

Interactive comment on “Ground based aerosol characterization during the South American Biomass Burning Analysis (SAMBBA) field experiment” by J. Brito et al.

Anonymous Referee #1

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Comments on "Ground based aerosol characterization during the South American Biomass Burning Analysis (SAMBBA) field experiment" by Brito et al.

General

This paper describes the results from the composition analysis and physical measurement (number size distribution) of submicron aerosol emitted from biomass burning (BB) during the ground based observations as a part of “SAMBBA” campaign. The temporal and qualitative variability of organic aerosols (OA) measured using Aerosol Chemical Speciation Monitor (ACSM) with respect to the chemical aging was in detail

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investigated in this study. Mass spectral analysis provided insights into the OA aging process such as SOA formation and changes in elemental composition. The major results and discussion in this study meet the scope of ACP. Despite the significance of this study, there are some points to be addressed before accepting the manuscript. Please consider the following comments.

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.

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 deltaOA. How does the constant CO_{bg} affect the calculation of the deltaOA/deltaCO ratio? As

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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.

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.

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.

Individual comments

P12284, L16-18.

What does this sentence actually mean?

P12285, L24 - P12286, L2.

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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?

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 0.4-0.6 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

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Middlebrook et al. (2012).

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).

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.

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.

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.

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

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into the new subsection.

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