Sources and geographical origins of fine aerosols in Paris (France)

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Responses to

Supplementary Material for

Response to Referee 1

The anonymous Referee 1 is thanked for his extensive comments on our manuscript. Please find below in italic the comments of the Referee 1, in default font the answers of the authors and in bold the changes made in the manuscript.

R1.1. 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.
A1.1. Regarding the approach and the methods, the authors agree with the referee that the PMF model used here has been widely reported in the literature. However, the authors also believe that the way PMF was applied here is innovative in some extent: using the bootstrap technique to determine the appropriate number of factors – which represents a major question in PMF studies – has not been reported elsewhere to our knowledge (see also the acknowledgement of Referee 2 in his comment R.2.11. on this subject). Regarding the results, the authors agree with the referee that the nature of the sources found in our study have also been reported elsewhere (although a thorough investigation of their chemical profiles was conducted in our case). However, the conclusions reached from our study are according to us novel and unexpected, such as for instance stating that on average, more than half of $PM_{2.5}$ mass measured in the city of Paris is due to mid- or long-range transport of secondary aerosols stemming from continental Europe, whereas local sources only contribute a quarter of the annual averaged mass. The contribution of PM wood burning which is equal (on a yearly basis) to PM related to traffic was also quiet unexpected for a megacity and in particular for Paris which is affected by diesel emissions (2/3 of the fleet). A robust database and source apportionment – as presented here - was needed to validate the few (time limited) studies which have highlighted this point before. Finally, the results of this study had strong implications on regional air quality strategic plans, which point is often raised as an objective of many papers dealing with PM pollution in urban areas. The results obtained in this paper have a large societal impact since local authorities have decided beginning of 2013 - for the first time in France - to forbid the use of open fire place in Paris city. This measure is currently undertaken by many other French cities. For these reasons (and those exposed in the paper), we believe that this paper does present original (and robust) conclusions that are worth being published.

R1.2. 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).
A1.2.The limitations stated by the Referee 1 regarding the metallic, traffic and mineral dust sources will be discussed in the comments A.1.22., A.1.3 and A.1.4., respectively.

R1.3.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 teh 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, whih 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).

A1.3. The limitations abovementioned regarding a mineral dust source will be discussed in the comment A.1.4., a focus will be made on the traffic source only in the current comment A.1.3. We agree with the referee that the traffic contribution seems at the first glance relatively low considering that the study location is Paris. First, let us recall that the location of our sampling site (roof platform) represents urban background conditions (i.e. not close to traffic and re-suspended dust). Second, this PMF road traffic factor represents primary emissions of vehicles only as stated in the manuscript (p 33258, l6-7; p 33259, l2) and as suggested by the absence of secondary inorganic species in its factor profile as well as its low OC to EC ratio of 1.2 (see p33258, l4-6, 20). As stated p33258, l26-27: "the secondary nature of road traffic related aerosols will be found in other factors" (e.g. the A.N. rich factor). The overall contribution of road traffic emissions to $PM_{2.5}$ mass including primary and secondary aerosols might thus be slightly higher (although ammonium nitrate is mostly imported in the region of Paris as suggested by the homogeneity of its levels at different sampling sites across the region, Bressi et al., 2013). Third, only fairly good agreement is observed between modelled and measured EC levels (r^2=0.56, see Table S1 and p33252, l4-5), which might lead to higher uncertainty on the road traffic source given the predominance of EC in this factor.

However, several evidences support the relevance of the contribution of the road traffic source estimated from PMF here. Comparisons with external compounds that are presumably primarily emitted by road traffic sources have been investigated. As discussed in details in the comment A.3.2., NOx has been monitored by the regional air quality agency (Aiparif) at the same urban background site (AIRPARIF and LSCE, 2012). According to emission inventories (see p33263, l21-29), NOx are primarily emitted by road traffic although other sources (e.g. biomass burning) might also contribute to its concentrations. In addition, 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 (BCff) and a wood burning (BCwb) 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., 2013). Results of these comparisons are shown in Fig. A.1.3, which can be found in the supplementary material of this comment. Fairly good correlation is observed between NOx and the road traffic source (r=0.50, n=329) although discrepancies are observed during wintertime, which might be related to wood burning emissions of NOx during this period (see comment A.3.2. for further interpretation). Interestingly, correlations between Road traffic and BCff are fairly good all along the year (r=0.50, n=327) including wintertime, while no correlation is found between BC_{ff} and the other sources (r typically equals to 0.2, see Table A.3.2.). These fairly good correlations with external tracers give further confidence on the relevance of the contribution of the road traffic factor. The relatively low contribution found here might also be due to the specific flat topography of the Paris region, which likely favours the dispersion of local emissions due to road traffic. Elemental Carbon levels - that mostly come from traffic emissions - are indeed lower in Paris than in other cities at urban background sites: Paris (1.3 µg/m^3, Bressi et al., 2013), Barcelona (1.7 µg/m^3, Reche et al., 2011), London (1.9 µg/m^3, Reche et al., 2011), Istanbul (2.9 µg/m^3, Theodosi et al., 2010) or Beijing (8.2 µg/m^3, Yang et al., 2011). (Note: this comparison presents some limitations as for some studies i) black carbon and not elemental carbon was measured and ii) PM_{10} and not $PM_{2.5}$ was sampled, although EC should mostly be in the fine mode.) In addition, although the average relative contribution of our road traffic source (14%) is lower than the median contribution (20%) found by Belis et al. (2013) in his review on SA studies for urban background sites, it remains in the 11-32% inter-quartile range.

Regarding the last part of your comment, stating that "*This would change their conclusions, as more mitigation strategies should then be local (and fewer of them external to the city)*", the authors do believe that our conclusions remain robust in spite of the abovementioned limitations. Other independent studies indeed support the importance of mid- long- range transport of PM in the city of Paris. First, Airparif estimated the proportion of local versus imported $PM_{2.5}$ mass based on Lenshow's methodology (Lenschow et al., 2001) at the sampling site described in our study for the same period (see AIRPARIF and LSCE, 2012 for the full report). They concluded that 32% of $PM_{2.5}$ measured at the urban site are produced by the agglomeration of Paris, whereas 68% are imported from outside the Ile-de-France region (see for instance p3 of the press release, Airparif, 2011). Second, from the one-year chemical characterization of $PM_{2.5}$ determined at the urban site and with the use of chemical transport models, Petetin et al. (2013) conclude that: "pollution reduction measures at the Paris scale alone are inadequate to prevent most exceedances of PM standards, thus underlying the necessity of integrated AQ management at the regional/continental scale". Third, from a one-month sampling campaign conducted in summer in the region of Paris, Freutel et al. (2013) conclude that: "the influence of the Paris emission plume onto its surroundings (…) [is] rather small". Fourth, from a one-month campaign conducted in winter in the region of Paris, Crippa et al. (2013) reports that: "The similarities in aerosol composition, total mass and temporal variation between the three sites [used in this study] suggest that particulate pollution in Paris is dominated by regional factors, and that the emissions from Paris itself have a relatively low impact on its surroundings." These studies therefore strongly support our conclusions.

R1.4.- 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 analythical methods (digestion wth 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.

A1.4. The authors agree with the referee it seems likely that a city dust source should be resolved in the analysis but it cannot be due to the lack of tracers (as stated p33266, l15-20). Limitations related to the analytical technique used have been more clearly 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 HNO_3), which might not be able to dissolve entirely these compounds (see e.g. Robache et al., 2000)."**

In addition, as discussed in the comment A.2.5. the specific location of our sampling site (20 m a.g.l.) might prevent from being highly affected by a road dust source. The following comment has been added accordingly:

P33242, l9: **"It should however be highlighted that this [sampling] site is at 20m a.g.l. which might prevent near ground sources (e.g. road dust) to be considered in our study.**"

Limitations related to the absence of a clearly identified road dust source have been reported in the manuscript as follow:

P33258, l10: **“Note that the lack of mineral tracers mentioned in Sect. 2.1.2 might prevent us from identifying a road dust fraction in this factor.”**
P33274, l5: **“As mentioned in Sect. 4.1.2, the absence of a road dust fraction might partly explain the relatively low contribution of our road traffic source.”**
P33266, l25: **“Further research should be conducted to better characterize mineral dust contribution to fine aerosols in the region of Paris.”**

To the best of our knowledge, only three studies have focused on mineral dust in fine aerosols in the region of Paris (Bressi et al., 2013; Favez, 2008; Guinot et al., 2007). Due to the abundance of calcium in mineral dust (e.g. see Putaud et al., 2004), these studies have used a non-sea-salt calcium (nss-Ca) to dust conversion factor (CF) to estimate the contribution of this source. In the region of Paris, Guinot et al. (2007) experimentally determined a nss-Ca to dust CF of ca. 6.5 based on mass closure exercises. Applying this CF, Favez (2008) found an average mineral dust contribution of 6% to fine aerosol mass from a 2 year sampling campaign performed at an urban background site of the city of Paris. Applying this CF, Bressi et al. (2013) report a mineral dust contribution of 3-4% depending on the site studied from a one-year campaign performed at five different sampling sites of the region of Paris. Note that Putaud et al. (2004) suggest using a CF of 5.6 at their sampling site during non-Saharan dust episodes; applying this conversion factor would thus lead to comparable mineral dust estimation in the region of Paris. The thorough chemical mass closure reported in Bressi et al. (2013) strongly suggest that a noticeable contribution of a mineral source in Paris is not found (since PM obtained from chemistry cannot be above PM obtained from gravimetric methods). The authors thus believe that the contribution from mineral dust to fine aerosols in Paris is per se small. The authors also believe the conclusions of the manuscript remain robust in spite of the lack of a mineral dust source (see second paragraph of the comment R.1.3.).

R.1.5. page 33239, line 9, USEPA 2011 b and a should be interchanged.
A.1.5. This has been modified accordingly.
R.1.6. same page, line 23: "possible exceedances...", with an annual mean of 14 ug/m3 the city doesn’t exceed any EU limit values, correct?
A.1.6. Given the comment of the referee “, or to possible exceedances of E.U. limit values” has been deleted from the manuscript.
R.1.7. page 33240, line 2: please enumerate briefly some of the measures, or at least the sources that were targeted
A.1.7. The sources that are targeted have been mentioned in the manuscript as follow:
P33240, l1: “At the present times, such strategies seem to be rather insufficient in this city. Despite the abatement policies implemented (e.g. prefectoral order n◦ 2011-00832 of the 27 October 2011 targeting sources such as wood burning, agricultural fertilizers, industrial emissions, etc.), $PM_{2.5}$ annual levels in Paris have remained rather stable during the last ten years (AIRPARIF, 2012).”
R.1.8. page 33241, line 1: "questions", I’d rephrase this, it doesn’t question it: it is evident that 19 days are not representative of a longer time series.
A.1.8. This sentence has been rephrased accordingly, taken also into account the comment R.3.5:
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.”**
R.1.9. 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.

A.1.9. First, sampling artefacts may be positive and/or negative. According to the authors, there is no scientific reasons suggesting sampling artefacts should be only positive and thus lead to an overestimation of OC mass. As mentioned p33242, l18-20 the reader is directed to the study of Bressi et al. (2013) for a thorough description of the chemical analyses. Briefly, to minimize sampling artefacts, before being sampled quartz filters (QMA, Whatman, 47 mm diameter) were baked at 480°C for 48h. Field blanks were taken every week for the whole duration of the campaign. Once sampled, filters were stored at -20°C in a freezer prior to chemical analyses. OC was detected in field blanks with an average value of 1.1µgC.cm^{-2}, which represents ca. 15% of sampled filters OC concentrations. Blank corrections have been performed by subtracting the blank average to the sampled filter values. As discussed in Sect. 3.4. of the aforementioned study, filter sampling OC concentrations were compared for a period of 70 days (6 January–15 March 2010) with semi-continuous hourly measurements of VOC denuded OC concentrations in PM2.5, obtained using an OCEC Sunset field instrument (Sunset Laboratory, Forest Grove, OR, USA; Bae et al., 2004). The default thermal program (National Institute for Occupational Safety and Health, NIOSH; Birch and Cary, 1996) was used in this instrument (whereas the EUSAARII protocol, Cavalli et al., 2010 was used for filter analyses). Measurement uncertainty given by the OCEC Sunset field instrument is poorly described in the literature and an estimate of 20% was considered following Peltier et al. (2007). Comparisons between filter sampling and semi-continuous OC measurements are exhibited in Fig. 4b of Bressi et al. (2013) and show good agreements (r^2 of 0.84) with a slope of 1.20 and a y-intercept of +0.17 µgm−3. Slope difference may partly originate from the different thermal programs used, having a nearly 200 C difference for the last temperature plateau under Helium (Cavalli et al., 2010).

R.1.10. 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 summarized (by around 20% would be my suggestion).

A.1.10. 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:

$$x_{ij} = \sum_{k=1}^{p} g_{ik} * f_{kj} + e_{ij} \qquad (1)$$

where x_{ij} is the measured concentration of the jth species in the ith sample, g_{ik} is the contribution of the kth source to the ith sample, f_{kj} is the concentration of the jth chemical species in the material emitted by the kth source and e_{ij} 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:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{e_{ij}}{\sigma_{ij}}\right)^2 \qquad (2)$$

where σ_{ij} is the uncertainty associated to the jth species in the ith sample. Different Q functions can be defined: Q_{true} calculated including all data and Q_{robust} calculated excluding outliers i.e. data for which the scaled residual (e_{ij}/σ_{ij}) is greater than 4. (Note that $Q_{theoretical}$ 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.1.11. page 33243, line 6: Linuma should have a capital "L"

A.1.11. The first author of the publication is Iinuma, with a capital "i".

R.1.12. page 33248, lines 9-11: repetition from the previous page

A.1.12. Although the parameters chosen for the calculation of backtrajectories are the same for CPF and PSCF, the model versions used are different; these two lines are thus according to the authors necessary (see also comment R.2.10 on this subject).

R.1.13. page 33250, line 20: if Qrobust=Qtheoretical+model error, values of the Qrobust/Qtheoretical ratio <1 should not be valid outcomes of fthemodel, correct? As far as I understand it, Qrobust should be >Qtheoretical, 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.

A.1.13. Please see comment R.2.7. given to the Referee 2 on this subject. Qtheoretical is not reported in the manuscript anymore.

R.1.14. page 33251, line 22: here the issue of the Q seems to be corrected, as Qrobust and Qtheo 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.

A.1.14. As mentioned in the comment A.1.13. Qtheoretical is not reported in the manuscript anymore.

R.1.15. page 33252, line 23: "biogenic source", the same is true for the mineral dust source, please make explicit reference to this.

A.1.15. Explicit reference to the mineral dust source has been reported:

P33252, l23: **"It could also be related to the absence of clearly identified biogenic and mineral dust sources in our study"**

R.1.16. page 33253: these 2 sections are a bit repetitive and only introduce what will be presented in section 4. They could be summarised.

A.1.16. The titles of the sub-sections 3.3.1 and 3.3.2 have been deleted (i.e. only the section 3.3 remains). The text has been shortened as follow:

P332523, l5: "**On the other hand**" has been removed

P33253, l9: "**the 5th, 25th, 50th, 75th and 95th**" have been replaced by "**different**"

Please precise if further changes are necessary (and if yes please precise which ones).

R.1.17. page 33256, lines 19 and 23, what is "2005 in Puxbaum et al 2007"? Please clarify.

A.1.17. 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.1.18. 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.

A.1.18. The values of 10.3 and 10.8 are taken from the literature and cannot be modified (note also that the value of 10.3 has been determined in the region of Paris). The values of 13-15 recommended in the conclusions of the Ghent workshop on biomass burning could be reported in our study as well if the referee provides more detailed references (e.g. title and authors of an oral presentation).

R.1.19. 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.*

A.1.19. We do agree with the referee that nitrate from traffic might be formed within the city scale as reported by Amato et al. (2011) in Barcelona (Spain). Although the topography of Paris is substantially different from Barcelona (flat region favouring the dispersion of pollutant in the former case versus complex topography and architecture favouring “the accumulation and resuspension of particulate matter deposited at ground”, Amato et al., 2011), we still believe that a fraction of nitrate is formed within the city scale of Paris. This fraction is estimated to be approximately 0.2-0.7 $\mu g.m^{-3}$ which corresponds to 3-5% of $PM_{2.5}$ mass (from a comparison between our urban site and 3 rural sites located from 50 to 65 km away, see Bressi et al., 2013). As stated p33256, l26-27, we believe that secondary nitrate stemming from traffic is found in the Ammoniun Nitrate rich factor in our study. As stated p33264, l8-11, we believe that the Ammonium Nitrate rich factor “stems from a large variety of sources, likely mainly being road and non-road transport, industrial activity, agriculture, and biomass burning”. The absence of nitrate in our road traffic factor suggests that this factor is primary, which is in agreement with its low OC to EC ratio of 1.2 (as well as the absence of ammonium and sulphate).

R.1.20. page 33259, line 2: "carbonaceous", please add "carbonaceous and metallic"

A.1.20. “metallic” has been added accordingly.

R.1.21. page 33259, line 24: the explanation that Cu and K may come from shipping seems far-fetched, althouh I don’t have a better one. If Cu and K have ever been linked to shipping emissions, please provide the reference.

A.1.21. We agree with the referee that the explanation that Cu and K may come from shipping is questionable. The text has been modified as follow:

P33259, l24: **“Shipping is a possible source of EC and Ni, because (…)”**

P33260, l1: **“(…) due to inland emissions (of e.g. EC, Ni, K, Cu) from combustion processes. “**

R.1.22. 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?

A.1.22. As mentioned by the referee, since this source presumably reflects a mesoscale background aerosol, one should expect the presence of secondary aerosol components. Although this factor has been identified thanks to metals (Cd, Pb, Cu, Fig. 1), this does not mean that secondary aerosol component are not present. It is actually clearly shown in Fig. 2 that most of the mass of this factor is due to nitrate, sulphate and OM (for which a noticeable proportion could be secondary given the OC to EC ratio of 1.8 found in this factor). The high contribution of these secondary components to the factor mass is stated p33262, l18-19.

R.1.23. page 3266, lines 14 to end: indeed, only here is the missing mineral source acknowledged.

A.1.23. Please see the additional acknowledgements mentioned in comments R.1.4 and R.1.15.

R.1.24. page 33271, line 7: 14% for traffic in a megacity such as Paris seems really low. Please describe the limitations of the model.

A.1.24. Please see the extensive answer to Comment R.1.3.

R.1.25. 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.

A.1.25. We agree with the referee that the comparison with Dunkirk has some limitations. We however consider this comparison relevant given the geographical origin of our heavy oil combustion source (mainly “north of France although a local contribution is not excluded”, see p33268, l3-20). Dunkirk is not only influenced by shipping emissions but also by a wide variety of industries. As stated p33273, l16-19: “industry-related sources identified in Dunkirk (such as metallurgical sintering plant, metallurgical coke plant, etc.) (…) represent 37% of PM_{10} mass". These emissions could plausibly influence levels measured in Paris as discussed in Sect. 4.2. In addition, regarding the possible influence of shipping emissions in our heavy oil combustion source, please see our answer A.1.28.

R.1.26. 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.

A.1.26. Once again, we do agree with the referee that comparing Toronto and Paris has some inevitable limitations. We however consider the results of Lee et al. (2003) worth to be reported, given their relatively low estimation of 18% of their road traffic source to $PM_{2.5}$ mass in a 5.6 million inhabitants city.

R.1.27. 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.

A.1.27. We agree with the referee that the oxidation of SO_2 to sulphate is enhanced in summer although photochemistry cannot be considered as the only factor driving ammonium sulphate concentrations. Seasonal variations of SO_2 show their highest values in winter (Airparif, 2014). Low boundary layer height observable in winter will also enhance the absolute concentration of every species, sulphate included. In the city of Paris, air mass origins will also play a significant (if not the main) role in the levels of sulphate measured. A comparison of the levels of sulphate measured during one year at 5 sites (urban, suburban and rural) in the region of Paris is reported in Bressi et al. (2013, Figs. 10 and 11); sulphate absolute concentrations are high during winter as well as during summer. Please also note that Crippa et al. (2013) report higher sulphate levels in PM_1 from AMS measurements performed at 3 sampling sites in winter 2010 (see their Fig. 1), than what is reported by Freutel et al. (2013) in the region of Paris during summer 2009 (see their Fig. 4).

R.1.28. page 33276, line 17: "vessel activities", again, in the descriptionof the source it was ststed that the influence of ships in this source is minor. The authors should revisit the interpretation ofthis source, is it industrial emissions or ships? Please clarify.

A.1.28. As stated p33260, l15-19: "Vanadium and nickel are primarily emitted by heavy oil combustion, whose sources are industrial boilers (e.g. used in refineries), electricity generation boilers (e.g. oil power stations), large shipping ports, etc. (Jang et al., 2007; Moreno et al., 2010; Pacyna et al., 2007). It is difficult to distinguish between these sources, and "heavy oil combustion" seems to be the most suitable label for this factor." We agree with the referee that from the V to Ni ratio of the heavy oil combustion factor, we concluded that it mainly stems from industrial instead of shipping emissions (p33261 l16-18). However, its geographical origin "suggests that maritime transport clearly affects the contribution of this factor. The low V to Ni ratio reported in our study (Sect. 4.1.4) might thus not be the best proxy to distinguish between industrial and maritime heavy oil combustion" (p33268, l13-15). The authors believe that both industrial emissions and maritime transport affect this factor. Given the comment of the referee, this interpretation has been more clearly stated in the manuscript and the paragraph p 33261, l6-24 has been replaced by the following:

P33261, l-6: **"The V/Ni ratio might give insights on the sources associated with oil combustion as suggested by Pandolfi et al. (2010) and Moreno et al. (2010). Pandolfi et al. (2010) managed to discriminate between shipping and industrial emissions in a study conducted in the vicinity of a port in southern Spain (Algeciras), and showed that the former source exhibit higher vanadium to nickel ratio (ca. 3.0, range 2.1–3.1) than the later (range 0.9–1.9 for a stainless steel plant). The same conclusions are reached by Moreno et al. (2010). In our study, the V/Ni ratio in the heavy oil combustion factor is 1.4 on average, suggesting that industrial emissions (e.g. oil power station, petrochemical complex, boilers and furnaces) are prevalent. However, the geographical origin of this factor (Sect. 4.2) indicates that shipping emissions cannot be neglected either."**

R.1.29. page 33277, 1st paragraph:the contribution in winter is very low compared to the rest of the year, and this is unlikely. If the aithors explain the higher AS contributions in winter as resulting from anticyclonic episodes, shouldn't the same occur for the traffic source? Please explain.

A.1.29. As suggested by the referee 3 (last comment) the low PMtraffic contribution could be partly due to an overestimation of EC content in the biomass burning factor and an underestimation in the road traffic one, 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 discussed in the comment A.1.3 and shown in Fig. A.1.3, BC_{ff} (determined from an independent method at another urban background sampling site of Paris) also exhibits lower levels during winter, suggesting that this pattern is real, although it is not fully explained. The higher contribution of the A.S. source is mostly due to the advection of air masses from continental Europe (ammonium sulphate is almost exclusively imported in the region of Paris, Bressi et al., 2013) although anticyclonic conditions will naturally enhance its contribution. The following comment has been added in the manuscript:

P33277, l1-3: **"The road traffic source exhibits rather stable concentrations all along the year (annual average of 2.1±2.1 µg.m^{-3}), with however a smaller contribution during winter (1.3±1.4 µgm^{-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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Response to Referee 2

The authors would like to thank the anonymous Referee 2 for his extensive and constructive comments, which will help clarifying and improving our manuscript. Please find below in italic the comments of the Referee 2, in default font the answers of the authors and in bold the changes made in the manuscript.

R.2.1. 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 $PM_{2.5}$ 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 $PM_{2.5}$ 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 $PM_{2.5}$ 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 $PM_{2.5}$ 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 $PM_{2.5}$ 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:

$$x_{ij} = \sum_{k=1}^{p} g_{ik} * f_{kj} + e_{ij} \quad (1)$$

where x_{ij} is the measured concentration of the j^{th} species in the i^{th} sample, g_{ik} is the contribution of the k^{th} source to the i^{th} sample, f_{kj} is the concentration of the j^{th} chemical species in the material emitted by the k^{th} source and e_{ij} 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:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{e_{ij}}{\sigma_{ij}}\right)^2 \quad (2)$$

where σ_{ij} is the uncertainty associated to the jth species in the ith sample. Different Q functions can be defined: Q_{true} calculated including all data and Q_{robust} calculated excluding outliers i.e. data for which the scaled residual (e_{ij}/σ_{ij}) is greater than 4. (Note that $Q_{theoretical}$ 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: Q_{true} calculated including all data and Q_{robust} calculated excluding outliers i.e. data for which the scaled residual (e_{ij}/σ_{ij}) is greater than 4. (Note that $Q_{theoretical}$ 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 $Q_{theoretical}$ 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 ($PM_{2.5}$): 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 $PM_{2.5}$ at other suburban or rural sites across the Paris metropolitan area (for financial reasons). However, during this campaign, co-located sampling of PM_{10} 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 PM_{10} 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 HNO_3), 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. $PM_{2.5}$ 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 $PM_{2.5}$ 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 (r^2=0.98, slope=1.00, n=335), assuming a mineral dust contribution calculated as [Ca^{2+}]/0.15 (Guinot et al., 2007). On average, the mineral dust fraction contributes 3% of $PM_{2.5}$ mass at this site using the above mentioned calculation.

As mentioned by the referee, by using $PM_{2.5}$ 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 PM_{10} 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 $PM_{2.5}$ 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, $PM_{2.5}$ mass reported in our study (14.7 $\mu g/m^3$) is lower than the measurement results reported by AIRPARIF for 2010 (18 $\mu g/m^3$) 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, $PM_{2.5}$ mass determined in our study agrees well with TEOM-FDMS measurements conducted at 30°C without taking into account semi-volatile materials (r^2=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 $\mu g.m^{-3}$, 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 $PM_{2.5}$ 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 $PM_{2.5}$ 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 SO_4 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/m^3, 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 $PM_{2.5}$ at the urban site described in our study and at the aforementioned traffic site. Their methodology is based on a coupling of the $PM_{2.5}$ 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 $PM_{2.5}$ 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 $PM_{2.5}$ 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/m^3, Bressi et al., 2013), Barcelona (1.7 µg/m^3, Reche et al., 2011), London (1.9 µg/m^3, Reche et al., 2011), Istanbul (2.9 µg/m^3, Theodosi et al., 2010) or Beijing (8.2 µg/m^3, 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 for more details.

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 $PM_{2.5}$. 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 $\mu g.m^{-3}$, 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 $PM_{2.5}$ mass is true (see Bressi et al., 2013, Fig. 2).

The referee is thanked once again for his valuable comments.

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Response to Referee 3

The authors would like to thank the anonymous Referee 3 for his extensive and constructive comments, which will help clarifying and improving our manuscript. Please find below in italic the comments of the Referee 3, in normal font the answers of the authors and in bold the changes made in the 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 quite well-written. I recommend publishing this work after the authors respond to the following comments.

General comments
R3.1. 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.
A3.1. 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 μgm^{-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 μgm^{-3}), Zurich (19.0 μgm^{-3}), Prague (19.8 μgm^{-3}), Vienna (21.8 μgm^{-3}), Barcelona (28.2 μgm^{-3}) (Bressi, 2012; Putaud et al., 2010). Implementing effective $PM_{2.5}$ 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 μgm^{-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, 2012 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 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.*

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, NOx 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 (BCff) and a wood burning (BCwb) 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 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 NOx 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 NOx and road traffic (r=0.71) than between NOx and biomass burning (r=0.36) (see also Fig. A.R.3.2.a of the supplementary material). Although NOx are primarily emitted by road traffic, according to emission inventories (see p33263, l21-29) NOx 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 BCff (r=0.50), whereas biomass burning and BCff 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 BCwb (r=0.89), while poor correlations are found for BCwb and road traffic (r=0.14) (see Fig. A.3.2.b.). Comparisons between our PMF sources and BCff or BCwb are further discussed in Bressi (2013).

The table A.3.2. has been added in the supplementary material and referred to in the manuscript as follow: p33253, l.17: "Correlations between factor time series and their presumable tracers are reported in Table S6". 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, l9-18. 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 (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 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 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 PM_1 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.

Figures 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.

- 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, l12: **“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, l14.**

- 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, l19: “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, l29: **“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, l16: **“to”** has been added accordingly.

- Page 33265 line 18: change “authors” with “literature study”

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 sulfate."**

– 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"

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 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±2.1 µg.m^{-3}), with however a smaller contribution during winter (1.3±1.4 µgm^{-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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Supplementary material for Referee 1

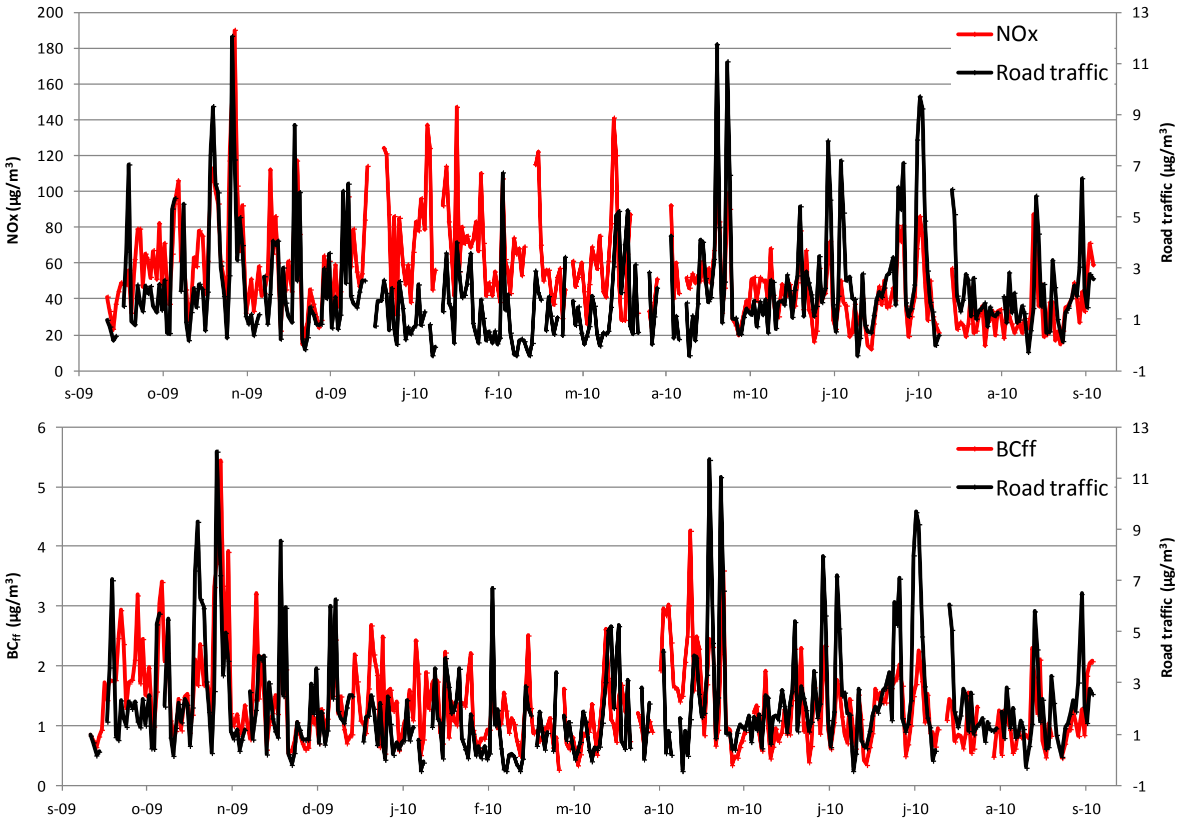

Fig. A.1.3. Comparison between the temporal variation of the PMF Road traffic factor and NOx (top) and BCff (bottom).

Supplementary material for Referee 2

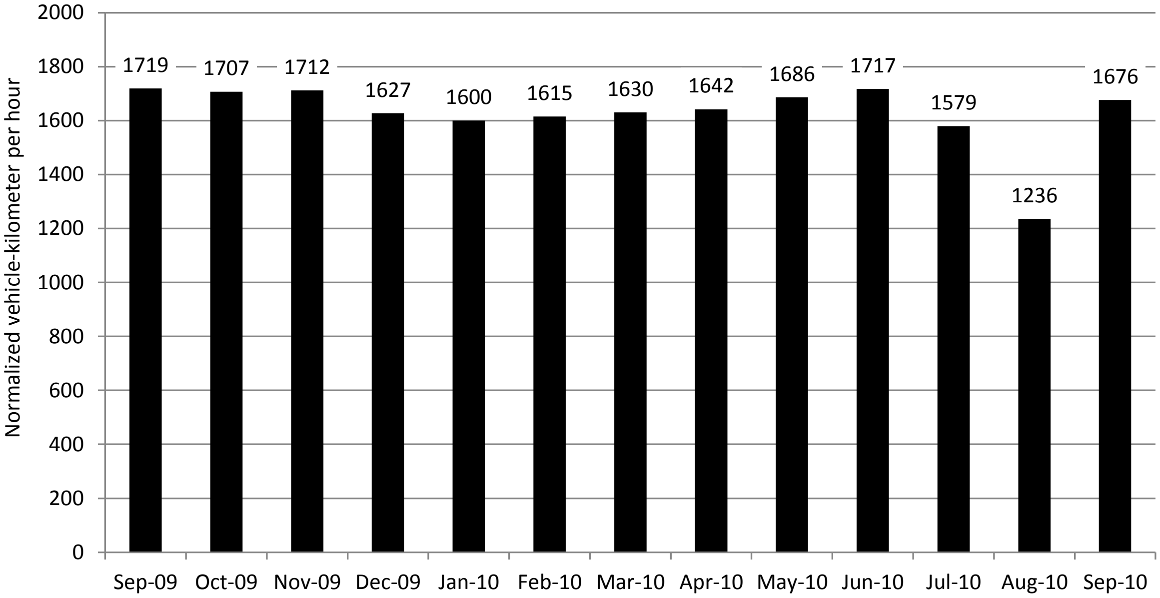

Fig. A.2.21.a. Monthly variation of the traffic flow in the city of Paris from September 2009 to September 2010 (adapted from Paris, 2010, 2009).

Note: the y-axis is in the unit of vehicle-kilometre per hour calculated between 7am and 9pm and normalized by the number of kilometres of road studied (see Fig. A.2.21.b).

Fig. A.2.21.b. Studied area (196km) used for Fig. A.2.21.a (adapted from Paris, 2010, 2009).

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Supplementary material for Referee 3

Table A.3.2. Correlation (Pearson coefficient, R) between PMF factor time series and their presumable tracers.

	SO_4	NO_3	V	NOx	BCff	Lev	Mann	BCwb	Na	Cl	Mg	Pb	Cd
A.S. rich factor	**0.85**	**0.58**	0.07	0.25	0.25	0.21	0.34	0.20	-0.30	-0.17	-0.32	**0.51**	0.40
A.N. rich factor	**0.73**	**0.99**	0.24	0.27	0.19	0.29	0.39	0.31	-0.24	-0.03	-0.26	0.43	**0.52**
Heavy oil combustion	0.35	0.11	**0.75**	0.09	0.22	-0.06	-0.05	-0.01	-0.25	-0.26	-0.16	0.29	0.27
Road traffic	-0.09	-0.15	0.08	**0.50**	**0.50**	-0.01	0.01	0.14	-0.25	-0.23	-0.19	0.23	0.20
Biomass burning	0.21	0.44	0.12	**0.61**	0.22	**0.99**	**0.98**	**0.89**	-0.12	0.18	-0.17	0.42	0.31
Marine aerosols	-0.45	-0.25	-0.25	-0.20	-0.22	-0.11	-0.15	-0.14	**0.91**	**0.82**	**0.88**	-0.35	-0.37
Metals industry	0.33	0.33	0.42	0.36	0.32	0.21	0.28	0.27	-0.19	-0.07	-0.20	**0.85**	**0.92**

Legend: BCff: fossil fuel black carbon, Lev: levoglucosan, Mann: mannosan, BCwb: wood burning black carbon. Pearson coefficients higher than 0.5 are indicated in bold.

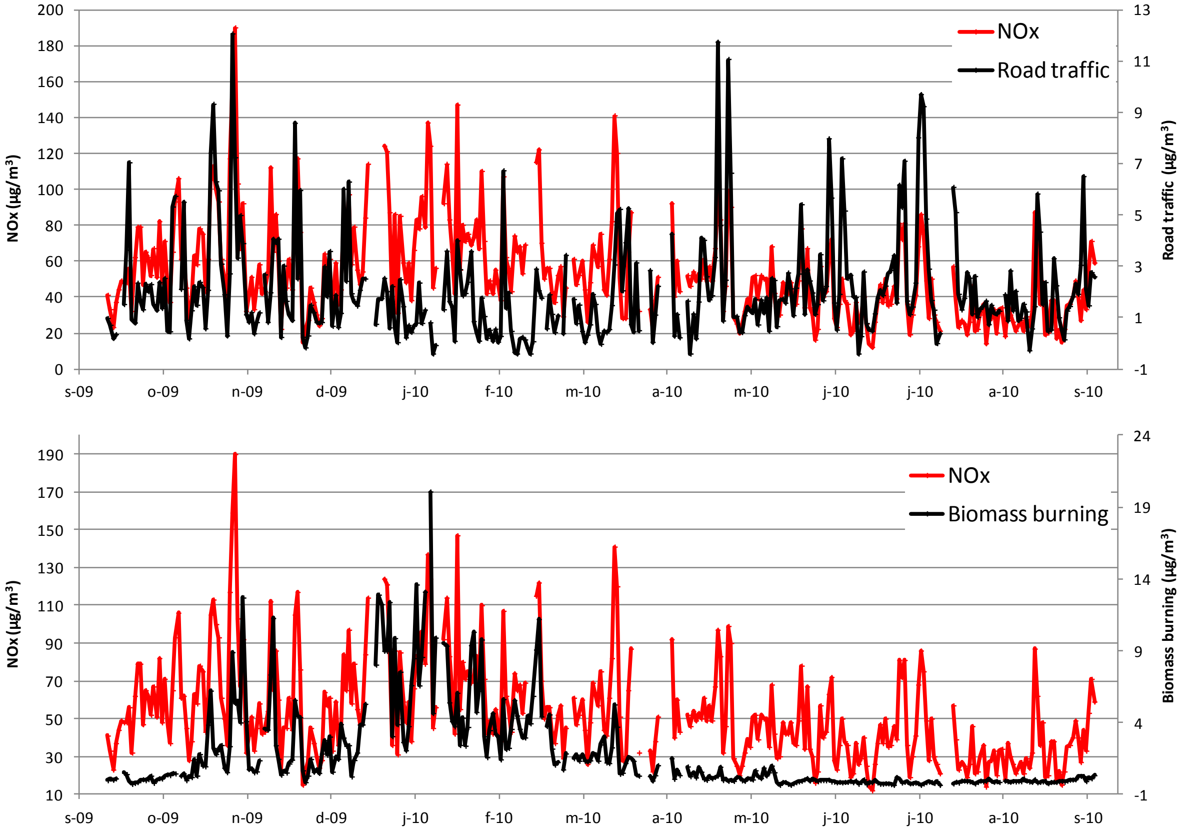

Fig. A.3.2.a. Comparison between NOx and Road traffic (top) or Biomass Burning (bottom) PMF factors.

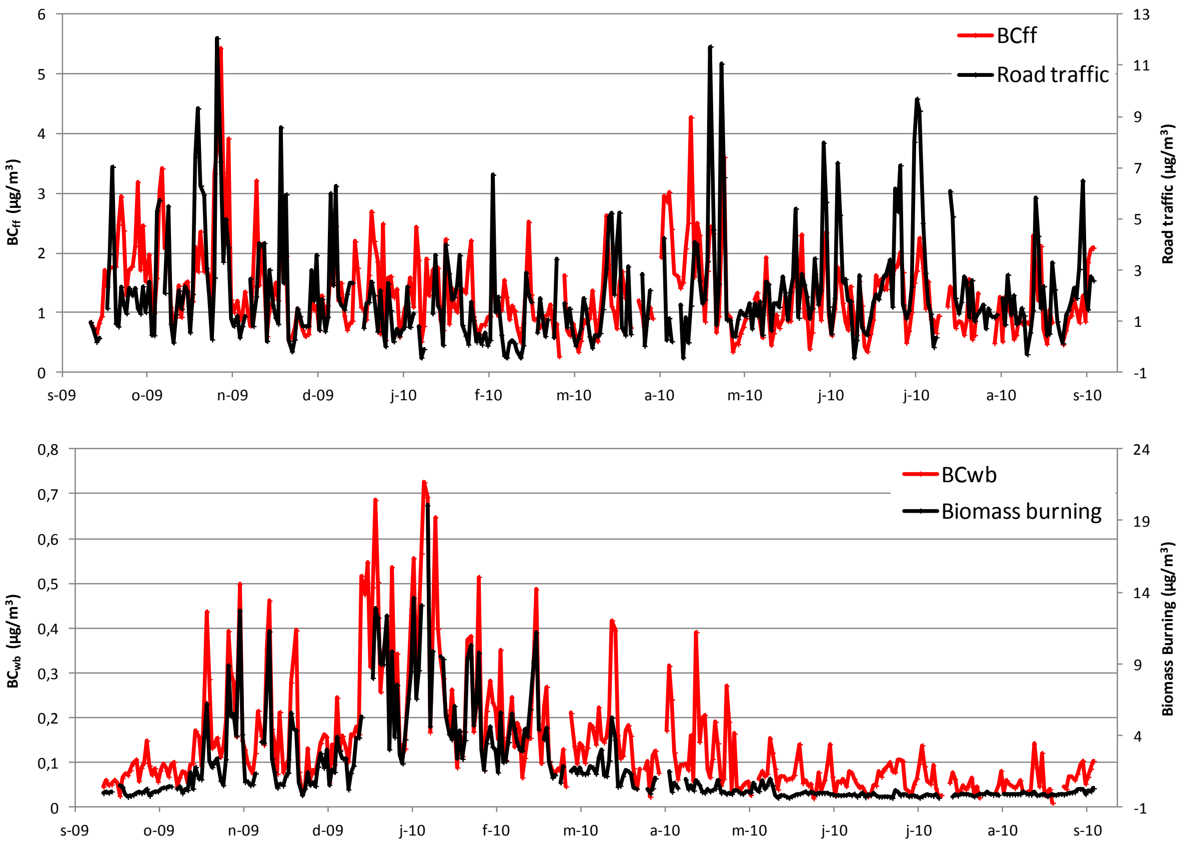

Fig. A.3.2.b. Comparison between BC_{ff} and Road traffic (top), and BCwb and Biomass Burning (bottom). Legend: BCff: fossil fuel Black Carbon, BCwb: wood burning Black Carbon (see comment A.3.2. for further explanations).

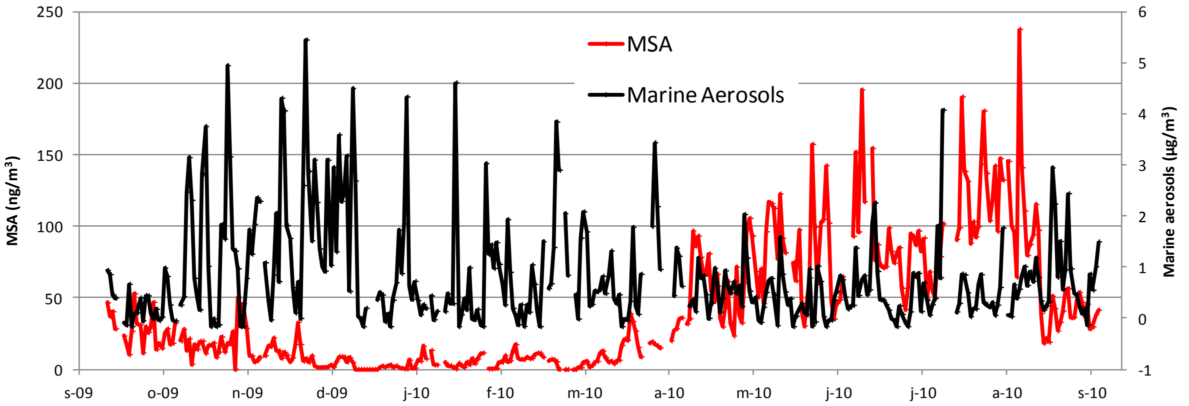

Fig. A.3.3.a. Comparison between methane sulfonic acid (MSA) and marine aerosols temporal variations (adapted from Bressi, 2013).

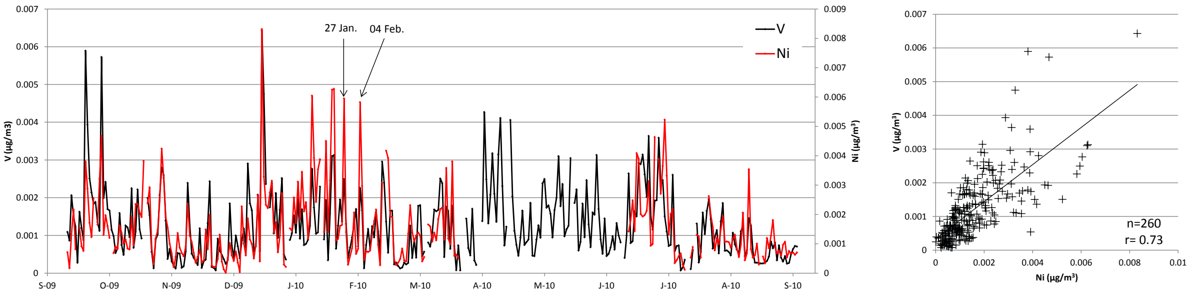

Fig. A.3.3.b. Comparison between Vanadium and Nickel temporal variations.

Note 1: for Ni, the period April-May is not shown here for clarity since most of the days have been replaced by the median of Ni concentrations (see Table S2 and comment p33251, l13-15). These values have been downweighted in our PMF analysis (see Sect. S1).

Note 2: some days exhibit lower V/Ni ratios but air mass origins are not sufficient to explain these observations (e.g. same V/Ni ratios of 0.4 observed on 27 January 2010 and 04 February 2010 but different air masses originating from Benelux and the Atlantic ocean, respectively).