

Interactive comment on “Sources and geographical origins of fine aerosols in Paris (France)” by M. Bressi et al.

Anonymous Referee #2

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This paper presents the result of a source apportionment study using EPA PMF 3.0 on a dataset containing daily samples of PM_{2.5} with speciation data collected for a period of one year at an urban site in Paris. The authors aim to identify the sources, the geographical origins of these sources and their contribution on the PM_{2.5} mass. The used tools and techniques in this paper are not new, only the use of bootstrapping to help identifying the number of factors is something I have not seen prior to this analysis. The results are interesting, but some questions remain. Several key species (tracers) were not available in this dataset, making the results harder to interpret. Expected sources could not be resolved, for example crustal material. The lack of a dust/crustal material profile in this study, primarily because the tracer species Al, Ti and Si were not available, is conspicuous. There are also some other comments which will be addressed

C11911

below.

General comment: Throughout the paper references are made to the PMF3.0 model. I suggest in changing these references to EPA PMF 3.0 as this eliminates the possibility of confusion between the EPA PMF 3.0 and PMF3 model.

Page 33238, line 2: the authors claim Paris is the largest megacity in Europe. It is unclear if definition of megacity is used for the city of Paris or the Paris metropolitan area known as Ile de France region. The metropolitan area is indeed listed as the second largest in Europe according to the definition of Larger Urban Zones as supplied by Eurostat Urban Audit. The text should be updated to reflect that information. See Bressi et al. (2013) for further information.

Page 33239, line 13-15: the authors again talk about the city of Paris with about 11 million inhabitants. The number of inhabitants is more consistent with the Paris metropolitan area. The city of Paris only has about 2.5 million inhabitants. This should be corrected to reflect the fact it is not the city but the metropolitan area of Paris.

Page 33242: line 6: The Paris metropolitan area is approximately 12,000 km² and PMF results from one urban area are used to describe this whole area. More sampling locations have been used by the research by Bressi et al. (2013). It would be nice if the authors could make a short comment on the comparison of the composition of different locations across the Paris metropolitan area. Depending on the location there could possibly be a large difference in both total PM_{2.5} mass and the composition, depending on the intensity of nearby local sources (e.g. traffic). Hence, the results of one urban station might not be representative for the PM_{2.5} contribution and composition in the whole Paris metropolitan area.

Page 33243, line 25: PMF is a well-known method, therefore section 2.2.1 can be shortened by using literature references.

Page 33245, line 5: Q_{theoretical} could be calculated by multiplying m with p, however

C11912

this only works with the assumption there are good estimates of the uncertainty matrix available. Bad estimates in the uncertainty matrix can lead to changes in Qrobust. If this is the case comparing the Qrobust against Qtheoretical does not make any sense. Interestingly, I found a similar comment regarding this issue later on page 33251, line 22-23.

The supplementary section contains important information in which the uncertainty matrix was optimized to satisfactory and robust PMF results among others. Initially the Qvalue will provide information about the used uncertainties. Changing the uncertainties to obtain a better Qvalue (e.g. ratio of 1 with Qtheoretical) is circular reasoning. Hence, the comparison between Qrobust and Qtheoretical cannot be made as it has been done in this paper.

Page 33246, line 1: The authors mention the measured concentrations of Al, an important tracer for dust/crustal material and one of the most abundant element, are mostly below the method quantification limit. This would suggest there is no dust/crustal material present in the Paris metropolitan region. The fact Paris has been and continues to be subjected to for example Saharan dust episodes makes this highly unlikely. Are Al-concentrations also that low at other sites across the Paris Metropolitan area? Is there a difference in the analysis used to determine the Al concentration compared to other studies?

Page 33246, line 16: The authors mention they have used 20 runs with random seeds. The reason random seeds are used is to prevent local minimum values when optimizing the Q-value. Can the authors comment on the fact if 20 runs are sufficient to assure a global minimum for the optimization of Q has been reached?

Page 33247, line 13-16: There seems to be a discrepancy between the section about the HYSPLIT model for the CPF function at Page 33247 (line 13-16) and a similar section for the PSCF function at page 33248 (line 9-11). Different references are used to denote the same model. From what I understand detailed meteorological measure-

C11913

ments were not available, hence the use of HYSPLIT. I think the section about obtaining the meteorological parameters should become a section and could be explained in more detail as this is the first paper I have seen to use this approach, at least for the CPF function.

Page 33251: The use of bootstrapping to assess the stability of the factors is interesting.

Page 33251, line 9-15: Information is given about the selection of species included in the PMF analysis. The authors mentioned an exception is made for Ni. However, it is unclear if Ni is used in the analysis as a weak or strong species. With the exclusion of Ti another important tracer for dust/crustal material is removed from the study. Since Si has not been measured no sufficient tracers are left to make an identification of dust/crustal material. Furthermore I am missing if PM2.5 mass was included in the analysis (probably as a weak variable). The comments on line 24-26 would suggest it was indeed included in the analysis. (I found this crucial information in the supplemental pages but should have been included in the primary paper.)

Page 33253, line 14: EPA PMF 3.0 supports the FPEAK parameter to assess the rotational freedom in the model. Have other FPEAK values than zero been examined to assess the rotational freedom? From the paper I understand the base run is used.

Page 33255: section 4.1: As expected no specific dust/crustal material profile was found in section 4.1 due to the lack of the tracers Al, Si and Ti. Contributions of this source are divided among other found sources, hence it is difficult to get 'clean' source profiles. It would be very interesting to see the results of mass closure for the species used in the PMF analysis. I suspect a rather large part of the PM2.5 mass cannot be explained by the concentrations of the species used in the analysis (see for example Almeida et al. 2006). If species, which were not measured, are strongly correlated to the measured species or if they represent sources that add negligible mass to the particulate matter samples the sum of the source contributions should be approximately

C11914

equal to the measured total mass (Juntto and Paatero, 1994; Hopke, 2003). If this is not the case, there should be some unexplained mass present in the study. However, by using PM_{2.5} as a total mass variable all available mass is apportioned across the factors. Depending on the sum of the concentration of the available species it is not unlikely part of the apportioned mass is actually unexplained.

Page 33256, line 21: Unclear reference: Schmidl (2005 in Puxbaum et al., 2007). Also mentioned later.

Page 33258, line 8: Road salt can consist of NaCl, but also CaCl₂, MgCl₂ and KCl or combinations of these salts. Without a reference about the composition of road-salt used in Paris I would refrain from making statement the composition is NaCl. Furthermore, the frequency of which road-salting has been applied within the measurement period might provide clues about the plausibility of the road traffic factor. If, for example, the roads have been salted on numerous days the fact no Cl is associated with this profile is conspicuous.

Page 33271: line 1-2: I find it interesting the PM_{2.5} mass (figure 7) is reported as 14.7 $\mu\text{g}/\text{m}^3$ whereas the measurements results for 2010 (Air Quality in the Paris Region 2010 (http://www.airparif.asso.fr/_pdf/publications/synthese_bilan_2010.pdf)) reports an urban background average of 18 $\mu\text{g}/\text{m}^3$. Although there is a slight shift in months (Sept-Sept vs Jan-Dec) I doubt if this could explain the difference of approx. 3 $\mu\text{g}/\text{m}^3$ between these measurements. I wonder why there is such a large difference between both measurements.

Page 33273, line 25: For the oil combustion the authors compare the contribution found in Paris with several other studies (Dunkirk (PM₁₀), Amsterdam and Copenhagen). Petrochemical activities are all reported in the vicinity of these cities, not to mention these cities are highly impacted by shipping. However, the contributions found in Paris are similar to those cities with a (nearby) harbor and nearby petrochemical activities. I would suspect the contribution in Paris to be lower compared to these cities, certainly

C11915

not equal.

Page 33273, line 28: The sampling site is located in the city centre of Paris (page 33242). Based upon the report Air Quality in the Paris region 2010 the European limits for PM_{2.5} were not exceeded at urban background locations but were exceeded at roadside locations. The contribution of traffic at the sampling site is calculated to be 2.1 $\mu\text{g}/\text{m}^3$, however, looking at the PM_{2.5} measurements for 2010 as given in the aforementioned report the difference between the urban background (on average 18 $\mu\text{g}/\text{m}^3$) and the roadside sites (30 $\mu\text{g}/\text{m}^3$) is much larger compared to the findings in this study. Judging from the measurements, I would have expected the traffic contribution from this study to be higher than the found contribution of 2.1 $\mu\text{g}/\text{m}^3$. The difference between the urban background and roadside sites cannot be entirely explained by traffic contributions, as the contributions at the roadside sites are dependent on the geographical structure (e.g. street canyon). It would be beneficiary if the authors could comment on the difference between their findings and the difference between PM_{2.5} measurements at urban and kerbside locations in Paris. Page 33275, line 4: Several studies, among Mooibroek et al (2011) have identified a gradient for marine aerosols with higher concentrations at the coast and lower concentrations inland. The found concentration of the marine aerosols is compared to a rural site in the Netherlands. Mooibroek et al found, depending on the distance from the coast, different concentrations of marine aerosols on the used sampling locations. Since the study used several rural sites it would be wise to compare the concentration of sites that have a similar distance from the coast as the sampling location in Paris has.

Page 33277, line 1: The road traffic source shows a stable concentration for the whole year. It would be interesting to compare this with either vehicle emissions during the seasons or with the number of vehicles in Paris during the seasons. If the contributions are the same it would suggest there isn't much change in the number of vehicles during the seasons. In general, traffic intensity increases if the weather conditions worsen (e.g. rain). Paris obviously has a solid public transportation, therefore it would be

C11916

interesting to see if the number of vehicles across the seasons stays approximately the same.

Page 33279, line 7: The conclusions are true if the assumption the sum of the measured species account for most of the PM_{2.5} mass is true.

References: Almeida, S. M., Pio, C. A., Freitas, M. C., Reis, M. A., & Trancoso, M. A. (2006). Approaching PM_{2.5} and PM_{2.5} – 10 source apportionment by mass balance analysis, principal component analysis and particle size distribution. *Sci. Total Environ.*, 368(2-3), 663–674.

Hopke P.K., (2003), A guide to positive matrix factorization, [Online] Available at: <http://people.clarkson.edu/~hopkepk/PMF-Guidance.htm>

Juntto, S., & Paatero, P. (1994). Analysis of daily precipitation data by positive matrix factorization. *Environmetrics*, 5, 127–144.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 33237, 2013.

C11917