Response to Anonymous Referee #2

We thank the reviewer for his/her thoughtful comments. The manuscript has been revised accordingly. Listed below are our point-by-point responses to the reviewer's comments, which are repeated in italic.

General comments:

(a) I think it is better not to show results for July 27 in Fig. 2, 3, 5, and 9. In page 30729, the authors stated that the data discussion was focusing only on the other three observations because the measurements on July 27 were largely influenced by generator exhaust (explained in detail in the supplementary). The presentations of contaminated data set in those figures parallel to the other observations are therefore unnecessary and would somewhat mislead the readers.

[Response]: We agree with the reviewer. All the results from July 27 were removed from the revised manuscript. They are now included in the supplementary material.

(b) The definitions of LV-OOA and SV-OOA imply a reverse relationship between volatility and O:C (Jimenez et al., 2009). With only information of O:C and m/z 43-to-44 ratios, it is not enough yet to conclude that one OOA is more volatile than the other. For example, Hildebrandt et al. (2010) identified two OOA factors in their study. Although the O:C ratios were estimated to be 0.9 for OOA-1 and 0.6 for OOA-2, OOA-1 were not found to be less volatile than OOA-2 (perhaps both are LV-OOA). Therefore, the identification of LV-OOA and SV-OOA in this study needs additional evidence to support. Moreover, the spectra of "LV-OOA" in the literature typically have more prominent m/z 44 and higher O:C (Jimenez et al., 2009). The LV-OOA spectrum shown in Fig. 7 for this study is indeed more similar to that of "SV-OOA" in DeCarlo et al., 2010 compared to typical "LV-OOA" spectra. Despite the lack of information about volatility, it seems that the two OOA components identified in this study could be both SV-OOA.

[Response]: We agree with the reviewer's comment that, for some cases, the O/C ratio and *m/z* 43/44 ratio of OA components might not be consistent with their volatilities, e.g., the observations by Hildebrabdt et al. (2010). Although no direct measurements of particle volatility (e.g., using a thermodenuder) were conducted during this study, we established the naming based on the fact that LV-OOA correlates well with sulfate and SV-OOA correlates with ammonium nitrate.

(c) Multiple aspects of the data on July 28, including the similar average bulk composition for less and more traffic periods (page 30730), the elevated HOA and ultrafinemode organics (page 30730) and the spikes in organic mass concentration (page 30731) during more traffic periods, and the weak correlation between HOA and BC (page 30735), were explained by the emissions from the MTA buses rather than from the vehicles in the LIE highway. The MTA bus stop, however, is in the north of the sampling site C (Fig. 1). The explanation of MTA bus emissions seems not supported by the wind direction that was stated by the authors as "persistent from the campus".

[Response]: The meteorology data shown in Fig. 1 were collected from the campus site because no measurements were available at the roadside site. The wind direction and wind speed might vary differently at these two sites due to microscale meteorology. The measurement heights at the two sites were also different (ground level for aerosol measurement vs. ~5 m for meteorology measurement). Thus, the MET data are only presented as a reference. We now make this point clear in the revised manuscript. Given that the MTA bus stop is very close, approximately 2 m, to our sampling inlet, it is very likely an important point source for our measurements. In addition, HOA has shown good correlation with BC in aerosol particles clearly influenced by the LIE traffic emissions (e.g., 30 July). For these reasons, we conclude that the possible explanation for the weak correlation between HOA and BC is likely due to the emissions from MTA buses nearby.

(d) In section 3.2, the authors showed that elevated concentrations due to traffic emissions were peaked at ~120 nm (D_{va}) and ~10 nm (D_m). While the 10-nm particles are too small to explain the elevated mass concentrations at 120 nm (D_{va}), the decrease of number concentrations in 30-70 nm (D_m) (Fig. 6b) seems to imply an opposite trend in mass size distributions. Additional discussion is therefore needed regarding to this point for data consistency, perhaps on the basis of the FMPS volume distributions, particle density, and shape factor.

[Response]: Although the 10 nm particles accounts for a large fraction of total number concentration, it contributes a minor fraction of the volume concentration. To address the reviewer's comment, we now present the volume distribution from the FMPS measurements in the supplementary (Fig. S9), and estimated the particle density as well. The estimated particle density based on the aerosol composition is ~1.4 g cm^{-3} assuming that the particles are spherical. D_{va} = 120 nm is thus approximately equivalent to D_m = 85 nm ($D_{va} \approx D_m \times \rho / \rho_0$). Indeed, we do see some enhancements at this size range in the particle volume distribution. However, significant differences are often observed between the SMPS volume distribution and the AMS mass-weighted size distribution (e.g., Zhang et al., 2005) since the comparison between D_{va} and D_m is largely affected by the particle shape, especially when the particle composition has a considerable contribution from BC, like in this study.

Specific comments:

(1) P30723, Line 14-16: This statement is misleading. It needs to be clarified that such "dominant" or "small" contributions are relative to the total vehicular emissions not the total emissions.

[Response]: We clarified this sentence. Now it reads "Gasoline-powered vehicles were found to contribute mostly to CO, aromatic and carbonyl species, and ammonia, but a small fraction of $PM_{2.5}$ and BC from the total vehicular emissions."

(2) P30727, Line 15-18: Because the meteorology data were not directly measured at Site C, it is important to evaluate the potential bias, especially in wind direction, on the basis of sampling height and topography etc. Also see the general comment (c).

[Response]: Since no meteorology data is available at the roadside site, we used those measured at the campus of QC as a reference. The measured height at the campus site was ~5 m, higher than that of at the roadside site (~2 m). Because of the different sampling height and surrounding environment, the meteorology might be different at these two sites. We now mention the potential bias when using the meteorology in the campus as a surrogate in the revised manuscript.

(3) P30727, Line 19: A recent understanding on the AMS mass concentrations is that the measured values correspond to the temperature and pressure conditions for the flow-rate calibration (personal communication with Jose Jimenez, CU). Also, because instruments were installed inside the ASRC-ML, sampling temperature might be different from ambient values. The authors should state clearly that how the various data sets were treated when converting the data to ambient conditions and provide the ranges of ambient temperature and pressure.

[Response]: The temperature inside ASRC-ML was almost identical to that in ambient environment since all the doors were open, thus free exchange of air between indoor and outdoor, during this study. In addition, sampling heights did not change during the whole sampling period either. We therefore think the impacts from temperature or pressure change on the reported mass concentrations would be negligible.

(4) Page 30728, Line 8-9: The authors stated that the four measurements were combined into one data set for the PMF analysis and the retrieved profiles were hence forced to be identical for all four experiments. Did the authors run PMF for each data set and obtain similar results? The question behind is in what ranges the factor profiles would change (e.g., in Fig. 7).

[Response]: Yes, we did run PMF for each dataset. The factor profiles of HOA and SV-OOA retrieved from the two morning datasets (28 July and 30 July) are very similar, and LV-OOA and NOA have some differences. The factor profiles from the evening dataset (1 Aug.) show more aged HOA and LV-OOA in comparison to the two morning datasets. To be consistent and better comparisons, we combined all the dataset and forced the retrieved the factor profiles to be identical for all experiments.

(5) Page 30728, Line 13: The authors showed a careful diagnosis about the large silicone contamination on July 27 in the supplementary. A component representing the contaminated OA was resolved from the UMR PMF analysis (Fig. S7). However, only four components (HOA, LV-OOA, SV-OOA, and NOA) were identified from the HR PMF analysis. Why is the exhaust component missing in the HR PMF analysis? Did the authors exclude ions like C_xH_y at m/z 73 in the analysis (seems not shown in Fig. 7)? Clarification is needed and the unsolved exhaust contribution to other factors (e.g., HOA) should be speculated in the main text.

[Response]: We did exclude major contamination ions such as $C_3H_9Si^+$ at *m/z* 73 from the mass spectra matrices before HR PMF analysis. The HR PMF analysis was also limited to *m/z* <120, mainly because of the difficulties to distinguish different ions at large *m/z*

due to the limitation of AMS mass resolution. Thus, other prominent contamination *m/z*'s larger than 120, e.g., 147, 201, 221, and 281 from the generator exhaust are not included in the HR PMF analysis either. Furthermore, the major exhaust peaks were also removed before the HR PMF analysis, which would lead to an even smaller contribution of exhaust factor to other PMF factors. For these reasons, the four factor solution didn't resolve the exhaust factor. However the impacts of the exhaust factor can still be seen in the dataset from July 27, which those from 28 July, 30 July, and 1 Aug. don't show (Fig. S7). Following the reviewer's suggestion, we clarified this point in the revised manuscript.

(6) P30730, Line 27: The wind direction plotted in Fig. 2 for July 28 is 120-240 degree, indicating an origin of wind from the south not the north.

[Response]: The wind direction was corrected to the south in the revised manuscript.

(7) Figure 5b: I can hardly see light gray lines on my screen.

[Response]: The light gray lines were removed and only the smoothed lines were kept for clarity in the revised manuscript.

(8) P30733, Line 8-15: It seems not appropriate to describe the "less traffic (i.e., before 7:30 a.m.)" distribution has a bimodal structure and the "more traffic (i.e., after 7:30 a.m.)" distribution has 3 modes. The two distributions (Fig. 6b) show at least 4 modes at 10, 18, 30, and 50 nm (and perhaps 120 nm). The mode structures are similar, although the relative height of each mode is different for the two distributions.

[Response]: We performed log-normal fit to the average number size distributions during LT and MT separately. Three modes were fitted with the peaks at ~11, 40, and 140 nm, respectively, during LT period. Comparatively, multiple modes were needed to get the best fit for the size distribution during MT period. With these results, we have revised our discussions accordingly.

(9) The works of Klems et al. 2010, 2011 are highly relevant to this study and could be cited perhaps in section 3.2.

[Response]: Klems et al. 2010, 2011 were cited in the revised manuscript

(10) The analysis in Page 30732, Line 23-25 is somewhat conflicting with the statement in Page 30734, Line 17-20.

[Response]: The "BC" referred in these two sentences came from different measurements, and the statement is not conflicting. For clarification, we have revised the sentence in the revised manuscript. Now it reads "HOA during MT on 28 July shows similar small particle mode to the BC measured along the LIE by a Soot-Particle AMS (SP-AMS)".

(11) P30736, Line 19-20: The suggestion of greater exposure to particle pollution of people living nearby the highways than living upwind or far from the highways is over-stated. Exposure assessment strongly depends on living style. Living close to the highway does not necessarily mean breathing more toxic air pollutants from the highway surroundings. Individual's indoor and outdoor activities and the toxicity of the pollutants are important aspects for the exposure assessment. I suggest deleting this statement from the conclusion.

[Response]: In response to the reviewer's comments, we have revised the discussions and the text now reads: "While this study does not provide direct information on particle toxicity and health impacts, our results might nevertheless have significant implications for near-highway air pollution characterization and exposure assessments. For example, the fact that the mass concentrations, chemical composition and size distributions of submicron particles change rapidly over short distances near highways suggests that people living in close proximity to the highways are regularly exposed to higher level of particle pollution than people living far away and that PM exposure assessments should take into account of the temporal and spatial variations in aerosol loading and properties near roadways. Since aerosol changes are found strongly dependent on wind direction and speed, mixed layer height, vehicle types, and traffic flow, additional studies are needed to thoroughly assess their impacts on near roadway air quality. In addition, long-term, continuous measurements of air pollutants near highways are necessary to characterize their diurnal, weekly, and seasonal variation profiles, all of which are important for interpreting epidemiological studies of ambient air pollution.

(12) P30737, Line 7: Be more explicit about "upwind areas".

[Response]: "upwind areas" is now specified as "the opposite direction of the highway".

Technical corrections:

P30722, Line 1: A missing space between "(FMPS)" and "measurements".

P30723, Line 9; P30724, Line 25 and 28: Missing comma for the use of "e.g.", "i.e." and "however" in a sentence.

[Response]: corrected

P30723, Line 17: "Most of previous studies" of what?

[Response]: It was revised as "Most previous roadside measurements"

P30723, Line 21-22: Remove "(> 10 nm)". In a previous sentence, the authors just stated that the particles at 30-m downwind were already greater than 10 nm (i.e., before growing up).

P30723, Line 26: Does "particle numbers" mean "particle number concentrations"?

P30724, Line 21: Be more explicit what "hydrocarbon characteristics" are.

P30724, Line 16-17: Does "diesel aerosol particles" mean "particles from diesel emissions"?

P30726, Line 11-13 and P30727, Line 14: Provide year for sampling periods.

P30727, Line 4: Delete the extra word "spectrometer".

P30730, Line 2-3: Redundant "and". Perhaps just say "which reflects ... in traffic flow, traffic type, and meteorology."

P30731, Line 7 and 11; P30732, Line 4; P30735, Line 22: Incorrect and redundant use of "also".

P30733, Line 5: Delete the extra comma after "(r^2=0.53)". Otherwise, the last part is confusing.

P30737, Line 11: Insert a comma between "vehicle types" and "and traffic flow".

P30752, figure legend: + signs for the ions are misplaced.

[Response]: Revised or corrected following the reviewer's suggestions

A general technical comment is that there are some ambiguous statements throughout the manuscript that need to be polished in the revised manuscript. Here are some examples: P30723, Line 8-10: ""driving modes, e.g., ... measurements, ... studies, ... sampling..."; P30723, Line 21-22: "number distributions ... to larger particles..."; P30730, Line 3: "The variation ... is flat ..."; P30731, Line 9: "compared ... measurements... and the site A ..."

[Response]: Following the reviewer's suggestions, we revised related texts to improve clarity.

References:

Hildebrandt, L., Engelhart, G. J., Mohr, C., Kostenidou, E., Lanz, V. A., Bougiatioti, A., DeCarlo, P. F., Prevot, A. S. H., Baltensperger, U., Mihalopoulos, N., Donahue, N. M., and Pandis, S. N.: Aged organic aerosol in the Eastern Mediterranean: the Finokalia Aerosol Measurement Experiment - 2008, Atmos. Chem. Phys., 10, 4167-4186, 10.5194/acp-10-4167-2010, 2010.

Zhang, Q., Canagaratna, M. C., Jayne, J. T., Worsnop, D. R., and Jimenez, J. L.: Time and size-resolved chemical composition of submicron particles in Pittsburgh - Implications for aerosol sources and processes, J. Geophys. Res., 110, D07S09, doi:10.1029/2004JD004649, 2005.