

Interactive comment on “Sources of carbonaceous aerosol in the Amazon Basin” by S. Gilardoni et al.

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Anonymous Referee #2

General comments: The overall goal of this manuscript is to obtain insights into properties and sources of carbonaceous aerosols in the Amazon region. A major concern, however, with the application of PMF to thermograms is that the number of variables is very limited and lower than, for example, in the case of IR and AMS data. This implies that detailed insights into sources of carbonaceous aerosols are very difficult to achieve; for example, biogenic and anthropogenic SOA cannot be differentiated. This reviewer already provided detailed comments on a previous version of this manuscript, which have been adequately addressed in the current version.

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We agree with the referee about the limitations of the thermogram PMF analysis. Nevertheless, this method offers the opportunity to differentiate among carbon classes that showed clearly different thermal behaviors, like biomass burning and secondary aerosol, which are expected to dominate the fine carbonaceous aerosol at the study site. The following sentence is added at page 29948 line 3:

“PMF analysis of thermograms was applied for the first time to the characterization of carbonaceous aerosol sources in ambient air. Although this approach can be used only to differentiate carbon classes that showed clearly different thermal behaviors, it allowed us to give an estimate of biomass burning and SOA contributions to fine carbon mass and of PBAP to coarse carbon mass”

Additional specific comments: Page 29926 – lines 19-21: The statement made about SOA is too simplistic and does not reflect the state of current knowledge: “SOA are formed by nucleation and condensation of oxidation products of VOCs, such as isoprene and terpenes, and they represent a significant fraction of fine aerosol mass.” Precursors for biogenic SOA in the Amazon region include terpenes, which give rise to SOA by nucleation and condensation of oxidation products. A major SOA precursor is isoprene; however, SOA from isoprene is formed through another mechanism, namely, heterogeneous reactions of volatile oxidation products (epoxydiols). See the recent article by Surratt et al. (Proc. Natl. Acad. Sci. USA 107, 6640-6645, 2010). SOA is definitely also from biomass burning origin, formed by oxidation of volatile compounds such as aromatic compounds emitted during the fires. See, for example, the article by Iinuma et al. (Environ. Sci. Technol. 44, 8453-8459, 2010). In addition, there is evidence that biomass burning is an important source of dicarboxylic acids, and that low-molecular-weight dicarboxylic acids have mainly a secondary origin in biomass smoke (Kundu et al. ACP 10, 2209-2225, 2010).

The authors thank the referee for pointing out the inaccurate description of SOA formation. The following sentence is introduced at page 29926 line 19:

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“SOA represents a significant fraction of fine aerosol mass. SOA is formed by nucleation and condensation of oxidation products of biogenic VOCs, such as isoprene and terpenes. In addition, particle-phase reactions (Surratt et al. 2010) and aqueous phase oxidation of organic precursors (Lim et al. 2011) from biogenic or anthropogenic sources, including biomass burning, could contribute to SOA formation in the Amazon region”

Page 29944 – lines 18-19: The statement “Over the entire campaign 43% of fine carbon mass was due to biomass burning, 34% to SOA, and 23% to volatile species” is misleading. Considering that part of the SOA has a biomass burning origin and the volatile species have a possible link with smoldering emissions it is likely that the fine carbon mass that is due to biomass burning is much higher than 43%.

We acknowledge the referee for pointing out the need for clarification. The following sentence was added at page 29944 line 21:

“Emissions from biomass burning can lead to the formation of SOA (quantified by factor 1), as well as to the presence of volatile species (quantified by factor 3); thus the biomass burning PMF factor represents a lower estimate of the total biomass burning contribution to the fine carbon content”

References

Lim, Y. B., Tan, Y., Perri, M. J., Seitzinger, S. P. and Turpin, B. J., Aqueous chemistry and its role in secondary organic aerosol (SOA) formation, *Atmospheric Chemistry and Physics*, 10, 10521-10539, 2010.

Surratt, Jason D., Chan, Arthur W. H., Eddingsaas, Nathan C., Chan, ManNin, Loza, Christine L., Kwan, Alan J., Hersey, Scott P., Flagan, Richard C., Wennberg, Paul O., Seinfeld, John H., Reactive intermediates revealed in secondary organic aerosol formation from isoprene, *Proceedings of the National Academy of Sciences*, 107, 6640-6645, 2010.

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