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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.

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The authors would like to thank the anonymous Referee 2 for his extensive and constructive comments, which will help clarifying and improving our manuscript.

R.2.1. (stands for Referee 2, 1st comment) This paper presents the result of a source apportionment study using EPA PMF 3.0 on a dataset containing daily samples of PM2.5 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 PM2.5 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 below.

A.2.1. We agree with the referee that used tools and techniques are not new (except for bootstrap). The objective of the paper was actually to use advanced and robust source apportionment techniques to deliver a robust scientific message regarding PM sources, which will have strong implications on regional air quality mitigation plans (please see the comment A.1.1. given to the Referee 1 for more details) We agree with the referee that the lack of key species leading to the lack of a dust/crustal source is a limitation of our study. We would please like to direct the referee to the comment A1.4. given to the Referee 1.

R.2.2. 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.

A.2.2. References to the PMF3.0 model have been changed to "EPA PMF 3.0" accordingly.

R.2.3. 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.

A.2.3. In order to clarify the text, the concept of Larger Urban Zone defined by Eurostat is the only one used in the new version of the manuscript. The text has been modified as follow: p33238, l1-3: "The present study aims at identifying and apportioning the major sources of fine aerosols in Paris (France) – the second most populated “Larger Urban Zone” in Europe –, and determining their geographical origins.”

R.2.4. 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.

A.2.4. The text has been modified accordingly as follow: p33239, l13-15: “First, about 11 million inhabitants (ca. 18% of the French population) are exposed to PM2.5 pollution in this “Larger Urban Zone” (LUZ), which is the second most populated in Europe (Eurostat, 2012).”

R.2.5. Page 33242: line 6: The Paris metropolitan area is approximately 12,000 km2 and PMF results from one urban 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 PM2.5 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 PM2.5 contribution and composition in the whole Paris metropolitan area.

A.2.5. As mentioned by the referee, more sampling locations have been used in Bressi et al. (2013) and it could be interesting to make a short comment on the comparison of both total PM2.5 mass and chemical composition across different sites of the region of Paris. The main conclusion of the aforementioned study of Bressi et al. (2013) is that PM2.5 mass and chemical composition is very homogeneous at the regional scale (e.g. see Fig. 8 to 10). The following sentence has been added in the manuscript accordingly: P33242, l9: “It is worthwhile noting that PM2.5 levels and chemical composition are very homogeneous in the Paris LUZ (Bressi et al., 2013). For instance, urban and

suburban sites (distant by 10 km) typically exhibit PM2.5 levels that are not statistically significantly different, whereas levels measured at rural locations (distant by 50 km) are ca. 25% lower than at the urban site. This urban sampling site is thus regarded as being representative of Paris metropolitan area. It should however be highlighted that this site is at 20m a.g.l. which might prevent near ground sources (e.g. road dust) to be considered in our study.”

R.2.6. Page 33243, line 25: PMF is a well-known method, therefore section 2.2.1 can be shortened by using literature references.

A.2.6. Section 2.2.1 has been shortened by using literature references accordingly. In particular, the text from p33243, l14 to p33245, l19 has been put in the supplementary material, and has been replaced by: P33243, l14: “A description of SA methods and receptor models can be found in the supplementary material (Sect. S1).

2.2.1. Positive Matrix Factorization (PMF)

The PMF model (Paatero and Tapper, 1994; Paatero, 1997) is used here (see Sect. S1). PMF is a receptor model that assumes mass conservation and uses a mass balance analysis to identify and apportion sources of PM; it aims at resolving the following equation: [please see Eq 3 of the current version of the manuscript p33244, l21] (1) where xij is the measured concentration of the jth species in the ith sample, gik is the contribution of the kth source to the ith sample, fkj is the concentration of the jth chemical species in the material emitted by the kth source and eij represents the residual element, or the PMF model error, for the species j measured in the sample i. Equation 1 is solved by minimising a Q function defined as: [please see Eq 4 of the current version of the manuscript p33245, l2] (2) where σij is the uncertainty associated to the jth species in the ith sample. Different Q functions can be defined: Qtrue calculated including all data and Qrobust calculated excluding outliers i.e. data for which the scaled residual (eij/σij) is greater than 4. (Note that Qtheoretical will not be studied here as explained in Sect. S1.) A standalone version of PMF using the second version of the multi-linear engine algorithm (ME-2; Paatero, 2000; Norris et al., 2009) has been developed by the United States Environmental Protection Agency (US-EPA) and is used in our study. This version will be named EPA PMF3.0 in the following and can be downloaded at http://www.epa.gov/heasd/products/pmf/pmf.html."

R.2.7. Page 33245, line 5: Qtheoretical could be calculated by multiplying m with p, however 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.q. 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.

A.2.7. Given your comment, we agree that the comparison between Qrobust and Qtheoretical should not have been performed in this paper. The manuscript has been modified accordingly. p33245, l5-7 the sentence has been modified as follow: "Different Q functions can be defined: Qtrue calculated including all data and Qrobust calculated excluding outliers i.e. data for which the scaled residual (eij/σij) is greater than 4. (Note that Qtheoretical will not be studied here as explained in Sect. S1.)" p33250, l11-12: the following sentence has been deleted "i) Qrobust/Qtheoretical ratios are significantly different from one (2.5, 1.9, 1.5, 0.5 and 0.4, respectively)" p33250, l19-20: the following sentence has been deleted "i) Qrobust/Qtheoretical respectively equal to 1.1, 0.9 and 0.7" p33251, l22-23: the following sentence has been deleted "Qrobust is slightly higher than Qtheoretical (6403 and 5569, respectively), which could stem from the procedure adopted to adjust species' uncertainties (Reff et al., 2007)." p2, l19 of the Supplementary material, the following sentence has been added: "However, [this

methodology] will prevent us from using the Qtheoretical function to assess the quality of our uncertainty matrix, since the latter was optimized as described above."

R.2.8. 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?

A.2.8. We indeed mention that the measured concentrations of Al are mostly below the method quantification limit. As noticed by the referee, it is unlikely that there is no dust/crustal material present in the Paris metropolitan region. However, a noticeable contribution of Saharan dust events on fine PM measured at ground level is not known by the authors (including LSCE and Airparif researchers working for several years on fine PM pollution in Paris) who would thus be interested in reading (and reporting) such studies if the referee could kindly provide references. The low Al concentrations found in our study are likely due to: i) the analytical technique used: acid microwave digestion procedure followed by Inductively Coupled Plasma Mass Spectrometry HF digestion, which - as suggested by the Referee 1 in his comment R.1.4. – does not use HF and might then loose a proportion of the minerals because it is unable to dissolve them ii) the location of our sampling site (20 m a.g.l.) thus preventing from the influence of road dust, iii) the size fraction studied (PM2.5): dust/crustal material and Al is expected to be found mostly in the coarse mode (e.g. see Keuken et al., 2013). Aluminium has not been analysed in PM2.5 at other suburban or rural sites across the Paris metropolitan area (for financial reasons). However, during this campaign, colocated sampling of PM10 has been performed at the same site during the same period and then chemically characterized (see AIRPARIF and LSCE, 2012 for more details).

Aluminium from PM10 sampling shows significantly higher concentrations (∼3 times higher on average) and is mostly higher than the MQL. The limitations related to the analysis used to determine the Al concentrations have been stated in the manuscript as follow: P33243, l3: “Note that some minerals (e.g. Al, Ti, etc.) might be underestimated due to the acid microwave digestion procedure used here (with HNO3), which might not be able to dissolve entirely these compounds (e.g. Robache et al., 2000).”

R.2.9. 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?

A.2.9. 20 runs have been chosen following the recommendations of Norris et al. (2008) in the EPA PMF3.0 user guide (page 12). We believe that in our study 20 runs with random seeds are sufficient to prevent local minimum values when optimizing the Q-value since very comparable factor profiles and factor contributions are found for each seed solution. 20 runs with random seeds were also sufficient to show unstable solutions in the case of inappropriate number of factors or too short datasets (e.g. see comment p33250, l13-15 and answer A.3.4 given to Referee 3). Naturally, choosing a higher number of runs (e.g. 50 or 100) would lead to even more robust results and could be suggested in case of sensitive solutions (e.g. if a specific run leads to a different solution) or after having chosen the appropriate number of factors and chemical species (to avoid time consuming computational analyses).

R.2.10. 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 measurements 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.

A.2.10. As mentioned by the referee, different references are reported when describing the HYSPLIT model applied for the CPF and PSCF functions. CPF was first applied given its relative simplicity and the web version of HYSPLIT (available at http://ready.arl.noaa.gov/HYSPLIT.php) was sufficient to determine the overall path of the air mass back trajectories. PSCF was then applied to further investigate the sources’ geographical origins, and calculation on a cell (or pixel) basis was this time required hence the use of a PC-based version (i.e. independent software) of HYSPLIT (v4.9). No significant differences should be found for air mass backtrajectories calculated with either the web or the PC-based versions of HYSPLIT. As suggested by the referee, further information has been given regarding the CPF function as follow: P33247, l16: “Backtrajectories were then defined according to their overall path in one of the sixteen θ directions separated by 22.5° (i.e. N, NNE, NE, etc.). This procedure allows curved backtrajectories to be binned in the appropriate direction, but is laborious and prone to user-approximations.” However, we are not sure that such information could represent a section on its own. Please let us know what exactly should be in the additional section you mention above in case the information added is not sufficient according to you.

R.2.11. Page 33251: The use of bootstrapping to assess the stability of the factors is interesting.

A.2.11. The referee is thanked for his comment.

R.2.12. 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.)

A.2.12. Ni is used in the analysis as a strong species since its signal to noise ratio is higher than 2. PM2.5 is indeed included in the analysis as a weak variable. As mentioned by the referee this information can be found in the supplementary material (Sect. S1). Given the comment of Referee 1 stating that: “In general, the paper is very long and should be summarized (by about 20% would be [his] suggestion)”, we would prefer letting this information in the supplementary material. However, if the Referee 2 insists and the Referee 1 agrees, the authors could include this section S1 in the manuscript (in the actual Sect. 2.2.2. Data preparation, p33245).

R.2.13. 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.

A.2.13. As mentioned by the referee, the base run has been used in our manuscript. FPEAK parameter to assess the rotational freedom in the model has been investigated in our solution. Fpeak values ranging from -5 to 5 (Norris et al., 2008) with a step of 0.5 (i.e. Fpeak values of -5, -4.5, . . . , 4.5, 5) have been applied. Fpkeak values lower than -2 and higher than +1.5 were not considered since the relative difference between Qrobust and Qtrue was higher than 10% indicating high numbers of outliers. By considering F peak values ranging between -2 and +1.5, very similar results were found. The standard deviation of the relative contribution of a given species to a given factor was typically lower than 2% for every Fpeak configuration (the median across all species is 1.2%) thus leading to the same factor identification. Factor contributions were also similar for every Fpeak configuration, with an average contribution to PM2.5 mass varying of ±1% for every factor. In addition, the G matrix has been examined by plotting factor contributions against each other (so-called G space plot) as suggested

by Paatero et al. (2005). No oblique “edges” have been observed in the G space plots when no Fpeak values was applied, suggesting that rotations were not necessary. Therefore, the authors decided not to apply any Fpeak values since no objective criterion was suggesting choosing a specific Fpeak solution. Results regarding our Fpeak solution could be reported in the supplementary material if the referee judges it necessary.

R.2.14. 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 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 PM2.5 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.

A.2.14. Results of mass closure for the major species used in the PMF analysis can be found in Bressi et al. (2013) (e.g. see Figure 2). Very good agreement is found between gravimetric and chemically reconstructed masses all along the campaign (r2=0.98, slope=1.00, n=335), assuming a mineral dust contribution calculated as [Ca2+]/0.15 (Guinot et al., 2007). On average, the mineral dust fraction contributes 3% of PM2.5 mass at this site using the above mentioned calculation. As mentioned by the referee, by using PM2.5 as a total mass variable all available mass is apportioned across the factors, although part of the apportioned mass might be unexplained. This information has been added in the supplementary material as follow: SM, p2, l31 – p3, l1: “PM mass was defined as a "Total Variable" i.e. was regarded as being the sum of all PM chemical components; all available mass was thus apportioned across the factors although part of this mass might still be unexplained. PM mass variable was categorised weak to lower its influence in the final PMF results.”

R.2.15. Page 33256, line 21: Unclear reference: Schmidl (2005 in Puxbaum et al., 2007). Also mentioned later.

A.2.15. The work of Schmidl (2005) has been written in German and is unfortunately not understandable by the authors. It has however been cited by Puxbaum et al. (2007). For clarification, we would suggest using the Harvard style reference (e.g. accessible at http://www.bournemouth.ac.uk/library/local-assets/how-to/docs/citing-references.pdf , see p5) and referencing the work of Schmidl (2005) as: “Schmidl (2005 cited by Puxbaum et al., 2007)”. If the referee still considers this type of reference unclear “Schmidl (2005)” can be used.

R.2.16. 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.

A.2.16. We agree with the referee that road salt can consist of different salts and not only NaCl. In Paris however, Le Priol et al. (2013) report (p23) that road salt are exclusively made of NaCl (60% of Cl, 39% of Na and 1% of other compounds), based on the chemical analysis of the road salt used in this city (this reference has been added p33258, l9). Road salting in the region of Paris has been investigated by AIRPARIF and LSCE (2012) during the same time period as our study. The influence

of road salting is found in PM10 and at a traffic site only (Porte d’Auteuil, mentioned in comment A.2.19.). In addition, comparison between sea salt levels at 5 sampling sites in the region of Paris has been estimated by Bressi et al. (2013) (see Fig. 10). No significant discrepancies are found between rural, suburban and urban sites (including the one used in the current study). The aforementioned results suggest that road salting is not significantly affecting PM2.5 levels measured at our urban background site of Paris.

R.2.17 Page 33271: line 1-2: I find it interesting the PM2.5 mass (figure 7) is reported as 14.7 μg/m3 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 μg/m3. Although there is a slight shift in months (Sept-Sept vs Jan-Dec) I doubt if this could explain the difference of approx. 3 μg/m3 between these measurements. I wonder why there is such a large difference between both measurements.

A.2.17. As mentioned by the referee, PM2.5 mass reported in our study (14.7 μg/m3) is lower than the measurement results reported by AIRPARIF for 2010 (18 μg/m3) which cannot be explained by the slight shift in months between these measurements. This difference is discussed in detail in Sect. 3.3 (p 7830) and Figs 2 and 3 of Bressi et al. (2013). Briefly, PM2.5 mass determined in our study agrees well with TEOM-FDMS measurements conducted at 30°C without taking into account semi-volatile materials (r2=0.94, slope=0.94, n=318). However, taking into account semi-volatile materials in TEOM-FDMS measurements leads to higher concentrations compared with the gravimetric method (18.4 versus 14.8 μg.m-3, respectively). In our study, gravimetric measurements are performed after PTFE filters are equilibrated for 48h at ambient T and below 30% relative humidity (RH) to minimize the influence of water adsorption (and perform more robust chemical mass closure studies), whereas standard European methods (EN 14907) suggest performing gravimetric measurements at 50% RH. Our gravimetric method thus underestimates PM2.5 mass compared to EU reference methods by ca. 20 % on average. This information has been added in the manuscript as follow: P33242, l23: “Note that the gravimetric procedure used underestimates PM2.5 mass compared to EU reference methods (EN 14907) by ca. 20% on average (see Bressi et al., 2013).”

R.2.18. 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 not equal.

A.2.18. We agree with the referee that the average contribution of the heavy oil combustion factor found in our study is high given the location of Paris. First, enhancement of this factor in Paris may result from increasing ammonium sulphate (and secondary OM) concentrations in the plume of upwind oil combustion sources. Second, it could also be explained by a mixing of different sources in this factor. As mentioned p33273, l16-22, different types of industry-related sources present in northern France (such as metallurgical sintering plant, metallurgical coke plant, etc.) could contribute to our heavy oil combustion factor, although they may not be distinguished since specific tracers (e.g. Rb, Cs, Bi, Th, etc.) are not analyzed in our study. As mentioned p33268, l15-20 (and p33278, l19-20) the influence of local industries cannot be excluded as well given the high number of industrial activities in the region of Paris. The relevance of this heavy oil combustion factor is on the other hand attested by i) its chemical profile, showing a high proportion of V, Ni and SO4 as reported in most European studies (Viana et al., 2008) and a reasonable V to Ni ratio (see p3326, l15) and ii) its seasonal variations, exhibiting higher concentrations during spring/summer as reported in other studies (Mooibroek et al., 2011). Nevertheless, the high contribution of this heavy oil combustion source is for the first time reported in Paris and should be the object of

future research. Regarding the referee’s comment, the following sentence has hence been added: P33273, l27: “Further research investigating the contribution of heavy oil combustion sources to fine aerosols should be conducted in the region of Paris, given the surprisingly high levels found in our study.”

R.2.19. 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 PM2.5 where not exceeded at urban background locations but where exceeded at roadside locations. The contribution of traffic at the sampling site is calculated to be 2.1 μg/m3, however, looking at the PM2.5 measurements for 2010 as given in the aforementioned report the difference between the urban background (on average 18 μg/m3) and the roadside sites (30 μg/m3) 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 μg/m3. 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 PM2.5 measurements at urban and kerbside locations in Paris.

A.2.19. We agree with the referee that one can reasonably expect the traffic contribution at an urban background site of the city of Paris to be higher than the one reported in our study (2.1 μg/m3, 14%). Difference evidence that will be detailed later on however support this surprising result. Regarding your specific question related to the substantial difference observable between one roadside site and one urban background site, the main reason for observing such high discrepancies between both sites is the specificity of the roadside site chosen in report Air Quality in the Paris region 2010. The authors know especially well this roadside sampling site (Boulevard périphérique porte d’Auteuil) since they have been performing sampling measurements during the period 2009-2010. The samplers are located less than one meter from this major road (ring road) of Paris, which has a traffic flow of more than 220 000 vehicles per day (please see picture of the site in AIRPARIF and LSCE, 2012 at p19). The influence of very local traffic processes (e.g. road dust, mechanical abrasion, etc.) thus highly affects this site without however necessary affecting the urban area of Paris. AIRPARIF attempted to estimate the main sources of PM2.5 at the urban site described in our study and at the aforementioned traffic site. Their methodology is based on a coupling of the PM2.5 chemical composition and emission inventories and is thus independent from the PMF approach (see AIRPARIF and LSCE, 2012 p17-18 for more details on the methodology). In addition, using a methodology introduced by Lenschow et al. (2001), AIRPARIF estimated the proportion of PM2.5 produced by local traffic, by the agglomeration or imported in the region. The main results of this study can be found in their press release at http://www.airparif.asso.fr/_pdf/publications/ communique_presse_particules_anglais_110914.pdf or in the aforementioned report. As inferred by the referee, at the roadside site 44% of the PM2.5 measured is estimated to come from local traffic and only 4% is estimated to come from the agglomeration traffic (p2 of the press release). On the other hand, at the urban site, the contribution of agglomeration and imported road traffic are 8 and 6%, respectively (p3 of the press release) i.e. the overall contribution of road traffic is estimated to be 14%, which is exactly our PMF estimation. In short, other evidences supporting the contribution of the road traffic source found in our study are: i) the fairly good agreement observed between the temporal variations of fossil fuel black carbon and our road traffic source (please see comment A.3.2. given to Referee 3), ii) the low levels of EC measured at our site compared to those measured in urban background sites of other cities: Paris (1.3 μg/m3, Bressi et al., 2013), Barcelona (1.7 μg/m3, Reche et al., 2011), London (1.9 μg/m3, Reche et al., 2011), Istanbul (2.9 μg/m3, Theodosi et al., 2010) or Beijing (8.2 μg/m3, Yang et al., 2011) (see A.1.3. given to Referee 1 for more details) and iii) the fact that in our study, road traffic refers to primary traffic emissions whereas secondary emissions can be found in the ammonium nitrate rich factor for instance. Please see comments A.3.2. and A.1.3. given to Referee 3 and 1, respectively more

detail.

R.2.20. 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.

A.2.20. We agree with the referee that choosing sites that have a similar distance from the coast as the sampling location in Paris has, would be more relevant to compare the contribution of marine aerosol sources to PM2.5. However, putting this constraint would prevent us from comparing our marine aerosol level with any site reported in our manuscript. In fact, our sampling site is located at approximately 300km from the coast in the north direction, and about 500km in the southwest direction (influencing significantly the contribution of this source, see Fig. 6 p33305). On the other hand, the distance from the coast for the other sites compared in our study are substantially lower: Helsinki (<10km), Athens (<10km), Schiedam (ca. 30km), Rotterdam (ca. 30km), Cabauw (ca. 50 km), Hellendoorn (ca. 80km) and Vredepeel (ca. 130km). Please also note that the distance from the coast does not seem to be the only factor governing the marine aerosol concentration, including in the study of Mooibroek et al. (2011); in fact, marine aerosol levels are for instance twice higher at Cabauw than at Rotterdam (1.6 and 0.8 μg.m-3, respectively) although the former site is further from the coast than the latter (approximately 50 and 30 km, respectively). Your comment has been taken into account in the manuscript as follow: P33275, l7: “This comparison however presents some limitations since the distance from the coast is substantially higher for our sampling site (ca. 300-500 km depending on the directions) compared to the aforementioned sites (around or below 100km).”

R.2.21. 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 interesting to see if the number of vehicles across the seasons stays approximately the same.

A.2.21. Looking at the number of vehicles in Paris during the seasons indeed appears interesting although linking emission and ambient measurements is often a complicated task. The monthly variation of the traffic flow in the city of Paris can be found in Paris, 2009 (p18) and Paris, 2010 (p 20) for the years 2009 and 2010, respectively. The main results are reported in Fig. A.2.21. (see supplementary material of this comment). From September 2009 to September 2010, the monthly variation of the traffic flow is low (relative standard deviation of 8%), indicating that there is not much change in the number of vehicles during the seasons. The only noticeable exception is found for the month of August (-27% compared to the annul mean). The low variation of the traffic flow according to seasons is in line with the stable concentrations observed for our road traffic source all along the year.

R.2.22. Page 33279, line 7: The conclusions are true if the assumption the sum of the measured species account for most of the PM2.5 mass is true.

A.2.22. As discussed in the comment A.2.14. the authors believe that the assumption the sum of the measured species account for most of the PM2.5 mass is true (see Bressi et al., 2013, Fig. 2).

The referee is thanked once again for his valuable comments.

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Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/13/C13491/2014/acpd-13-C13491-2014-supplement.pdf

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