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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #3

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General comments The paper titled "Sources and geographical origins of fine aerosols in Paris (France)" by Bressi et al. deals with the fine aerosols source apportionment in Paris, making use of one year filter data and positive matrix factorization as source apportionment method. The impact of local emission sources vs. the regional contribution of secondary aerosols is also assessed. The investigation of PM sources and their geographical origin is a relevant topic in the atmospheric science field and therefore it is suitable for ACP. The overall quality of this work is good and the manuscript is quite well-written. I recommend publishing this work after the authors respond to the following comments.

General comments

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Interactive Discussion
Discussion Paper

In the introduction you mention that Paris is highly concerned about air quality issues. However, it would be interesting to report a brief comparison with other worldwide (or European) megacities to highlight whether the air pollution levels in Paris are really high or not. Compared to other megacities, Paris is rather clean, so it would be interesting to contextualize the importance of your work in a broader view. One of the main results of your work is that the pollution observed in Paris is actually formed and transported from outside the city, so, as you mentioned, the goal of your work is more policy related (local vs. regional reduction measurements) than health related.

In order to evaluate the interpretation of the identified sources, the authors could provide time series correlation with external data, like gas phase measurements etc. For example the road traffic factor could be correlated with NOx which is often measured by permanent monitoring stations. It would be interesting to report in a table or with some graphs the time series correlation of the retrieved sources and their tracers. For example you might report the correlation coefficient between the marine factor and Na time series. Some events might be characterized by weak correlations and then you could complete your validation using the trajectory analysis you did.

An interesting result is associated with the separation of a primary marine factor. Do the authors have any evidence of secondary marine production in an additional factor? What does it happen when considering a more factor solution? Is there a split of the marine component? Were methane sulfonic acid measurements performed during the campaign? If so, they could be used to investigate the presence of secondary marine production in Paris (as already shown during the MEGAPOLI field campaigns happened in Paris in the same year).

How good is the correlation between the marine aerosols time series and the heavy oil combustion ones? During some events they might correlate when both sources had the same “marine” origin (ship emissions), while during other periods they might not be correlated. It would be interesting to have further discussion on that.

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What is the V/Ni ratio during marine influenced air masses? You could report this ratio for different back trajectories groups.

It would be interesting to see the source apportionment results obtained running different experiments on each season. In this way you might be able to separate different ammonium nitrite/sulfate factors to better characterize the secondary components. Another option would be to use a much higher number of factors in one experiment. Have you tried to compare PMF results from the run on the yearly data with results from separate runs?

Similarly to ammonium nitrate, also ammonium sulfate can be produced by a variety of sources and aging processes. Sulfate can have both anthropogenic and natural (sea, etc.) origin. Have you tried to split this factor into more components based on tracers? For example, can be the presence of Pb, Cd etc. in the ammonium sulfate factor an indication of coal burning? It might be that air masses affected by coal combustion emissions are transported from Eastern Europe to Paris. Also in this case the analysis of the back trajectories might help in assessing this point. Moreover, adding more factors in PMF (or running separate experiments for seasons/reference periods/events) might produce the split of the A.S. factor into several components.

In general, the authors should cite more recent literature (e.g. in chapter 4 in addition to the ones they report). Moreover, there is quite a recent literature about Paris and especially related with the MEGAPOLI project happened in the year you covered with your measurements. The authors should cite already published papers instead of Beekmann et al. 2012 (still in preparation). For example at page 33265 and 33270, you could quote Crippa et al. (2013), but there is a quite broad literature on this topic.

Specific and technical comments - Figures 1 and 2 are not readable since all the graphs are very small. You might try to regroup the graphs. - In Table 3 it is not completely clear why there are negative concentrations for sources retrieved with PMF (which has as a constraint that both F and G are positive). - I would move Fig. 5 to the sup-

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plementary material. - Page 33249, line 19: too few factors will result not only in a mixing of different sources in a factor, but also they could lead to high residuals - Page 33251, line 11: categorise as bad (please add “as”) - Page 33252, line 2: remove ‘ to good” - Page 33253, line21: is the unaccounted fraction corresponding to the residuals? Please clarify it also in Fig.4. - Page 33243: cite Zhang et al. (2011) together with Belis et al. (2013) - Page 33245: cite also Ulbrich et al., (2009) when referring to PMF2 and Canonaco et al. (2013) for ME-2 - Page 33255: 4.1 Source identification (remove F matrix) - Page 33256, line 12: here you could cite results from other measurement campaigns performed in the Parisian area finding the regional feature of biomass burning - Page 33256: how was the OM/OC ratio for biomass burning calculated/assumed? Please, cite the corresponding literature - Page 33263 line 3: 1.98$\pm$0.23 etc. In your version you report only the plus symbol and not the minus - Page 33263, line 19: change “have much varied sources” with “ are produced/emitted by a variety of sources” - Page 33263, line 29: since you list a lot of very different activities, it is not clear to which activities are you referring to with the expression “come from these activities” - Page 33265 line 16: “due to the chemical conversion”. “To” is missing - Page 33265 line 18: change “authors” with “literature study” - Page 33266, lines 3-5: reformulate the sentence since it is not clear. - Page 33266: paragraph 4.1.8 could be easily included into the previous discussion of each factor. - Page 33266, line 13: replace “more recently in SA studies” with “in more recent SA studies” - Page 33267, line 17: “which compound. . .”, check the correctness of the sentence - Page 33271: Source contribution (remove G matrix) - Page 33271, line 21: “reported in a SA study” and not “in an SA study” - Page 33271-33272. . .: it is not clear why the authors focus so much on the comparison between their results (absolute values etc.) with data obtained for different sites and years (which are not necessarily comparable among them). There are indeed a lot of other studies related to the same region and the same time period, so it would be more interesting to make a comparison with those studies than with the one presented. If the authors want to make a broader overview about Europe, they should explain why they selected the reported literature and how

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those studies are comparable with their work. - Page 33272, line 16: " based on one year measurements. . .", remove "a" - The authors should comment the results shown in Table S6 - How do the authors explain the lower concentration observed for PM2.5 in the road traffic source during wintertime (Fig. S3)? Is it due to an overestimation of the EC content in the biomass burning factor and an underestimation in the road traffic one?

References

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