Answers to Reviewer 1:

This manuscript reports the source apportionment results from three aerosol mass spectrometers and two black carbon analyzers deployed at three stationary sites in the Paris metropolitan area during winter 2010. It presents a thorough analysis of the chemical characteristics and the sources of aerosol particles in the Paris region during winter season. The subsequent discussions on the effect of Paris on local and regional air quality are very interesting. The overall quality of this work is good and the manuscript is well-written. I thus recommend its publication after the authors respond to a few comments.

- I'd like to suggest that the authors are more specific about the spatial contexts of the regional and local sources. For example, what region (spatial coverage) does "local" correspond to? Does it refer to the city center and nearby vicinity only or to the broad metropolitan area? Speaking of Paris emissions, it seems that the latter is more relevant. According to Wikipedia: "A metropolitan area is a region consisting of a densely populated urban core and its less-populated surrounding territories, sharing industry, infrastructure, and housing." (http://en.wikipedia.org/wiki/Metropolitan_area). So, what if all three sites are located within the Paris metropolitan area and neither GOLF nor LHVP is a background site strictly speaking? The similarities in the time series of primary aerosol species (HOA, COA, BBOA, and BC) seem to suggest so. Their diurnal patterns correlate very well with human activities (e.g., rush hour, meal times, and wood burning for heating) and there are no time shifts in the temporal variation profiles among the three sites. If urban emissions are the main sources of primary aerosols in Paris, the fact that they together constitute a significant fraction (~30% or more according to reading Fig. 12) of the aerosol loading indicates emissions from Paris is a significant contributor to aerosol pollution in its metropolitan region. It was mentioned on page 22562 that aethalometer measurements at a remote rural site located 58 km east/northeast from the center of Paris show no significant differences from the BC levels detected at GOLF and LHVP. I agree with the authors that this could be interpreted as an evidence for the regional infuences on Paris aerosol. However, another interpretation is also possible – Paris emissions control BC loading in the region. These points need to be clarified.

The urban core of Paris is ~20 km in diameter, while the greater Parisian metropolitan area is ~ 40 km in diameter; the SIRTA and GOLF sites are located near the edges of this metropolitan area. While we agree with the reviewer that homogeneity in terms of chemical composition, mass concentration and sources among the three sites could theoretically indicate a dominant impact of Paris on its surroundings, the following evidence indicates the opposite conclusion.

1. The MEGAPOLI project showed that the yearly impact of Paris emissions is on average equal to only 30% of PM2.5, while the remaining 70% is advected to Paris and impacted by long-range transport of continental pollution (Beekmann et al., 2012). These findings included one year of PM2.5 chemical composition measurements at 4 rural sites,

covering a spatial distribution of 140km (and overlapping with measurements performed during the summer phase of MEGAPOLI). Together with meteorological measurements that indicate which of these rural stations is upwind of Paris, these measurements indicate the dominance of regional sources over the Paris plume.

2. PM1 levels measured at the GOLF site were much higher for air masses coming from central Europe than for air masses with SW winds, which contain the urban emissions from Paris (Beekmann et al., 2012).

3. During the MEGAPOLI summer 2009 campaign, a marine aerosol factor was identified in PMF analysis of high-resolution AMS data. This marine aerosol contributed on average ~16% of total OA in Paris, even in the urban core (Crippa et al., 2012b). The ability of the Atlantic Ocean to significantly influence air quality in the Paris urban core (200 km from the ocean) suggests that particulate emissions from Paris are unlikely to be the major regional influence.

4. Finally, major increases in PM1 concentrations occurred when the Paris area was influenced by transported air masses (e.g. during the event on the 26-28th of January 2010, the high pollution event at the end of the winter campaign and the continental period observed in the beginning of the summer 2009 campaign (Freutel et al., 2012)). The strong effects of these events, coupled with the predominance of secondary/aged organic components during these periods, further demonstrate the dominant effect of regional air quality effects relative to primary and secondary sources in Paris.

We acknowledge the Reviewer for pointing out the importance of the definition of the local vs. regional sources in Paris. This topic is developed into several papers, therefore now we included a brief section in the manuscript to clarify the impact of Paris emissions and the regional sources.

“The observed homogeneity in terms of chemical composition, mass concentration and sources among the three sites could theoretically indicate a dominant impact of Paris on its surroundings, however the reduced impact of Paris itself on the surrounding air quality indicates the opposite conclusion. The urban core of Paris covers ~20 km in diameter, while the greater Parisian metropolitan area is ~ 40 km in diameter; the SIRTA and GOLF sites are located near the edges of this metropolitan area. Beekmann et al. (2012) showed that the yearly impact of Paris emissions is on average equal to only 30% of $PM_{2.5}$, while the remaining 70% is advected to Paris and impacted by long-range transport of continental pollution. Moreover PM_1 levels measured at the GOLF site were much higher for air masses coming from central Europe than for air masses with SW winds, which contain the urban emissions from Paris during the summer and winter MEGAPOLI campaigns (Beekmann et al., 2012;Freutel et al., 2012). Crippa et al. (2012b) showed also the ability of Atlantic Ocean air masses to significantly influence air quality in the Paris urban core (200 km from the ocean), suggesting that particulate emissions from Paris are unlikely to be the major regional influence. Our results agree with the conclusions of Sciare et al. (2010) who reported long-range transport to be the major source of $PM_{2.5}$ secondary aerosol in Paris during springtime.”

- The OOA2-BBOA factor is ambiguous and related discussions vague. Was PMF performed on the high-resolution mass spectra? The aerosol loading was high during this study, so the high resolution spectra should have good signal-to-noise ratios. Analyzing the high resolution spectra will likely give less ambiguous PMF solutions and better separated factors.

PMF was here performed using the unit mass resolution MS since the HR information was not available for all the three sites (e.g. the GOLF site deployed a C-TOF-AMS). From our analysis we were not able to determine whether the OOA_2-BBOA at LHVP was a real secondary OA factor deriving from primary BBOA emissions, an atmospheric mixture of primary BBOA and secondary OOA, or a mathematical mixture due to the inability of PMF to clearly separate sources with similar temporal variation (i.e. peak concentrations at night). These uncertainties prevent us from precisely defining this factor. However, PMF analysis of combined UMR AMS data and simultaneous PTRMS measurements at the LHVP site (Crippa et al., in preparation) provided clearly separated BBOA and nighttime SV-OOA factors. This suggests that the OOA_2-BBOA factor presented in this study is mostly a mathematical mixture of factors that could not be completely separated by PMF. This is a limitation of AMS PMF and will be discussed in detail in the forthcoming publication.

- Page 22539, line 23, a comprehensive review of factor analysis approaches of aerosol mass spectrometry of ambient aerosol was published [Zhang et al., 2011]. It seems an appropriate reference to cite for this sentence.

This reference is now added to the manuscript.

- Page 22558, 2nd paragraph, for COA in Beijing, Sun et al. [2010] and Huang et al. [2010] should be cited since they actually reported the observations of cooking aerosols based on analyzing ambient AMS measurement data while He et al. [2010] primarily discuss the spectral profiles of various cooking OA. In addition, significant quantities of COA were also determined in New York City [Sun et al., 2011] and Fresno [Ge et al., 2012]. Fig. 1, it would be helpful to show the comparisons of the average loadings of total PM1 among three sites too.

Sun et al. 2010 and Huang et al. 2010 references are now cited instead of He et al. 2010 for the Beijing case and Sun et al. 2011 and Ge et al. 2012 for New York and Fresno, respectively are additionally cited.
The average loadings of total PM1 are already reported in Fig.12 for the three sites, so we decided not to show it here again.

- Fig. 2 and 8, the medians are used for making the diurnal profiles at here. Often times, the means are also shown. It will be interesting to know how the diurnal patterns differ if the mean values are plotted. The difference between the median and the mean is usually larger for a set of data that is more deviated from normal distribution. Primary species are more influenced by spikes, thus might show bigger differences between the mean and the median.

The mean values are now added in Fig.2 and Fig.8.

Answers to Reviewer 2:

This manuscript summarizes wintertime observations of aerosol composition in downtown Paris and two suburban areas. Factor analysis has been performed on the aerosol mass spectrometer data to assess contribution of different sources to the submicron aerosol mass at these sites. The results are sound and consistent with the summertime measurements, indicating a significant regional contribution to the aerosol mass in the area. However, there are few points that need clarification or a better explanation. I suggest publishing this work after authors respond to the following comments:

Technical comments:

1. Regarding the CE in AMS: for the data obtained at the LHVP site a CE=0.4 applied at all times. This is lower than what is typically applied for ambient data and it contradicts the data shown in figure SI-2e. Based on the SI data, there's definitely a NO3-effect apparent in the dataset, so applying a constant, low CE factor appears to not be justified. If the DMPS data are valid, authors should revisit the CE determination. Note also that the explanation that's given in terms of the higher density particles affecting AMS CE doesn't apply here- with higher densities, particles appear larger in the vacuum aerodynamic sense and it's more likely that they fall out of the 'good' transmission region of the AMS lens; in such case AMS mass decreases compared to other instruments whereas the data in SI-2e at high NO3 fraction times indicate that the apparent AMS mass is actually higher compared to the other times. In general, I believe now that parameterization of composition dependent CE is available, it makes sense to apply such parameterizations and then evaluate the closure between estimated AMS mass and other measurements; if this is not done in this order, the estimated CE appears to be more of a 'fudge' factor to make sure 'consistent' results (but may be low or high depending on errors in the other measurements) are obtained.

While a CE of 0.5 is frequently assumed for ambient measurements, the CE of 0.4 proposed here for the LHVP falls within the range of CEs typically found when this estimation is attempted with ambient data. For example, Takegawa et al. (2005) reported AMS/PILS ratios of 0.81 to 1.26 for inorganic species after assuming CE = 0.5. We further note that the CE = 0.4 value does not come from the comparison of the AMS with a single instrument (in which case errors in the other measurement might be expected to play a large role), but is rather selected to for consistency with a suite of instruments (SMPS, PILS, ATOFMS, FDMS-TEOM). A detailed analysis of these intercomparisons

will be presented in a forthcoming paper (Poulain et al., in preparation), and the results are briefly summarized as follows:
- AMS+MAAP vs. SMPS: slope of 1.06 R^2 0.77
- PILS SO4 (slope 0.68 $R^2 = 0.84$)
- PILS NH4 (slope 0.53 $R^2 = 0.65$)
- PILS NO3 (slope 0.81 R^2 0.79)

Moreover, LHVP used an aerosol dryer in the inlet line, that might result in a reduced CE.
Publication of the CE analysis for the LHVP dataset (Healy et al., 2012) predates the publication of the CE parameterization by Middlebrook et al. (2012) paper. Therefore, while an assessment of the NO3 dependence of the CE was performed, we retain the constant CE = 0.4 in this paper to maintain consistency with the previous publication. The Middlebrook calculation for this dataset provided differences between ~10-15% for the different AMS species quantification compared to the application of a CE=0.4.

After applying a CE = 0.4, remaining discrepancies between AMS and PILS inorganics are believed to mainly result from the instrument size cuts (~PM_1 of the AMS and $PM_{2.5}$ of the PILS). We observed an increase in the coarse mode concentration when AMS underestimated the PILS concentrations (Poulain et al., in preparation).

Concerning the density explanation shown in Fig. SI-2c, we want to underline that it is referred to the SIRTA case and not to the LHVP one as mentioned by the reviewer.

2. Discussion on angstrom exponent values for traffic and biomass burning BC is circular- in section 2.3.2 it's mentioned that the choice of the parameters is discussed in section 3.3 while in sec. 3.3 it's referred back to 2.3.2! It appears these values come directly from another study. Maybe the best is to explain a bit in Section 2.3.2 how these factors were determined. Related to the BC mass- the concentrations reported here are awfully high! Are there any wintertime direct BC mass measurements (e.g., by SP2) to confirm such high concentrations in Paris or could this be due to the choice of mass absorption efficiencies used to estimate BC mass? Also, in the apportionment of BC only biomass burning and traffic related BC are considered. Isn't cooking also a source of BC? Do we know how small this source is to justify the assumptions here?

The discussion on the choice of the angstrom exponent is now entirely reported in section 2.3.2 as suggested by the Reviewer.

"Literature values of the BC absorption Ångstrom exponent vary between 1.9 and 2.2 for wood burning (Sandradewi et al., 2008) and 1±0.1 for traffic (Bond and Bergstrom, 2006;Bond et al., 2004). In the present study absorption Ångstrom exponent values for traffic and wood burning were chosen as $\alpha_{tr}=1$ and $\alpha_{wb}=2$ coherently with Sciare et al. (2011). The choice of such α values is in agreement with previous sensitivity analyses performed in the Paris region in order to evaluate the influence of different absorption exponent values on the aethalometer model (Sciare et al., 2011)."

Below are some additional comments on the choice of angstrom exponent and the BC mass concentration measured in Paris.

In Sciare et al. (2011), α_{wb} was taken equal to 2, based on values previously reported for wood burning aerosols (Lewis et al., 2008;Clarke et al., 2007) and used by Favez et al. (2010). This value is also close to the highest a value of 1.92 obtained by Sciare et al. (2011) for a suburban site of Paris.

The BC mass concentrations are obtained from a constant mass absorption efficiency (MAE = 5.78m^2 gC^{-1} at 880 nm) that has been calculated from on-line EC measurements performed with a Sunset Field instrument. Quality control of EC measurements were performed using off-line EC measurements performed in parallel during the campaign. Comparison of EC measurements between the LHVP site and another background urban site located at 3-4km are very consistent (Bressi et al., ACP, 2012 submitted), suggesting that the site is not particularly impacted by high local traffic emissions. For these reasons, there is no reason to believe that BC measurements are awfully high (considering that the French car fleet is mainly composed of diesel cars). We applied a similar MAE for biomass burning and traffic because of the poor influence of biomass burning on the MAE as observed in Sciare et al. (2011), whose MAE is mainly driven by coating.

We do not expect cooking to be a source of BC since BC is produced mainly from combustion processes, which do not typically occur during cooking. Recent smog chamber studies performed in our laboratory showed also the absence of BC from cooking emissions. Allan et al. (Allan et al., 2010) also found very bad correlation between their PMF cooking factor and BC measurements.

3. P. 22551: Comparison of MAAP and aethalometer results in a slope of 1.16- is this considered a good closure? In this case, reasoning of instrument differences for different BC mass estimates is not valid; I suggest removing that sentence.

We do consider that 1.16 agreement between MAAP and aethalometer being quite good; however in agreement with the Reviewer's suggestion we removed that sentence.

4. P. 22555: indicate what the correlation coefficients are when comparing PMF BBOA related factors and filter-based levoglucosan measurements.

The corresponding R^2 values are added to the manuscript.

5. P. 22559: Sciare (2010) analysis indicated OM/OC =0.9- does this OM include only HOA or other PMF factors as well? If it included other factors then it's not surprising that HOA/BC(tr) is lower than 0.9.However some explanation is needed as to why compared to Chirico (2010) work a factor of two higher HOA/BC is obtained in this work.

In the manuscript we referred to the OM/EC and not to the OM/OC ratio being equal to 0.9. However the reported OM/EC value was wrong (the correct value was OC/EC=0.7), so the sentence was reformulated as following:

"The average ratios of HOA to BC_{tr} found from the slopes in Figure 11 are 0.37 (intercept=0.33) and 0.61 (intercept=-0.12) for the two sites, and the BBOA to BC_{wb} ratios are 3.16 (intercept=0.11) and 3.62 (intercept=-0.12) both ratios are thus quite consistent between the two stations. Considering the SIRTA case, a positive intercept is found both for the HOA vs. BC_{tr} and BBOA vs. BC_{bb} comparison (0.33 and 0.106, respectively), representing an underestimation of BC_{tr} and BC_{bb} for this site. For the LHVP case the opposite situation is observed: negative intercepts are found for the HOA and BBOA vs. the corresponding BC fractions (-0.12 for both sources), meaning that the BC associated to these two sources is slightly overestimated. The observed HOA to BC_{tr} ratio is smaller than values reported for tunnel measurements at low OA concentrations (Chirico et al., 2011) because of different measurement conditions (lower temperatures during wintertime), different vehicles fleets and in urban areas stop-and-go traffic might be different than constant speed driving in a tunnel."

6. Why isn't OOA compared to sulfate? Since NH4 can be associated with SO4 and NO3 and so it can have a diurnal temperature-dependent variation, it's better to compare the regionally influenced OOA factor with SO4.

For completeness, in the manuscript the R^2 values for all the secondary inorganic species with the respect of OOA are now reported.

"The R^2 values for NH_4^+, NO_3^- and SO_4^{2-} with the respect of OOA component are 0.64, 0.53 and 0.65 for the LHVP site, 0.36, 0.32 and 0.21 for the GOLF site and 0.67, 0.53 and 0.63 for the SIRTA site, respectively."

7. Figure 5- in COA factor, m/z 43, 55, 57 seemed to have been mis-marked. Please double check.

Figures 3 and 5 are now corrected

8. Figure 9- please clarify 'avg' lines are the 'reference' lines.

The caption of Fig.9 includes now the definition of COAavg and HOAavg as reference lines:

Figure 9. Mass fraction of *m/z* 55 and 57 (f_{55} and f_{57}) for primary organics at the LHVP site. Reference lines for pure COA and HOA (defined as COA_{avg} and HOA_{avg}, respectively) are shown.

9. Figure 11- These fitted lines shouldn't be forced to go through zero. I believe the nonzero intercepts actually are an indication for the extent that the 'factorization' doesn't work. For example, at SIRTA, the positive intercept of HOA may indicate that BC(tr) is underestimated and that is consistent by getting a positive intercept for BC(BB) when looking at BBOA vs. BC(BB).

We accepted the Reviewer's suggestion and we did not force the fit through zero. The new fitting parameters are now shown in Fig. 11 and reported in the manuscript.

The following sentence was added to the manuscript:

"Considering the SIRTA case, a positive intercept is found both for the HOA vs. BC_{tr} and BBOA vs. BC_{bb} comparison (0.33 and 0.106, respectively), representing an underestimation of BC_{tr} and BC_{bb} for this site. For the LHVP case the opposite situation is observed: negative intercepts are found for the HOA and BBOA vs. the corresponding BC fractions (-0.12 for both sources), meaning that the BC associated to these two sources is slightly overestimated."

10. Figure SI-2d: is there an explanation of why comparison of SO4 and NO3 between AMS and PILS is so off during the 'high' period while NH4 looks ok? Is this good agreement for the wrong reason (wrong RIE(NH4)).

During the high concentration periods the agreement between the AMS and the PILS is not so good because of the different size cut of the two instruments (PM1 vs. PM2.5), as shown also in Fig. SI-2f for the LHVP case. In fact, we observed an increase in the coarse mode concentration when AMS overestimated the PILS concentrations. However we agree with the Reviewer that in Fig. SI-2d the good agreement of NH4 between AMS and PILS and the discrepancy observed for NO3 and SO4 might be partially associated with wrong RIE values.

11. Figure SI- 3a-b: again- you shouldn't force the lines to go through zero. There are enough data points close to (0,0) that the line should be fitted freely.

We agree with the Reviewer that there might be still small errors in the zero of the instruments (air corrections), therefore we allowed the intercept of the fit to vary to check the quality of the data. New graphs are now reported in Figs. SI-3a, SI-3b, SI-3c and SI-3d.

12. Figure SI-4: how is OC calculated from AMS measurements? There was no reference to what OM/OC was assumed; this of course will change the comparison shown here!

The OM/OC ratio is this figure was calculated from high resolution AMS analysis performed at the LHVP and SIRTA sites. The HR- analysis provided an average OM/OC ratio equal to 1.58±0.11 for LHVP and to 1.79±0.10 for SIRTA. This was stated in the text accompanying SI-4 and the OM/OC values will be added.

13. Figure SI-5: there was no reference to this plot in the main text. I suggest either adding a ref. to it or removing it.

In the main text the following sentences have been added:

"In addition, Fig. SI-5.1 shows that the identified organic sources at the three sites are grouped in different regions of the triangular space defined by Ng et al. (2010). Some of the differences within each group of sources are probably due to the deployment of different types of instruments (e.g. C-ToF vs HR-ToF-AMS), different ion transmission and fragmentation etc."

14. Figure SI 6.6.2- what is the explanation for not having high f55/f57 at peaks of COA time series at SIRTA?

The SIRTA site is considered an urban background location since not directly exposed to primary emission sources and 20 km far from the core of Paris. The diurnal pattern retrieved at this had a much less distinct diurnal pattern than at other locations. This is consistent with the strongest cooking emissions being located near the city center rather than near SIRTA. While Figure SI-6.6.2 shows some high COA events yielded high f55/f57 ratios, the proportionally stronger contributions of non-COA factors to m/z 55 and 57 was proportionally stronger at SIRTA.

The following sentence was added in the supplementary information:

"The relative fraction of cooking is never reaching as high values at SIRTA compared to LHVP. In rural areas, the f55/f57 approach will be much more uncertain as also OOA contributes to a higher degree to these mass fragments compared to urban areas."

Answers to Reviewer 3:

This paper presents the AMS/PMF analysis from the winter MEGAPOLI campaign in Paris. This analysis has now become commonplace in urban intensive studies for the identification of primary sources in particular. The results are not particularly controversial, as they compare well with other datasets obtained in other studies in other cities. However, there is real novelty in how the authors have used the Aethalometer data and how they have compared the different measurement sites within the campaign. The end result is a largely well written paper that presents some useful, if not earthshattering, statistics regarding primary aerosols for Paris. I would recommend that this be published in ACP subject to the following minor comments:

- There is some crossover between this paper and Freutel et al., which I was asked to review in tandem with this. I refer specifically to the technical details such as the scaling of the organics by a factor of 1.3. But curiously, the authors do not refer to this paper, which is unfortunate because it covers details regarding the intercomparison in more detail. This paper would benefit from referring to this directly.

The Freutel et al. paper is now referenced at page 22544 at line 20 and in the supplementary material section about the AMS intercomparisons.

"A scaling factor of 1.3 was applied to the organic concentrations measured at the stationary GOLF site after comparison with the mobile laboratory deployed at the same location (Freutel et al., 2012)."

"The low ion transmission efficiency of the C-ToF-AMS deployed at the GOLF stationary site has been taken into account with a scaling factor of 1.3 for the organic concentrations after the comparison with contemporary measurements performed with the HR-ToF-AMS deployed at the same location (Freutel et al., 2012)."

- As a matter of taste, I do not see it necessary that equations 2-4 need repeating, as these have already been presented numerous times in the papers cited by this work.

We prefer to keep the PMF equations in our manuscript.

- Page 22543, line 16: 'Lee et al.' should be outside the parentheses.

Modified in the manuscript.

- Page 22544, line 20: I don't particularly like the explanation for the scaling factor of 1.3 applied to the data. Is it not more likely that this could be caused by something to do with the vaporiser (e.g. different temperature) rather than the ion transmission function of the mass spectrometer?

Two different heater temperatures were used during the summer campaign for the C-TOF-AMS (800°C during the first two weeks and 600°C during the last two weeks) and no difference was observed in the identified scaling factor (Freutel et al., 2012). So, the different ion transmission efficiency is probably the cause in the observed differences of the C-ToF-AMS and MoLa-AMS mass spectra.

- Page 22545, line 8: Rather than Weingartner et al. (2003), the authors should consider using the updated correction presented by Collaud Coen (2010, doi:10.5194/amt-3-457-2010).

In the Collaud Cohen et al. paper, the authors recommended that the new correction algorithm should be used only when nephelometer data was available; otherwise, they recommended that the Weingartner et al. (2003) algorithm should be adopted. In the present study, nephelometer data were not available and so the Weingartner et al. (2003) correction was applied.

- Page 22547, line 23: How many variables were downweighted as 'weak'?

Actually the number of weak variables was very low: zero for SIRTA and LHVP and 1 for GOLF (m/z 279).

- Page 22549, line 14: The use of a solution set with a nonzero fpeak for LHVP must be justified further. According to Paatero et al. (2002, doi:10.1016/S0169-7439(01)00200-3), while fpeak can be used to explore the amount of rotational freedom within a potential solution space, solutions associated with nonzero values can only be considered physically meaningful for certain systems. Unless there is a specific reason to prefer the fpeak=-0.1 solution, it would be safer to use the fpeak=0 version.

The following sentence was added to the manuscript

“The selection of the fpeak=-0.1 solution was motivated by the clearer separation obtained for several sources as discussed in section SI-6.3.”

In section SI-6.3 the following clarification is reported:

“For the LHVP site, the fpeak=0 solution provided a BBOA MS with a very small contribution at m/z44 (which should instead contribute to biomass burning sources). Additionally, a clearer distinction between the OOA and OOA_2-BBOA factor was retrieved (major differences both in the time series and mass spectra). For this reason we decided to discuss within this paper the fpeak=-0.1 solution.”

- Page 22551, line 9: Replace ‘pretty good’ with something less informal.

This sentence has been removed accordingly with the suggestion of reviewer 1

- Page 22557, line 9: Another explanation for the org60 could be that it reflects a portion of the BBOA that is prevalent during a particular burn phase, hence the separation from the BBOA factor.

As clarified in the answers to Reviewer 1, from our analysis we were not able to determine if the OOA_2-BBOA was a real secondary OA factor deriving from primary BBOA emissions, or if it was an atmospheric mixture of primary BBOA and secondary OOA or if it was a mathematical mixture of PMF due to the impossibility to clearly separate the OA sources peaking at night based on their temporal variation. Therefore we could not be too precise in the definition of this factor. However, an additional study was performed combining the UMR AMS data with contemporary PTRMS measurements performed at the LHVP site (Crippa et al., in preparation), where a clear separation of a BBOA factor and a nighttime SV-OOA was obtained. This means that our OOA_2-BBOA factor was mainly a mathematical mixture of factors which were not completely separated by PMF. This represents one of the limits of PMF and therefore the publication of this additional work will help to compare results from different source apportionment techniques and their uncertainties.

The reference to Crippa et al. paper (in preparation) is now reported in the manuscript.

"Additional source apportionment analysis performed combining gas (PTRMS) and particle phase (AMS) measurements for the LHVP site allowed a clearer discrimination of a pure BBOA source and a secondary semi-volatile OOA (SV-OOA) (Crippa et al., in preparation)."

- SI-6.3: The authors did not explore a particularly wide range of fpeak values. It would have been informative to have seen at what point the algorithm failed to converge, produced unacceptable profiles or suffered a large increase in Q.

A new fpeak analysis was performed in order to explore a wider fpeak (-10,+10) and Q/Qexp range and results of these analyses are reported in section SI-6.3.

"Figure SI-6.3.1 shows the variation in the factor relative contribution in the fpeak range -10 to +10 for the LHVP site. Negative fpeaks reduce the split between the BBOA and OOA_2-BBOA factors, while the HOA mass spectrum receives higher contributions from mass 44. This is especially the case for fpeak values below -3 (corresponding to a Q/Q_{exp} variation around 10%). Similarly, positive fpeaks > 4 incorporated the OOA_2-BBOA factor into the BBOA and OOA ones and a non-meaningful mass spectrum was obtained for highly positive fpeaks."

"Figure SI-6.3.2 shows the variation in the factor relative contribution in the fpeak range -10 to +10 for the SIRTA site. Negative fpeaks affect the separation of a cooking factor, in fact below fpeak=-4 the COA contribution disappears. Strongly positive fpeaks affect the separation of OOA and BBOA, providing a split of the BBOA factor and not a separation of two OOA components."

"Figure SI-6.3.3 shows the variation in the factor relative contribution in the fpeak range -10, +10 for the GOLF site. The separation of the OOA_2-BBOA and OOA factors is fpeak dependent and affected by both negative and positive fpeaks. Negative fpeaks produced often unreasonable time series, while positive fpeaks influenced mainly the mass spectra."

- Figure SI-6.4.1: The scaling on the Q/Qexp graph is not much use, as it doesn't show how much variation there is in the red line.

A new scale is now used for the left axis of Fig. SI-6.4.1 in order to highlight the variation of Q/Qexp with seeds (which is very small).

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