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Interactive comment on "Organic aerosol components derived from 25 AMS datasets across Europe using a newly developed ME-2 based source apportionment strategy" *by* M. Crippa et al.

Anonymous Referee #1

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This manuscript provides an overview of organic aerosol components obtained from AMS data across 25 European sites. The new aspect of this work is that it uses multilinear engine (ME-2) to specifically extract and apportion the observed OA due to primary organic aerosol components from traffic (HOA), burning (BBOA) and cooking (COA). Marine organic aerosol is also extracted in certain sites.

I commend the authors on their work compiling all this information together and I think the subject matter is suitable for ACP. However, I think that there are several details that the authors need to fully address before the manuscript is published. These details deal with the two goals that the authors lay out in the abstract: 1) To propose a standardized methodology for using ME-2 for extraction of organic aerosol components from AMS data 2) to provide a large overview of organic aerosol sources in Europe.

STANDARDIZED METHODOLOGY

1)Justification of a-priori information

Since the ME-2 method depends on a-priori information, the authors need to provide more justification for how and why they constrained the ME-2 runs in the ways they did. More information needs to be provided about how the standard spectra were chosen, the uncertainties associated with them, and also how the a-values were chosen. Currently there is very little context for this. Even if this information is in other manuscripts that are referenced, it should be summarized in this one to make this manuscript clearer.

a) The authors choose the Crippa et al. HOA and COA and the Ng et al. BBOA. How do the results change if a different set of starting mass spectra was used? For example, how stable are the results if the Ng et al. or other HOA was used? How similar are the Crippa et al. HOA to direct vehicle emission source mass spectra that have been observed for European vehicles?

b)There is very little discussion of the a-factor and why the values that are used were chosen. Are the a-factor values chosen for HOA reasonable when considering the variabilities among HOA across the world? In the spectral database, Ng et al. HOA reference spectrum mass spectrum, for example, has reported standard deviations for most of the key ions that are at least 20% if not greater of the measured ion intensity. How does the a-factor that you have chosen compare to these variabilities? Why were the a-factors that are being used chosen? What happens to the extracted HOA loadings if the a-factors are increased to 0.3 as in the case of the BBOA a-factors?

c) Is the a-factor applied uniformly across the entire mass spectrum? What uncertainties does this introduce given that the spectral database reference mass spectra clearly

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have standard deviations that vary with m/z.

d) The authors mention in the first paragraph of p. 23348 that the a value of 0.3 is found to be reasonable for the BBOA MS. They further mention that it is hard to determine when the BBOA factor fails when increasing the a-value. This sounds troubling. If you don't have a metric to judge whether the results that are obtained from the analysis is reasonable, how do you choose an a-value? Could you not use the variabilities observed in source BBOA signatures to define the a value? The authors mention the variabilities, but don't quantify them in any way. How does the a-value that you have chosen for BBOA compare with the observed variabilities in BBOA, particularly since the BBOA factor accounts for all kinds of burning.

2) Validity of extracted components

a) The technical guidelines for standardized strategy should include more details on how to validate whether the extracted components are reasonable or not. In step 3 the authors recommend looking at diurnal increases to identify wood burning. Isn't this going to be complicated by the diurnal changes in boundary layer height as well? Also, if PMF could not extract the components distinctly, there is likely not enough distinct temporal variation between the components to start with that can be used for this purpose.

Since the extracted components are primary components, couldn't source emission ratio information be used to help guide the interpretation of whether component apportionments are reasonable or not? For example, HOA/CO, HOA/BC and HOA/NOx values for source emissions exist in the literature. What are these ratios for the extracted components in the various sites and how do they compare to these known values? A compilation of at least some of these values for the dataset you have could be very useful.

Similar information can likely be obtained for BBOA/CO and BBOA/BC. The f60/BBOA ratio is a good start at this as well, but isn't this correlation primarily driven by the allowed variability in the f60 of the assumed BBOA mass spectrum in the first place?

3) Uncertainties in Source Apportionment

The sensitivity analysis to the a-value that is presented in section 4.5 does not seem to be complete since it really only tests for the sensitivity to small changes in the a-value used for HOA and does not really address the variability in the BBOA factor MS which could be large at many of the sites.

Some suggestions on how the uncertainties could be quantified:

a) It is not clear to me how the mass is redistributed as additional factors are added to the analysis. For example, how much cross talk is there between HOA and BBOA vs. HOA and SV-OOA etc.. It would be good to see for at least one case study (likely Barcelona) how the mass apportionment changes as you go through the steps suggested in your methodology.

b) In Figure S1-3.2 it seems that the agreement between ME-2 HOA and HR-PMF HOA and BBOA is quite poor (correlation slopes of 1.9 and 0.5 respectively). There is a similar difference in the SV-OOA and LV-OOA assignments as well. It is clear that the apportionment is not unique and there is quite a bit of variability. How do the authors pick which is the most likely apportionment? In this manuscript the authors want to make the case that ME-2 is capable of extracting the more "reasonable" answer. The authors should discuss this in more detail and explain why they think the ME-2 solution is better than the one obtained with HR-PMF.

c)In the extreme case, one limit on the apportionment would be to always run an ME-2 with the constrained (with reasonable a-factors) HOA and BBOA factors and see what comes out compared to what you get from PMF and your methodology for ME-2 that you propose. This would give information about how much error one could potentially have if you constrained both factors in a situation where you didn't need to and would also provide information on how coupled these factors are and also on uncertainties

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with which zeros in concentrations of either of these factors could be obtained.

d) The reference MS of the HOA and BBOA appear to correspond to "fresh" emissions. Due to the constraints of the a-factor, presumably aged HOA could appear as an OOA factor rather than as an HOA factor with a larger f44. Similarly aged BBOA could appear as BBOA and another OOA. In order to disentangle this, it would be useful to check correlations between the OOA components and the primary components. Do you see evidence for this at any of the sites?

INTERCOMPARISON ACROSS EUROPE

4) While data is presented for all the sites, The discussion regarding the overview of organic aerosol sources in Europe is limited. There is a lot of information and it would be useful if the authors presented more context to the trends between sites given overall meterorology and source regions that impact the various sites.

a) One way of giving context could be ordering the sites in tables and figures according to some consistent metric along which trends could be discussed (Rural/Remote/Urban or North/South or Loading levels)

b) It is surprising that Puijo has nearly the largest HOA fraction even though it is classified as rural. Does this make sense with respect to the source regions that are close to this site? A similar question for the rural VHL site where HOA when 0.2 of the mass and CBW where it is 0.15 of the mass. The issue behind these questions is whether HOA is overestimated at these sites perhaps due to misapportionment from BBOA or SV-OOA or due to the chosen HOA profile. As mentioned earlier, one way to check whether the HOA is in a reasonable range would be to check against CO or NOx and see whether the HOA/CO ratios are consistent with vehicle emissions.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 23325, 2013.