

The authors would like to thank both reviewers for their comments and suggestions, which have been very helpful in improving the quality of this manuscript. In order to guide the review process we have copied below each of the reviewer comments in *black italics*. Our responses follow each comment in bold black font and any changes to the text of the manuscript are in bold blue.

Review of “Characterizing the impact of urban emissions on regional aerosol particles; airborne measurements during the MEGAPOLI experiment” by Freney et al.

This paper describes the evolution of organic matter in the outflow from the city of Paris; it involves airborne measurements and contributes to the general knowledge in the secondary aerosol formation field. Correctly capturing changes in organic mass concentration and its evolution remains a challenge for regional models, which highlights the importance of this study. The paper is generally well written, but some questions concerning the PMF solutions and methodology should be answered before this manuscript is suitable for publication.

General comments:

R2.1: A comparison with ground based measurements (Freutel et al., 2013) would add more value to the study, differences and similarities should be discussed.

EF: More detailed comparison with Freutel et al., 2013 and other studies is now included in the text.

Updated/additional text:

Page4, Line 90: Freutel et al. (2013) and Crippa et al., (2013a) described the chemical composition of aerosol particles measured at the ground based sites during the summer and winter campaigns, respectively.

Page 9, Line 259: In Freutel et al., (2013), air masses were classified into three categories: Central Europe, Atlantic Polluted, and Atlantic Clean. In this work, research Flights only took place during Atlantic polluted or Atlantic Clean periods. “Atlantic polluted” were generally classified as air masses that spent more time over land and correspond to flights: N16, N21, and N29. Average temperature measured on the ground during these meteorological events were $22^{\circ}\text{C} \pm 4^{\circ}\text{C}$ for Atlantic polluted and $18^{\circ}\text{C} \pm 3^{\circ}\text{C}$ for Atlantic Clean. Similar differences in ambient temperatures were measured aboard the aircraft with $21^{\circ}\text{C} \pm 0.89^{\circ}\text{C}$ for Northern sector flights (Atlantic polluted) and $17^{\circ}\text{C} \pm 0.5^{\circ}\text{C}$ for eastern sector flights (Atlantic clean).

Page 10, Line 281: These observations are similar to those made at the ground based sites (Freutel et al., 2013), where the highest mass concentrations were measured when air masses arrived from the continent and when wind speeds were lower. Freutel et al., (2013) measured lower mass concentrations when air masses arrived from the Atlantic with higher wind speeds. The higher wind speeds lead to greater dilution of primary species such as BC, HOA, and NOx.

Page 13, Line 367: The combination of inorganic and organic aerosol measurements during different meteorological periods shows that aerosol mass concentrations measured in Paris are

strongly influenced by regional air mass history, as was already observed from ground based measurements (Freutel et al., 2013).

Page 14, Line 427: Compared with Mexico City (DeCarlo et al., 2010, Kelinman et al., 2008) these values of CO and Δ CO are low, but recent measurement made in London (McMeeking et al, 2012) show that low CO values appear to be representative of European air masses.

Page 16, Line 469: Corresponding values of Δ NO₃/ Δ CO give 23, 19, 10, and 17 $\mu\text{g m}^3/\text{ppm CO}$ for N16, N20, N21, and N29 using a maximum value of $-\log(\text{NO}_x/\text{NO}_y)$ of 0.6, 0.3, 0.7, and 0.3, respectively. McMeeking et al., (2012) reported values varying between 20 and 30 $\mu\text{g m}^3/\text{ppm CO}$ in London. However, DeCarlo et al., 2008 reported decreases in the same ratio as a result of dilution. These measurements show that the formation of secondary OA are almost three times more important than the formation of other aerosol species through secondary processes.

Values of Δ OA/ Δ CO calculated from Paris emissions are similar to those measured in a number of different research environments all over the world (Table 3) even though there are large differences in the absolute CO values observed. deGouw and Jimenez (2009), report enhancement ratios of OA relative to CO for fresh and aged urban emissions and stated that although there are large differences in the absolute CO concentrations and emission properties the Δ OA/ Δ CO did not show significant variations ($70 \pm 20 \mu\text{g m}^3/\text{ppm}$). These conclusions were also recently confirmed from London measurements (McMeeking et al., 2012).

Page 19, Line 582: Similar to observations made by Freutel et al., (2013) we observe that largest differences in aerosol concentration and BC levels are related mainly to air-mass origin.

Page 19, Line585: Similar findings were reported during the REPARTEE (Harrison et al., 2012) and the EM25 (McMeeking et al, 2012) experiments in London. However, the increase of Δ OA/ Δ CO with photochemical age measured aboard the ATR-42, as well as results during MILAGRO demonstrates that it is necessary to take into account a larger geographical area when assessing the formation of SOA from urban emissions.

R2.2: PMF solutions and especially HOA seems different from this study and much more comparable with previous HOA's found in the literature. I guess it is expected result as airborne measurements are by default further from the source, but then it should be clearly discussed in the text.

EF: In Figure S7 we compare the retrieved PMF MS with those published on the AMS website (<http://cires.colorado.edu/jimenez-group/AMSsd/>). The authors have changed the correlations provided in the original table with correct ones and apologise for the incorrect values. The correct correlations result in a slightly higher R² correlation between HOA_{RESOLVED} vs HOA_{LITERATURE} of 0.88 (instead of 0.86), compared with 0.82 and 0.8 with SV-OOA and LV-OOA respectively. In the authors opinion, we feel that the HOA measured in this study is comparable with HOA measured in other ambient studies.

R2.3: Actually, I have doubts that authors separated a clear HOA factor in this study (see specific comments), maybe it is not justified to push for HOA if it isn't exist. Otherwise, more convincing

evidence should be provided for HOA separation. PMF, in general, raises many questions: time trends of different factors doesn't show significant difference and seems correlating quite well between each other, is there really a reason to separate 3 factors? Correlations with reference spectrums or external tracers are not so unambiguous as well...

EF: Given that there are distinct increases in primary BC aerosol particles within the pollution plume, the authors believe that it is reasonable to assume that there should be a primary HOA factor. BC particles contribute approximately $7\pm 5\%$ to the overall aerosol mass concentration (C-ToF-AMS+BC) measured during MEGAPOLI, and the resolved HOA factor contributes approximately $11\pm 6\%$ to the total sub micron aerosol. At the ground sites a cooking OA was resolved and contributed between 10 and 30% to the total measured OA. At altitude we were unable to separate these factors but they could possibly be included within the HOA.

Although, the method was not used in this study, the new multi-linear Engine (ME-2) method is used to constrain the statistical separation of organic mass spectra in order to resolve species that would not otherwise be resolved with PMF alone (as a result of low mass concentrations or low contribution from certain organic types (Crippa et al., 2013). In this study we use unconstrained PMF-2, and a HOA species was separated providing smaller residuals and a smaller Q/Q_{EXP} as well as improving correlations with external time series and mass spectra. The authors feel that a three factor solution best represents the measured organic aerosol.

Organic aerosol components derived from 25 AMS datasets across Europe using a newly developed ME-2 based source apportionment strategy. M. Crippa, F. Canonaco, V. A. Lanz, et al., Atmos. Chem. Phys. Discuss., 13, 23325-23371, 2013

Additional figures, including correlations with reference mass spectra as a function of fpeak are included.

Updated text:

Page 13, Line 398: Above the city of Paris we might not expect to resolve a primary organic species, however the measured contribution of the resolved HOA species to the total aerosol mass concentration (C-ToF-AMS + BC) is $11 \pm 7\%$, which is similar to the contributions of primary BC particles measured in the Paris plume ($7\pm 5\%$). At the ground sites a cooking OA was resolved and contributed between 10 and 30% to the total measured OA. At altitude we were unable to separate these factors but they could possibly be included within the HOA.

R2.4: In addition, one would expect HOA to increase within the plume, but it is not obvious in the current Fig. 2 or S2. Moreover, $\Delta HOA/\Delta BC$ would likely to decrease with photochemical aging (Fig.6) due to evaporation of volatile compounds or at least remain constant, but not increase as stated in text. This could also indicate problems in resolving HOA factor.

EF: Figure 2 and S2 have been changed so that the BC increases are in the background and those of PMF are in the front. The authors would like to highlight that data are subject to a low signal to

noise ratio as we used a very short averaging time to obtain the best temporal resolution during airborne measurements.

The text has been updated below to discuss the change in the $\Delta\text{OA}/\Delta\text{CO}$ ratios as a function of photochemical age.

Updated text:

Page 15, Line 484: The averaged ratio of $\Delta\text{LV-OOA}/\Delta\text{BC}$ and the $\Delta\text{SV-OOA}/\Delta\text{BC}$ for all flights showed increases of 2.12 and 1.98 respectively when the $(-\log(\text{NO}_x/\text{NO}_y))$ increased from 0.1 up to 0.6. The $\Delta\text{HOA}/\Delta\text{BC}$ varied slightly with an average ratio of 1.28, which is thought to be within the error of the measurements (Figure 6). Several studies have shown that for urban HOA, evaporation is thought to occur on a timescale of minutes (Zhang and Wexler, 2004) and therefore urban HOA/(BC/CO) should not be affected by additional evaporation. In Paris, the ratio appears to be stable, similar to observations made in Mexico City (DeCarlo et al, 2008) where the HOA was conserved over the timescale of the study (~6 hrs). However, as suggested by recent observations in the Mediterranean, the HOA might decrease after much more oxidation (Hildebrandt et al., 2010).

R.2.5: Concept of scaling ΔOA to ΔBC needs more rezoning, if one assumes that ΔOA is constant and ΔBC is decreasing with going further from the source, which is very reasonable as BC would eventually go to the background level, then the ratio of ΔOA to ΔBC would increase as you have in the paper, but it won't be resulted from an increase in OA mass, it would be rather due to decrease in BC. Plausibility for OA to remain constant vs. increase with decreasing BC should be elaborated and explained in more details.

EF: BC is used as a marker for the pollution plume, in a similar way as that of CO, and is used as a conservative tracer to remove the effect of dilution of polluted airmasses with cleaner background air far from sources. The only removal mechanisms for BC are through rainfall and with air mass dilution. The removal mechanisms for CO are with air mass dilution. Since we did not have rain events during these measurements we consider that the only changes in BC concentration are a result of aerosol dilution. Increases observed in both $\Delta\text{OA}/\Delta\text{BC}$ and $\Delta\text{OA}/\Delta\text{CO}$ are assumed to be a result of the formation of OA through secondary processes.

Updated text:

Page 15, Line 444: As noted by Park et al. (2005), BC mixed with soluble aerosol particles could be susceptible to wet deposition. In good weather conditions, wet deposition is not likely to be important and the BC concentration should be conserved. For this study, we believe that any decrease in BC concentration with photochemical age of the pollution plume is a result of air mass dilution.

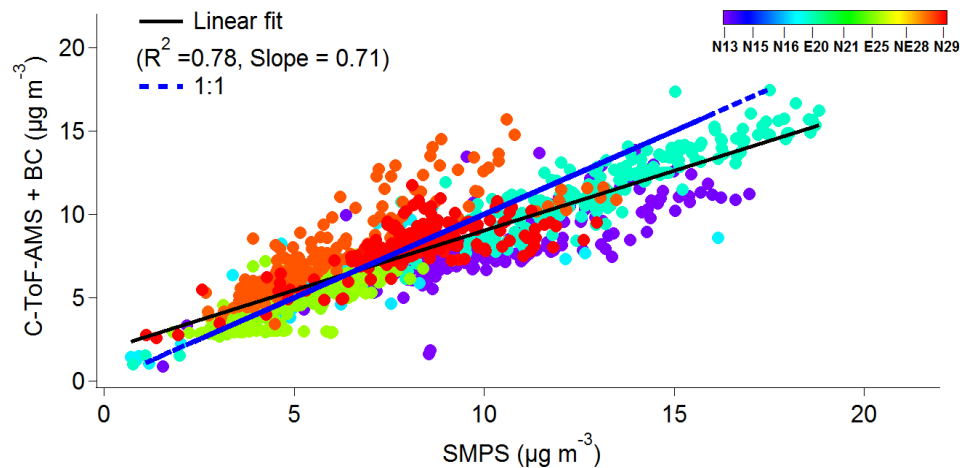
Specific comments:

R2.6: Page 24891 lines 17-20: mass, not volume, was compared in Figure S1. Why constant densities were selected for SO₄, NH₄, NO₃? Ammonium nitrate density differs from that of sulphate or sulphuric acid. Correlation or slope or line 1:1 or all of them should be presented in Figure S1.

EF: We would like to thank the reviewer for pointing out this error. We compared the total mass concentration measured by the AMS with the total mass concentration measured from the SMPS. To convert the SMPS number concentration to $\mu\text{g m}^{-3}$ we used a density of 1.8 g cm^{-3} . In the presented figures we only reported r^2 and slope values for 2 flights. The data and figures are now updated to provide average r^2 and slope values for all flights.

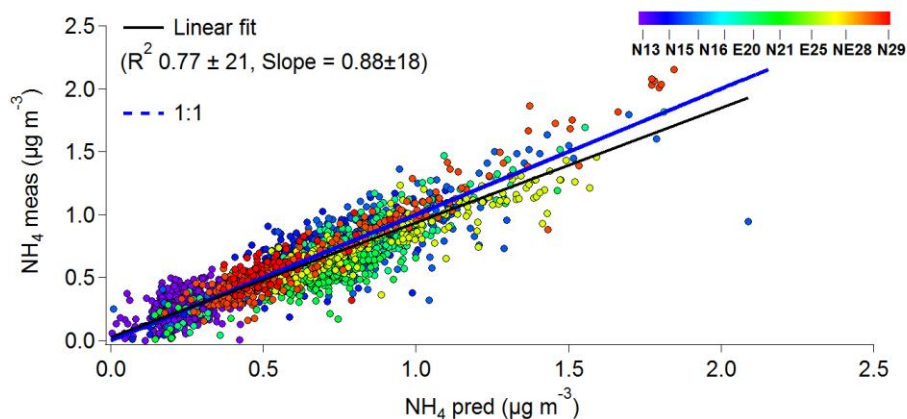
Updated text:

Page 6, Line 155: In order to validate our chosen CE, we compared the total mass concentrations of aerosol particles sampled by the C-ToF-AMS and BC with that sampled by a scanning mobility particle sizer (SMPS). The SMPS number concentrations were converted to mass concentrations using a density of all aerosol particles of 1.8 g cm^{-3} . Comparing the total mass concentration measured by the C-ToF-AMS and BC for all research flights with the corresponding SMPS measurements we obtain a correlation with an average r^2 and slope of 0.78 ± 0.13 and 0.71 ± 0.15 respectively (Figure S1).



R2.6: Why only two flights were selected (although, text states that all flights were compared), single graph on all flights with all statistical parameters would be much more informative. How neutralization of sulphate affected CE, did authors exclude that for a reason, if yes, it should be explained in the manuscript as original algorithm by Middlebrook et al. (2011) includes that.

EF: Figure S1 is updated to include the comparison between the mass concentrations SMPS and AMS for all flights. In addition, a figure (Figure S2) is included in the supplementary material to illustrate that the aerosol particles measured during MEGAPOLI were always neutralized. According to Middlebrook et al., (2011) accounting for aerosol acidity is only performed if $\text{NH}_4\text{meas}/\text{NH}_4\text{pred}$ is < 0.73 . In our study, this average ratio ($\text{NH}_4\text{meas}/\text{NH}_4\text{pred}$) for each flight ranged from 0.77 to 0.82. Therefore we did not account for aerosol acidity.



Updated text:

Page 6, Line 144: A CE for aerosol acidity is not included in this study since the $\text{NH}_{4\text{MEAS}}/\text{NH}_{4\text{PRED}}$ was always between 0.77 and 0.82. In Middlebrook et al., (2011), a CE for aerosol acidity was included only when the ratio of $\text{NH}_{4\text{MEAS}}/\text{NH}_{4\text{PRED}}$ was <0.73.

R2.8: Page 24892 lines 22-23: Separate Fig. S3 into a b c d and refer accordingly in the text, otherwise, it takes some time to understand to which graph authors are referring.

EF: This figure and the corresponding text has been updated.

Updated text: Page7, Line184: The background value of BC was determined from the lowest values of Org/BC ratio ($\sim 0.5 \mu\text{g m}^{-3}$ in Fig S3c). This value was then confirmed based on up-wind BC mass concentrations (Fig S3a).

R2.9: Page 24894 line 4: update HYSPLIT citation to the newest one.

EF:A more recent reference has been added to the reference list.

Draxler, R.R. and Rolph, G.D." HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model access via NOAA ARL READY Website (<http://ready.arl.noaa.gov/HYSPLIT.php>)."
NOAA Air Resources Laboratory, Silver Spring, MD (2013).

R2.10: Figure 2: BC time trends overlap and hide Organic's and PMF factor's trends; authors should work on that figure a bit more. Periods of "in" and "out" of the plume referred in table 2, figure 3 and elsewhere, have to be identified in figure 2. As it is presented now, the difference between organics in the plume and organics out of the plume seems to be within the variations, could you present averages with standard deviations or maximums, minimums in table 2.

EF: The figures are now updated so that the BC time trends are behind those of the organics and PMF factors.

R2.11: Page 24896 line 25: oceanic source rather than oceanographic (oceanography is a marine science).

EF:This is corrected.

R2.12: Page 24897 lines 16-21: SO₂ or NO₂ reactions with OH will result in sulphuric or nitric acid, which further be neutralized by existing ammonia and form ammonium sulphate or nitrate. In fact, sulphuric acid is a stronger acid; therefore, it would be neutralized first and preferably form ammonium sulphate particles. Degree of aerosol neutralization by ammonium would give more information. If aerosol was acidic or fully neutralized, one can estimate from AMS data. Maybe, another explanation could be included as well: sulphate formation through secondary processes from anthropogenic SO₂, which would result in higher background SO₄ concentrations if compared to the plume ones. The latter would add up to marine sulphate and form elevated background sulphate concentrations. Reduction of sulphate in the plume due to competition is not so convincing if considered remote and not local (marine) source of SO₂.

EF: Figure S2 has been included in the supplementary material to illustrate that the aerosol measured during MEGAPOLI was always neutralized.

In laboratory studies, Berndt et al., (2008) showed that at high NO_x, the formation of new particles from the reaction of OH radicals with SO₂ is inhibited.

Berndt, T, F. Stratmann, S. Bräsel, J. Heintzenberg, A. Laaksonen, M. Kulmala. SO₂ oxidation products other than H₂SO₄ as a trigger of new particle formation – Part 1: Laboratory investigations Atmos. Chem. Phys. 8, 6365-6374, doi:10.5194/acp-8-6365-2008, 2008

Additional text:

Page 11, Page 328: Other possible explanations could be sulphate formation through secondary processes from anthropogenic SO₂ arriving from Le Havre (the nearby shipping port), which would result in a higher background of SO₄ aerosol particles.

Page 11, Line 350: However, given the high concentrations of NO₂, NO, and VOC species within the plume it is likely that NO, NO₂, and VOC species play an important role in the gas-phase chemistry, reacting preferentially with OH to form HNO₃ and organic aerosol particles, at the expense of H₂SO₄. This hypothesis could be supported by observations from laboratory studies (Berndt et al., 2008), showing that at high NO_x concentrations, the formation of new particles from the reaction of OH radicals with SO₂ is inhibited. However, once formed H₂SO₄ is a stronger acid than HNO₃, and will be neutralised preferentially to (NH₄)₂SO₄.

Berndt, T, F. Stratmann, S. Bräsel, J. Heintzenberg, A. Laaksonen, M. Kulmala. SO₂ oxidation products other than H₂SO₄ as a trigger of new particle formation – Part 1: Laboratory investigations Atmos. Chem. Phys. 8, 6365-6374, doi:10.5194/acp-8-6365-2008, 2008

R2.13: Page 24898 line 16: figure S5 not S6?

EF: This is updated

R2.14: Page 24899 lines 17-19: Sentence is not clear.

EF: This sentence has been removed.

Updated text:

Page 14, Line 401: Increasing the number of factors to four neither improved the weighted residual analysis nor resulted in any substantial gain in the correlations of the factor time series and factor profiles with to external species. (Table S2,S3,S4).

R2.15:Page 24899 lines 20-22: Only for the 3 factor solution not 4. Reasoning of why 0.2 was selected instead of 0 should be provided, not only stated.

EF: The text has been updated and more information included.

Original text: Reasonable solutions lay in the fpeak range from -0.2 to 0.2, the solution with fpeak=0.2 was chosen to best represent the data.

Updated text:

Page 14, Line 408 : Correlations of the three factor solution as a function of three fpeak (0.2, 0, and -0.2) values are listed in Table S6. Q/Qexp values as a function of fpeak values show lowest values for an f-peak of 0 but only vary between 0.005 for an f-peak of -0.2 and 0.2 (Figure S10). More significant differences between mass spectral correlations were observed for an f-peak solution of 0.2 compared with those with an f-peak of 0 and -0.2.

R2.16: Tables S2 and S3: references for the reference mass spectra should be provided in the captions.

EF:Text is updated.

Additional text: Reference mass spectra were taken from the AMS mass spectral data set (<http://cires.colorado.edu/jimenez-group/AMSSd/>).

R2.17: Page 24899 line 25- page 24900 line 6: HOA mass spectrum is quite different from the reference HOA or other HOA's presented in the literature. Correlation of 0.86 is not so good and not so different from the 0.82 or 0.8 correlations with ref BBAO and SV_OOA. It should be addressed and explained in the text. I'm not convinced that the factor, which authors identify as HOA is really HOA, more evidence should be provided. Do correlations provided in tables S2-4 correspond to one flight (which?) or all flights? As for some flights HOA could be more real (N29) than for others (N21). Did you do separate PMF on all flights or combined?

EF: As stated in response to comment 2.2, the original correlations were updated with correct values, however the mass spectra have not changed. The correct R^2 correspond show a slightly higher r^2 correlation of 0.88 (instead of 0.86) for HOA_{MEAS} vs $HOA_{LITERATURE}$. The authors apologise for this error. Figure S6, S7, and S8 illustrate the resolved PMF factors with reference mass spectra. As stated in the methods section PMF analysis was preformed for all research flights together in order to determine the variation between flights.

Page 14, Line 397: PMF analysis was performed on the ensemble organic mass spectra of all flights in order to extract different groups of organic aerosol.

The correlations provided in table S2 to S4 are therefore the correlations calculated from all flights.

Lower time series correlations are linked with variations in aerosol particle concentrations during all flights. Additional text has been added to the table captions to make this more clear. **Supplementary material: Pearsons R (P_R) correlation values with particle and gas phase species for a two factor PMF solution with an f_{peak} of 0.2 for all flights. Reference mass spectra were taken from the AMS mass spectral data set (<http://cires.colorado.edu/jimenez-group/AMSsd/>).**

R2.18: Figure S2. Why SO₄ didn't drop to 0 during filter periods? (Assuming that concentration drop ~14:30 is the filter period).

EF: This is not a filter period. During this time the aircraft made a vertical profile and therefore sampled in the free troposphere where aerosol concentrations are very low. An additional legend is included in each of the figures to make this more clear.

R2.19: Page 24901 lines 24-25: Provide R² and slope for $\Delta NO_3/\Delta BC$ as well, now it is not obvious if it is really insignificant or scaling problem in the graph S2.

EF: The increase $\Delta NO_3/\Delta BC$ with photochemical age is now included in the text along with comparison with other studies. By comparing with NO₃ we wanted to illustrate the difference with $\Delta OA/\Delta BC$ which is clearly being formed through secondary processes as a function of photochemical age.

Updated text: Page 18, Line 521: Corresponding values of $\Delta NO_3/\Delta CO$ give 23, 19, 10, and 17 $\mu g m^{-3}/ppm$ CO for N16, N20, N21, and N29 using a maximum value of $-\log(NO_x/NO_y)$ of 0.6, 0.3, 0.7, and 0.3, respectively. McMeeking et al., 2012 reported values varying between 20 and 30 $\mu g m^{-3}/ppm$ CO in London. These measurements show that the formation of secondary OA are almost three times more important than the formation of other aerosol species through secondary processes.

R2.20: Page 24902 lines 14-16: Discussion why CO doesn't increase over the background is needed.

EF: The CO mixing ratios were generally low when compared with those measured in Mexico City or in New England. However, they were similar to those measured in London during the EM25 experiment.

We measure background values of CO of between 90 ppb and 110 ppb and increases above background of 10 ppb up to about 60 ppb, with average variations of about 25 ppb. From figures presented in McMeeking et al., 2012, background concentrations of CO varied from 100 to 120 ppb and increases above background ranging from 20 up to 80 ppb with average variations of around 30 ppb. Therefore our CO measurements are in agreement with those of London but are much lower than those in New England or in Mexico city.

Updated text. Page 16, Line 457: During our measurement period the difference between the background and local CO measurements were small and the absolute CO concentrations were low (<150 ppb). In Paris, background values of CO ranging between 90 ppb and 110 ppb are measured, and increases above background range from 10 ppb up ~ 60 ppb, with average variations of about

25 ppb. Figures presented in McMeeking et al., (2012), suggest background concentrations of CO varied from 100 to 120 ppb, and increases above background ranging from 20 up to 80 ppb with average variations of around 30 ppb. Compared with Mexico City (DeCarlo et al., 2010, Kleinman et al., 2008) these values of CO and Δ CO are low. However, compared with London, these measurements appear to be representative of European air masses.

R2.21: Page 24904 line 14: correct figure numbers, I assume it is only S2d, which is relevant.

EF: This error has been corrected

Original text: coloring Figs. 1d and 2d (Figs. S1d and S2d) by the ratio of C8-aromatics / CO.

Updated text: coloring Fig S2d by the ratio of C8-aromatics / CO.

R2.22: Page 24905 lines 6-7: " Higher values in N21 may be related to the sparse data points available for N21 (Fig. S2)." Elaborate more as significant difference in data point availability from N29 (figure 2) is not really obvious.

EF: Original text: Higher values in N21 may be related to the sparse data points available for N21 (Fig. S2).

Update text: Higher values in N21 may be related to the sparse data points (70% availability) for N21 compared to N29 (100% availability).

R2.23: Figure 1. Color scale is not clear, should be elaborated more in the caption.

EF:Additional text is included:

Figure 1. Air mass backward trajectories calculated using HYSPLIT for 72 hrs. HYSPLIT trajectories are calculated at a height of 700 m in the hysplit model. Color scale is used to indicate the log of the number of backtrajectories that pas over a given area (pixel) of $0.5^{\circ} \times 0.5^{\circ}$.

R2.24: Figure 4. SO₄ (b,d) not (b,c)

EF:This has been corrected:

Figure 4: Flight tracks around the Paris region for N13 ((a) and (b)) and E25 ((c) and (d)) colored by SO₄ ((b) and (e)) and OA ((a) and (d)). c) and f) show OA vs longitude and Latitude vs SO₄, respectively.

R2.25: Figure 5. Consistency with the text should be maintained in the legend: flights should be named according to classification (N16, N21 and so on) instead of the real flight number.

EF: Flight nomenclature has been verified throughout the manuscript.

R2.26: Figure S2: Δ OA/ Δ BC is nondimensional in d.

EF: This has been corrected.

R2.27: Figure S6: correct a or d caption, now it is both SV-OOA1

EF: The caption is updated.

Figure. S6 Four factor PMF solution a) SV-OOA b) HOA c) LV-OOA, and d) SV-OOA1. Black mass spectra correspond to reference mass spectra for a) SV-OOA, b) HOA, c) LV-OOA, and d) SV-OOA. Reference mass spectra were taken from the AMS mass spectral data set (<http://cires.colorado.edu/jimenez-group/AMSSd/>).

In addition to the requested changes, the authors would also like to highlight some other changes made to the manuscript.

Equation 6 was missing a term “ γ ” which is now included and the text following the equation is updated.

$$\left(\frac{\Delta O A}{\Delta C O}\right)_{predicted} = \left(\frac{\sum \gamma_i * \Delta V O C_i}{\Delta C O}\right) \quad (\text{Eq.6})$$

Updated text: VOC_i corresponds to the each VOC species (i) used. To calculate the aerosol formation from benzene, toluene, C8-aromatics, and C9-aromatics, we used the yields (γ_i) for low concentrations of both NO_x and Δ hydrocarbons (ΔHC) determined by Ng et al. (2007) (Table S4).

Page 21, Line 641:

Original text: Simultaneous AMS, NO_x/NO_y and VOC measurements were available during two research flights. Using only four anthropogenic marker species and organic aerosol formation yields reported for low NO_x conditions we were able to predict ~ 50% of the organic aerosol measured in the plume. This good agreement between predicted and measured values is a result of the improved knowledge of aerosol formation properties from laboratory studies on gas-to – particle reaction processes.

Updated text: Simultaneous AMS, NO_x/NO_y and VOC measurements were available during two research flights. By Using major anthropogenic SOA precursors (C6-C9 aromatics) and their corresponding organic aerosol formation yields reported for low NO_x conditions we were able to predict ~ 50% of the organic aerosol measured in the plume. This value is consistent with studies using a similar approach in urban environments including Paris (de Gouw, 2005; Aït-Helal et al., 2013). However, since a significant fraction of SOA remains unexplained, predicting its formation is still challenging for future research.

References that have been added to the manuscript:

- Bond, C. M., Anderson, T. L., Campbell, D., and Bond, T. C. (1999). Calibration and intercomparison of filter-based measurements of visible light absorption by aerosols. *Aerosol Science and Technology*, 30(6), 582–600.
- Berndt, T, F. Stratmann, S. Bräsel, J. Heintzenberg, A. Laaksonen, M. Kulmala. SO₂ oxidation products other than H₂SO₄ as a trigger of new particle formation – Part 1: Laboratory investigations *Atmos. Chem. Phys.* 8, 6365-6374, doi:10.5194/acp-8-6365-2008, 2008
- Deguillaume, L, M. Beekmann, C. Derognat Uncertainty evaluation of ozone production and its sensitivity to emission changes over the Ile-de-France region during summer periods. *J.Geophys. Res-Atmos*, 113, D2, DOI: 10.1029/2007JD009081, 2008.
- de Gouw, J and J.L. Jimenez. Organic Aerosols in the Earth's Atmosphere. *Environ. Sci. Technol.*, 43, 7614–7618, 2009. DOI: 10.1021/es9006004
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