

Interactive
Comment

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

M. Bressi et al.

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The authors would like to thank the anonymous Referee 3 for his extensive and constructive comments, which will help clarifying and improving our manuscript.

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

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quite well-written. I recommend publishing this work after the authors respond to the following comments.

R.3.1. (stands for Referee 3, 1st comment) General comments 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.

A.3.1. (stands for Answer to Referee 3, 1st comment) As the reviewer mentioned, it would indeed be interesting to report a brief comparison between the air pollution levels in Paris and other cities. We do agree with the reviewer that Paris is rather clean compared to other European (and worldwide) urban environments. However, we still believe that our work is relevant regarding health issues since - as mentioned in the introduction - reducing PM_{2.5} levels in Paris to the recommended World Health Organisation (WHO) value of 10 $\mu\text{g m}^{-3}$ would lead to a gain in life expectancy of ca. half a year in this city (Aphekom, 2011). In addition air quality issues are not only related to PM concentrations but also depend on the population which is affected (exposition and number of persons exposed). Policies are according to us a mean to tackle these health issues, which are even emphasized in other European urban environments. In order to take into account your comments, the text has been modified as follow: p33239, l13: the word “highly” has been removed. P33239, l27: the following sentence has been added: “It should be mentioned that in a broader context, PM_{2.5} levels measured in Paris are generally lower than in other European urban environments: Paris (17.8 $\mu\text{g m}^{-3}$), Zurich (19.0 $\mu\text{g m}^{-3}$), Prague (19.8 $\mu\text{g m}^{-3}$), Vienna (21.8 $\mu\text{g m}^{-3}$), Barcelona (28.2 $\mu\text{g m}^{-3}$) (Bressi, 2012; Putaud et al., 2010). Implementing effective

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PM2.5 abatement strategies is thus not only necessary in Paris but also in most European urban environments.” Note to the referee: the level of $17.8 \mu\text{gm}^{-3}$ reported here for Paris has been calculated using the same assumptions applied for chemical mass closure by Putaud et al. (2010) to the aforementioned studies (see Bressi, 2013 for more details).

R.3.2 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 NO_x 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.

A.3.2. Time series correlation of the retrieve sources and their tracers would indeed be an interesting way to evaluate the interpretation of the identified sources. The following tracers have been selected directly from the input PMF matrix: sulfate, nitrate, vanadium, levoglucosan, manosan, sodium, chloride, magnesium, lead and cadmium (see Sect. 2.1 and references therein for information on the analytical techniques used). In addition, NO_x has been monitored by the regional air quality agency (Aiparif) at the same urban background site used in our study (AIRPARIF and LSCE, 2012). Finally, black carbon (BC) has been measured with an Aethalometer (AE-31, e.g. see Sciare et al., 2011 for a description of the instrument) at a similar urban background site of Paris (see Favez, 2008 for a description of the site) from 11 September 2009 to 10 September 2010. BC has been apportioned to a fossil fuel (BC_{ff}) and a wood burning (BC_{wb}) fraction, following the methodology introduced by Weingartner et al. (2003) and described by Favez et al. (2010) for instance. BC_{ff} is often regarded as a tracer of primary traffic emissions (e.g. see Crippa et al., 2013b). The Pearson correlation coefficient of the retrieved source and their tracers is reported in Table A.3.2, which can

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be found in the supplementary material of this comment (see also Figures associated). In general, tracers correlate very well with their retrieved sources (e.g. biomass burning and levoglucosan, $r=0.99$; marine aerosols and Na: $r=0.91$). When looking at NO_x as suggested by the referee, fairly good correlation is observed with the road traffic source ($r=0.50$), although good correlation is also observed with the biomass burning source ($r=0.61$). When focusing on spring and summer, correlations are however far better between NO_x and road traffic ($r=0.71$) than between NO_x and biomass burning ($r=0.36$) (see also Fig. A.R.3.2.a of the supplementary material). Although NO_x are primarily emitted by road traffic, according to emission inventories (see p33263, l21-29) NO_x could also be emitted by biomass burning during winter, explaining the fairly good correlation observed. Another explanation would be that during wintertime, our model slightly underestimates EC from road traffic and slightly overestimate EC from biomass burning, as suggested by the referee in his last comment of his review, which could explain the fairly good agreement only observed between modeled and measured EC levels ($r^2=0.56$, see Table S1 and p33252, l4-5). Interestingly, the road traffic source correlates fairly well with BC_{ff} ($r=0.50$), whereas biomass burning and BC_{ff} are poorly correlated ($r=0.22$). This suggests that the lower concentrations observed during wintertime are real, although they are not fully explained by the authors (see also our response to your last comment). On the other hand, good correlations are observed between biomass burning and BC_{wb} ($r=0.89$), while poor correlations are found for BC_{wb} and road traffic ($r=0.14$) (see Fig. A.3.2.b.). Comparisons between our PMF sources and BC_{ff} or BC_{wb} are further discussed in Bressi (2013). The table A.3.2. has been added in the supplementary material. The figures A.3.2.a. and b. are only intended to the referee comment.

R.3.3 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. What is the V/Ni ratio during marine influenced air masses? You could report this ratio for different back trajectories groups.

A.3.3 Unfortunately the authors have no evidence of secondary marine production in an additional factor. When considering 8 factors, a factor with high proportions of Fe, Cu and Mn is found in addition to the 7 factor solution described in our manuscript, and contributes ca. 3% of total PM_{2.5} mass. When considering 9 factors, ca. 80% of potassium is found in a single factor, whereas no potassium is found anymore in the biomass burning source (which is very unlikely); in addition technical problems arise as detailed p33250, I9-18. Therefore a split of the marine factor is not visible (even for the 10 factor solution). Methane sulphonic acid (MSA) measurements were performed during the campaign although the dataset was not ready at the time the publication was written. For information, a comparison between MSA and the marine aerosol source is shown in Fig. A.3.3.a. (see supplementary material of this comment). No correlation is found between both datasets ($r=-0.18$). As mentioned by the referee, it would be interesting to investigate the presence of second marine production in Paris including the MSA dataset in the input PMF matrix and this work might represent a future publication. The correlation between the marine aerosol time series and the heavy oil combustion one is very poor during the whole campaign ($r^2=0.10$, $n=335$). Unfortunately no clear periods have been found for which both sources correlates. This might be due to their different geographical origins (Fig. 6, p33305): the heavy oil combustion source has high contribution when air masses come from north of France and the English Channel, whereas the marine aerosol source has –most of the time - high contribution when air masses come from the Atlantic ocean (southwest direction of Paris). Substantial sources of the heavy oil combustion factor are also industrial

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(e.g. oil power station, petrochemical complex, boilers and furnaces). Preliminary tests show that the medians of the V/Ni ratio are similar during marine (including Atlantic and English Channel) and non-marine influenced air masses (ca. equals to 0.8). Good agreement is observed between V and Ni temporal variations most of the year (see Fig. A.3.3.b, $r=0.7$, $n=260$) suggesting a relatively constant V/Ni ratio, independently or air mass origins.

R.3.4. 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.

A.3.4. We agree with the reviewer that it would be highly interesting to see the source apportionment results obtained running different experiments on each season. Unfortunately, the time resolution of our study (24h) does not allow us to perform such experiments, since the number of samples for one season (ca. 90) is too low to reach robust results given the number of species taken into account in our study (18). In fact, we attempted to perform PMF on the first three months of the campaign, when our dataset was not fully constructed yet. High seed variability was observed leading

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to substantial differences in factor profiles and contributions according to runs; scaled residuals and the output of bootstrap test were not satisfactory either. Performing PMF on the first six months of the campaign led to results comparable to the one presented in this manuscript, at the exception that our current Ammonium Sulfate (AS) and Ammonium Nitrate (AN) rich sources were found in one unique factor. This is likely due to the high correlations observed between ammonium, nitrate and sulfate during the first six months (Sept. 2009 – Feb. 2010) thus not allowing their apportionment; on the other hand, the substantial discrepancies observed between AS and AN during summertime (almost null concentrations for the later component) allows the apportionment into two factors when performing PMF on one entire year. Performing PMF on a seasonal basis in the region of Paris is a perspective of research mentioned in Bressi (2013) and would require a high time resolution instrument (e.g. 30 min with the Aerosol Chemical Speciation Monitor, Ng et al., 2011) or a larger number of chemical species (i.e. about 70 species to reach the same matrix dimension as ours). Regarding your specific comment on ammonium sulfate, we have intensively tried to split the AS rich source into two or more factors in vain. As mentioned in the comment A.3.3. increasing the number of factors in PMF runs does not split the AS factor. As mentioned p33279, l12-14 we believe that studying the isotopic composition of individual elements such as sulfur or nitrogen could provide valuable information which would help better apportioning the AN and AS rich factors (see for instance Kundu et al., 2010; Nriagu et al., 1991; Pichlmayer et al., 1998; Yeatman et al., 2001 for an illustration of the added value of isotopic composition analyses). As suggested by the referee, the presence of Pb, Cd, Ni and Fe could indeed be an indication of coal burning emissions that would have been transported from Eastern Europe to Paris. This suggestion has been included in the manuscript as follow: p33265, l5: “Ni, Cd, Fe and Pb might also come from coal burning emissions (Junninen et al., 2009) which could have been transported from Central/Eastern Europe to Paris (see Sect. 4.2).”

R.3.5. 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

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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.

A.3.5. As suggested by the referee, the authors have cited more recent literature - especially related to the MEGAPOLI project - in the new version of the manuscript as follow: p33266, l2: "(see also Crippa et al., 2013a, 2013b on this subject)" p33240, l26: "Freutel et al. (2013) reaches the same conclusion, reporting the highest PM1 levels in the region of Paris when air masses are advected from continental Europe." p33240, l28-29, p33241, l1-2: "Nevertheless, the results reported by Sciare et al. (2010) and Freutel et al. (2013) on fine aerosols were based on few weeks periods (19 and 30 days, respectively) occurring during late spring/summer and thus suffer from a lack of representativeness on a longer time scale."

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."

Figure 1 has been split into two figures accordingly (same for Figure 2).

"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)."

As reported in Belis et al. (2014), although a constraint of non-negativity is applied to F and G matrices, the ME-2 solver (used in EPA-PMF3.0) actually "allows a certain degree of negativity in the source/factor contributions for the sake of better rotational uniqueness (Norris et al., 2009)".

"I would move Fig. 5 to the supplementary material."

Fig. 5 has been moved to the supplementary material accordingly.

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- "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"

This information has been added accordingly: p33249, l19 "as well as high residuals"

– "Page 33251, line 11: categorise as bad (please add "as")"

"as" has been added accordingly.

- "Page 33252, line 2: remove ' to good' "

"to good" has been removed accordingly.

- "Page 33253, line21: is the unaccounted fraction corresponding to the residuals? Please clarify it also in Fig.4."

The unaccounted fraction corresponds to the proportion of a chemical species that is not attributed to any PMF factors i.e. equal to 100 minus the sum for every factor of the relative contribution of a given species to a given factor. This has been clarified as follow: p33253, l20-21: "The contribution of the unaccounted fractions (i.e. proportion of a chemical species that is not attributed to any factor) is below 5% for most species" Fig. 4: "Legend: Lev: Levoglucosan, Man; Mannosan; Unaccounted: proportion of a chemical species that is not attributed to any factor."

- "Page 33243: cite Zhang et al. (2011) together with Belis et al. (2013)"

p33243, l22: Zhang et al. (2011) has been cited.

- "Page 33245: cite also Ulbrich et al., (2009) when referring to PMF2 and Canonaco et al. (2013) for ME-2"

p33245, l12: Ulbrich et al. (2009) has been cited p33245, l13 : Canonaco et al. (2013) has been cited

- "Page 33255: 4.1 Source identification (remove F matrix)"

p33255, l2: "F matrix" has been removed.

- "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"

The following references have been added: P33256, I12: "and the literature (Crippa et al., 2013a, 2013b)"

- "Page 33256: how was the OM/OC ratio for biomass burning calculated/assumed? Please, cite the corresponding literature."

The OM/OC ratio used here is the one determined in the study of Bressi et al. (2013) at this site. It is not specific for the biomass burning source. This reference has been added p33256, I14.

- "Page 33263 line 3: 1.98 ± 0.23 etc. In your version you report only the plus symbol and not the minus"

This has been modified accordingly.

- "Page 33263, line 19: change "have much varied sources" with " are produced/emitted by a variety of sources" "

P33263, I19: "have much varied sources" has been replaced by "are produced 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" "

This has been clarified as follow: p33263, I29: "The heavy metals present in this factor presumably come from some of the aforementioned activities such as road transport, manufacturing industry, energy transformation, etc."

- "Page 33265 line 16: "due to the chemical conversion". "To" is missing

P33265, I16: "to" has been added accordingly."

- "Page 33265 line 18: change "authors" with "literature study" "

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This has been modified accordingly.

- "Page 33266, lines 3-5: reformulate the sentence since it is not clear."

The sentence has been reformulated as follow: P33266, l3-5: "To summarize, this factor is primarily made of secondary aerosols, which stem from a variety of sources including agriculture, industrial activities, non-road transport and biomass burning, to name a few."

- "Page 33266: paragraph 4.1.8 could be easily included into the previous discussion of each factor."

Most of this paragraph could indeed be included into the previous discussion of each factor. However, this paragraph allows us to mention the absence of a mineral dust source in our study, compared to what is generally found elsewhere. Additional information has also been reported given the comments of referee 1 and 2 (e.g. see comment A.1.4). This is why we would prefer letting this paragraph as it is if the referee agrees.

- "Page 33266, line 13: replace "more recently in SA studies" with "in more recent SA studies" "

This has been modified accordingly.

– "Page 33267, line 17: "which compound ...", check the correctness of the sentence"

This sentence has been modified as follow: P33267, l15-18: "Interestingly, these geographical regions are amongst the major emitters of sulfur dioxide in Europe (Pay et al., 2012), which is - with ammonia - a precursor of ammonium sulphate."

– "Page 33271: Source contribution (remove G matrix)"

"G matrix" has been removed accordingly.

- "Page 33271, line 21: "reported in a SA study" and not "in an SA study" "

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This has been modified accordingly.

- "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 those studies are comparable with their work."

To the best of our knowledge, there is no study reporting the source apportionment (SA) of PM_{2.5} mass from one year measurements in the region of Paris. As mentioned by the reviewer in his comment R.3.5., several studies have been conducted during the MEGAPOLI project in the region of Paris but they represent several limitations for comparison here. First, in these studies SA is mostly performed with aerosol mass spectrometers (AMS) on the organic fraction only, and not on fine aerosol mass. Second, these studies were performed during one month in two seasons of the year only. On the other hand, the studies chosen for comparison in our manuscript are i) focused on the apportionment of PM mass and ii) conducted during one year minimum, thus taking into account every season. Moreover, for sources imported to the region of Paris, SA studies have been chosen according to their presumable geographical origins (e.g. continental Europe for A.S. and A.N. rich sources). Given the comment of the referee, the choice of the source apportionment studies reported in this section 4.3.1 has been more clearly justified as follow: p33271, l8-10: "These [factor] contributions were compared with different source apportionment studies (see Fig. 8 and Table S6), chosen according to the following criteria: i) SA is performed on PM mass (PM_{2.5} in most of the cases) ii) Each SA study is representative of one year minimum iii) When possible, SA studies have been chosen according to their presumable geographical origins (e.g. continental Europe for A.S. and A.N. rich sources) iv) Similar

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source categories (i.e. factor identifications) are reported”

- "Page 33272, line 16: “ based on one year measurements ... ”, remove “a” ”

“a” has been removed accordingly.

- "The authors should comment the results shown in Table S6”

The results shown in Table S6 are commented throughout Sect. 4.3.1. All the figures reported from p33271 to p33275 come from this Table S6 (which is cited in the manuscript p33271, l9). In case these comments are not sufficient, could you please precise which additional information should be included?

- "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?"

The lower concentration observed for the road traffic source during wintertime is not fully explained by the authors. As suggested by the referee it might be due to an overestimation of the EC content in the biomass burning factor and an underestimation in the road traffic source given that EC is only fairly well modelled by PMF ($r^2=0.56$, $n=337$ when comparing measured versus modelled EC concentrations, Table 1). However, as shown in Fig.A.3.2.b BCff also exhibits lower levels during wintertime, suggesting that this pattern is real. Given your comment, the following paragraph has been modified in the manuscript: P33277, l1-3: “The road traffic source exhibits rather stable concentrations all along the year (annual average of $2.1 \pm 2.1 \mu\text{g}\cdot\text{m}^{-3}$), with however a smaller contribution during winter ($1.3 \pm 1.4 \mu\text{g}\cdot\text{m}^{-3}$). An overestimation of EC content in the biomass burning factor and an underestimation in the road traffic one could explain this observation. However, fairly good correlations between fossil fuel black carbon and the road traffic source are observed all along the year ($r=0.50$, $n=327$, Table S6 and Bressi, 2013), suggesting that this pattern is real although it is not fully explained by the authors.”

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References of the referee Canonaco, F., Crippa, M., Slowik, J. G., Baltensperger, U., and Prévôt, A. S. H.: SoFi, an Igor based interface for the efficient use of the generalized multilinear engine (ME-2) for source apportionment: application to aerosol mass spectrometer data, *Atmos. Meas. Tech.*, 6 3649–3661, 2013. Crippa, M., DeCarlo, P. F., Slowik, J. G., Mohr, C., Heringa, M. F., Chirico, R., Poulain, L., Freutel, F., Sciare, J., Cozic, J., Di Marco, C. F., Elsasser, M., José, N., Marchand, N., Abidi, E., Wiedensohler, A., Drewnick, F., Schneider, J., Borrmann, S., Nemitz, E., Zimmermann, R., Jaffrezo, J.-L., Prévôt, A. S. H., and Baltensperger, U.: Wintertime aerosol chemical composition and source apportionment of the organic fraction in the metropolitan area of Paris, *Atmos. Chem. Phys.*, 13, 961-981, 2013. Ulbrich, I. M., Canagaratna, M. R., Zhang, Q., Worsnop, D. R., and Jimenez, J. L.: Interpretation of organic components from positive matrix factorization of aerosol mass spectrometric data, *Atmos. Chem. Phys.*, 9, 2891-2918, 2009. Zhang, Q., Jimenez, J., Canagaratna, M., Ulbrich, I., Ng, N., Worsnop, D., and Sun, Y.: Understanding atmospheric organic aerosols via factor analysis of aerosol mass spectrometry: a review, *Anal. Bioanal. Chem.*, 401, 3045-3067, 2011.

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