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Atmospheric Chemistry and Physics Discussions

Interactive comment on "New insights into $PM_{2.5}$ chemical composition and sources in two major cities in China during extreme haze events using aerosol mass spectrometry" *by* M. Elser et al.

Anonymous Referee #2

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Authors present a very interesting AMS (Aerosol Mass Spectrometer) data set obtained in two major cities in China (Xi'ian and Beijing) during winter 2013/2014. The field campaign was characterized by 2 (or 3) extreme haze events with PM2.5 concentrations up to 1000 μg/m3. During these haze events about 40% of the PM2.5 mass concentration is in the 1-2.5 μm size fraction, which underscore the relevance of PM2.5 aerodynamic lens inlet in such heavily polluted environments. The authors adopt a rigorous source apportionment strategy in order to reduce the subjectivity of the choices that must be done at different steps of the data treatment and to improve the representability of the solutions.

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Discussion Paper

Overall the paper is well written, well-illustrated, the methodology is robust and the results present a real interest for the scientific community. One can just regret the lack of ancillary measurements such as offline chemical PM analyses (OC/EC, major ions) or SMPS/OPC measurements. This paper should be accepted on completion of the minor revisions/clarification requested below.

P30134 line 25. Clarify the position of the nafion drier (ie. in the 4 L/min primary line or after the split to the AMS line, not clear)? As RH can play an important role in terms of aerosol size distribution what was the RH after the nafion dryer during haze events?

P30135 line 10-20, comparison with offline gravimetric measurements. I'm surprised by the difference between offline gravimetric measurements and AMS+Aethalometer observed during the haze period which cannot be explained by "deposition of dust and waters on the filters"(remove). The samples were collected at ambient temperature or in heated shelter?

P30138 line 19. What is j in eq (3)?

P30138 line 2: Among traffic sources, Diesel LDV/HDV are obviously the main BC emitter, but I suggest to change "diesel engines" by "traffic".

P30138 line14-30 and P30139 first §. This section is very interesting and I suggest to go further in the discussion and to provide more details. Regarding the multilinear approach did the authors mix the two data sets (Xi'ian and Beijing)? If yes, as coal could be quite different from one region to another, is there a difference in terms eBC/CCOA between the 2 cities (by applying the same methodology to each dataset separately)? Why did the authors choose to keep the results from the Aetholometer models instead of the results obtained from the multilinear approach which should provide the eBC contribution from coal combustion, fossil fuel combustion (derived from HOA) and wood burning?

P30139 line 8. Considering DeWitt et al 2015 (ACP) a ratio BC/HOA of 0.79 should

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correspond to a % of diesel fuel consumption of about 30-40%. Does this make sense in China?

P30139. PAH quantification. About half of the m/z listed are not molecular ions. Moreover as PAHs are a vast family of compounds (without considering alkylated PAHs nor oxygenated nor nitrated PAHs) and as the PAH concentrations reported here are very high (!) and one of the main point developed in the discussion, more details are necessary in this section. Especially it's important to establish a rough correspondence between the ions considered for the quantification and the PAHs or nitroPAHs (lot of common fragments between these two subfamilies). Such correspondences are not easy to get precisely and will be subjected to uncertainties, but it's important in order to compare with the literature and to fix the limits of the compounds actually quantified or not considered here in the quantification (again the PAH family is vast).

P30140-30145 Source apportionment Optimization (general). This section is undoubtedly the most innovative part of the paper. The methodology adopted by the authors to minimize the subjectivity of the solutions is scientifically robust and interesting from a conceptual point of view. My main question is what are the differences in terms of source contributions or external parameters correlations (ie. eBC) between the 5 factors unoptimized solution and the optimized one? In others words, are the differences significant?

P30141 line 15 and 22. HOA and COA profiles used to constrain the ME2 model were obtained in Paris with an unconstrained PMF approach. Can you add few words discussing the representativity of those source profiles considering that the vehicular fleet is potentially significantly different as well as cooking activities (despite the COA profile was obtained in the Paris Chinatown). The use of a values allows to minimize this potential issue of representativity of the source profile, but here I'll provide more information about how the use of a values improved this representativity under the light of the results obtained with the unoptimized solution.

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P 30145 line 25. This information ("analyses were conducted separately for the four periods") should be given at the beginning of this section. I assume that the discussion and the illustrations (fig 1, 2 and 3) related to the solutions optimization is for one of the period. Specify which one or clarify. Also the haze period in Beijing is really short, do you observe any discontinuity with the non-haze period?

P30147 line 27. A standard deviation can be considered as an error only if we expect equal values (which is not the case here).

P30148 line3. True considering absolute concentrations, but the relative contribution of NH4 decrease during haze events. How about the ionic balance? In such environments and conditions, acidic properties of aerosol is of great interest (SOA formation pathways etc.).

P30152-P3053. Not sure that evolutions of the absolute concentrations or contributions vs RH are useful here. From the results presented here it seems that the aerosol acidity strongly increase during haze periods. I suggest to add the ionic balance of the aerosol in figure 8b. Also such conditions (high RH, high SO4 and very high OA concentrations) are ideal to have a careful look to the organo-sulfur fragments. Do you observe any of those specific fragments during the field campaign and especially during haze periods?

P 30154 PAH sources. As stated above the PAH concentrations reported here are very high. A rough calculation shows that PAHs contribute to few % of the OA mass concentration (1-5%) which is really high (!). In Europe or US, PAHs contribution to OA is typically in the range 0.01-0.1 % maximum. It seems also that the PAH contribution to OA is much higher in Beijing than in Xi'ian, most probably due to coal emissions. I suggest to the authors to put the PAH concentration and/or PAH contribution to OA into perspective with literature data (ambient and source).

I guess, it isn't possible to extract the PAH signatures (f(m/z)) of the different sources (traffic, coal and BB) from your analysis. But if I'm wrong, this information could be very interesting.

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P30156 line9-11: This sentence is apparently in contradiction with the sentence P 30145 line 25 (“analyses were conducted separately for the four periods”)

Figure 4. I’d add the visibility shown in fig S9.

Fig 6 (A) : Difficult to see the comparisons with “external” parameters. Try to make these figs clearer.

Fig6(B) Add the total OA concentration above each pie chart

Fig 7 Legend not readable in my printed version

Fig 10B Add the total PAH concentration above each pie chart.

In the SI or in the main text, I’d add a table summarizing all relevant concentrations (OA, NO3, SO4, NH4, BC, HOA, BBOA, CCOA, COA, OOA, PAH, ..).

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 30127, 2015.