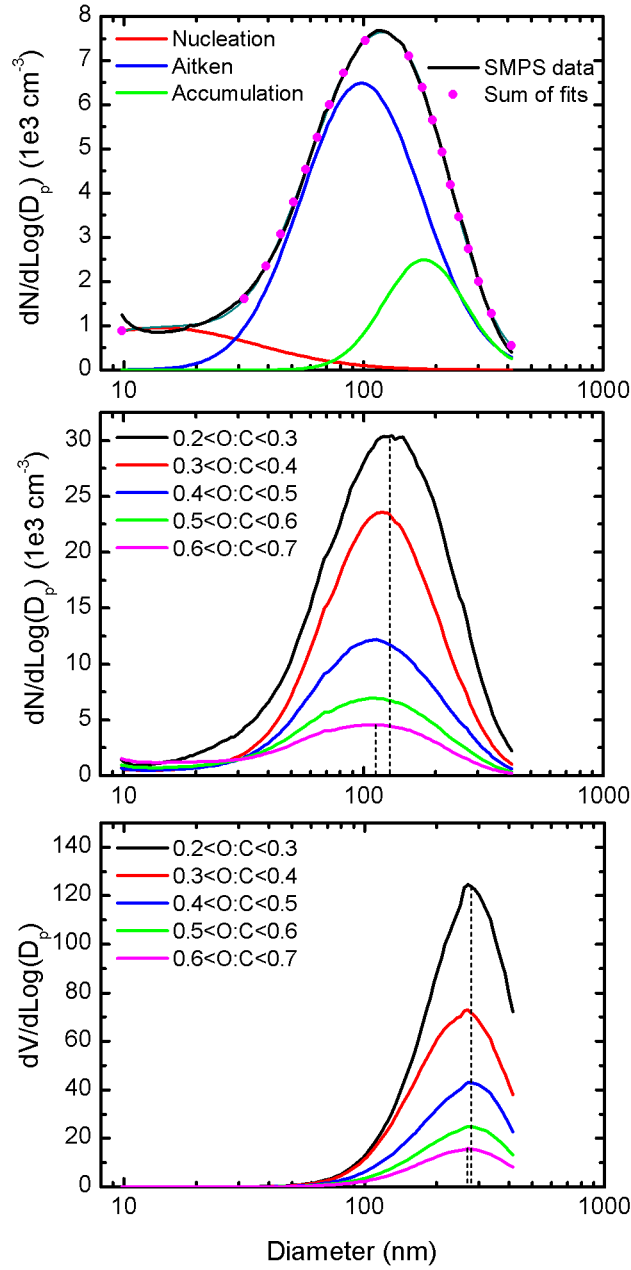


Fig. 9. Top: average aerosol size distribution observed at ground level during SAMBBA. The average particle number concentration and diameter were $5\,800\,\text{cm}^{-3}$ and $94.4\,\text{nm}$, respectively. Log-normal fit parameters are shown in Table 2. Middle: solid line shows the average number size distribution based on different O:C ranges. The dashed line indicates size distribution centroid for fresh ($130\,\text{nm}$) and aged aerosol ($110\,\text{nm}$). Bottom: solid line shows the average volume size distribution based on different O:C ranges. The dashed line indicates size distribution centroid for fresh ($278\,\text{nm}$) and aged aerosol ($267\,\text{nm}$).

Supplemental material

Figure S8

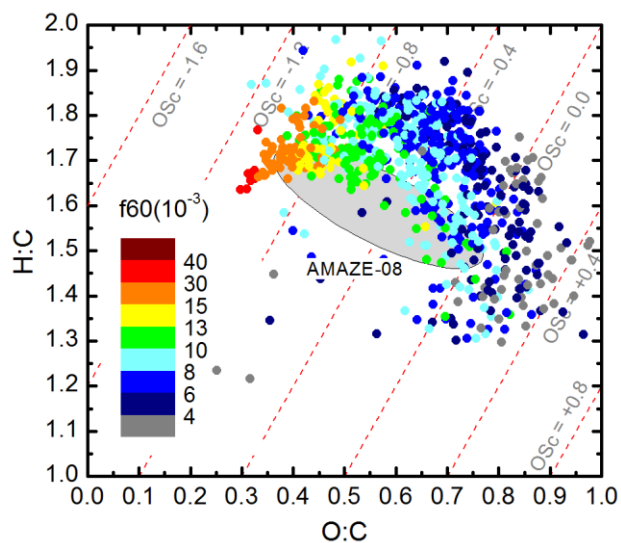


Fig. S8. Similar to Fig. 07, taking into consideration the newly proposed corrections for HR-AMS O:C and H:C ratios according to Canagaratna et al., (2014). For consistency the AMAZE-08 V-K area based on Heald et al., (2010), shown here, has been corrected by similar factors.