

Interactive comment on “Source Apportionment of Urban Particulate Matter using Hourly Resolved Trace Metals, Organics, and Inorganic Aerosol Components” by Cheol-Heon Jeong et al.

Anonymous Referee #2

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General comments The paper is focused on source apportionment by Positive Matrix Factorization of hourly resolved particulate matter sampled in Toronto (Canada). The topic is interesting, the data set is large. Obviously the joint use of metal, BC and ACSM data should give better results respect to the use of data from a single instrument; however, the joint use of data coming from different instruments in the PMF analysis it is not trivial and there are many inconsistencies in the reported factor profiles and in the interpretation of the results; furthermore, many important information are missing and the results of the PMF analysis for the identification of the sources is less clear respect to a traditional 24 h based study. Therefore, I cannot suggest the article for publication.

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Specific comments: 1) The uncertainty of each metal is normally related to the square root of the number of counts in the peak; it depends on the number of counts in the peak, therefore it is not fix for each element, it varies from spectra to spectra and it cannot be obtained from the measurements on a standard (formula (1) S.I. pag.1); this method clearly underestimate the uncertainty (which is a crucial parameter for PMF analysis) 2) PM2.5 Mass concentrations were measured at the 4th floor rooftop. Nothing is said about the positioning of the other instruments except they are 15 m apart from a main road. At which height? This is a crucial information. If the PM2.5 monitor and the other instruments are not positioned at the same height, the bias in the quantitative PMF results is relevant. 3) In my opinion, the combination of PM1 data for ACSM and PM2.5 data may introduce artefacts in PMF analysis. The reduction of the CE down to 0.5 may be arbitrary. The PM2.5 to PM1 may be compound or day dependent. The authors present the regression for the winter period, but the results could be different in the summer period. 4) The combination of stable markers like the metals and the organics (that maybe unstable at higher temperature) is an open problem in the PMF analysis; this may produce artefacts like the presence of factors which are simply related to the difference in the ratio between some markers in different seasons. The authors do not discuss such a problem that can be the origin of some of the problems in their results 5) Some important information are missing to have an idea of the quality of the PMF results. The graph of the measured PM2.5 mass vs the reconstructed PMF mass must be inserted at least in the SI. Another necessary information is a table with the percentage of the mass of each element or compound explained by the PMF (by all the identified sources) respect to the measured element/compound mass (which is something different from what is reported in table 2). Furthermore, which is the percentage of the mass of each of the identified sources explained by the measured elements/compounds? If all the elements/compounds present in the source are measured this percentage must be 100%, otherwise less (e.g. for the road dust the oxygen from the oxides of Si, Ca, Fe, Ti is not measured) but sometimes PMF overestimates the mass of the source with a percentage above 100%. The sum of

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the percentage due to all the sources reported in tables 2 and 3. is 100%. Which is the percentage of the unexplained mass? It is quite strange if PMF reconstruct 100% of the mass. Without all these information, it is impossible to understand how good the PMF results are. PMF is a powerful instrument but it must be used with caution and many checks must be made before giving the results. All these information are missing. Finally, looking at fig. 1 I can see that the error bars for the contribution of all the typical markers are very high; this is unusual; I suppose therefore that the error on the contribution of each source, too, is very high, but it is not reported in any of the tables. 6) The identification of the road dust source is questionable. The error bars for all the characteristic markers are very high. The presence of V (50% of its mass in this source is unusual). The percentage of OA is very high compared to what is normally found in many articles (and higher respect to the primary vehicle emission source). No BC is present in this source at variance with the articles reported as references. The time trend is not similar to that of traffic or of the typical traffic related gases. There is a similarity just at the beginning of the morning, but no decrease after the rush hour and it start decreasing at 3 pm while traffic start decreasing at 6 pm. Finally, it is known that the contribution of the road dust is mainly in the coarse fraction while the exhaust component is mainly in the PM2.5 fraction. It seems quite unreliable that the road dust contributes similar to the exhaust in the PM2.5 fraction or less if we consider also the tire wear source (see below for the problems in the identification of that source). This is probably due to the weakness of the profiles of the identified sources or to the fact that this source is not a traffic related source. In the reference Amato et al (2016) a similar source was identified but it was attributed to a local soil source related to anthropogenic activities, among which construction works. The weekend/weekday ratio would be the same as that of the traffic source. Which are the Enrichment Factors respect to Si of the crustal elements? 7) The identification of the Primary vehicle Emission source is more sounded, looking at its time trend in comparison with that of the traffic related gases or of the traffic counts; however also in this case the profiles shows some problems. Again the errors for the main markers are very high; a comparable contribution

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(micrograms/m³) from Si, Ca, Fe and BC is unreasonable in this source, where OC and BC are the main components (see many articles on PMF identification of traffic sources, e.g. the references reported in the article like Amato et al., 2016). This could be due to the combined use of data coming from different instruments, to the quality of the data or to the error estimate. The percentage of OC in this source is also too low (27%). 8) The identification of the Tire Wear source is not clear at all. In fig. 2 there is a maximum common with that of NO_x or CO but the time pattern is completely different and the weekday/weekend ratio may be typical also of industrial activities. I would not say that $r^2 = 0.32, 0.43, 0.17$ means "a strong correlation between the Tire Wear factor and traffic related pollutants" (p. 8 l.8). Furthermore, as reported in literature, most of the mass from the tire wear is due to the carbon component, Zn is a typical marker but it is a trace, not a main constituent (and Fe gives even a higher contribution). 9) From what I have reported above, the conclusion (p. 14 l. 7) "the traffic related sources accounting for 17% of PM_{2.5}" is not justified. 10) The Industrial sector source is identified by Pb and As, but the time trend of those two elements shown in fig. S4 is completely different. 11) Which are the metals that help in the identification of the cooking source as reported at p. 9 l. 3? There are plenty of articles that identify the cooking source with only organics. 12) In the Biomass Burning source the absolute contribution of K (a tracer) is higher than that of BC, which is quite strange looking at the literature 13) Why the nitrates source has such a correlation with SO₂, which is far higher than that of the sulphate source? 14) All the discussion on the PMF with only metals and ACSM data is not very useful. The quality of the metal data is not very good. Si has background problems with a high noise (and Al is not measured, like Na and Cl), V, Ni, As, Sr are present only in very few cases and also Se is present in less than 30% of the cases. I am not sure they can be considered as good variables (no information on good or weak variables is reported). With only 11 "good" elements, they found 8 factors! In the discussion about the identification of the different sources, there are problems similar to the ones reported for the "full" analysis. The identification of 4 traffic sources is arbitrary. I understand that S was not used in the PMFfull analysis because there is the

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ACSM sulphate, but there is no reason for not using it in the PMFmetals analysis. Finally, the authors contradict themselves saying “PMFfull analysis did not always enable better resolution of factors”. 15) p 13 l- 29-35 The combination in the PMF of stable markers like K with unstable markers (at higher temperatures) like the organics may be problematic. This could explain the different correlations.

Minor comments: 1) Which program was used to fit the X-Ray spectra? 2) How were the raw aethalometer data were corrected (e.g. which mass absorption coefficient was used?) 3) What does it mean that "only good species" (p 5 l. 13) were taken into account when calculating the Q/Qexp ratio? This is a number that is given by the program considering the results as a whole

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