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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Technical Note: A new approach to discriminate different black carbon sources by utilising fullerenes and metals in Positive Matrix Factorisation analysis of High-Resolution Soot Particle Aerosol Mass Spectrometer data" *by* Zainab Bibi et al.

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This paper presents a new approach by using fullerenes and metals in PMF analyses of HR-SP-AMS data and shows that BC from more than one biomass burning sources can be separated. The work can be a good contribution to the aerosol chemistry, however, there are quite a number of important issues to be addressed first before considering its possible publication in ACP. Please see below:

(1) This work uses fullerenes and metals to help the BC source apportionment via SPAMS, however, it is surprised that the authors seem to completely overlook a few important papers very closely related with your current analyses. Necessary citations and incorporation of findings from these studies has to be added in your work. And I also suggest the authors to do a more thorough search on the recent SPAMS studies. These papers at least include: Distribution of carbon cluster ions in different BC types (Aerosol Sci Tech 2015;49:409-422); detection, quantification and source apportionment of fullerenes by using SP-AMS (Environ Sci Technol Lett 2016,3,121-126); Characterization of OA via BC fragments and metals detected by SP-AMS (Atmos. Chem. Phys., 20, 5977–5993, 2020); Source identification of BC by using SP-AMS (Atmos Environ 2018,185,147-152); Another study combining SP-AMS and SP2 data to apportion BC into different sources (Atmos Chem Phys 2019,19,6749-6769).

Ans: Thank you so much for your feedback and suggesting me some new research articles to include in my journal. I will consider your suggestion and add these citation work in my manuscript.

Onasch, T.B., Fortner, E.C., Trimborn, A.M., Lambe, A.T., Tiwari, A.J., Marr, L.C., Corbin, J.C., Mensah, A.A., Williams, L.R., Davidovits, P. and Worsnop, D.R., 2015. Investigations of SP-AMS carbon ion distributions as a function of refractory black carbon particle type. Aerosol Science and Technology, 49(6), pp.409-422.

Wang, J., Onasch, T.B., Ge, X., Collier, S., Zhang, Q., Sun, Y., Yu, H., Chen, M., PreÌĄvoÌĆt, A.S. and Worsnop, D.R., 2016. Observation of fullerene soot in eastern China. Environmental Science & Technology Letters, 3(4), pp.121-126.

Carbone, S., Onasch, T., Saarikoski, S., Timonen, H., Saarnio, K., Sueper, D., Rönkkö, T., Pirjola, L., Worsnop, D. and Hillamo, R., 2015. Characterization of trace metals with the SP-AMS: detection and quantification. Atmospheric Measurement Techniques Discussions, 8(6).

Liu, D., Joshi, R., Wang, J., Yu, C., Allan, J.D., Coe, H., Flynn, M.J., Xie, C., Lee, J., Squires, F. and Kotthaus, S., 2019. Contrasting physical properties of black carbon in urban Beijing between winter and summer. Atmospheric Chemistry and Physics, pp.6749-6769.

(2) Line 115: What is the role of a catalytic stripper? And why data under this mode is not used then?

Ans: Thanks for asking. The catalytic stripper was used in the setup and has already published in Danton Liu's paper. It was not included for this research work publication. The only reason to quote in my manuscript is, it's been used in another study. I have also tried analysing catalytic stripper data but did not get sensible results. So, I have excluded that data.

(3) Line 120: Why no ionization efficiency was not obtained? Due to what difficulty? If other studies can determine the IE, then why this work cannot? It is not explained clearly to the readers. If the IE or RIE of different species, especially fullerenes, are not determined, this is a fatal issue. This reviewer doubts the credibility of PMF results and subsequent analyses.

Ans: Thank you so much for your comment. At the time, we tried but it did not work properly so we cannot go back and recalibrate. Also, RIE did not affect PMF analysis results and it's not at all fatal issues.

(4) Section 2.2: Some necessary technical details are still missing here: what is the chemical resolution, and how does this allow you to detect the ions with low signals? For example, different metals? What is the time resolution? And what are the detection limits of different species by using SP-AMS? At last, did the SP-AMS here only detect BC-containing particles? Then it is not clearly specified that the SP-AMS is operated with only laser vaporizer? Or with laser and tungsten vaporizer? Is the thermal vaporizer physically removed? The different modes significantly influenced the aerosol components detected (Check and cite if necessary: Atmos. Meas. Tech. 2014, 7, 4507−4516; Atmos Chem Phys 2019,19,447-458)

Ans: In terms of technical details, High Resolution SPAMS detected the metal signals such as Sr, Fe, Cs and Ti. The time resolution, as I have mentioned in my manuscript was from 29th Oct-11th Nov 2014 (line 94). Also, I have clearly stated that in this study, the SP-AMS was only operated with the laser vaporiser only in combination with the electron impact source and measures both refractory and nonrefractory components along with metal nanoparticles and it's also cited (Onasch et al., 2012).

(5) Section 2.3: m/z up to 3200 was determined, however, the m/z range of your PMF results is only up to 1000. Can you explain?

Ans: Thank you so much for your comment. The reason for 1000 m/z range of this PMF analysis is that above m/z 1000 the signals were not approachable and it was only noise so that's why we have ignored them and only added m/z up to 1000 by clearly showing the separate BC sources.

(6) Figure 4. More details are needed. How did you determine BrC for example?

Ans: The Aethalometer AE31 was used to determine BrC and it has already published in Reyes et al., 2018.

(7) Figure 5. How to determine PON etc? A citation of previous work is not enough.

Ans: Particulate organic oxides of nitrogen (PONs) were estimated using the m/z 46 : 30 ratios from aerosol mass spectrometer (AMS) measurements, according to previously published methods (published in Reyes et al., 2018).

(8) Figure 6. You have a high-resolution SP-AMS, why not use different colors to differentiate different ion categories? (Figure 7 is redundant). This helps justification of your PMF results and better for readers to judge and understand your results. Also, the justification of your PMF results is not clear. I am not sure for example, why factor 5 is fullerenes, and why factor 1 is BC and HULIS, and so onïij§ The O/C and H/C are too high, and they seem to be wrong. This has to be addressed well, otherwise, analyses based on the PMF are not trustworthy.

Ans: Thank you so much for the suggestion. Sure, I will use different colours to differentiate the ion categories. I have added some more details to explain why factor 1 is BC and HULIS and others as well. Yes, you are right, I have mistakenly added the wrong values of O:C but H:C values are correct. Thank you so much for pointing this, I have now added the correct values.

(9) How about the diurnal patterns of your factors?

Ans: The data for this study is only for few days, and it is already obvious from the time series that the signals were very high during the bonfire night specifically, when the fire log burning, and fireworks started. And there is no point of doing diurnal plots because it is obvious that it will be dominated by bonfire night.

(10) Figure 9 is not clear, please replace with a high resolution one Ans: Thank you so much for pointing this out. I have replaced figure 9 in manuscript.

Please also note the supplement to this comment:
https://acp.copernicus.org/preprints/acp-2020-890/acp-2020-890-AC1-supplement.pdf

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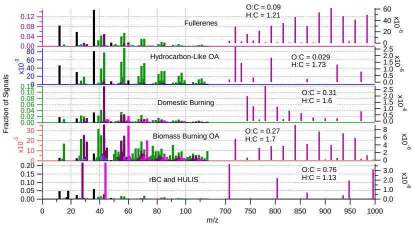

Fig. 1. PMF five factors source profile (factor 1 = BC and HULIS, factor 2 = BBOA, factor 3 = Domestic burning OA, factor 4 = Hydrocarbon-Like OA, factor 5 = Fullerene). Note the difference in scales of the f

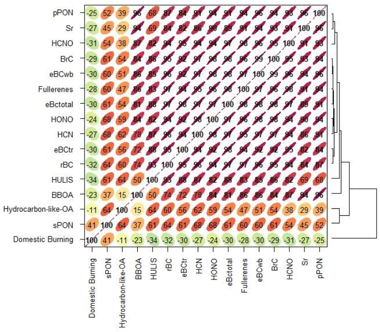

Fig. 2. The similarity between different pollutant time series through hierarchical cluster analysis (HCA)