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

Anonymous Referee #1

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This MS presents a source apportionment analysis using PMF of a 1-year dataset of chemical speciation data. The authors describe the need for this work based on the lack of detailed source apportionment studies for Paris in the literature, and I concur with them. However, despite it being necessary, the work is not novel regarding the approach or the methods used, nor regarding the results. The study is certainly interesting, but it is also certainly not innovative. In general, some results are difficult to interpret, such as the nature of the metallic source or the too low traffic contribution. The main limitation of the work is the lack of mineral tracers (and thus mineral dust contributions). A number of issues to be addressed are described below:

- Traffic contribution: the contribution from this emission source seems really low (14% of PM2.5) considering that the study location is Paris, a megacity (11 million inhabitants) and the second megacity in Europe (stated by the authors). If the model is unable to provide a more realistic estimate, then the authors should describe the limitations of the model, given that I find it very unlikely that the contribution is so low. I believe this could be related to another of the limitations of the study, which is the absence of a mineral dust source. The database used lacks the major mineral matter tracers, and as a result no mineral dust source is resolved by the model. This could also be impacting the traffic source, which should probably have a larger road dust component which might then increase its contribution to >20% of PM2.5, which would probably be more realistic. This would change their conclusions, as more mitigation strategies should then be local (and fewer of them external to the city).

- Mineral dust source: as the reader works through the paper this issue arises, given that few mineral tracers are considered in the database (Ca, Mg and K, but only as the water-soluble fraction) and then most of them are excluded from the PMF analysis (e.g., Al, Ca, Ti, because they are weak variables). It seems very likely that a "city dust" source should be resolved in the analysis, but it cannot be due to the lack of tracers. The authors only acknowledge this limitation far into the MS (page 33266), and then they state that this source would represent at most 3% of PM2.5 and that it is therefore negligible. Firstly, I find it hard to believe that in a megacity such as Paris the city dust would have such a low contribution. Perhaps the low mineral dust levels obtained in Bressi (2013) could be related to the analytical methods (digestion with microwave), which does not use HF and which would then lose a large proportion of the minerals because it is unable to dissolve them? Secondly, even if the contribution from the mineral dust source per se were small, the authors are missing the contribution from mineral dust to the traffic source (road dust), which as stated above would increase the traffic contribution to levels more representative of this type of city. - page 33239, line 9, USEPA 2011 b and a should be interchanged. - same page, line 23: "possible exceedances...", with an annual mean of 14 µg/m3 the city doesn’t exceed any EU limit values, correct? - page 33240, line 2: please enumerate briefly some of the measures, or at least the sources that were targeted - page 33241, line 1: "questions", I’d rephrase this, it doesn’t ques-
tion it: it is evident that 19 days are not representative of a longer time series. - page 33242. Sampling: from the text it is not possible to learn whether sampling artefacts for OC were taken into account. With the use of low-vol samplers, if the artefacts were not taken into account then the OC mass may have been overestimated, and therefore the biomass burning source may have been overestimated too. - page 33244: the theory of PMF is well-known, and therefore this page and the next could be summarised using references. - In general, the paper is very long and should be summarised (by around 20% would be my suggestion). - page 33243, line 6: Linuma should have a capital "L" - page 33248, lines 9-11: repetition from the previous page - page 33250, line 20: if $Q_{\text{robust}} = Q_{\text{theoretical}} + \text{model error}$, values of the $Q_{\text{robust}}/Q_{\text{theoretical}}$ ratio <1 should not be valid outcomes of the model, correct? As far as I understand it, $Q_{\text{robust}}$ should be $>$ $Q_{\text{theoretical}}$, otherwise it suggests that the run should be refined. This is the case for the 7 and 8 factor solutions (.9 and 0.7 according to the authors). Please comment on this. - page 33251, line 22: here the issue of the Q seems to be corrected, as $Q_{\text{robust}}$ and $Q_{\text{theo}}$ are 6403 and 5569, according to the authors. If these data are correct, please correct the previous. Then the 7 factor solution is more robust. - page 33252, line 23: "biogenic source", the same is true for the mineral dust source, please make explicit reference to this. - page 33253: these 2 sections are a bit repetitive and only introduce what will be presented in section 4. They could be summarised. - page 33256, lines 19 and 23, what is "2005 in Puxbaum et al 2007"? Please clarify. - same page, line 26: "10.3 and 10.8", the use of values closer to 13-15 was recommended in the conclusions of the recent Ghent workshop on biomass burning (2-3 December, 2013) by Maenhaut and other authors. - page 33258, line 6: "local origin", the paper Amato, F., Viana, M., Richard, A., Furger, M., Prévôt, A. S. H., Nava, S., Lucarelli, F., Bukowiecki, N., Alastuey, A., Reche, C., Moreno, T., Pandolfi, M., Pey, J., and Querol, X.: Size and time-resolved roadside enrichment of atmospheric particulate pollutants, Atmospheric Chemistry & Physics, 11, 2917-2931, 2011. reports that NO3- from traffic may be formes within the city scale, and therefore it is surprising that no NO3- is found in this source in Paris. It could be related to the lower photochemical activity in Paris (lower oxidation rates), but still some NO3- should be expected in the traffic source. The same is true for mineral dust, as stated above. - page 33259, line 2: "carbonaceous", please add "carbonaceous and metallic" - page 33259, line 24: the explanation that Cu and K may come from shipping seems far-fetched, although I don’t have a better one. If Cu and K have ever been linked to shipping emissions, please provide the reference. - page 33262, line 23: "mesoscale", if this source is mesoscale then it should also have a secondary aerosol component, and not only primary (metals), correct? How do the authors explain this? - page 33266, lines 14 to end: indeed, only here is the missing mineral source acknowledged. - page 33271, line 7: 14% for traffic in a megacity such as Paris seems really low. Please describe the limitations of the model. - page 33273: the comparison with Dunkirk doesn’t seem valid, given that it is a large harbour area and the authors stated above that this source has almost no (or very little) influence from shipping. - page 33274, line 7: the data from Canada (5.6 million inhabitants) are hardly comparable with the data from Paris (11 million). The traffic contribution in Paris must be larger. - page 33275, line 25:"high absolutely concs. along the year", this doesn’t seem to make sense: oxidation of SO2 to sulphate is enhanced in summer, therefore absolute and relative values of AS should be lower in winter. - page 33276, line 17: "vessel activities", again, in the description of the source it was stated that the influence of ships in this source is minor. The authors should revisit the interpretation of this source, is it industrial emissions or ships? Please clarify. - page 33277, 1st paragraph: the contribution in winter is very low compared to the rest of the year, and this is unlikely. If the authors explain the higher AS contributions in winter as resulting from anticyclonic episodes, shouldn’t the same occur for the traffic source? Please explain.

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