We thank the referee for his/her time to provide us with extensive and valuable input. Please find below our responses to the raised comments, questions and suggestions. In the following, raised **comments / suggestions are in red** and respective **responses in green**, while **alterations to the manuscript text are indicated in blue.**

General Comment

This paper presents measurement of ambient carbonaceous submicron particulate matter in a roadside site in urban Hong Kong. Multilinear regression analysis was performed with the observed HOA and EC concentrations and real traffic data to interpret the contribution of different vehicles to the carbonaceous aerosols. The results would be helpful to the local authority on traffic emission controls and be of interest to the community. The writing and construction of the paper need to be further improved before considering for acceptance, and the extremely long paragraph and sentences could confuse the reader and make the meaning ambiguous. Some parts in the introduction, methodology, and discussions also require more clarification and refinement.

There are always long paragraphs and long sentences without appropriate breaks, and the lack of organization confuse the reader and diminish the intent of the discussions. For example, Line 81 to 112, single long paragraph for Section 3.1, line 279 to 317, and also some extremely long sentences in many places in the text.

We agree that there are occasional long sentences but we do not consider these to be too detrimental to the overall readability. We generally insert paragraph breaks if there is a thematic change, and keep contextually-related parts arranged together. In the revised manuscript, we have split longer paragraphs wherever possible to reduce the amount of long sections as suggested.

-- Insertion of paragraph breaks throughout the main manuscript --

Introduction. Since the present study made substantial analysis and discussion of the primary emission of HOA from different vehicles, the author needs to provide some review of the finding and current understanding of HOA from traffic emission in the introduction section, to better present what is new and significant in the present study.

We have substantially revised the introductory part to reflect a more general discussion on the relevance and impacts of traffic emissions on primary and secondary aerosol pollution in urban environments and the aims of our work. Due to the extent of the changes, the introductory section in its entirety is appended at the bottom of this document.

-- See end of document for revised introductory section --

Methodology. More details on the multilinear regression would be necessary, including inputs and outputs of the regression, any assumptions were made in the analysis, and the uncertainties raised by different factors, three-point box smoothing, constrained intercept, etc. For the sake of clarity, this information should be included to show the validity of the regression and analysis after that.

The multiple linear regression (MLR) is discussed in more detail in the corresponding result/discussion section (3.3.1.), since the immediately preceding discussion on the characteristics and variabilities in the measured carbonaceous species and traffic composition are critical in setting the context for the MLR analysis.

While we mentioned input and output information within the discussion, we more explicitly state these information at the beginning of section 3.3.1 in the revised manuscript.

Smoothing was performed due to the brevity of the data set (72h), where individual concentration or vehicle count spikes would have skewing effects on the regression analysis. We opted for box smoothing over three points as described in the methodology section to retain the general trends as close to the original data series as possible.

The choice of a zero intercept is based on the following considerations:

1. HOA and EC are predominantly local pollutants originating from the direct vicinity of the sampling site. A further significant source of EC (and HOA) could be shipping, however, the nearest coastline is at a considerable distance (~ 2km) and shielded by complex, dense and tall urban building geometry. We consider a persistent influence from shipping emissions on ground-level pollution at this measurement site as unlikely.
2. Organic species from farther sources (other city areas or regional pollution) are very likely to have undergone some form of atmospheric oxidation and would be accounted for in PMF-resolved oxygenated species (SOA – secondary organic aerosol).
3. Traffic in the vicinity of the sampling site is largely homogenous (dense inner city traffic) and short-range transport through the street canyons should have little effect on the measured composition and concentration of carbonaceous PM_1 species.
4. With a constant (non-zero) intercept, we would physically assume a constant background level (of either HOA or EC), regardless of changes in surface wind, air mass influence or possible diurnal changes in (background) source strength, which would not be sufficiently realistic.

We have expanded the discussion on the considerations for the MLR analysis in the revised manuscript.

Section 2 (Methodology)

[…] Measured HOA and EC during the traffic counting period were decomposed by multilinear regression (MLR) based on the time series of these engine count data, i.e. the hourly time series of HOA and EC were regarded as functions of the count of diesel, gasoline and LPG powered vehicles representing the independent regression variables. To reduce the skewing impact of scattered spikes in vehicle count number, HOA and EC concentrations, the time series were subjected to three-point box smoothing prior to regression analysis. Multiple linear regression was performed for two and three factor solutions and with a constrained zero intercept. A non-zero intercept may not be physically meaningful as it requires the assumption of a constant background level of HOA and EC regardless of changes in surface wind, air mass influence or diurnal changes in background source strength. Contrarily, traffic is largely homogenous in the vicinity of the sampling site (dense inner city traffic) and other possible combustion sources (i.e. shipping) were sufficiently removed (~2km of straight-line distance to the nearest coastline) and shielded (complex and tall urban geometry) from the ground-level sampling site, i.e. impacts of transport on measured traffic-related carbonaceous constituents can be assumed to be of minor influence. MLR solutions were assessed per statistical significance of resolved regression coefficients, adjusted R^2 of the regression and the distribution of residuals. Further details are discussed in the corresponding discussion section 3.3.1. […]

Section 3.3.1 (Results & Discussion)

[…] Utilizing the detailed information on average daily traffic composition at the measurement site from the counting exercise, contributions of exhaust from different engine types to overall ambient HOA and EC concentrations can be evaluated. Multiple linear regression was carried out with the concentration time series of HOA and EC during the traffic counting period as the dependent variables, the pooled sum of counted diesel, gasoline and LPG vehicles as the independent variables, and a constrained zero intercept, assuming negligible transport of HOA and EC (see section 2). Two factor solutions (diesel + gasoline), as well as three factor solutions (diesel + gasoline + LPG), were considered, as LPG vehicles are expected to contribute less to particle-phase and more to gas-phase emissions (Faiz et al., 1996). The key statistical output parameters (adjusted R^2, regression coefficients, p-values) of the multiple linear regression analysis for HOA and EC for two and three factor models are presented in Table S1 in the Supporting Material. […]

Results. Line 161-170, Line 182-186, and Line 189-193. The general descriptions of HOA during the campaign, relation with other species (e.g., NOx), the EC/HOA ratios, and the Sunday reductions of HOA and EC, all have been previously presented in the published paper by the same author (Lee et al., 2015). Thus it is suggested to condense the already published results and try to focus on what is new from the analysis in the present study.

While we agree that parts of the methodology and underlying data are similar as in the mentioned overview paper, this manuscript is not a companion paper and should remain readable as a separate analysis. The concerned parts of the analysis are essential in providing the context for the discussion of the work of this study and should therefore be retained.

Figure 1c, the author has already reported similar results from the same campaign in his previously published papers, i.e., Figure 4c in Lee et al., 2015, except that the current one uses EC in PM1 with an empirical ratio of 0.8 from PM2.5. It is also surprised that the same dataset for HOA gave different results between Lee et al., 2015 and the present study, see below comparison figures. The author needs to clarify this, otherwise, there are reasons to doubt the validity of the data and analysis in the present study.

While the base data are the same for both studies, we have employed a different analysis procedure: in the overview paper (Lee et al., 2015) the entire available dataset of EC and HOA data was used in the calculation of the Weekday-Sunday difference.

As data gaps (e.g. due to instrument maintenance) may bias average concentrations - especially if diurnal variations are well pronounced as is the case for traffic-related pollutants at this sampling site - we excluded days where data availability was < 75% (i.e. more 6h of data missing) from both HOA and EC datasets in this work. This leads to slight differences in nominal concentrations (and thus relative reductions) between this work and the cited work but does not affect the overall conclusions in either study. We consider the method employed here an improvement in accuracy compared to the previous work and we will point this out more specifically in the revised manuscript for clarification.

[…] Both HOA and EC exhibited significant reductions in overall mass concentrations on Sundays between 14% and 27%, compared to the rest of the week (Monday to Saturday) as shown in Figure 1d. Days with less than 75% (>18h) data availability were excluded from both time series in the analysis to obviate bias in the daily averaging from their strong diurnal patterns, as opposed to the original data presented by Lee at al. (2015) where the entire unabridged time series were used. […]

Same problem for Figure 1a, the diurnal variations of EC and HOA in the summer campaign had also been published in the previous paper, i.e., Lee et al., 2015, as a supplement figure, Figure S5. Also, the Figure1Sb in this manuscript is also similar to Figure 4d in Lee et al., 2015.

The analysis of traffic related carbonaceous aerosol components in this work has been substantially expanded compared to the overview paper (Lee et al., 2015). As discussed above, to ensure readability as a standalone paper, we include previously analyzed data to provide sufficient background information required for the context of the discussions in this paper.

Section 3.3.2 and Conclusion. The discussion on the control strategies did not show too many links with the results obtained in the present study, some of the statements seem too speculated without direct results or evidence to support, e.g., Line 332-335. I would suggest the author perform further analysis with other related species and data, and emphasize the indications from the results, and highlight what is new regarding findings in the present work in comparison to previous studies conducted in other places in the world.

A direct evaluation of control strategy impacts requires a long-term database of measurement data in terms of vehicle-related ambient species as well as detailed traffic count and composition data, which are unfortunately not available. The local specificity of both traffic characteristics and regulatory control strategies similarly limits direct inferences to be made from studies from other parts of the world.

Nonetheless, as presented in this manuscript, we examined findings from various recent studies on the emission characteristics of vehicles, including high EC fractions in GDI gasoline vehicles, efficient total PM reduction in diesel vehicles equipped with exhaust after treatment devices (such as DPF, EGR) as well as overall changes in ambient concentrations observed at the roadside in Hong Kong which altogether are consistent with the results obtained from our analysis of the measurement data. Apart from the mentioned species (VOCs, criteria gas pollutants, NR-PM_1, EC, OC, meteorological data), no other supporting data were available in the captioned time period and we note that "carbonaceous particulate matter" is the focal point of this manuscript.

Specific Comments

Comment	Line 34-39. The author stated that the previous investigations typically rely on source inventories with models, then how about the previous field measurements at the roadside environment? It is better to perform more comprehensive review regarding similar field measurements.
Response	We have substantially revised the introductory part and include further references and discussions to both field and laboratory measurements.
Alteration	*-- See end of document for revised introductory section --*
Comment	Line 40. It is not clear how different the approach in the present study compared to previous studies in the literature. The author should review the literature methods first and then can come up with the statement that the approach here is different and better.
Response	As mentioned above, we have included further references and discussions to both field and laboratory measurements. The aim of this paper is to present results from one possible approach to investigate the role of different exhaust types and their influence on ambient species concentrations (as opposed to emission factor or exhaust characterization studies) utilizing more detailed traffic data. "Different" as used in our introductory section refers to the previous description of other commonly used methods and the evaluation of the performance of our method to other methods is beyond the scope of this manuscript.
Alteration	*-- See end of document for revised introductory section --*
Comment	Line 40-47. I would suggest the author re-locate the position of this paragraph to the place more fits the content, for example, the end of the Introduction section, where the description seems have some connection to this paragraph.
Response	We have revised the introductory part with added discussions on the role of motor-vehicle derived particulate matter, improving the contextual relationship of this paragraph to the introductory section.
Alteration	*-- See end of document for revised introductory section --*
Comment	Line 63 to 67. Was the data measured in the present study also affected by the non-local pollution sources? Then how to differentiate the effects of local emission from non-local influence? Anyassumptions were made, and any uncertainties would be raised? The author should clarify this in the results and discussion section.
Response	We refer to our response in the general comment section with regard to the methodology part. HOA is freshly emitted POA with corresponding mass spectral characteristics. There are no other significant sources of fossil fuel combustion in the area, except for shipping. As mentioned earlier, the location of the measurement site within a street canyon and radially surrounded by dense and high building structures make a consistent influence of shipping emissions on our ground-level measurements unlikely. The same applies to EC. Other non-local pollutants are likely to have undergone atmospheric processing during transport and would be resolved as other factors (e.g. SOA factors in the PMF analysis), which are not discussed in this work.
Alteration	N.A.
Comment	Line 61-67 and Line 75-80. The author listed many results from previous studies, however, the simple enumeration without refinement makes the descriptions confused. Another example can be found in Line 84-86, it is not clear about the purpose to mention the contribution to ozone formation, since nothing was discussed in this manuscript regarding the ozone issues.
Response	Studies on traffic emissions from the South East Asian region are relatively scarce and we therefore provide a more detailed overview of the research efforts in Hong Kong to provide a context for the reader to understand the location-specific circumstances, especially in contrast to Europe and the USA (lower building and population density) or the mainland of China (differences in vehicle composition/technology). While the ozone issue may not be directly related to this work, we included

	such references for a more complete overall picture. We offer a more compact discussion of studies using similar methodologies, e.g. the ambient EC and OC measurement based studies. For gas-phase components and emission factor studies, methodologies differ widely across the reviewed papers and we therefore summarize their key findings on a more individualistic basis.
Alteration	N.A.
Comment	Line 93-94, How about the share of OC in diesel emissions, since the emission factors of OC is about 8 times higher than that for non-diesel vehicles?
Response	The same study reported a 26% share of OC in the $PM_{2.5}$ emissions of diesel vehicles.
Alteration	N.A.
Comment	Line 97-99. Many studies reviewed in this section were conducted around or after 2010, e.g., Ning et al., 2011; Ho et al., 2013; Huang et al., 2014; Cheng et al., 2010; Yuan et al., 2013; Sun et al., 2016; Lee et al., 2015, etc. The author needs to clarify more on why '*they are unlikely to reflect the contemporary… over the last 15 years*' and the advantages of the present study to make progress on this issue.
Response	While it is correct that these studies were published in fairly recent years, most of the measurements reporting EC and OC concentrations or emissions explicitly in these studies were undertaken in the early 2000s (e.g. Cheng et al, 2010 reports on measurements from 2003 as mentioned in the same paragraph in the manuscript, while Yuan et. al., 2013 covered filter samples up to 2008 only as mentioned in the introductory section). Changes in traffic mix and engine technology / exhaust treatment since then are highly likely to have taken place and our more recent measurements would yield a more up-to-date evaluation of traffic-exhaust contributions to ambient PM.
Alteration	N.A.
Comment	Line 185-186. Did the concurrent measurement of hydrocarbon in the gas-phase show any pieces of evidence to support the hypothesis of more partitioning of HOA in gas-phase in summer?
Response	Only measurements of VOC compounds were available, which do not include higher molecular weight species that form part of the semi-volatile (SV) fraction of HOA.
Alteration	N.A.
Comment	Line 206-208. The purpose and links of these two sentences with the following discussion are not clear. The number of counted vehicles during the three-day counting exercise should be more useful here.
Response	As discussed in the methodology section, the traffic counting exercise did not continuously monitor all lanes of traffic at the site and therefore, we here elaborate on the relationship between the counted traffic numbers and the expected total number of vehicles from official government statistical data.
Alteration	N.A.
Comment	Section 3.3.1. The discussion of the selection of two-factor or three-factor models here seems clogged and could be largely condensed and refined. Also, as mentioned in methodology comments, more details on the validity and uncertainties of the analysis should be clarified.
Response	*See response to General Comments (Methodology).* We consider the placement of a more detailed discussion on the multiple linear regression at this place in the manuscript more appropriate as the preceding discussion on the trends in both carbonaceous primary PM and traffic composition are vital in providing the context for the discussion of the MLR analysis. Validity and uncertainties from the MLR analysis are discussed in response to a subsequent comment (→ *see response to comment on Line 267-270 further below*).
Alteration	N.A.
Comment	Line 255-257. It is better to discuss the figure in the main text, i.e., Figure 3, and use the supporting figure as a supplementary discussion, otherwise, it makes the reader confused about the relationship and difference between Figure S5 and Figure 3.
Response	We have amended the wording and added a reference to Figure 3 in the said sentence to clarify the relationship between the main and supplementary figures.
Alteration	[…] The time series of measured and reconstructed HOA and EC concentrations based on the regression coefficients are depicted individually in Figure S5a and S5c in the Supporting Material and in combination in Figure 3 to represent the sum of motor vehicle related primary carbonaceous particulate compounds […]
Comment	Line 267-270, The multilinear regression is a statistical analysis that may not necessarily represent good physical meanings, it is necessary to compare the regression resolved emission factors with previous laboratory or field measured emission factors in Hong Kong or other regions, to validate the regression results.
Response	The aim of this work is the evaluation of contributions of traffic emissions to ambient concentrations in contrast to emission factor studies, which are based on the composition and concentrations from the tailpipe. Comparability to emission factors is limited due to the lack of either CO_2 concentration & fuel consumption data (*concentration per fuel consumed*), or travelling distance of vehicles (*concentration*

per mileage) as well as the influence of dispersion (*sampling at 3m of height, to the side of the road*) to determine emission factors from our measurement data.
We have demonstrated the considerations of our MLR analysis (statistical significance, traffic data, reference to previous studies) in section 3.1.1. validating its mathematical soundness.
While mathematically sound and physically meaningful solutions cannot automatically be equated, we have provided a comprehensive discussion of our results with both ambient and laboratory studies of more recent years in Section 3.2.2 (similar ambient studies, exhaust characterization studies, exhaust control devices). As they widely agree with the observations from our study - considering the specific characteristics of the vehicle mix and the sampling site environment – we are confident that our results are also physically meaningful.

Alteration N.A.

Comment Line 270-272, it is hard to understand what the author wants to interpret by only reading the text.

Response We have revised the wording for clarification.

Alteration […] Each diesel and gasoline vehicles contributed 75-85% more carbonaceous PM_1 mass than LPG vehicles, which despite making up ~30% of total counted traffic accounted for less than 13% of traffic-related primary PM_1 (Figure 3e). […]

Comment Please give a sense of the uncertainty of the obtained values, providing error bars in Figure 3b and 3e, and the uncertainties should be at the least qualitatively noted in the main text.

Response Standard deviations of the fit parameters have been included in Table A1 in the Supporting Material and we include error bars in Figure 3b (main text) and standard deviations derived from the fit parameter uncertainties (Table S1, Supporting Material) in the pie charts of Figure 3e (main text) and Figure S5b,d (Supporting Material).

Alteration

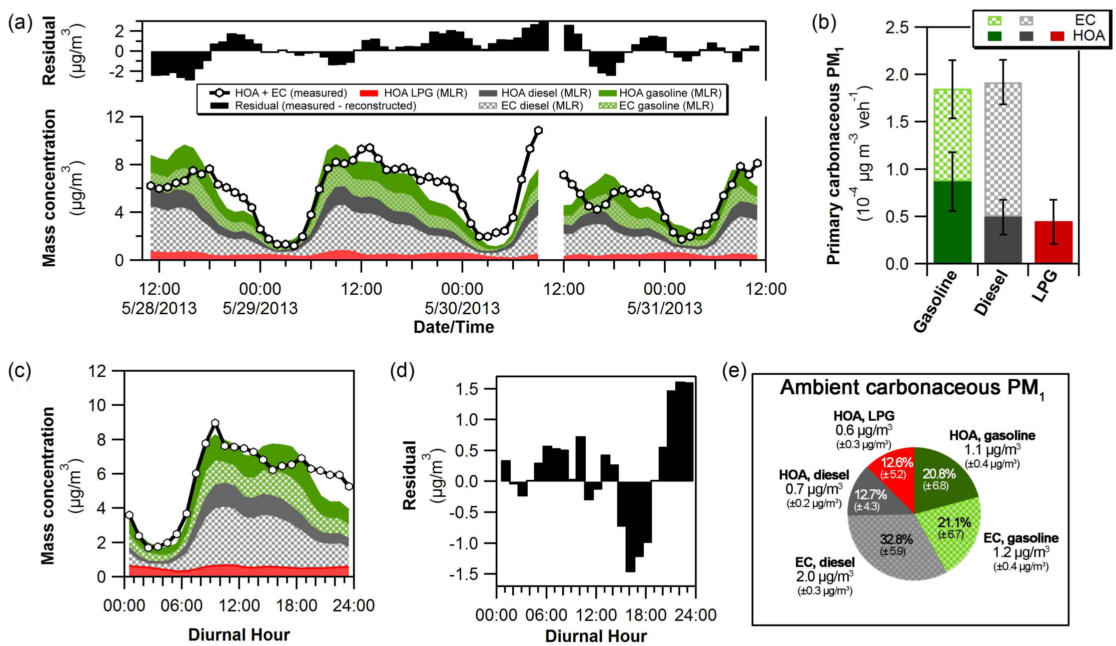

Figure 3. […]

Comment Line 290-291. It is not clear how the high fraction of EC in gasoline engine was related to the explanations here. More clarification is required to support the author's statement.

Response Lines 290-91 are to be seen in context with the following two sentences and associated references detailing how unstable engine loads (e.g. acceleration phase due to the sampling site location at a crossing / junction) lead to higher fractions of EC in gasoline vehicle exhaust compared to studies next to open (i.e. straight) roadways or in tunnels, where vehicles travel at relatively constant speed and thus at more constant engine loads. Further to this, recent studies also indicate that changes in vehicle technology (e.g. gasoline direct injection, GDI) are likely to shift tailpipe composition of gasoline vehicles more in favor of elemental carbon. We have added this discussion in the revised manuscript.

Alteration […] It has also been noted that changes in engine technology, i.e. the move from port fuel injection (PFI) to gasoline direct injection (GDI), may shift gasoline vehicle exhaust characteristics in favor of elemental carbon. Higher particulate matter mass emissions of GDI vehicles compared to PFI vehicles by a factor of 2 have been reported, which were mainly due to enhancements in EC emissions (Saliba et al., 2017). Similar observations were made in comparisons of PFI and direct injection spark ignition (DISI – a derivative of GDI) vehicles over both cold and hot-start conditions with higher total carbon (TC) emissions and higher EC/TC ratios for the DISI vehicles (Fushimi et al., 2016). […]

Comment	Line 295-300. Most of the previous studies reported higher particles for diesel vehicles compared to gasoline, is there any possibility that the different result in the present study was artifacts resulting from the statistics analysis lacking real physical meaning? Any other studies of direct emission measurement to support the similar low particles from diesel vehicles with DPF as the gasoline vehicles?
Response	We would like to point out that the reported measurements in this study are in terms of particle mass which does not imply that diesel vehicles emitted a smaller number of submicron particles as this would depend on the characteristics of the particle size distributions of each engine type. Low particle mass emissions for diesel vehicles with fitted DPFs have been reported in previous studies (Li et al., 2014;Quiros et al., 2015;Mathis et al., 2005) where emission factors were either comparable (or lower) than those from port-fuel injection (PFI) gasoline vehicles. In contrast, gasoline direct injection (GDI) vehicle often exhibited larger PM emissions than both PFI gasoline and DPF-equipped diesel vehicles, as noted previously.
Alteration	N.A.
Comment	Line 313-317. What do these numbers mean and how can be linked to the results presented above? More discussions are needed here.
Response	As noted previously, advanced exhaust after treatment has been shown to greatly reduce particulate mass emissions in diesel vehicles. The vehicle data available in this study comprised information on the year of manufacture of the vehicle, but no details on the engine specifications (except for fuel type) or built-in exhaust after treatment devices. The requirement to comply with emission standards (e.g. Euro or LEV standards) have led to strict PM emission limits in newer vehicle models and are typically achieved through such exhaust after treatment devices. The vehicle manufacture year (compared to the introduction year of the respective Euro standard) can be regarded as a predictor for the fitting with advanced exhaust after treatment devices, given that most diesel vehicles in Hong Kong are imported from Europe, the US, Japan or Korea where manufacturers comply with such respective standards. We have modified the section to further stress its relationship with the discussion of our results.
Alteration	[…]At the same time, various control schemes targeting diesel vehicle emissions have been introduced in recent years. In Hong Kong these included inter alia an incentive scheme […] […] New engine technologies for diesel vehicles, such as DPFs, have been shown to greatly reduce both EC and OC emissions (Alves et al., 2015) thus leading to overall little total particle mass emissions (Li et al., 2014;Quiros et al., 2015) partially due to shifts in the particle size distributions towards a greater fraction of particles in the ultrafine mode (Giechaskiel et al., 2012). The Euro III, IV and V standards for trucks and buses were introduced in late 2000, late 2005 and late 2008 respectively. While the year of manufacture does not directly infer compliance to a specific emission standard, an approximation of the fraction of vehicles fulfilling a certain standard can be made by assuming that vehicles produced between 2001 and 2005, between 2006 and 2008 and between 2009 and 2013 very likely comply with the Euro III, IV and V standards respectively. In this case, it is assumed that emission standards were immediately or had already been adopted in vehicle models in the corresponding year of manufacture. Figure S6 in the Supporting Material depicts the number and proportion of vehicles of certain years of manufacture and their assumed Euro standard. For goods vehicles, 52% of counted vehicles were built between 2005 and 2013 (i.e. likely fulfilling Euro IV and V), while for buses the proportion was slightly lower at 33%. With these two vehicle groups representing the bulk of diesel powered vehicles, an estimated 40% of diesel vehicles complied with Euro IV and Euro V standards during the time of our ambient measurements, further rationalizing the relatively low per-vehicle contribution of diesel vehicles to ambient exhaust-derived carbonaceous PM_1 in this study. [...]

Technical corrections:

Comment Line 68-71, grammar issues in the long sentences. Please rewrite it.

Response We could not identify any obvious grammatical mistake in the captioned sentence. However, we have split the sentence to improve its clarity in the revised manuscript.

Alteration […] Generally, low overall organic carbon (OC) to elemental carbon (EC) ratios (0.6-0.8) which are typical for locations in direct proximity to primary combustion sources were observed. In comparison, samples from urban rooftop sites exhibited lower contributions of carbonaceous constituents (<50%) and were impacted more by oxidized secondary organic species with correspondingly higher overall OC/EC ratios ~1.9 (Louie et al., 2005;Cheng et al., 2010;Lee et al., 2006). […]

Comment Line 76, Where was the open roadside located, the same site as the present study?

Response The referenced studies were conducted at different locations, which also included the site of this present study. We note that open roadside here is used in contrast to "enclosed" tunnel studies which are also discussed in the same paragraph.

Alteration N.A.

Comment Line 81, which contributions did you refer to?

Response We have revised the sentence for clarification.

Alteration […] Contributions of traffic emissions to ambient gas-phase species were evaluated utilizing VOC canister samples collected in locations with different dominant-vehicle types in Hong Kong in 2003 (Ho et al., 2013). […]

Comment Line 99-103, reference needs to be provided

Response We have moved the captioned reference accordingly.

Alteration […]A mobile measurement platform with an array of on-board PM and gas monitors was deployed in early 2012 for a more up-to-date characterization of fuel-based emission factors of PM, NO_x, and butane by chasing vehicle plumes on major roadways and highways (Ning et al., 2012) […]

Comment Line 175. 'was is' should be 'was'.

Response We have rectified this mistake in the revised manuscript.

Alteration […] 90% of the daily HOA was accumulated between the start of the rush hour and midnight.[…]

Comment Line 209, the Figure S2a citing here is not the correct figure.

Response The Figure reference has been amended in the revised manuscript.

Alteration […] The diurnal variation of the counted vehicle number, resolved by broad vehicle classes, and their varying contribution to the vehicle mix during the day are depicted in Figure B2c in the Supporting Material. […]

Comment Line 286, uncompleted sentence.

Response We have removed an erroneous "and" at the end of the captioned sentence.

Alteration […] Previous measurements of vehicle emissions in a highway road tunnel in the San Francisco Bay Area (Dallmann et al., 2014) employing a soot-particle aerosol mass spectrometer (SP-AMS) yielded a similar characteristic ratio of black carbon to organic aerosol (BC/OA~2.6) for diesel truck plumes and a smaller ratio of BC/OA<0.1 for gasoline vehicle plumes, while filter samples at the same location from light duty vehicles location exhibited BC/OA ratios of ~0.7 for light-duty vehicles and ~2.1 for medium and heavy diesel trucks (Ban-Weiss et al., 2008). […]

Changes to main text

1.1. Particulate matter from motor vehicles in urban areas

Emissions from on-road motor vehicles comprise gas-phase species (CO, CO_2, NO_x, SO_2) and volatile organic compounds among them important precursors of secondary organic aerosol (SOA), as well primary particulate matter predominantly in the fine ($PM_{2.5}$) and ultrafine particle size range ($PM_{0.1}$) mostly as carbonaceous species which encompass primary organic aerosol (POA) components and elemental carbon (Giechaskiel et al., 2014). Increased acute occurrence and risk of chronic development of cardiovascular and pulmonary diseases are important epidemiological effects of particulate matter inhalation (Davidson et al., 2005;Pope, 2007;Valavanidis et al., 2008;World Health Organization, 2011). In urban areas with high population and building densities, proximity to vehicle emissions poses a significant public health risk and renders paramount importance to the characterization and quantification of vehicle emissions (Kumar et al., 2014;Uherek et al., 2010).
Their contribution to total ambient concentrations however remains elusive with considerable variability in measured emission rates and species composition between as well as within vehicle classes from exhaust measurements in laboratory settings, vehicle chase or portable emission measurement systems (PEMS) studies (Franco et al., 2013;Kwak et al., 2014;Karjalainen et al., 2014;Giechaskiel et al., 2014;Alves et al., 2015) . These differences have been attributed to numerous influencing factors such as vehicle age, fuel use, operational parameters, environmental

conditions and the subsequent introduction of more advanced engine technology (e.g. gasoline direct injection / GDI) and exhaust after-treatment (e.g. diesel particulate filters / DPF) (May et al., 2014). Apart from combustion-related compounds, species originating from lubricating oils have been identified as major components in primary gasoline and diesel exhaust and may be responsible for more than half of the total in-use POA emissions (Worton et al., 2014).

Post-emission oxidation of gas-phase organic constituents leads to atmospheric formation of secondary organic aerosol. Recent renewed interest in the SOA forming potential of motor vehicle exhaust through ambient observations, oxidation chamber and flow reactor studies (Ensberg et al., 2014;Gentner et al., 2012;Gentner et al., 2017;Platt et al., 2013;Zhao et al., 2015) affirms its potency in contributing significantly to urban SOA burdens however with still considerable quantitative uncertainties (Gentner et al., 2017).

Investigations on the impacts of motor vehicle emissions on ambient air pollution and related effects such as health implications (Kheirbek et al., 2016;Zhang and Batterman, 2013;Levy et al., 2010) typically rely on combinations of source inventories with dispersion modelling and combined physical-chemical models with significant uncertainties arising from the complexity of the urban built environment (Kumar et al., 2011;Belcher et al., 2013), inventory currency and accuracy of emission factors (Simon et al., 2008;Fuzzi et al., 2015). The latter is critically affected by the design of dynamometer test cycles with discrepancies having been noted between type approval procedure derived emissions and on-road measured driving emissions, which underlines the continued need to evaluate and monitor contributions of traffic emissions to urban air pollution by direct ambient measurements (Harrison and Beddows, 2017). A dominant factor that affects both gas- and particle-phase species distribution is the engine type used (Alves et al., 2015;Franco et al., 2013;Jang et al., 2016;Kam et al., 2012;Karjalainen et al., 2014;Kwak et al., 2014), i.e. the distribution of engine types in the traffic mix will have significant influence on ambient concentrations. In the US gasoline fuel use in light duty vehicles is wide spread contributing to a greater dominance of gasoline-related VOC and particle-phase organic emissions, whereas in Europe diesel-vehicle use prevails leading to higher ambient burdens of black carbon (BC) (Zotter et al., 2014;Gentner et al., 2017). Evaluating the contribution of different vehicle groups to exhaust related species in ambient measurements is therefore of vital interest to gauge their current importance to urban air pollution and to acquire a baseline for assessing the efficacy of control policies and future effects of traffic mix and vehicle technology changes.

In this study, we evaluated the contributions of the three predominant engine-type vehicle groups (gasoline, diesel, LPG) in Hong Kong to primary carbonaceous aerosol by combining time-resolved ambient measurements by aerosol mass spectrometry (AMS) and ECOC analysis with vehicle count data. Characterization studies on road traffic emissions in Hong Kong are sparse due to the complexity of the urban built environment and the encountered transient engine loads which make emission factor and dispersion modeling approaches difficult to implement. Measurements were undertaken within a street canyon at a typical inner-city location where urban driving patterns with frequent stop-and-go traffic are prevalent, which may not be adequately reflected in dynamometer or cruising speed chase studies, but are more representative of pedestrian exposure levels particularly in view of growing concerns about exposure to air pollutants and their public health impacts in densely populated and built-up environments.

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