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.**

The manuscript describes aircraft-based measurements of aerosol composition performed in the vicinity of Paris during the MEGAPOLI campaign in 2009. The paper focuses on a characterization of non-refractory particulate matter (NR-PM) in the Paris plume as a function of photochemical age. The NR-PM measurements are compared with measurements performed in similar environments near large urban areas and also used to examine the ability of recently reported organic aerosol (OA) formation yields to predict OA loadings from simultaneously-measured volatile organic compound (VOC) mixing ratios.

The paper is well written and the topic within the scope of ACP. The field measurements reported in this paper are a valuable addition to the growing collection of SOA formation studies in the atmosphere. I recommend it for publication once the following minor revisions have been addressed.

General comments
R1.1: One general comment is that it would be interesting to see more detailed comparisons to the ground measurements in Freutel et al. (2013) as well as some of the results (e.g., OA/CO) we reported for London during a similar period (McMeeking et al., 2012).

EF: Additional and updated discussions including the ground based measurements (Freutel et al., 2013) and those from London (McMeeking et al., 2012) are 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°C ± 4 °C for Atlantic polluted and 18 °C ± 3 °C for Atlantic Clean. Similar differences in ambient temperatures were measured aboard the aircraft with 21°C ± 0.89°C for Northern sector flights (Atlantic polluted) and 17°C ± 0.5 °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 there was lower wind speeds. 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 and smaller concentrations of primary species such as BC, HOA, and NOx.

Page 12, Line 361: The combination of inorganic and organic aerosol measurements during different meteorological periods shows that aerosol mass concentrations measured in Paris are influenced strongly by the 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 ΔCO 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 $\Delta NO_3/\Delta CO$ give 23, 19, 10, and 17 μg 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 μg 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.

Values of ΔOA/ΔCO 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 ΔOA/ΔCO did not show significant variations (70 ± 20 μg m^3/ 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 ΔOA/ΔCO 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.

R1.2: While the writing is in general quite clear, there are minor grammatical and/or typographical errors in places that need to be corrected prior to publication (e.g., "in the New England, USA" [24903, line 25]; "OOA missions" [24906, line 22]).
EF: OK, these errors and any others are corrected

R1.3: Please state somewhere if concentrations have been corrected to standard temperature and pressure.

EF: No, all data is at the temperature and pressure of the plane. The C-ToF-AMS is measuring behind a PCI that maintains a pressure of ~400 mbarr at all times. The C-ToF-AMS data is corrected to the pressure of the plane.

Updated text:

Page 5, Line 116: Data acquired from the C-ToF-AMS, as well as all other measurements aboard are corrected to temperature and pressure (950 hPa) of the plane.

Specific comments

R1.4: 24890, 9: Please give an estimate of the upper size range sampled by the PSAP and C-ToF-AMS, including transmission through the aerodynamic lens in the case of the AMS.

EF: The text has been updated to respond to this comment.

Additional text:

Page 4, Line 101: Aerosol particle species were sampled through a forward-facing inlet installed in place of a side window of the aircraft, the community aerosol inlet (CAI). This is an isokinetic and isoaxial inlet with a 50% sampling efficiency for particles with a diameter of 4.5 μm.

Page 4, Line 108: The PCI ensured a constant pressure at the inlet of the C-ToF-AMS (~400 hPa) and avoids pressure changes to the aerodynamic inlet of the C-ToF-AMS during airborne sampling (Bahreini et al., 2008). The aerodynamic lens of the C-ToF is reported to have a 100% transmission efficiency between 40 nm and 500 nm when using a 100 μm orifice at 1016 mBarr (Liu et al., 2007). Bahreini et al., (2008) illustrated when using a PCI between ~400 and 654 hPa mbarr (assuming that ambient pressure is greater than that of the PCI), and an orifice >100 μm that the transmission efficiency of the lens is not changed. Bahreini et al., (2008), tried a number of different critical orifices ranging from 120 μm up to 180 μm, an orifice of 130 μm diameter was used in this study.

Page 5, Line 118: For BC measurements, a particle soot absorption photometer (Radiance research ® (PSAP), measured the particle absorption coefficient. The sampling flow rate of the PSAP was ~1.2 L min^{-1} instrument time resolution of the PSAP was < 10 seconds. Light absorption coefficient was corrected according to the Bond et al., (1999) method. Black carbon concentration was calculated using the light absorption coefficient at 650 nm and a mass specific absorption coefficient of QBC = 6.6m^2/g. Filters were changed prior to each flight to ensure that transmission efficiency was greater than 80%.

Page 5, Line 130: There was no impactor placed upstream of the PSAP, CPC or C-ToF-AMS, however given the length of tubing and the presence of several bends in the sampling line, the 50% sampling efficiency for aerosol particles is calculated to be approximately 2.5 μm (McFarland et al., 1997).

R1.5: 24890, 15: Please state whether the standard corrections were applied to the PSAP data (e.g., Bond et al., 1999)? If not, please estimate the magnitude of any biases that may result from using the un-corrected measurements and their potential impacts on defining the plume and background removal. This is especially important for the results discussed later in Section 3.2.2, since changes in the aerosol composition could affect aerosol optical properties and the PSAP measurements of BC used to normalize the other aerosol components.

EF: Yes, the PSAP data were corrected according to Bond et al., 1999, the text is updated to include this information.

Updated text:

Page 5, Line 122: For BC measurements, a particle soot absorption photometer (Radiance research ® (PSAP), measured the particle absorption coefficient. The sampling flow rate of the PSAP was ~1.2 L min^{-1}. Instrument time resolution of the PSAP was < 10 seconds. Light absorption coefficient have been corrected according to the Bond et al., (1999) method. Black carbon concentration have been calculated using the light absorption coefficient at 650nm and a mass specific absorption coefficient of Q_{BC} = 6.6m^2/g. This calculation is done in accordance with conclusions from a workshop (EUSAAR 2007) on the comparison of different measurements of absorption coefficient (MAAP, PSAP and aethalometer) with the assumption that BC always interacts the same way with the light whatever the BC particle's size. It has been illustrated in several studies that the majority of BC mass is measured in the submicron size mode (Sellegri et al., 2003).

Filters were changed prior to each flight to ensure that transmission efficiency was greater than 80%. A scanning mobility particle sizer (SMPS) measured the mobility diameter of aerosol particles from 30 to 500 nm with a resolution of 84 s. The total number concentration measured by the SMPS was compared with that of the CPC to ensure that the two instruments were coherent for comparison with the C-ToF-AMS.

There was no impactor placed upstream of the PSAP, CPC or C-ToF-AMS, however given the length of tubing and the presence of several bends in the sampling line, the 50% sampling efficiency for aerosol particles is calculated to be approximately 2.5 µm (McFarland et al., 1997).

R1.6: 24890,18: An 84-second SMPS sample time translates to a large horizontal distance, so changes in aerosol concentrations will lead to errors in the measured size distributions if they occur on similar timescales. How homogeneous was the aerosol around Paris, both inside and outside of the plume? Did the validation of the AMS collection efficiency include both plume and non-plume SMPS samples?

EF: The total number concentration of the SMPS was validated against a CPC measuring on the same sampling line. For this study, we only used the total mass concentration of the SMPS to compare with the AMS. Both in-plume and non-plume concentrations were used to validate the CE of the C-ToF-AMS.

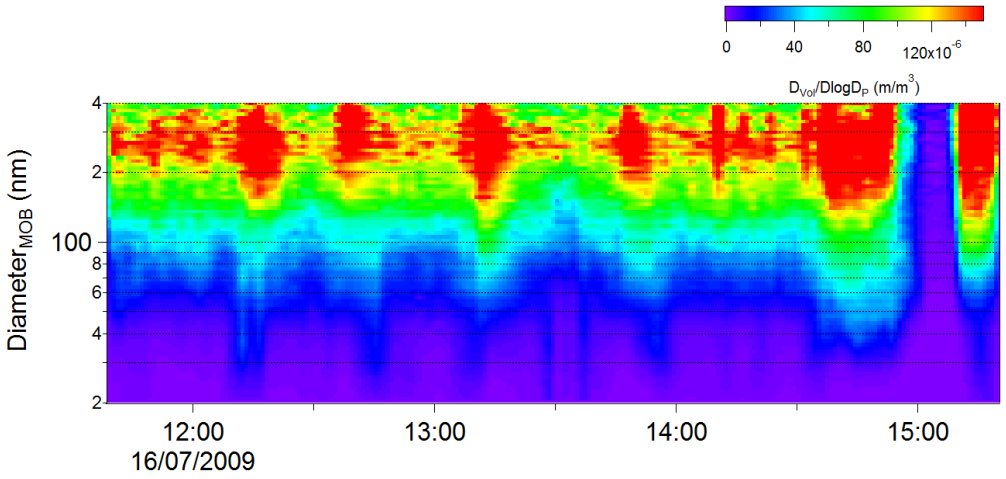
Additional text:

Page 5, Line 129: The total number concentration measured by the SMPS was compared with that of the CPC to ensure that the SMPS size distribution was acquired when there was little variation in aerosol concentrations during an SMPS scan.

Page 6, Line 161: The SMPS number concentrations were converted to mass concentrations using a density of all aerosol particles of 1.8 g cm^{-3}.

R1.7: 24891, 16-23: The observed slope of ~1 implies that the majority of aerosol mass/volume was below 500 nm, assuming the AMS measured efficiently up to the typically-reported aerodynamic diameter of about 1 µm. Do the SMPS volume distributions confirm this? The text also states that aerosol volume concentrations measured by the SMPS and AMS were compared, but Figure S1 is labeled as a mass concentration comparison. If BC volume was included in the volume comparison please also provide the density used.

EF: AMS size distributions are not shown (Low signal to noise ratio due to short data acquisition time), but they show maximum concentrations at aerodynamic diameters ranging from 200 nm up to 500 nm with average values of 350 ± 100 nm for all flights. This aerodynamic diameter is similar to the SMPS volume distributions which show maximum concentrations between 200 and 400 nm.



Although, the AMS is capable of sampling aerosol particles up to 1 micron, the aerodynamic lens of the AMS is reported to have a 100% transmission efficiency between only between ~80 and ~550 nm (at 760 torr using a 100 µm orifice) (Liu et al., 2007). The inlet transmission efficiency decreases sharply after 550 nm. The authors do not consider that during these measurements that there is a large contribution of aerosol particles after D_{VA} of 500 nm. The SMPS used in this study only measures between 30 and 450 nm.

We compared the AMS total mass concentration with the SMPS mass concentration (using an assumed density of 1.8 g cm^{-3}, and not the volume concentration as stated in the text. We would like to thank the reviewer for pointing out this error. In addition, the text stated that the calculated slopes and regression were for all flights. However, the data only showed results from

two flights. The values are now updated to include data from all flights. Figure S1 is updated to include all data. The text is corrected and updated below.

Updated text:

Page 6, Line 159: 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).

R1.8: 24892, 25: I am a little confused on how plume boundaries were defined. The text states plume boundaries were defined based on when concentrations increased above the background level by more than 0.15 µg m^{-3}, but the next sentence states the differences varied from 0.15-0.58 µg m^{-3}. Do these values represent different values used to define the plume boundaries or the range of observed differences with respect to the background? Also suggest changing "Subtracting too large background. . ." to "Subtracting incorrect background. . ." in the last sentence of the section.

EF: The text now states that the increase above the background level must be ≥ 0.15 µg m^{-3}. The values listed represent the observed differences with respect to the background.

Updated text:

Page 7, Line 183: Plume boundaries were defined as when the difference in BC above background (ΔBC) was ≥ 0.15 µg m^{-3}

Page 7, Line 190: Subtracting incorrect background values can skew these ratios to either very large or very small values.

Page 7, Line 186: For all RF, the ΔBC within the urban plume was observed to vary from 0.15 and 0.58 µg m^{-3}, with lowest values measured during eastern flights and highest during northern flights.

R1.9: 24893, 12: define MONA acronym

EF: The text is updated to include the definition of MONA "Measurement Of Nitrogen on Aircraft "

Updated text:

Page 7, Line 196 : "NO and NO_2 were sampled through a separate rear-facing pressure controlled inlet at a 30 s time resolution and measured using the "Measurement of Nitrogen on Aircraft (MONA)" instrument based on ozone chemiluminescence and developed by the Laboratoire Interuniversitaire des Systems Atmospheres (LISA), Paris."

R1.10: 24893, 15: "inox tube cover of gold"

EF: Test is updated.

Updated text:

Page 7, Line 199: The air then passes through a gold converter (8 mm gold coated inox tube) heated to 200°C with H_2 as a reagent to convert nitrogen species into NO.

R1.11: 24894, 14-16: Please also change "northerly, north-easterly, and easterly direction" to "northern, north-eastern and eastern directions" here and throughout the manuscript to avoid confusion with wind direction (e.g., the "easterly" research flights were associated with westerly winds).

EF: These have been changed throughout the manuscript.

R.1.12: 24895, 7-12: It may also be worth commenting on the different photochemical environments in the two locations (Paris versus Mexico City).

EF:

Updated text:

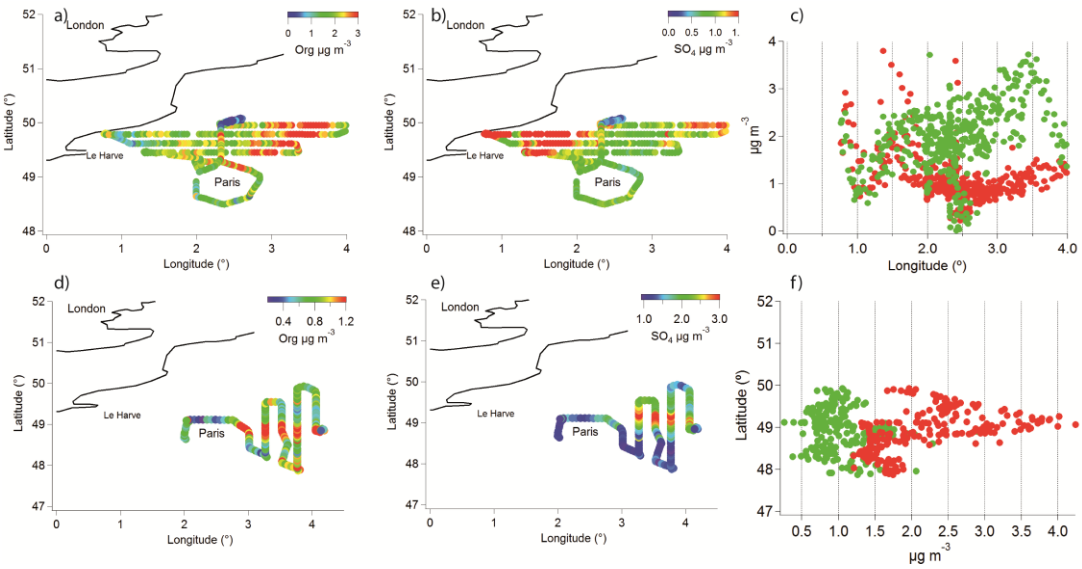
Page 9, Line 260: Photochemistry within both Paris and Mexico City are thought to be VOC limited (Song et al., 2010, Deguillaume et al., 2008). As the plume becomes chemically aged, the photochemistry is thought to shift to an NO_x-limited regime.

R1.13*: 24896, 26: The wording here is a little confusing "SO4 mass concentrations appear to decrease as soon as they encounter the urban plume". I believe the authors mean SO4 concentrations are observed to decrease when the aircraft encounters the urban plume, not the air mass itself, which originated to the west, not north, of the plume in the example being discussed. It is difficult to see the changes in sulfate relative to other aerosol components in Figure 4d. The sulfate and OA plumes appear to be in slightly different locations based on this graph. A separate figure showing OA and sulfate concentrations versus latitude might help.*

EF: The text and figures are updated.

Updated text:

Page 10, Line 289: However, during airborne measurements these SO_4 mass concentrations are observed to decrease as soon as the aircraft encounters the urban plume, and when organic, nitrate, and BC aerosol begin to increase. In order to illustrate this more clearly we included plots of SO4 and Org concentrations as a function of longitude (Figure 4c Concentration vs Longitude) and latitude (Figure 4f Latitude vs Concentration).



R1.14: 24901, 11: Can the authors comment further on the low differences between background and local CO mixing ratios? Was this true even for the flight leg closest to Paris? Were observed excess BC / excess CO ratios consistent with similar measurements downwind of other urban regions?

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. 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. Therefore our CO measurements are in agreement with those of London but are not similar to those in New England or in Mexico City.

The text is updated to include this information.

Updated text:

Page 15, Line 441: 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 ΔCO are low. However, compared with London, these measurements appear to be representative of European air masses.

R1.15: 24905, 21-25: The similar concentrations of isoprene inside and outside of the Paris plume suggest Paris itself does not represent a major biogenic VOC source (at least for isoprene), but the

regional biogenic emissions may still play a role in SOA formation in the plume through interactions with anthropogenic VOCs (e.g., Spracklen et al., 2011).

EF: Text has been updated.
Updated text:

Page 18, Line 593: Since biogenic VOC emissions (isoprene) were similar within and outside of the Paris urban plume we do not believe that biogenic VOC had a strong impact on the formation of secondary OA. However, biogenic VOC emissions may still play a role in secondary OA formation in the plume through interactions with anthropogenic VOCs (Spracklen et al., 2011, Shilling et al., 2013).

R1.16: 24906, 24-26: It is also worth commenting on different temperatures at the surface compared to aircraft sample height given that some of the OA is semi-volatile.

EF: The text has been updated to include information on the temperature.

Updated text:
Page 9, Line 244: 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°C ± 4 °C for Atlantic polluted and 18 °C ± 3 °C for Atlantic Clean. Similar differences in ambient temperatures were measured aboard the aircraft with 21°C ± 0.89°C for Northern sector flights (Atlantic polluted) and 17°C ± 0.5 °C for eastern sector flights (Atlantic clean).

Page 21, Line 625: As described in section 3.0, the vertical temperature gradient between the ground sites and that of the aircraft were small, for this reason we do not believe that we can make any conclusions on the impact of temperature on the formation of SV-OOA vs LV-OOA.

R1.17: General comment on tables and figures: Both "OA" and "org" are used in the Tables and Figures. . .should be consistent unless there is a reason for the distinction.
EF: All figures and tables are now updated.

R1.18: Table 1: Units for the total aerosol mass concentration are listed under the OA percentage column label. Also give units for BC.
EF: This is corrected now.

R1.19: Table 2: Give units for mass concentrations. Picky point, but first column is date, not the research flight numbers listed in Table 1.
EF:Thank you for pointing this out. The table is now updated.

R1.20: Table 3: Could also list OA/CO values reported for London here as a column in this table as it is probably the most comparable study location.

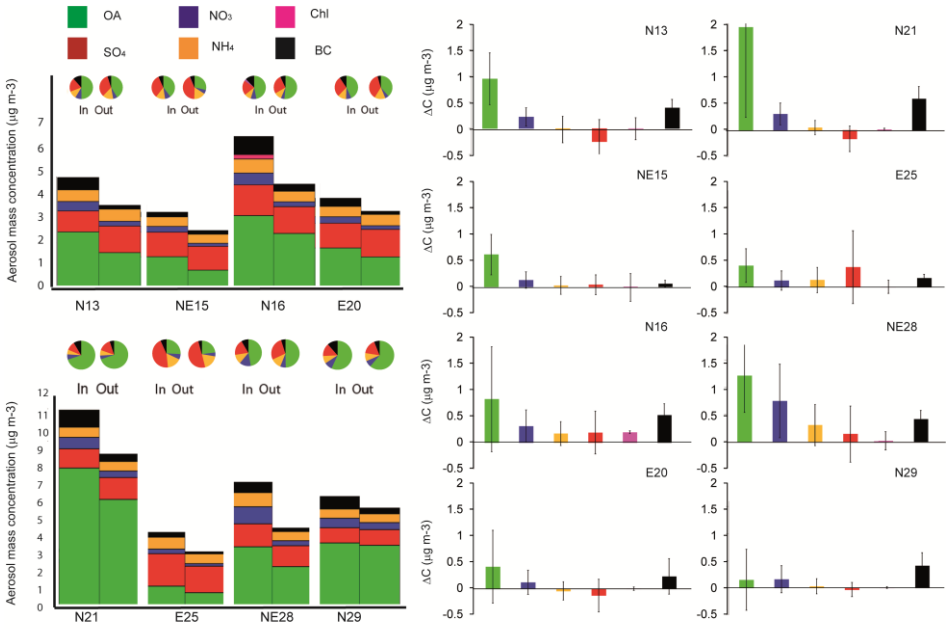
EF: More discussion of the EM25 dataset is included in the manuscript. Table 3 is updated to include the measurement data.

ΔOA/ΔCO μg m^{-3} ppm CO

Photochemical age	N16	E20	N21	N29	Mexico City[a]	NEAQS 2002[b]	NEAQ/ITCT 2004[d]	EM25
Near source	40	37	62	32	10	37	6.6	9±3
After 1 day*	98	101	133	97	73	103	70	50
Change	58	64	71	65	63	66	63	41
Absolute CO	152	138	150	150	2500	325	325	~120

R1.21: Figure 3: Including error bars on the right-hand side panels would help show if observed differences between plume and non-plume are significant.

EF: Figure 3 is updated with error bars. The error bars are calculated from the average of the standard deviation calculated for the average data within and outside of the plume.



G.M: 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.

G.M: Spracklen, D. V., Jimenez, J. L., Carslaw, K. S., Worsnop, D. R., Evans, M. J., Mann, G. W., . . . Forster, P. (2011). Aerosol mass spectrometer constraint on the global secondary organic aerosol budget. Atmospheric Chemistry and Physics, 11, 12109–12136. doi:10.5194/acp-11-12109-2011 Interactive comment on Atmos. Chem. Phys. Discuss., 13, 24885, 2013.

EF: References are now included.

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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.

$$\left(\frac{\Delta OA}{\Delta CO}\right) predicted = \left(\frac{\sum \gamma_i * \Delta VOC_i}{\Delta CO}\right) \quad \textbf{(Eq.6)}$$

<u>Updated text:</u>

<u>Page 19, Lin 561</u>: 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 Δhydrocarbons (ΔHC) determined by Ng et al. (2007) (Table S4).

Page 21, Line 641:

<u>Original text:</u> 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.

<u>Updated text:</u> 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.

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