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Interactive comment on "Source apportionment of organic aerosol from two-year highly time-resolved measurements by an aerosol chemical speciation monitor in Beijing, China" by Yele Sun et al.

Anonymous Referee #2

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Review of Sun et al. "Source apportionment of organic aerosol from two-year highly time-resolved measurements by an aerosol chemical speciation monitor in Beijing, China"

The authors present two-year measurements on organic aerosols (OA) in Beijing with an aerosol chemical speciation monitor (ACSM). Source apportionment was performed using multilinear engine (ME-2). Five factors, including fossil fuel organic aerosol (FFOA), biomass burning organic aerosol (BBOA), cooking organic aerosol (COA), less oxidized oxygenated organic aerosol (LO-OOA), and more oxidized oxygenated

organic aerosol (MO-OOA). Based on the source apportionment results, seasonal variations, loading dependence (Section 3.2, see suggestion on change of section title below), RH/T dependence, and potential source regions, were thoroughly discussed in view of emission and formation of OA in Beijing. The dataset is highly valuable in view of its long duration and deep analyses. The analyses are rigorous and the manuscript is generally well written. This work is certainly within the scope of ACP and of interest to readers of ACP. I do, however, have a few points for the authors to address, as below.

Major comments:

1. Mixing of factors. It is understood that statistical models like positive matrix factorization (PMF) suffer from this problem, even with ME-2, and especially with less chemically resolved ACSM. The mixing of hydrocarbon-like organic aerosol (HOA) and coal combustion organic aerosol (CCOA), resulting in FFOA, is a well justified one. But there are still a few other complications. For example, in page 9 line 10 and line 20, the authors admitted that FFOA/COA and BBOA/LO-OOA pairs might not be well resolved. Since the entire paper is based on the quantitative fractionation of OA into those factors, one might be skeptical about how well PMF/ME-2 can separate those distinct primary sources (FFOA/COA) and changing primary/secondary OA (BBOA/LO-OOA). In order not to let readers take those numbers (i.e., mass concentrations and mass fractions of different OA factors) for granted, at lease a caveat has to be mentioned about the potential uncertainties in separating OA factors using ACSM data and PMF analysis.

Better yet, further exploration can provide some quantitative estimates on the uncertainties. Take the FFOA/COA pair for example. FFOA (either as HOA or CCOA) has associated markers (BC and/or Chl) that might be strongly correlated with it (HOA or CCOA) according to previous studies. If using data points from periods with minimal COA contribution (e.g., morning rush hours or after midnight), one can get good correlations and scale factors between FFOA and BC/Chl in non-cooking periods. In the case of FFOA in summer, which is suspiciously mixed with COA, one can use the

scale factors to estimate the "real" FFOA during cooking periods. Although the so-called "real" FFOA is even less rigorously obtained compared to that from PMF/ME-2, one can at least have a quantitative understanding on how much difference can it be between the FFOA obtained from these two methods, serving as an uncertainty for PMF analysis.

2. PSCF analysis. The potential source analysis on primary (assuming unchanged) is reasonable, but might be easily over-interpreted. For instance, the author stated that COA should not be regional, but its "potential source" can extend to very far a distance from the sampling site (to the Bohai Sea in Sp12 and S12 in Figure 8, and distinct hot spots southwest of the city in F11 and Sp13). The same analysis applied to secondary factors might be even more difficult to apprehend. In that type of analysis, does it mean the precursors are from those regions or the oxidation occur in those regions? Again, some precautions should be mentioned to avoid over-interpretation of PSCF results, which only provide a very rough estimate on the coupling of air mass transport and chemical species.

3. Conclusions. With a number of aspects discussed, the manuscript presents a number of conclusions. It is difficult to pin-point what new findings were obtained with this two-year dataset (very valuable, indeed), and how much this work is different from other AMS studies in Beijing, many of which were done by the authors. I strongly suggest the authors to distill the conclusions into one or two major leaps that this work achieves compared to other one-month or even one-year measurements.

4. Some obstacles in smooth reading. There are a number of places that requires careful grammatical check. Below in the minor comments are a few examples I spotted. More thorough checking will help increase the readability of this paper.

Minor comments:

1. Page 2, line 22. "real-time" to "real time".

2. Page 2, line 24. I don't think PCA/PMF/ME-2 can "differentiate" OA factors from sources/processes. They can resolve the OA matrix into different factors, which correspond to different sources/processes.

3. Page 2, line 30-31. Two sentences here that need splitting.

4. Page 3, line 8. "one year" to "one-year".

5. Page 3, line 9. "season variation" to "seasonal variation".

6. Page 3, line 11. "this study". Which study? Sun et al. (2015)? Zhang et al. (2013)? Or Hu et al. (2017)? Are all these three studies lack of seasonal variations of OA?

7. Page 3, line 13. "two years'" to "two-year", and other few places using the same form.

8. Page 4, line 9-11. It is ambiguous here. Should be "...and default relative ionization efficiencies (RIE) were used, except for ammonium whose RIE was determined from measurements with ammonium nitrate."?

9. Page 4, line 18. "mass resolution" to "low mass resolution".

10. Page 5, line 11. "the seasonal ME2-ACSM reports" looks odd. Should be "the seasonally average SOA concentrations are overall 16% higher using PMF/ME-2 analysis with the ACSM dataset compared to those using conventional PMF analysis."?

11. Page 5, line 14. "ME-2 analysis" to "from ME-2 analysis".

12. Page 5, line 15. "during the first eight months' measurements" to during the first eight months".

13. Page 5, line 16. "during the rest of months" to "during the other months"; "with differences less than 3%" to "with differences of less than 3%".

14. Page 6, line 14. "which is much higher" to "which are much higher".

15. Page 7, line 1-2. This can be incorrectly understood as COA is more important

than traffic emissions in all time in Beijing. Suggest to put "in non-heating seasons" to the second half of the sentence.

16. Page 7, line 15. "much differences" to "many differences".

17. Page 8, line 3 – 22. The first and the last sentences of this paragraph looks contradicting. Does MO-OOA have a pronounced seasonal variation or not?

18. Page 9, Section 3.2. Suggest to change the section title to "Loading-dependent OA composition" because it is basically what this section is about.

19. Page 10, line 14. "during lunch and dinner times respectively" to "during lunch and dinner times, respectively".

20. Page 11, line 25. "at $\sim 50 - 60\%$" to "to $\sim 50 - 60\%$".

21. Page 12, line 17. FFOA emissions or FFOA formation? I believe FFOA is primary (emission).

22. Page 12, line 24. "dependence" to "dependences".

23. Page 14, line 6. "surprising" to "surprisingly".

24. Page 14, line 7. Delete "we".

25. Page 15, line 10. The previous discussion stressed on the constant mass concentrations of COA, not constant mass fractions. And the authors stated previously that COA mass fractions increased during clean periods. Should this be modified to be consistent with the points made in the discussion?

26. Page 15, line 26. "correlated"? The authors used a present tense almost throughout the whole discussion. How come a past tense is used here suddenly? Is it "is correlated"?

27. Page 25, Figure 1-c1. "FOA" to "FFOA".