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 reports measurements of carbonaceous particles at a busy street canyon. The authors focus on primary hydrocarbon like (HOA) and elemental carbon (EC) fraction of the aerosol. They combine the concentration data with engine-type specific vehicle counts to obtain the contribution of diesel, gasoline, and LPG vehicles. The changes in emission regulations and the rapid development in engine and aftertreatment techniques make efforts such as this necessary in evaluating the effect of the policies and development on the air quality. The measurement site is ideal for this kind of study because of the high contribution of traffic and the local measures taken for decreasing the traffic emissions. The paper brings new information on an important question and merits publication, but there are some issues that should be treasted, as discussed below.

I understand that there are so many aspect on carbonaceous aerosols that the discussion needs to be limited somehow. However, I find the approach especially in the introduction too limited to the Hong Kong primary aerosol case. The introduction should shortly discuss the relative importance of the primary and secondary aerosol in urban settings and not just mention the secondary aspect in the last sentence of the paper. The authors should make a case why it is still important to study the primary components. In presenting and discussing the results, the authors should clearly, preferably in a table, report the primary and overall concentrations. The effect of renewing fuels and vehicle fleet on the urban concentrations has been studied also outside Hong Kong. These studies should be discussed, starting with the Harrison and Beddows, Nature 2017 paper. There are recent papers on the same measurement cite, partly by the same authors. The authors should clearly state what is new here, especially compared to the Lee at al. 2015 JGRA and Huang et al ACP 2014 papers. There is an important methodological difference to the Huang paper in treating the traffic related OC. The difference in methodology and results, as well as the meaning of those should be discussed more explicitly. Finally, the methodology and the analyses appear solid but they are treated so concisely that the reader needs to deduce what data was used. The methods are partly the same as used in the papers mentioned above, but this paper should also be readable separately. As this media allows some volume, the methodology should be described in more detail.

We have substantially revised the introductory part (→1) to reflect a more general discussion on the relevance and impacts of traffic emissions on primary and secondary aerosol pollution in urban environments. Due to the extent of the changes, the introductory section in its entirety is appended to the bottom of this document.

A clarifying sentence (→2) has been added to describe the differences to previous work.

The comparison of the AMS measurements to those using the EC/OC method has been extended (→ 3).

The methodology section has been revised to include all data analysis and processing steps that are relevant to the discussion of this work (→ 4) and an additional table is provided to give an overview of measured PM_1 and $PM_{2.5}$ mass concentrations and the traffic-related components as reported in this study (→ 5).

1 […]

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

1 […]

2 […]

We aim to complement previous studies which only estimated overall traffic-related aerosol contributions from online and offline ECOC measurements at different time resolutions (from hourly to 24h samples) (Louie et al., 2005;Huang et al., 2014) and online mass spectrometric methods (Sun et al., 2016;Lee et al., 2015) at the same location by combining resolved traffic-related organic aerosol concentrations from factor analysis of aerosol mass spectrometer (AMS) data, online EC measurements and traffic count data to gain an improved understanding of the influence of traffic composition on traffic-related submicron carbonaceous aerosol.

2 […]

3 […]

Contributions of vehicle emissions to ambient organic aerosol have been derived from one-year long measurements at the same site based on analysis of semi-continuous ECOC measurements (Huang et al., 2014). The EC tracer method (Turpin and Huntzicker, 1991;Lim and Turpin, 2002) was employed which evaluates minimum OC/EC ratios, and OC and EC measurements during time periods of high primary source strength and low photochemical activities when OC is mainly of local origin and attributable to the dominant primary sources in the vicinity. A

characteristic vehicle-related OC/EC value ($OC/EC_{vehicle}$) of 0.5 was derived from the 5% lowest summer OC/EC ratio data to approximate organic vehicular emissions. The reported monthly average primary organic matter concentrations from traffic in $PM_{2.5}$ (using an OM/OC conversion factor of 1.4) ranged between 2.7 – 3.4 µg m^{-3} with little seasonal variation. To compare to our measurement base of PM_1, we use the previously mentioned average ratio of $PM_1/PM_{2.5}$ of 0.8 for conversion. The OC/EC approach would thus yield an approximate 2.2 – 2.7 µg m^{-3} of traffic-related primary organics in PM_1 which compares well with the overall average concentration of traffic-related HOA (2.8 µg m^{-3}) covering the entire AMS measurement campaign (March – July 2013). However, we observed significantly different concentrations depending on the season with much higher concentrations (3.5 µg m^{-3}) in spring than in summer (2.0 µg m^{-3}). The lack of a clear seasonal variation in OC/EC derived traffic-related organics is due to the use of a fixed conversion factor directly dependent on ambient EC concentration which did not exhibit a strong seasonal behavior and thus could not capture the same trends observed from the AMS measurements. The derivation of the $OC/EC_{vehicle}$ ratio is subject to further uncertainty due to the substantial influence of cooking-related organics at the site (Lee et al., 2015;Sun et al., 2016), i.e. traffic emissions may not be the dominant source of "local" organics at the site, as well as the influence of background SOA which is likely to persist even in time periods of low OC/EC ratios.

Nonetheless, the agreement of the whole-campaign average HOA concentrations (this study) and $OC/EC_{vehicle}$ derived traffic organics mass concentration (Huang et al., 2014) suggests that the long-term average primary organic aerosol contribution from vehicles is reasonably well-approximated with the OC/EC approach but may lead to over- and underestimation of actual organic concentrations in shorter time frames, e.g. different seasons and different times of the day.

3[…]

4 […]

Elemental and organic carbon (EC, OC) were measured (Semi-continuous thermo-optical ECOC analyzer RT-3131, Sunset Inc., USA) at the AQMS at 1h time resolution and at similar sampling height (~2m) employing a modified National Institute for Occupational Safety and Health (NIOSH) method 5040 protocol which has been described in a previous study (Huang et al., 2014).

AMS data were treated according to general AMS data treatment principles (DeCarlo et al., 2006;Jimenez et al., 2003) with standard software packages (SQUIRREL v1.53G, PIKA v1.12G). Analysis of the unit-mass resolution mass spectra yielded non-refractory submicron particle species concentrations of major inorganic constituents (SO4, NO3, NH4, Chl) and total organics at a base time resolution of 10 min. Positive Matrix Factorization (PMF) was used to deconvolute high-resolution organic mass spectra acquired at 10 min time resolution following recommended PMF guidelines for AMS data (Zhang et al., 2011) with the AMS PMF analysis toolkit (Ulbrich et al., 2009). Six organic aerosol (OA) factors were identified encompassing three secondary organic aerosol (SOA) and three primary organic aerosol (POA) factors of which one was attributed to traffic emissions and two to cooking activities. OA factor concentrations were established using the fractional factor contributions from the high-resolution PMF analysis and the total submicron organics concentrations from the unit-mass resolution analysis. Both unit- and high-resolution mass spectral derived data were averaged to 1h time resolution to match ECOC measurements and traffic count data (see below). The latter were assessed from 28 May to 31 May, 2013 and thus data presented in this study mainly cover said time period. Average species concentrations measured in this time as well as seasonal averages (spring, summer) from the whole measurement campaign are summarized in Table 1.

Further specific details on instrument parameters, data analysis and an overview of the general characteristics of submicron particulate matter from the same measurement campaign can be found in a previous publication (Lee et al., 2015) .

4 […]

5 […]

Table 1. Species concentrations from AMS and total $PM_{2.5}$ mass from TEOM (a), and EC, OC and organic aerosol constituent concentrations from AMS PMF analysis (b); average concentrations during the traffic counting period (May 28-31, 2013) and seasonal averages (Spring: Mar – May 2013, Summer: May – Jul 2013 (Lee et al., 2015)) at the Mong Kok site

(a)

µg/m³ Mean ± *SD*	**Org** (AMS)	**SO4** (AMS)	**NO3** (AMS)	**NH4** (AMS)	**Chl** (AMS)	**NR-PM_1** (AMS)	**$PM_{2.5}$** (TEOM)
May 28-31	5.9±3.0	4.8±1.2	0.3± 0.1	1.5±0.3	0.1±0.1	12.5±3.7	16.0±5.3
Spring	12.8±7.6	7.0±3.8	2.5±2.1	2.5±1.5	0.4±0.3	25.3±13.1	32.3±12.4
Summer	7.9±5.4	3.4±2.0	0.4±0.4	1.1±0.5	0.1±0.1	12.7±3.6	17.3±7.5

(b)

µg/m³ Mean ± *SD*	**EC** ($PM_{2.5}$)	**OC** ($PM_{2.5}$)	**EC** (PM_1)	**OC** (PM_1)	**HOA** (PM_1)	**COA**[a] (PM_1)	**SOA**[b] (PM_1)
May 28-31	4.2±2.3	3.0±1.2	3.2±1.7	2.3±0.9	2.4±1.4	2.0±1.2	1.6±1.1
Spring	4.3±2.6	7.6±3.9	3.2±1.9	5.7±2.9	3.5±2.4	4.4±4.3	4.9±3.4
Summer	4.3±2.5	4.1±2.1	3.2±1.8	3.0±1.6	2.0±1.3	3.6±3.4	2.2±2.2

Notes: [a] Sum of two cooking-related PMF factors
[b] Sum of three SOA-related PMF factors

5 […]

Specific Comments

Abstract:

Comment The abstract should highlight the new findings of this paper and report them quantitatively.

Response The abstract has been modified to include more of our quantitative results.

Alteration […]On an average per-vehicle-basis, gasoline vehicles emitted 75% and 93% more organics than diesel and LPG vehicles respectively, while EC emissions from diesel vehicles were 45% higher than those from gasoline vehicles. LPG vehicles showed no appreciable contributions to EC and thus overall represented a small contributor to traffic-related primary ambient PM_1 despite their high abundance in the traffic mix (~30%). Total carbonaceous particle mass contributions to ambient PM_1 from diesel engines were only marginally higher (~4%) than those from gasoline engines, which is likely an effect of recently introduced control strategies targeted at commercial vehicles and buses. Overall, gasoline vehicles contributed 1.2 µg/m³ of EC and 1.1 µg/m³ of organics, LPG vehicles 0.6 µg/m³ of organics and diesel vehicles 2.0 µg/m³ of EC and 0.7 µg/m³ of organics to ambient carbonaceous PM_1. […]

Methodology:

Comment The methodology part should be rewritten to work on its own for this paper. Reading just this paper, it remains unclear what instrument data was used for, e.g., the engine specific contributions. Was this done on hourly basis as is indirectly indicated on the vehicle flow chapter? Obviously AMS data was used for the HOA, but what about EC: Aethalometer or the hourly EC measurement? And how did the latter two compare?
Huang et al., 2014 should be cited already in the methodology part for the hourly EC/OC. An obvious measurement missing from the study is CO2. This inhibits the calculation of emission factors and should be clearly stated.

Response We have revised the methodology part to include more information on the data acquisition and treatment which are relevant to this work. As the reviewer correctly points out, CO_2 measurements were indeed not available thus preventing emission factor calculations, and we include a statement

to emphasize this circumstance. We provide an estimated per-vehicle contribution for each engine-type category in Figure 3b – while this was previously based on the vehicle numbers from the counting days we have scaled the contributions by the expected total daily vehicle number (average annual daily traffic – AADT) in the revised manuscript.
We mention the aethalometer in the methodology as it was integral to the instrumental setup used during the campaign, but was not operational continuously. There was significant overlap between BC and EC measurements in later parts of the sampling campaign (June-July) with good temporal agreement between the ECOC analyzer and the aethalometer (see below). We note a significant positive bias in the BC mass measurements (intercept of 1.7 $\mu g/m^3$) which may be caused by filter loading effects (Virkkula et al., 2007) or the presence of other light absorbing species, e.g. brown carbon (BrC) (Olson et al., 2015;Lin et al., 2015;Zhang et al., 2013). Due to these uncertainties and the lack of BC measurement data during the captioned time period covered in this work (late May 2013), we only discuss the hourly EC measurements.

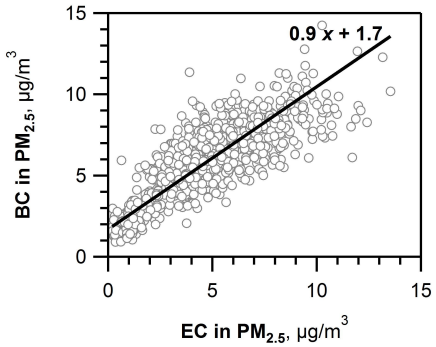

Figure R1. Scatter plot of hourly BC (aethalometer) and hourly EC (ECOC analyzer) measurements (*open circles*) with orthogonal distance regression (*solid line*) between June 1 and July 19, 2013 at the Mong Kok site

Alteration

[…] Elemental and organic carbon (EC, OC) were measured (semi-continuous thermo-optical ECOC analyzer RT-3131, Sunset Inc., USA) at the AQMS at 1h time resolution and at similar sampling height (~2m) employing a modified National Institute for Occupational Safety and Health (NIOSH) method 5040 protocol which has been described in a previous study (Huang et al., 2014). Concentrations of EC and OC in PM_1 were estimated by converting the raw $PM_{2.5}$ concentrations using a PM_1 to $PM_{2.5}$ ratio of 0.8 representing the overall average of PM_1 to $PM_{2.5}$ ratios from previous roadside sampling studies (0.75-0.85) in Hong Kong (Cheng et al., 2006;Lee et al., 2006).
AMS data were treated according to general AMS data treatment principles (DeCarlo et al., 2006;Jimenez et al., 2003) with standard software packages (SQUIRREL v1.53G, PIKA v1.12G). Analysis of the unit-mass resolution mass spectra yielded non-refractory submicron particle species concentrations of major inorganic constituents (SO4, NO3, NH4, Chl) and total organics at a base time resolution of 10 min. Positive Matrix Factorization (PMF) was used to deconvolute high-resolution organic mass spectra acquired at 10 min time resolution following recommended PMF guidelines for AMS data (Zhang et al., 2011) with the AMS PMF analysis toolkit (Ulbrich et al., 2009). Six organic aerosol (OA) factors were identified encompassing three secondary organic aerosol (SOA) and three primary organic aerosol (POA) factors of which one was attributed to traffic emissions (hydrocarbon-like organic aerosol, HOA(Zhang et al., 2005)) and two to cooking activities. OA factor concentrations were established using the fractional factor contributions from the high-resolution PMF analysis and the total submicron organics concentrations from the unit-mass resolution analysis. Further specific details on instrument parameters, data analysis and an overview of the general characteristics of submicron particulate matter from the same measurement campaign can be found in a previous publication (Lee et al., 2015) and are not directly relevant to this work. […]

[…] Vehicle emission factors are typically based on the concentration of emitted species per unit of consumed fuel (derived from CO_2 measurements) or per driven distance. As concurrent CO_2 measurement data were not available, emission factors were not evaluated in this study. Instead, the

relative emission behavior of different vehicle groups and their impacts on ambient PM concentrations were approximated by normalizing the resolved engine-specific carbonaceous primary aerosol concentrations by the total number of vehicles in each category (Figure 3b). […]

Comment Vehicle identification method to gain engine-type specific emissions seems to have been used at least in tunnels, although not necessarily documented very well. The authors should cite those, such as Cui et al., STOTEN 2016. They might also discuss the individual-vehicle specific approach of Wang et al., AtmEnv 2016.

Response We have included said references in the revised manuscript.

Alteration […] Details on traffic composition are usually obtained by concurrent video image recordings and subsequent manual assessment of vehicle numbers and identification of vehicle types with classifications e.g. by vehicle function (Wang et al., 2016) or broader groups such as fuel types (Cui et al., 2016). In this work, vehicle data were obtained from the Hong Kong government, which conducted a counting exercise during the sampling period over three weekdays (noon 28 May 2013 to noon 31 May 2013, Tue to Fri) using automated license plate recognition (ALPR) with four infrared cameras on-site. […] Non-personalized data on registered vehicles were obtained, including license class (*vehicle type*), year of manufacture (*vehicle age*) and engine type (*gasoline*, *diesel*, *LPG*, *others*), thus providing a direct linkage to actual vehicle inventory data and circumventing the need for manual assessments. […]

Results:

Comment The paper reports the contributions of the vehicle types, but also PM2.5, PM1, EC and OC concentrations should be given, maybe in a table gathering the results. Those reported earlier with a citation. At least Zhang et al., ACP 2005 should be cited for using HOA as a surrogate for combustion POA. In discussing the seasonal variations and gaseous/particulate phase partitioning, Robinson et al., 2007 should be cited, maybe together with comparison to some other studies, such as Budisulistrioni et al., ACP 2016; Huffmann et al., ACP 2009.
The paper reports exceptionally high EC fractions (EC/HOA ratios) for gasoline vehicles. This is an interesting and potentially important finding, but also subject to controversy. While the site-specific driving patterns could explain some of this, other aspects should also be discussed. A high fraction of gasoline direct injection (GDI, DISI) vehicles could possibly affect this, as they have been found to exhibit high primary EC fractions (e.g. Karjalainen et al., ACP 2016; Fushini et al., AtmEnv 2016). On the other hand, the HOA concentration of this study comes from AMS, while the EC does not. The low sampling efficiency of the AMS for sub-50 nm particles could cause low HOA concentration as a measurement artefact, especially if a high mass fraction of the POA is within the nucleation mode.

Response A table has been included *(see reply to general comment above: →5)*.
We have also added the suggested references in the revised manuscript. We further mention the possibility of GDI engines contributing to the high observed EC fraction. The vehicle data available in this study however are not specific enough to deduce the proportion of GDI vehicles among gasoline vehicles in this study.
Another paragraph has been added to discuss the portioning/semi-volatility of HOA and related studies (see below).
We also further stress the limitations of the AMS inlet in sampling nucleation mode particles. While this is relevant in terms of particle number concentration, the mass fraction of nucleation mode particles is low and we therefore believe that AMS inlet losses should have limited effects on the measured mass concentrations.

Alteration […] The semi-volatility of HOA in this study is inferred from the observed trend in ambient mass concentrations and affirms more recent studies on the partitioning behavior of POA components. Evaporation of semi-volatile components upon dilution decrease ambient POA concentrations considerably and are expected to vary with temperature (Robinson et al., 2007) as is the case in this study. Measurements of thermally denuded particles of primary origin by TD-AMS indicate that POA components can exhibit properties of semi-volatility on a scale similar to SOA in urban environments (Huffman et al., 2009a;Huffman et al., 2009b), and more recent studies suggest that HOA possesses a relatively wide volatility distribution with up to 50-60% of its mass consisting of semi volatile organic compounds (Cappa and Jimenez, 2010;Paciga et al., 2016). As noted earlier,

in this study the seasonal decrease in submicron particle-phase organic mass concentrations (HOA) by 40% was associated with a substantial increase in mean temperature by 7°C between spring (23°C ± 3°C) and summer (30°C ± 2°C). […]

[…] Dynamometer studies mostly reported EC/OC values for gasoline powered vehicles < 1 (Alves et al., 2015;Geller et al., 2006) indicating overall a greater importance of organics in particulate emissions. The relatively high fraction of EC in gasoline related PM in our work likely stems from a combination of factors. HOA mass concentrations are derived from AMS measurements which are subject to limitations of inlet lens transmission, i.e. a standard lens as used in this study is expected to transmit efficiently (~100%) between 90nm and 700nm D_{va} (Williams et al., 2013), and thus may cause a low bias in measured ambient HOA concentrations. However, these inlet losses are expected to only have limited effects on total measured submicron particle mass concentrations as mass- and volume-based particle concentrations from vehicle exhaust typically peak in the region of ~ 100 – 300nm (Fushimi et al., 2016;Ban-Weiss et al., 2010). The observed seasonal variability of particle-phase organic concentrations due to partitioning of semi-volatile components into the gas-phase would also decrease particle-phase HOA concentrations in warmer seasons, thus elevating the observed ambient EC/HOA ratio with our measurements taking place in summer. While this would affect all engine types, the extent may vary with the distribution of semi-volatile species in the respective tailpipe emissions of different vehicle groups. Due to the characteristics of the sampling site (inner-city urban road traffic and proximity to both a road junction and pedestrian crossing) sampled emissions include a considerable fraction of variable and higher engine loads during the acceleration phase whereas road tunnel environments are characterized by largely constant engine loads and traveling speeds. As gasoline vehicles are typically not equipped with particle filters, they have been shown to emit significant amounts of EC under unstable engine loads (Karjalainen et al., 2014). 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). At the same time, various control schemes targeting diesel vehicle emissions have been introduced in recent years. In Hong Kong these included […]

Details:

Comment In section 3.3.2. it would be good to explicitly state that the reconstructed mass does not include SOA. The Environmental Protection Department (?) is differently named in the reference list. Kirchtetter et al. is misplaced in the alphabetical reference list.

Response We have included a statement on the reconstructed mass as recommended. The reference list has been corrected as well.

Alteration […] The time series of measured and reconstructed HOA and EC concentrations are depicted in Figure S5a and S5c in the Supporting Material respectively, representing the sum of primary carbonaceous particulate compounds and not including SOA species that may be formed through subsequent atmospheric processing of gas- and particle-phase species. […]

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;WHO, 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 currentness 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 significant 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 over time the 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 different 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. Measurements were undertaken within a street canyon at a typical inner-city location where urban driving patterns with transient engine loads and 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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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

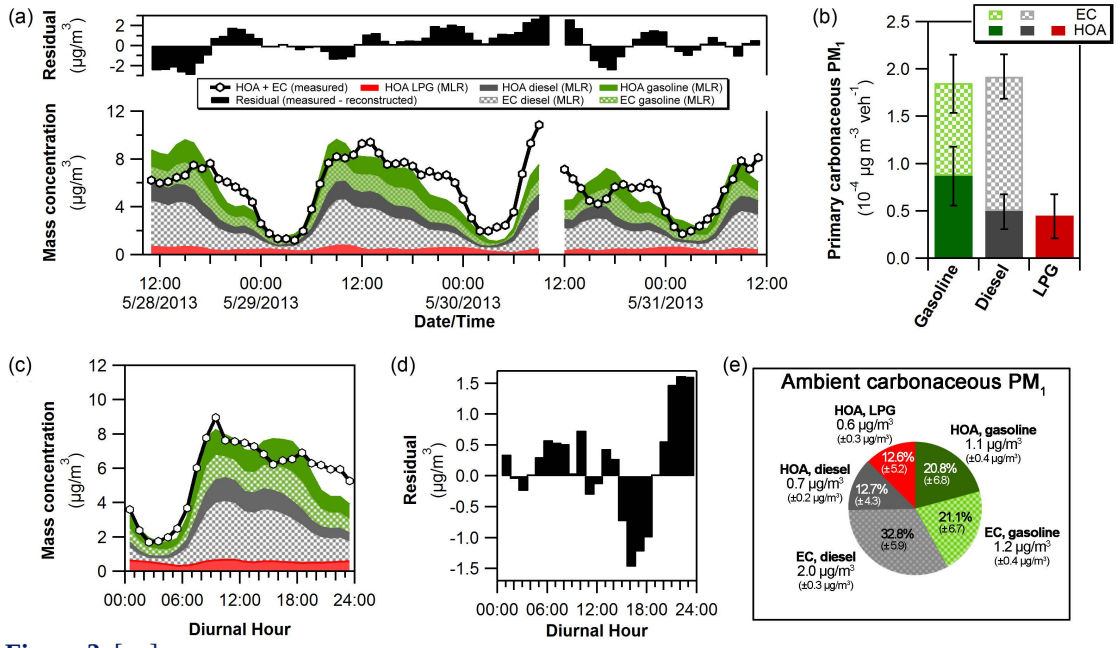

Figure 3. […]

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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Evaluation of traffic exhaust contributions to ambient carbonaceous submicron particulate matter in an urban roadside environment in Hong Kong

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Abstract. Road traffic has significant impacts on air quality particularly in densely urbanized and populated areas where vehicle emissions are a major local source of ambient particulate matter. Engine type (i.e. fuel use) significantly impacts the chemical characteristics of tailpipe emission and thus the distribution of engine types in traffic impacts measured ambient concentrations. This study provides an estimation of the contribution of vehicles powered by different fuels (gasoline, diesel, LPG) to carbonaceous submicron aerosol (PM_1) based on ambient aerosol mass spectrometer (AMS) and elemental carbon (EC) measurements and vehicle count data in an urban inner city environment in Hong Kong with the aim to gauge the importance of different engine types to particulate matter burdens in a typical urban street canyon. On an average per-vehicle-basis, gasoline vehicles emitted 75% and 93% more organics than diesel and LPG vehicles respectively, while EC emissions from diesel vehicles were 45% higher than those from gasoline vehicles. LPG vehicles showed no appreciable contributions to EC and thus overall represented a small contributor to traffic-related primary ambient PM_1 despite their high abundance (~30%) in the traffic mix. Total carbonaceous particle mass contributions to ambient PM_1 from diesel engines were only marginally higher (~4%) than those from gasoline engines, which is likely an effect of recently introduced control strategies targeted at commercial vehicles and buses. Overall, gasoline vehicles contributed 1.2 $\mu g/m^3$ of EC and 1.1 $\mu g/m^3$ of organics, LPG vehicles 0.6 $\mu g/m^3$ of organics and diesel vehicles 2.0 $\mu g/m^3$ of EC and 0.7 $\mu g/m^3$ of organics to ambient carbonaceous PM_1.

1. Introduction

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.,

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1.2. Background of vehicle emission studies in Hong Kong

Despite a low vehicle-to-population ratio (<10%), private and public road transport of passengers and goods is extensive throughout Hong Kong. In the year of this study (2013) 680,914 vehicles were registered in the Hong Kong Special Administrative Region (HKSAR) which accounted for an average of 35.17 million vehicle kilometers per day across all modes of road transport such as heavy goods haulage, commercial transport, private and public transportation. Passenger journeys on Hong Kong's three major franchised bus companies in 2013 were in excess of 1.1 billion (Transport Department, 2014). Road traffic has been recognized as a major local emission source and subjected to various regulatory efforts over the years, such as a large-scale fuel switch of taxis to a virtually all-LPG fleet, partial conversion of minibuses to LPG engines, ex-gratia incentive schemes for vehicle replacement or control device retrofitting and increased stringency in new-vehicle imports (Environmental Protection Department, 2013;Lyu et al., 2017;Ning et al., 2012).

Characterization studies on particle-phase species associated with traffic emissions in Hong Kong have mainly focused on semi-continuous online and offline filter analysis with carbonaceous components (sum of elemental carbon and organic matter) typically constituting the bulk (45-70%) of particulate mass in $PM_{2.5}$ at roadside sampling sites and up to 82% in road tunnel environments (Cheng et al., 2010;Huang et al., 2014;Lee et al., 2006;Louie et al., 2005;Research Centre of Environmental Technology and Management, 2005). Among major primary and secondary aerosol sources of ambient particulate matter which were identified from speciated long-term filter samples (1998-2008) of $PM_{2.5}$ by source apportionment analysis utilizing

positive matrix factorization, vehicle emission exhibited substantial contributions: at general measurement sites (urban, rooftop), vehicle emissions accounted for 8-9 μg m^{-3} equivalent to 11-25% of total $PM_{2.5}$ with lower fractions in the fall and winter seasons primarily due to the masking influence of regional and long-range transport. Higher fractions (22-47%) and concentrations (19-23 μg m^{-3}) were resolved from roadside samples (street level) and with seasonal variations similar to those at the general sites attributed to similar effects of non-local pollution sources (Yuan et al., 2013).

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). Impacts of long-range transport of continental aerosol were evident in both higher total PM and higher OC concentrations in the winter months. By contrast, seasonal variations of EC concentrations were weak. Higher EC concentrations were typically associated with rush hour traffic and confirm the local nature of EC and its relation to mainly on-road traffic (Lee et al., 2006;Cheng et al., 2010;Huang et al., 2014;Louie et al., 2005). While earlier studies (*samples dated pre-2005*) reported EC concentrations in the range of 11-25 μg C m^{-3} in $PM_{2.5}$ at the open roadside (Louie et al., 2005;Cheng et al., 2010;Lee et al., 2006), more recent measurements (*2012-2013*) indicate a significant reduction in overall roadside EC burdens with concentrations ~ 5 μg C m^{-3} (Huang et al., 2014;Sun et al., 2016;Lee et al., 2015). Vehicle-related OC and organic matter (OM) concentrations at a roadside measurement site were estimated to ~2 μg C m^{-3} and ~7 μg m^{-3} respectively in 2012, accounting for one-third of total OC in $PM_{2.5}$ and one-quarter of total $PM_{2.5}$ (Huang et al., 2014).

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). Propane, n- and i-butane accounted for over 80% of identified species in the LPG vehicle dominated samples, while toluene was the most abundant species in the gasoline vehicle dominated samples. Ethene and ethyne were enriched in diesel exhaust dominated samples. Contributions of vehicle-related VOCs to ozone formation were evaluated from the maximum incremental reactivity, with toluene (15-17%) and propene (7-8%) accounting for the largest shares (Ho et al., 2013).

Vehicle emission factors were estimated by a mass-balance model with differential measurements near the entrance and exit of a tunnel bore in 2003 and manual vehicle counts. Diesel vehicles were found to contribute substantially more to NO_x, $PM_{2.5}$, and PAH than gasoline vehicles which dominated CO emissions. Diesel and LPG-fueled vehicles dominated total measured VOCs and total measured carbonyl compounds. EC emission factors of diesel vehicles (131 mg veh^{-1} km^{-1}) were ~40 times higher than those of non-diesel (i.e. gasoline and LPG-fueled) vehicles (3.2 mg veh^{-1} km^{-1}). For OC, the difference amounted to a smaller factor of ~8 with emission factors of 67.9 mg veh^{-1} km^{-1} and 8.5 mg veh^{-1} km^{-1} for diesel and non-diesel vehicles respectively. In terms of fractional contribution, OC made up 51% of $PM_{2.5}$ in non-diesel emissions, while EC made up 51% of $PM_{2.5}$ in diesel emissions. Contributions of inorganic species including ions and trace metals in traffic emissions were minor amounting to only ~10% (Cheng et al., 2010;Research Centre of Environmental Technology and Management, 2005). While these emission factors compared fairly well with those from similar studies undertaken in the 1990s (Gertler et al.,

2001;Lowenthal et al., 1994;Kirchstetter et al., 1999) in other parts of the world, they are unlikely to reflect the properties of contemporary mobile road traffic emissions in Hong Kong with substantial changes in vehicle fleet composition and the emergence of more advanced engine and emission control technologies over the last 15 years.

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). The measurements focused on the evaluation of heavy duty diesel trucks (HDDV), franchised diesel buses (DB) and LPG fueled light buses (LB) with plumes sampled at cruise speed to mitigate biases from engine load-dependent emission variability. HDDVs were the highest emitters of BC (1.6 g kg^{-1} of fuel) followed by diesel buses (1.1 g kg^{-1} of fuel) and LPG light buses (0.1 g kg^{-1} of fuel). Analogously, average particle number emission factors were 3.1 x 10^{15} kg^{-1} of fuel, 3.1 x 10^{15} kg^{-1} of fuel and 0.9 x 10^{15} kg^{-1} of fuel for HDDVs, diesel buses and LPG light buses respectively.

On-road remote sensing measurements of gas-phase emissions showed that gasoline vehicles had higher CO and HC emissions and lower NO emissions compared to diesel vehicles, while LPG vehicles overall emitted more gaseous pollutants than their light-duty petrol and diesel powered vehicle counterparts. (Lau et al., 2012). The higher LPG vehicle emission factors were attributed to the higher usage rate of mostly LPG-powered taxis and light buses combined with insufficient vehicle maintenance, imploring a gap between tightened regulations and effectiveness in the reduction of on-road emissions. Overall, lower emission factors in newer vehicles within the same engine category were attributed to the efficacy of fitted emission control technologies.

Despite road traffic representing a major local source of ambient particulate matter in Hong Kong, characterization studies on vehicle emissions in Hong Kong remain sparse. While most previous studies evaluated traffic exhaust contributions in confined environments (tunnels), focal locations (e.g. bus terminus, taxi stand) or by plume chase, this work provides a quantitative estimation of vehicle exhaust related carbonaceous aerosol contributions and their relationship to traffic flow characteristics from ambient measurements in a more complex urban environment. We aim to complement previous studies which only estimated overall traffic-related aerosol contributions from online and offline ECOC measurements at different time resolutions (from hourly to 24h samples) (Louie et al., 2005;Huang et al., 2014) and online mass spectrometric methods (Sun et al., 2016;Lee et al., 2015) at the same location by combining resolved traffic-related organic aerosol concentrations from factor analysis of aerosol mass spectrometer (AMS) data, online EC measurements and traffic count data to gain an improved understanding of the influence of traffic composition on traffic-related submicron carbonaceous aerosol.

2. Methodology

Sampling of ambient particulate matter took place in spring 2013 (7 March 2013 to 15 May 2013) and summer 2013 (16 May 2013 to 19 July 2013). A ground shelter housing a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS, Aerodyne Inc.), a fast mobility particle sizer (FMPS, TSI Inc.) and an aethalometer (Magee Scientific Inc.) was located on a pedestrian island in the center of Lai Chi Kok Road, a bidirectional urban main road branching off the north-south Nathan

Road corridor at the south-east end of the sampling site. Located in the Mong Kok (MK) district in the densely populated and built-up Kowloon peninsula, the measurement site represents an inner-city environment typical for the central city areas of Hong Kong with dense and relatively slow moving traffic and tall bordering buildings. The sampling inlet on top of the shelter at a height of ~3m was equipped with a $PM_{2.5}$ cyclone (flow rate: 16.67 L min^{-1}). A four-way sampling port supplied the HR-ToF-AMS at a flow rate of 0.08 L min^{-1} while the remaining sampling volume was distributed among co-sampling instruments and an auxiliary pump. The AMS sampling stream passed through a diffusion drier (BMI, San Francisco CA, USA, length 1m, silica gel as drying agent) for the removal of gas- and particle-phase water prior to entering the instrument.

The Environmental Protection Department (EPD) of the HKSAR Government operates an air quality monitoring site (AQMS) on the same road divider island and provided additional data, such as meteorological data (wind, temperature, relative humidity, solar irradiation), concentrations of volatile organic compounds (VOCs), as well as standard trace gases such as NO_x, SO_2, and O_3, Details on employed instrument model and sampling methodologies are available in other dedicated publications (Lee et al., 2015;Environmental Protection Department, 2016;Sun et al., 2016). Elemental and organic carbon (EC, OC) were measured (semi-continuous thermo-optical ECOC analyzer RT-3131, Sunset Inc., USA) at the AQMS at 1h time resolution and at similar sampling height (~2m) employing a modified National Institute for Occupational Safety and Health (NIOSH) method 5040 protocol which has been described in a previous study (Huang et al., 2014). Concentrations of EC and OC in PM_1 were estimated by converting the raw $PM_{2.5}$ concentrations using a PM_1 to $PM_{2.5}$ ratio of 0.8 representing the overall average of PM_1 to $PM_{2.5}$ ratios from previous roadside sampling studies (0.75-0.85) in Hong Kong (Cheng et al., 2006;Lee et al., 2006).

AMS data were treated according to general AMS data treatment principles (DeCarlo et al., 2006;Jimenez et al., 2003) with standard software packages (SQUIRREL v1.53G, PIKA v1.12G). Analysis of the unit-mass resolution mass spectra yielded non-refractory submicron particle species concentrations of major inorganic constituents (SO4, NO3, NH4, Chl) and total organics at a base time resolution of 10 min. Positive Matrix Factorization (PMF) was used to deconvolute high-resolution organic mass spectra acquired at 10 min time resolution following recommended PMF guidelines for AMS data (Zhang et al., 2011) with the AMS PMF analysis toolkit (Ulbrich et al., 2009). Six organic aerosol (OA) factors were identified encompassing three secondary organic aerosol (SOA) and three primary organic aerosol (POA) factors of which one was attributed to traffic emissions (hydrocarbon-like organic aerosol, HOA (Zhang et al., 2005)) and two to cooking activities. OA factor concentrations were established using the fractional factor contributions from the high-resolution PMF analysis and the total submicron organics concentrations from the unit-mass resolution analysis. Both unit- and high-resolution mass spectral derived data were averaged to 1h time resolution to match ECOC measurements and traffic count data (*see below*), assessed from 28 May to 31 May, 2013. Average species concentrations measured during this time as well as the seasonal averages (spring, summer) from the whole measurement campaign are summarized in Table 1. Further specific details on instrument parameters, data analysis and an overview of the general characteristics of submicron particulate matter from the same campaign discussing the whole measurement period can be found in a previous publication (Lee et al., 2015).

Details on traffic composition are usually obtained by concurrent video image recordings and subsequent manual assessment of vehicle numbers and identification of vehicle types with classifications e.g. by vehicle function (Wang et al., 2016) or broader groups such as fuel types (Cui et al., 2016). In this work, vehicle data were obtained from the Hong Kong government, which conducted a counting exercise during the sampling period over three weekdays (noon 28 May 2013 to noon 31 May 2013, Tue to Fri) using automated license plate recognition (ALPR) with four infrared cameras on-site. Traffic along Lai Chi Kok road was monitored with three cameras monitoring all three lanes in one direction (out- or inbound) and the remaining camera monitoring one lane of the opposite direction. Camera positions and the monitored single lane were swapped in 2-3h intervals. Non-personalized data on registered vehicles were obtained, including license class (*vehicle type*), year of manufacture (*vehicle age*) and engine type (*gasoline, diesel, LPG, others*), thus providing a direct linkage to actual vehicle inventory data and circumventing the need for manual assessments. The obtained data were pooled by engine type (i.e. type of fuel used) to construct an hourly time series of diesel, gasoline and LPG powered vehicles.

Measured HOA and EC during the traffic counting period were decomposed by multiple linear 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 were sufficiently removed (i.e. shipping with ~2km of straight-line distance to the nearest coastline) and shielded by 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.

3. Results

3.1. Chemical and temporal characteristics of traffic-related carbonaceous species

Organic aerosol components were resolved by PMF from the organic part of the measured high-resolution mass spectra acquired during the measurement campaign and scaled to the measured total organic species concentrations based on the unit-mass resolution acquisition mode (Lee et al., 2015). Hydrocarbon-like organic aerosol (HOA) was resolved and identified as the traffic exhaust related factor, exhibiting chemical characteristics typically associated with freshly emitted primary organic aerosol from fossil fuel combustion (Zhang et al., 2005). Its mass spectrum (Figure S1a in the Supplement) is dominated by ions of saturated hydrocarbons (C_nH_{2n+1} series) and its elemental composition is largely void of oxygenated components with

correspondingly low O/C ratio of 0.066 and a high H/C ratio of 2.073. These findings are in line with most ambient studies that have identified a similar factor, e.g. (Sun et al., 2012;Sun et al., 2011;Mohr et al., 2012;Lanz et al., 2007;Huang et al., 2010;Sun et al., 2010;Aiken et al., 2009;Setyan et al., 2012).

Over the whole sampling campaign (March to July 2013), HOA correlated appreciably with NO_x (R_{pr}=0.64) as well as i- and n-Pentane (R_{pr}=0.66-0.67). The correlation with CO was much weaker (R_{pr}=0.37) indicating the presence of other significant, non-traffic related sources of CO in the vicinity of the Mong Kok measurement site. The highest fractions of HOA among total organic particulate matter in PM_1 were observed with southeasterly and northwesterly surface winds following the street canyon axis along Lai Chi Kok Road (Figure S2a in the Supplement). Average HOA mass concentrations varied between spring (3.5 µg m^{-3}) and summer (2.0 µg m^{-3}), with an overall campaign average of 2.8 µg m^{-3}. Diurnal variations of HOA were substantial, with mass concentrations rising sharply in the early morning at 06:00, increasing almost linearly during the morning rush hour at rates between 0.6 and 0.8 µg m^{-3} h^{-1} and leveling off beyond 09:00 (Figure 1a). 90% of the daily HOA was accumulated between the start of the rush hour and midnight. Minimum HOA concentrations in the low traffic period (00:00 to 06:00) amounted to ~1.7 µg m^{-3} in spring and ~0.7 µg m^{-3} in summer. Average post rush-hour concentrations (09:00 to 00:00) were 4.1 µg m^{-3} and 2.6 µg m^{-3} in spring and summer respectively.

Elemental carbon (EC) and organic carbon (OC) in $PM_{2.5}$ were measured semi-continuously at the adjacent AQMS following Huang et al. (Huang et al., 2014) and converted to PM_1 as described in the methodology section. In the following, we refer to the PM_1 concentrations for discussion. Diurnal variations of EC were similar to those of HOA, however, nominal concentrations were not affected by the seasonal transition. This led to significantly different EC/HOA ratios of 0.86 in spring and 1.78 in summer (Figure S1b in the Supplement). As the measurement location falls within the Kowloon urban area, which is characterized by uniform annual traffic flows (Transport Department, 2014), the observed decrease in HOA concentrations was more likely due to significant amounts of HOA being semi-volatile and partitioning in the gas-phase portion of the exhaust emissions in summer than changes in traffic flows. This is also consistent with the trend in relative diurnal variation of HOA (depicted cumulatively in Figure 1b) which remained constant across seasons despite the seasonality in nominal concentrations, and the overall negative correlation of HOA mass concentration and temperature (Figure S2b in the Supplement). The semi-volatility of HOA in this study is inferred from the observed trend in ambient mass concentrations and affirms more recent studies on the partitioning behavior of POA components. Evaporation of semi-volatile components upon dilution decreases ambient POA concentrations considerably and is expected to be temperature-dependent (Robinson et al., 2007), as is the case in this study. Measurements of thermally denuded particles of primary origin by TD-AMS indicate that POA components can exhibit properties of semi-volatility on a scale similar to SOA in urban environments (Huffman et al., 2009a;Huffman et al., 2009b), and more recent studies suggest that HOA possesses a relatively wide volatility distribution with up to 50-60% of its mass consisting of semi-volatile organic compounds (Cappa and Jimenez, 2010;Paciga et al., 2016). As noted earlier, in this study the seasonal decrease in submicron particle-phase organic mass concentrations (HOA) by 40% was associated with a substantial increase in mean temperature by 7°C between spring (23°C ± 3°C) and summer (30°C ± 2°C).

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. Mean reductions in nominal mass concentrations were generally quite similar in the range of 0.8 – 1.0 μg m^{-3} for EC and ~0.5 μg m^{-3} for HOA. The greater overall reduction in EC indicates a greater reduction in the number of diesel vehicles on Sundays, owing to reduced commercial traffic and lower bus frequency. Contributions of vehicle emissions to ambient organic aerosol have been derived from one-year long measurements at the same site based on analysis of semi-continuous ECOC measurements (Huang et al., 2014). The EC tracer method (Turpin and Huntzicker, 1991;Lim and Turpin, 2002) was employed which evaluates minimum OC/EC ratios, and OC and EC measurements during time periods of high primary source strength and low photochemical activities when OC is mainly of local origin and attributable to the dominant primary sources in the vicinity. A characteristic vehicle-related OC/EC value ($OC/EC_{vehicle}$) of 0.5 was derived from the 5% lowest summer OC/EC ratio data to approximate organic vehicular emissions. The reported monthly average primary organic matter concentrations from traffic in $PM_{2.5}$ (using an OM/OC conversion factor of 1.4) ranged between 2.7 – 3.4 μg m^{-3} with little seasonal variation. To compare to our measurement base of PM_1, we use the previously mentioned average ratio of $PM_1/PM_{2.5}$ of 0.8 for conversion. The OC/EC approach would thus yield an approximate 2.2 – 2.7 μg m^{-3} of traffic-related primary organics in PM_1 which compares well with the overall average concentration of traffic-related HOA (2.8 μg m^{-3}) covering the entire AMS measurement campaign (March – July 2013). However, we observed significantly different concentrations depending on the season with much higher concentrations (3.5 μg m^{-3}) in spring than in summer (2.0 μg m^{-3}). The lack of a clear seasonal variation in OC/EC derived traffic-related organics is due to the use of a fixed conversion factor directly dependent on ambient EC concentration which did not exhibit a strong seasonal behavior and thus could not capture the same trends observed from the AMS measurements. The derivation of the $OC/EC_{vehicle}$ ratio is subject to further uncertainty due to the substantial influence of cooking-related organics at the site (Lee et al., 2015;Sun et al., 2016), i.e. traffic emissions may not be the dominant source of "local" organics at the site (Table 2), as well as the influence of background SOA which is likely to persist even in time periods of low OC/EC ratios.

Nonetheless, the agreement of the whole-campaign average HOA concentrations (this study) and $OC/EC_{vehicle}$ derived traffic organics mass concentration (Huang et al., 2014) suggests that the long-term average primary organic aerosol contribution from vehicles is reasonably well-approximated with the OC/EC approach but may lead to over- and underestimation of actual organic concentrations in shorter time frames, e.g. different seasons and different times of the day.

3.2. Traffic and vehicle flow characteristics

During the three-day counting exercise, a total of 21,048 vehicles were registered (~7016 vehicles per day). This accounts for approximately 18% of the annual average daily traffic (A.A.D.T.) of 38,220 vehicles estimated in the 2013 Annual Traffic

Census (Transport Department, 2014). 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 S2c in the Supplement. The maximum hourly vehicle number occurred towards the end of the rush-hour (~9:00) for most vehicle classes (except "*other vehicles*", which mostly comprised motorcycles). For the evening rush-hour (17:00-19:00), only the private car category exhibited a peak in vehicle number. Overall, the post rush-hour total vehicle count remained stable throughout the day, and only fell off after 19:00, largely due to a strong decrease in the number of goods and private vehicles. The variation in the fractional contribution of different vehicles types over the day resembles that assessed by the Transport Department (Transport Department, 2014) at the closest core traffic counting station at Nathan Road (Figure S2c in the Supplement) and affirms that the measured traffic composition during the 72h counting period at Mong Kok was largely representative. At the Mong Kok site, smaller vehicles (*cars and taxis*) accounted for about 60% of total registered vehicles, while heavier vehicles (*buses, vans, trucks*) made up most of the remainder.

Total HOA concentrations showed good correlations (R_{pr}>0.6) with most of the measured vehicle-related VOCs, NO_x and EC concentrations as well as the total number of passing vehicles during the counting period (Figure 2). As with the overall campaign data, CO did not correlate with either HOA or EC, indicating the impact of significant non-traffic-related CO sources in the Mong Kok area. Among the counted buses, light buses and goods vehicle, over 96% were running on diesel engines, while 99.9% of counted taxis utilized LPG engines. Private vehicles were almost exclusively powered by gasoline engines (99.4%) due to government restrictions on the import and use of diesel-powered private vehicles at the time of this study.

3.3. Estimation of engine-type contributions

3.3.1. Multiple Linear Regression of HOA and EC

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 2. Methodology*).

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 Supplement. For HOA, both two and three factor models yielded acceptable results with all resolved coefficients statistically significant at the 95% confidence level ($p<0.05$). Exclusion of LPG vehicles (two factor model) led to additional apportionment of HOA to both gasoline (+25%) and diesel (+18%) vehicles. Overall model performance improved only slightly when moving from the two factor solution (R^2_{adj}=0.86) to the three factor solution (R^2_{adj}=0.90). Comparative studies on particulate vehicle emissions have reported non-negligible particle

mass contributions of LPG vehicles. Estimated particle mass emission factors from LPG passenger vehicles were similar (Chan et al., 2007) or ~30% lower (Ristovski et al., 2005) compared to gasoline powered models in chassis dynamometer runs. In on-road environments, absolute particle number concentrations in bus exhaust plumes were lower in samples from CNG and LPG fueled buses compared to diesel and DME (dimethyl ether) fueled buses, but still 10 times higher than typical ambient background particle concentrations (Kwak et al., 2014). Evaluating HOA concentrations measured between 0:00 and 5:00 when LPG vehicles dominated the total vehicle population (45-60% of total counted vehicles) in our study shows a clear positive relationship between ambient HOA and LPG vehicle count (Figure S3a in the Supplement). The two factor MLR model tends to underestimate measured HOA mass concentrations with the distribution of absolute residuals (*difference of reconstructed and measured HOA mass concentration*) biased to negative values whereas the three factor model yielded more normally distributed residuals (Figure S3b, c in the Supplement). We thus consider the three factor solution including LPG vehicles as a more appropriate representation for the deconvolution of the ambient HOA mix from our measurements. For EC, a three factor representation could not resolve variable coefficients that were all statistically significant at the 95% confidence level yielding $p>0.05$ for the gasoline vehicle factor. While both diesel and gasoline vehicle number correlated appreciably with EC (Figure S4c in the Supplement), the number of LPG vehicles lacked a corresponding relation and stayed almost constant over the range of measured EC concentrations, rendering the two factor model the more appropriate representation in deconvoluting measured ambient EC.

3.3.2. Analysis of engine-type reconstructed carbonaceous components

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 Supplement and in combination in Figure 3 to represent the sum of motor vehicle related primary carbonaceous particulate compounds, not including possible additional SOA species formed through subsequent atmospheric processing of gas- and particle-phase species in tailpipe exhaust. Gasoline vehicles accounted for more than 45% of total HOA, while diesel and LPG-fueled vehicles were responsible for similar shares of 25-29% (Figure S5b in the Supplement). Over 60% of total EC in PM_1 was accounted for by diesel vehicles (Figure S5d in the Supplement). The sum of HOA and EC in PM_1 represent approximately the total submicron primary carbonaceous aerosol contributions from road traffic. Despite the limited time frame of the counting exercise, the MLR factors adequately resolved the overall trend in EC and HOA concentration (Figure 3a) and diurnal variation (Figure 3b). Relative residuals from the reconstructed sum of HOA and EC were $\leq$25% with overestimation (*Residuals* < 0) predominant in the later afternoon hours between 14:00 and 18:00, while significant underestimation (*Residuals* > 0) was observed mostly in the late evening hours between 19:00 and 0:00. The semi-volatile character of HOA would contribute to the observed reduced afternoon particle-phase concentration and conversely enhanced nighttime particle-phase concentrations. Averaged over the whole 72h period, measured and reconstructed carbonaceous submicron PM_1 differed by only 4% indicating that the overall reconstruction was not affected

considerably by these diurnal trends in residuals. Vehicle emission factors are typically based on the concentration of emitted species per unit of consumed fuel (derived from CO_2 measurements) or per driven distance. As concurrent CO_2 measurement data were not available, emission factors were not evaluated in this study. Instead, the relative emission behavior of different vehicle groups and their impacts on ambient PM concentrations were approximated by normalizing the resolved engine-specific carbonaceous primary aerosol concentrations by the total number of vehicles in each category (Figure 3b). On a per-vehicle basis (Figure 3b), gasoline vehicles emitted 75% more HOA than diesel vehicles and 93% more HOA than LPG vehicles, while EC emissions from diesel vehicles were 45% higher than those from gasoline vehicles. Therefore, on average gasoline and diesel vehicles emitted similar total amounts of primary carbonaceous PM_1 and each about three times more than LPG powered vehicles. In terms of ambient concentrations, gasoline vehicles accounted for 78% more ambient HOA than diesel vehicles and 56% more ambient HOA than LPG-fueled vehicles. As opposed diesel vehicles accounted for 62% more ambient EC than gasoline-powered vehicles. 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).

It must be noted that these data represent the total average of the encountered diesel, gasoline and LPG vehicle fleet, notwithstanding that each of these broad classes is inhomogeneous consisting of a wide variety of vehicles of different size, engine power and age, and thus different individual emission characteristics.

As the fractional contributions of diesel and gasoline vehicles to ambient HOA and EC varied substantially, i.e. diesel vehicles emitted almost three times as much EC as HOA (EC/HOA=2.85) and gasoline vehicles contributed similar mass concentrations to both HOA and EC (EC/HOA=1.02). This caused significant diurnal variations in the EC/HOA ratio (Figure S1c in the Supplement) with low values in the nighttime hours, where LPG- and gasoline-fueled vehicles were more dominant and highest values occurring at the tail of the morning rush-hour where fractions of goods vehicles and buses among overall traffic were highest. 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). Dynamometer studies mostly reported EC/OC values for gasoline powered vehicles < 1 (Alves et al., 2015;Geller et al., 2006) indicating overall a greater importance of organics in particulate emissions.

The relatively high fraction of EC in gasoline related PM in our work likely stems from a combination of factors. HOA mass concentrations are derived from AMS measurements which are subject to limitations of inlet lens transmission, i.e. a standard lens as used in this study is expected to transmit efficiently (~100%) between 90nm and 700nm D_{va} (Williams et al., 2013), and thus may cause a low bias in measured ambient HOA concentrations. However, these inlet losses are expected to only have limited effects on total measured submicron particle mass concentrations as mass- and volume-based particle concentrations from vehicle exhaust typically peak in the region of ~ 100 – 300nm (Fushimi et al., 2016;Ban-Weiss et al.,

2010). The observed seasonal variability of particle-phase organic concentrations due to partitioning of semi-volatile components into the gas-phase would also decrease particle-phase HOA concentrations in warmer seasons, thus elevating the observed ambient EC/HOA ratio with our measurements taking place in summer. While this would affect all engine types, the extent may vary with the distribution of semi-volatile species in the respective tailpipe emissions of different vehicle groups. Due to the characteristics of the sampling site (inner-city urban road traffic and proximity to both a road junction and pedestrian crossing) sampled emissions include a considerable fraction of variable and higher engine loads during the acceleration phase whereas road tunnel environments are characterized by largely constant engine loads and traveling speeds. As gasoline vehicles are typically not equipped with particle filters, they have been shown to emit significant amounts of EC under unstable engine loads (Karjalainen et al., 2014).

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).

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 in 2010 for the replacement of diesel commercial vehicles, the retrofitting of diesel particulate filters (DPF) on pre-Euro IV buses and subsequent replacement of older buses with Euro IV, V and VI standard models (Environment Bureau, 2013;Environmental Protection Department, 2013). $PM_{2.5}$ emission factors determined from dynamometer test runs of various vehicle types in Hong Kong were generally higher for diesel vehicles compared to gasoline and LPG vehicles (Chan et al., 2007), while we observed similar total particle mass emission on a per-vehicle basis for diesel and gasoline vehicles in this study. The dynamometer tests were conducted on relatively old vehicles (*newest date of manufacture: 2001*) and are unlikely to adequately reflect the ambient vehicle mix in our study, particularly with regard to the implementation of recent emission control programs. 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 Supplement 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.

The specific characteristics of the Mong Kok measurement site as well as the continual changes in vehicle technology are not represented well in typical dynamometer driving cycles and tunnel studies and are at the least partially responsible for the observed differences to other studies reporting relative abundances of elemental carbon to organic mass concentrations.

4. Conclusion

Primary submicron carbonaceous aerosol was measured by combined HR-ToF-AMS and ECOC measurements, supplemented by various VOCs and other gas-phase species, at an inner-city urban roadside sampling site in the Mong Kok District in Hong Kong. Organic species concentrations (HOA) were of similar magnitude as traffic-related organic matter estimated previously, but with strong seasonal dependency with on average 40% lower concentrations in summer (2.0 $\mu g/m^3$) compared to spring (3.5 $\mu g/m^3$) likely due to greater fractions of semi-volatile species remaining in the gas-phase in the warmer season. EC concentrations remained at similar levels throughout the sampling period, leading to significantly different average EC/HOA ratios in spring (0.86) and summer (1.78). Sunday concentrations of both HOA and EC were 14-27% lower than during the rest of the week (Mon-Sat) reflecting significant reductions in traffic volume on Sundays. Larger relative reductions in EC compared to HOA indicate a greater reduction in the number of diesel vehicles, e.g. commercial trucks and buses.

Detailed vehicle counting data acquired during a 72h counting exercise in late May 2013 were utilized to decompose HOA and EC concentrations into vehicle-type factors (gasoline-, diesel- and LPG-powered vehicles) by multiple linear regression analysis. Gasoline vehicles contributed similar amounts of EC and HOA, while diesel vehicles emitted predominantly EC and to a lesser extent HOA. On an average per-vehicle basis, contributions of diesel and gasoline vehicles to carbonaceous PM_1 were similar, contrary to previous studies which attributed higher particulate matter emissions to diesel vehicles. This clear reduction is likely due to recent control strategies targeted at commercial vehicles and buses, which represent the bulk of diesel powered vehicles at the measurement site. This becomes especially important in view of the growing number of higher Euro standard diesel vehicles which, while effectively reducing overall emitted particle mass may emit similar number concentrations of particles with particle sizes having been shown to shift more into the ultrafine region (Tartakovsky et al., 2015). The number of LPG-vehicles did not exhibit a significant correlation with EC concentrations and was thus considered to be a negligible EC source. Its contributions to ambient primary traffic-related PM_1 were small (13%) despite representing about one-third of total counted vehicles.

While this study has focused on primary emissions, gas-phase emissions from the tailpipe can lead to subsequent condensation of organics or atmospheric oxidation to form secondary organic aerosol, significantly enhancing post-emission particulate matter concentrations (Gentner et al., 2012;Platt et al., 2013) and should be subject to further future investigation.

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Table 1. Species concentrations from AMS and total $PM_{2.5}$ mass from TEOM in (a); EC, OC and organic aerosol constituent concentrations (AMS PMF analysis) in (b); average concentrations during the traffic counting period (May 28-31, 2013) and seasonal averages (Spring: Mar – May 2013, Summer: May – Jul 2013) based on Lee at al. (2015) at the Mong Kok site

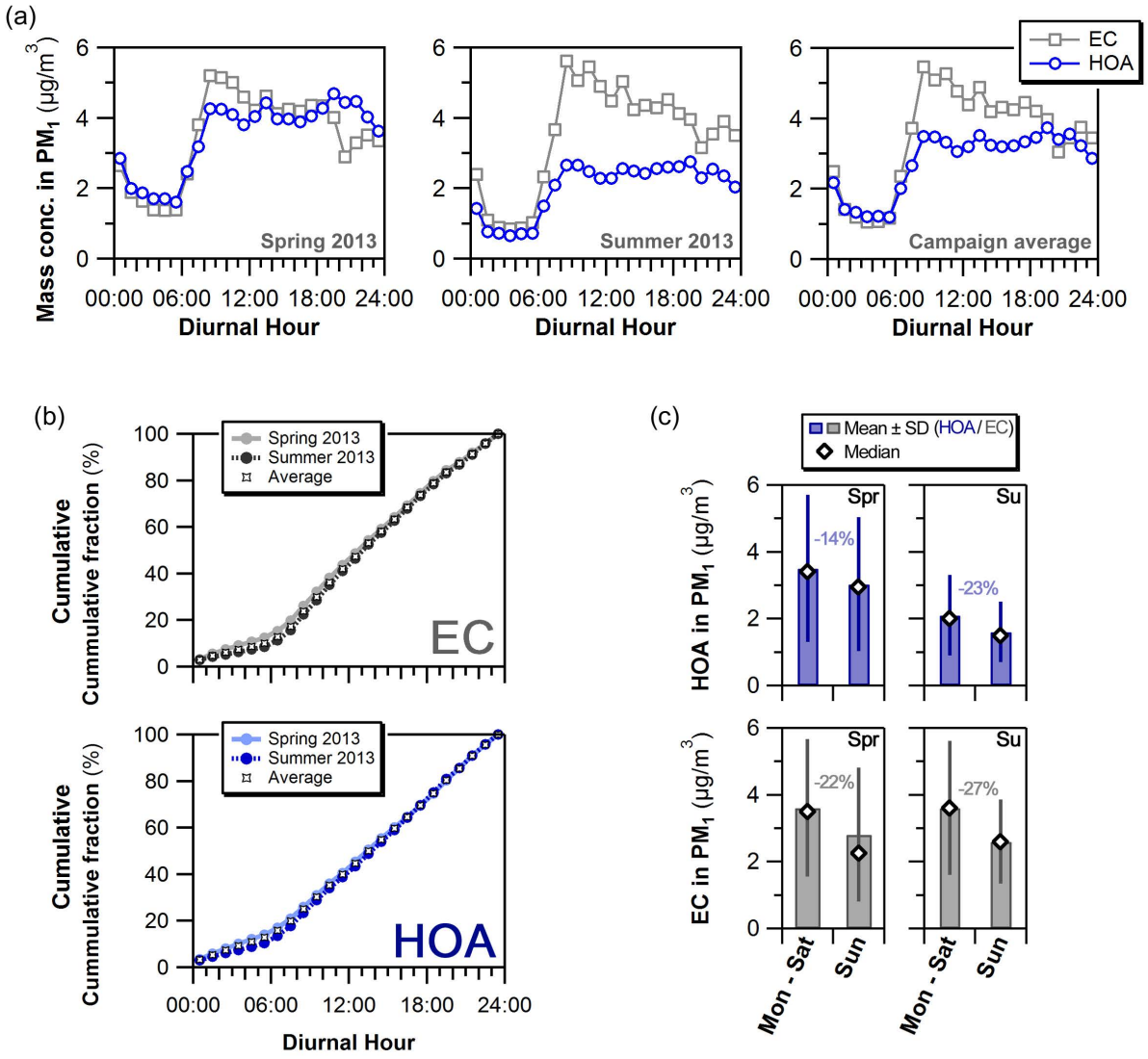
(a)

µg/m^3 Mean ± *SD*	**Org** (AMS)	**SO4** (AMS)	**NO3** (AMS)	**NH4** (AMS)	**Chl** (AMS)	**NR-PM_1** (AMS)	**$PM_{2.5}$** (TEOM)
May 28-31	5.9±3.0	4.8±1.2	0.3± 0.1	1.5±0.3	<0.1	12.5±3.7	16.0±5.3
Spring	12.8±7.6	7.0±3.8	2.5±2.1	2.5±1.5	0.4±0.3	25.3±13.1	32.3±12.4
Summer	7.9±5.4	3.4±2.0	0.4±0.4	1.1±0.5	<0.1	12.7±3.6	17.3±7.5

(b)

µg/m^3 Mean ± *SD*	**EC** ($PM_{2.5}$)	**OC** ($PM_{2.5}$)	**EC** (PM_1)	**OC** (PM_1)	**HOA** (PM_1)	**COA**[a] (PM_1)	**SOA**[b] (PM_1)
May 28-31	4.2±2.3	3.0±1.2	3.2±1.7	2.3±0.9	2.4±1.4	2.0±1.2	1.6±1.1
Spring	4.3±2.6	7.6±3.9	3.2±1.9	5.7±2.9	3.5±2.4	4.4±4.3	4.9±3.4
Summer	4.3±2.5	4.1±2.1	3.2±1.8	3.0±1.6	2.0±1.3	3.6±3.4	2.2±2.2

Notes: [a] Sum of two cooking-related PMF factors
[b] Sum of three SOA-related PMF factors



730 **Figure 1.**

(a) Diurnal variation of HOA and EC mass concentrations in spring 2013 (7-Mar-2013 to 15-May-2013), summer 2013 (16-May-2013 to 19-Jul-2013) and averaged over the entire sampling campaign (7-Mar-2013 to 19-Jul-2013)

(b) Diurnal variation of the cumulative fraction of total daily HOA and EC mass concentrations in spring 2013, summer 2013 and averaged over the entire sampling campaign

735 (c) Mean HOA and EC concentrations (*bar with standard deviations, open marker depicts median*) from Monday through Saturday and on Sunday in spring 2013 (Spr) and summer 2013 (Su); days with >25% (>6h) of missing data were excluded

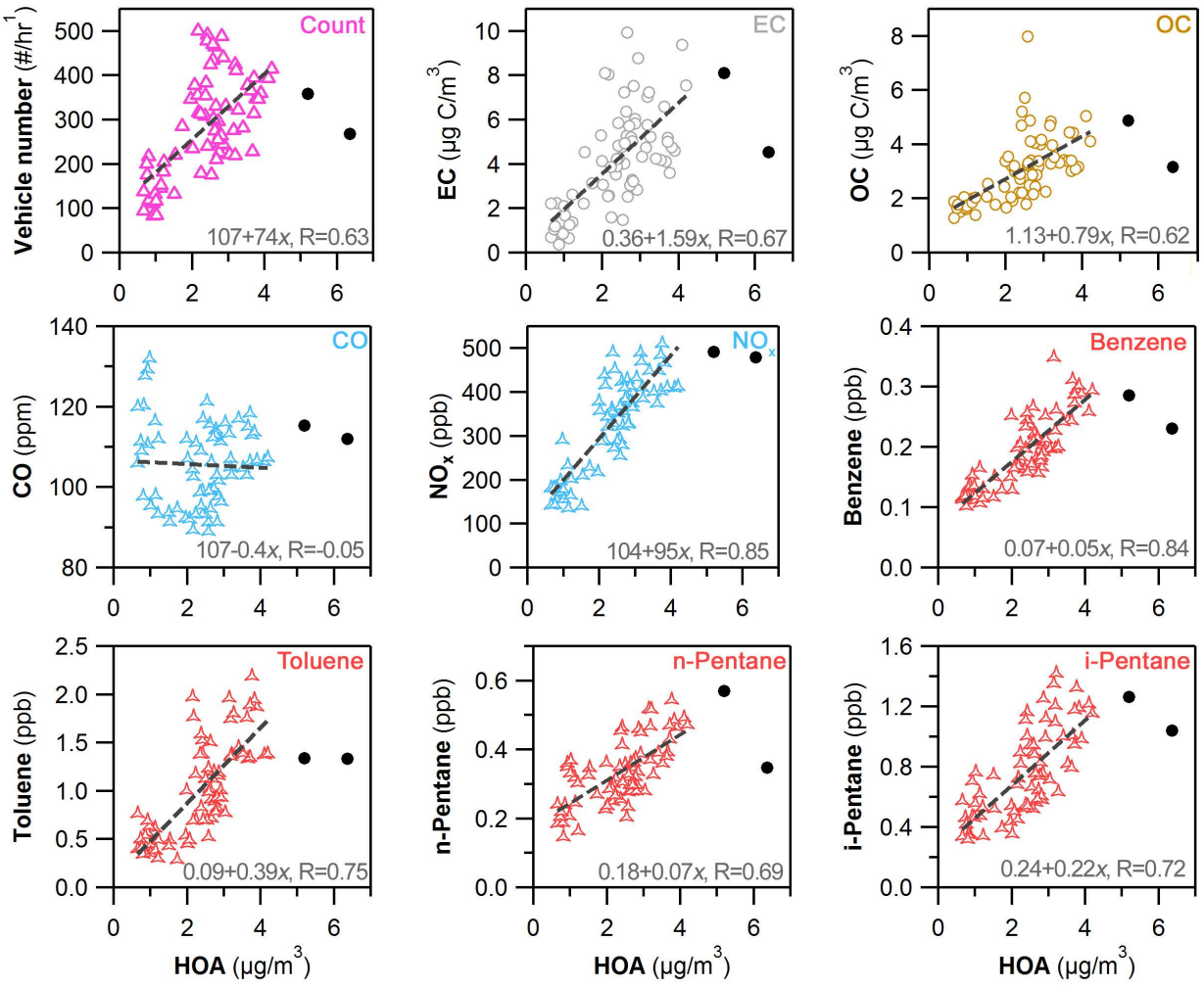


Figure 2.

Scatter plot of vehicle count, mass concentration of EC, OC and gas-phase concentrations of various VOCs, NO_x, and CO against HOA during 28-31 May 2013, *linear least squares fits with equations and Pearson's R values in grey, black dots signify outlier values excluded from the curve fits*

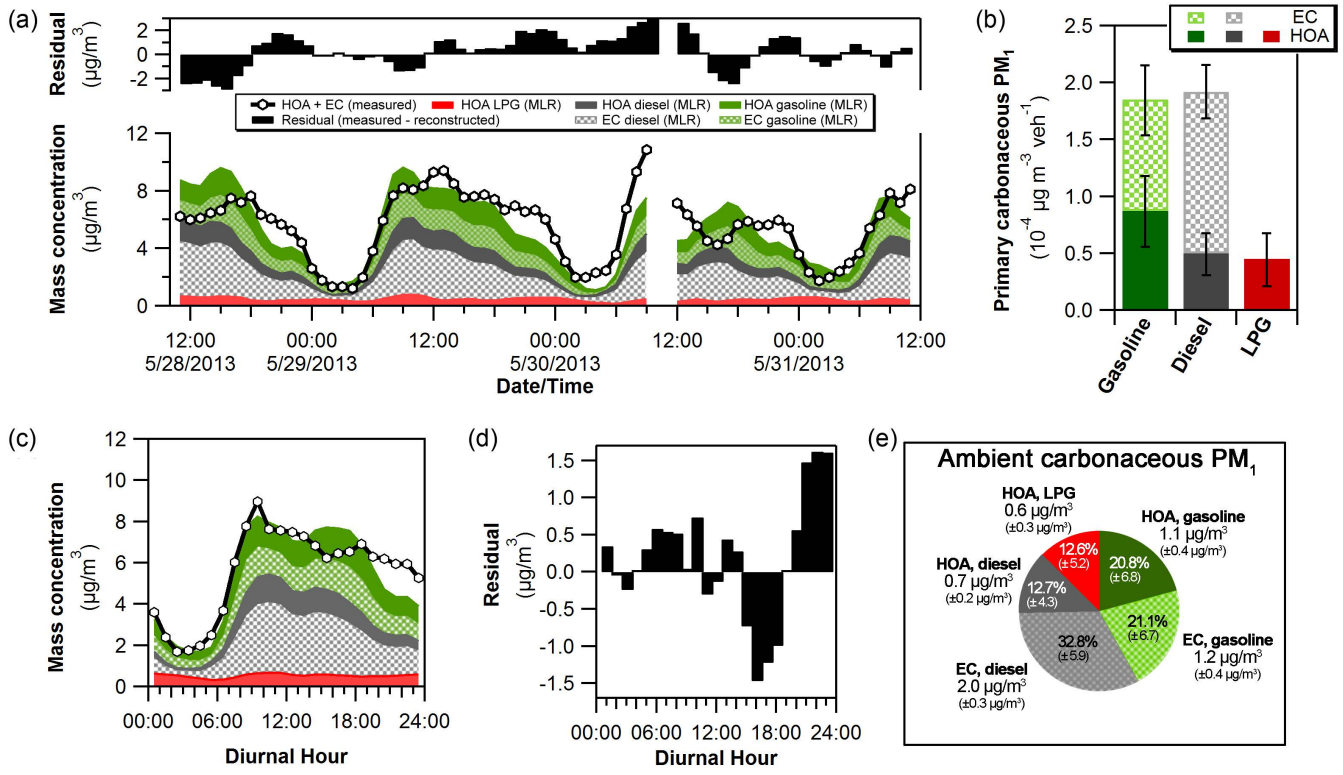


Figure 3.

(a) Time series of engine-type separated HOA and EC in PM_1, sum of actual measured HOA and EC in PM_1, as well as residuals as the difference between resolved HOA+EC and measured HOA+EC

(b) Contribution of engine-type separated HOA and EC to PM_1 normalized by the total number of vehicles per engine category; vehicle composition based on traffic count (28 – 31 May, 2013) and vehicle number scaled to average annual daily traffic (Transport Department, 2014), uncertainties from resolved fit parameters (MLR) included

(c) Diurnal variation of vehicle-type separated HOA and EC concentrations as well as sum of actual measured HOA and EC in PM_1

(d) Diurnal variation of residuals as the difference between resolved HOA+EC and measured HOA+EC in PM_1

(e) Average contribution of engine-type separated HOA and EC in total reconstructed HOA+EC in PM_1, uncertainties from resolved fit parameters (MLR) included

Supplement of

Evaluation of traffic exhaust contributions to ambient carbonaceous submicron particulate matter in an urban roadside environment in Hong Kong

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I. Additional Tables

Table S1 – Regression statistics of two and three factor multiple linear regression analysis of HOA and EC mass concentrations as functions of gasoline (G-veh.), diesel (D-veh.), and LPG (L-veh.) vehicle number counts

Regression statistics		**HOA, 3 Fac**	**HOA, 2 Fac**	**EC, 3 Fac**	**EC, 2 Fac**
$\mathbf{R^2_{adj}}$	Total	0.90	0.86	0.92	0.92
P-Value	G-veh.	5.63×10^{-3}	1.53×10^{-5}	0.63	1.51×10^{-3}
	D-veh.	5.47×10^{-3}	1.77×10^{-3}	5.04×10^{-9}	6.41×10^{-8}
	L-veh.	4.86×10^{-3}	N.A.	3.39×10^{-3}	N.A.
Var. coeff.	G-veh.	1.10×10^{-2} $\pm 3.86 \times 10^{-3}$	1.40×10^{-2} $\pm 3.01 \times 10^{-3}$	2.12×10^{-3} $\pm 4.40 \times 10^{-3}$	1.12×10^{-2} $\pm 3.40 \times 10^{-3}$
	D-veh.	7.05×10^{-3} $\pm 2.46 \times 10^{-3}$	8.33×10^{-3} $\pm 2.56 \times 10^{-3}$	1.91×10^{-2} $\pm 2.86 \times 10^{-3}$	1.82×10^{-2} $\pm 3.00 \times 10^{-3}$
	L-veh.	6.17×10^{-3} $\pm 3.08 \times 10^{-3}$	N.A.	1.06×10^{-2} $\pm 3.48 \times 10^{-3}$	N.A.

II. Additional Figures

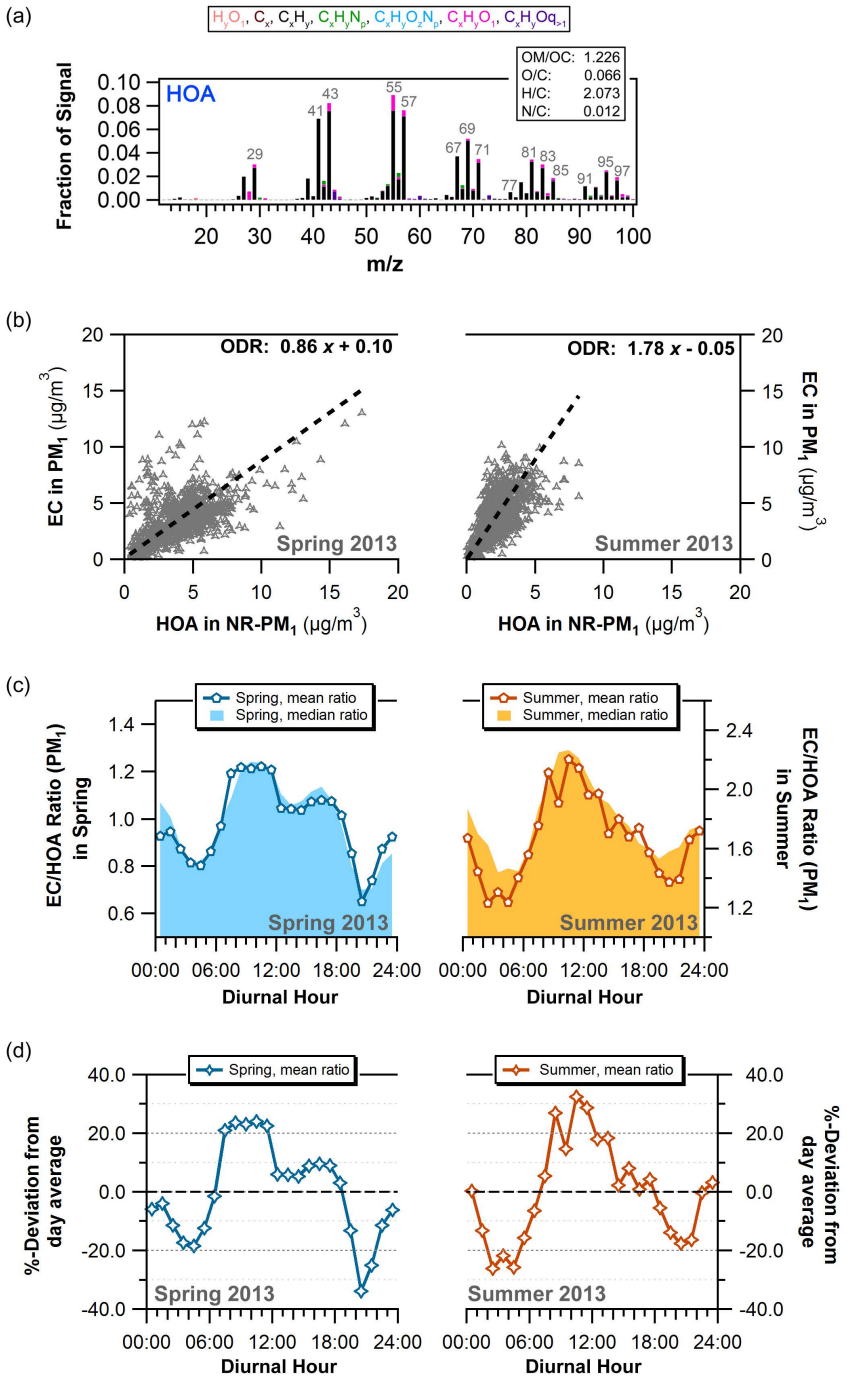

Figure S1

(a) Mass spectrum and elemental characteristics of HOA (Lee et al., 2015)

(b) Scatter plot of EC and HOA in PM_1 at the Mong Kok site in spring (Mar to May) and summer (May to Jul) 2013

(c) Diurnal variation of EC/HOA ratio (in PM_1) based on mean concentrations *(solid line and open markers)* and based on median concentrations *(shaded area)*

(d) Relative diurnal change in EC/HOA ratio in PM_1 in relation to daily average, in %

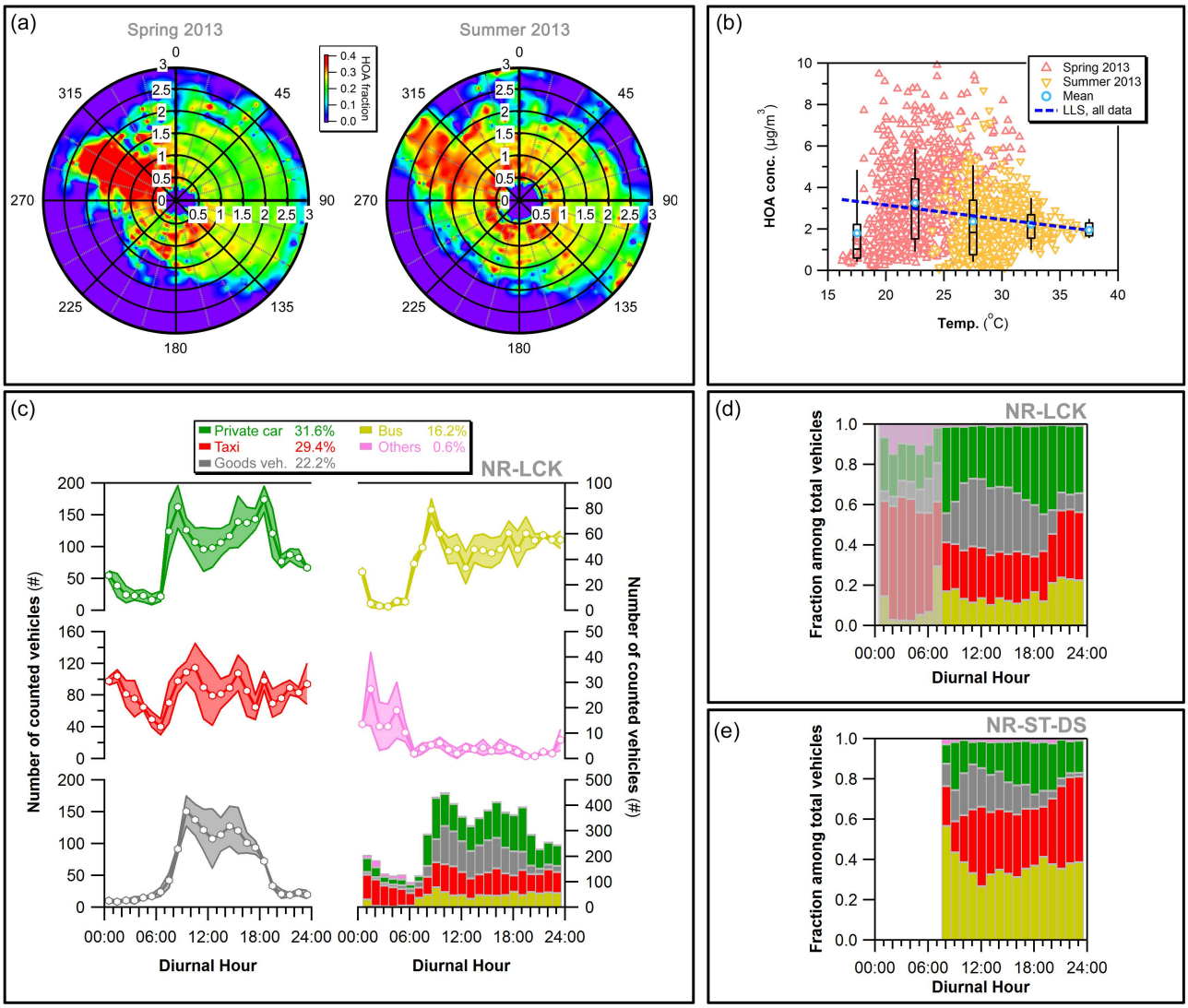

Figure S2

(a) Wind rose plots of HOA fraction in total organics (NR-PM_1) in spring (*left*) and summer (*right*) 2013 at the Mong Kok site

(b) HOA mass concentration as a function of ambient temperature at the Mong Kok site; with linear least squares fit *(dark blue hashed line)* and box/whisker plots (*10^{th}, 25^{th}, 75^{th}, 90^{th} percentile, median as line, mean as blue circle)* for temperature bins from 15-40°C at fixed bin width of 5°C for all data

(c) Diurnal variation of number of vehicles (grouped by main use) passing by the measurement site *(NR-LCK: junction of Nathan Road and Lai Chi Kok Road)* between 28 May and 31 May 2013, with average fraction of total counted vehicles (vehicle mix) given in the legend box

(d) Fractional diurnal variation of vehicle groups among total counted vehicles passing by the measurement site *(NR-LCK: junction of Nathan Road and Lai Chi Kok Road)* between 28 May and 31 May 2013

(e) Fractional diurnal variation of vehicle groups among total counted vehicles at the closest major vehicle counting station (*NR-ST-DS: Nathan Road, between junction with Shantung Street and Dundas Street*), data based on Annual Traffic Census, 2013 (Transport Department, 2014)

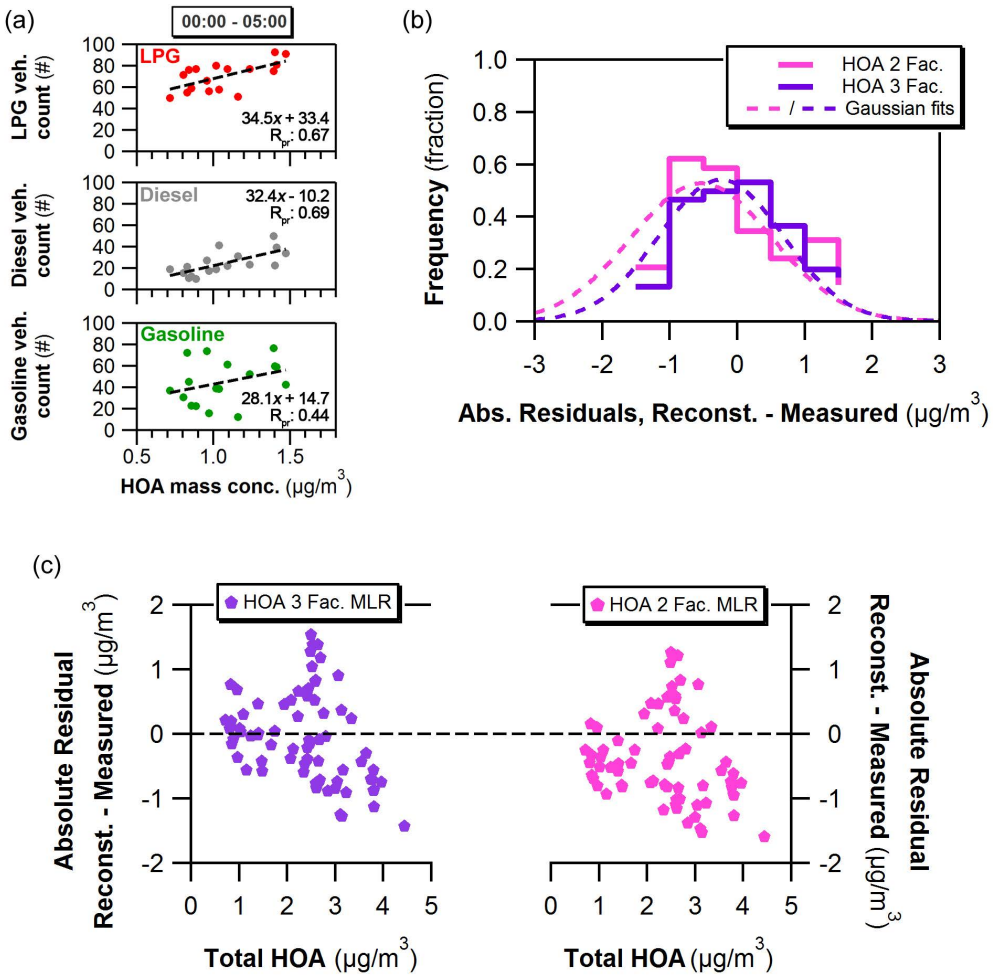

Figure S3

(a) Number of counted gasoline, diesel and LPG powered vehicles as a function of total HOA mass concentration during the low traffic hours *(00:00 to 05:00)*

(b) Frequency distribution of absolute residuals *(in µg/m^3)* and their Gaussian fits for three factor and two factor MLR solutions

(c) Scatter plot of absolute residuals *(in µg/m^3)* for three factor and two factor MLR solutions against total HOA mass concentrations

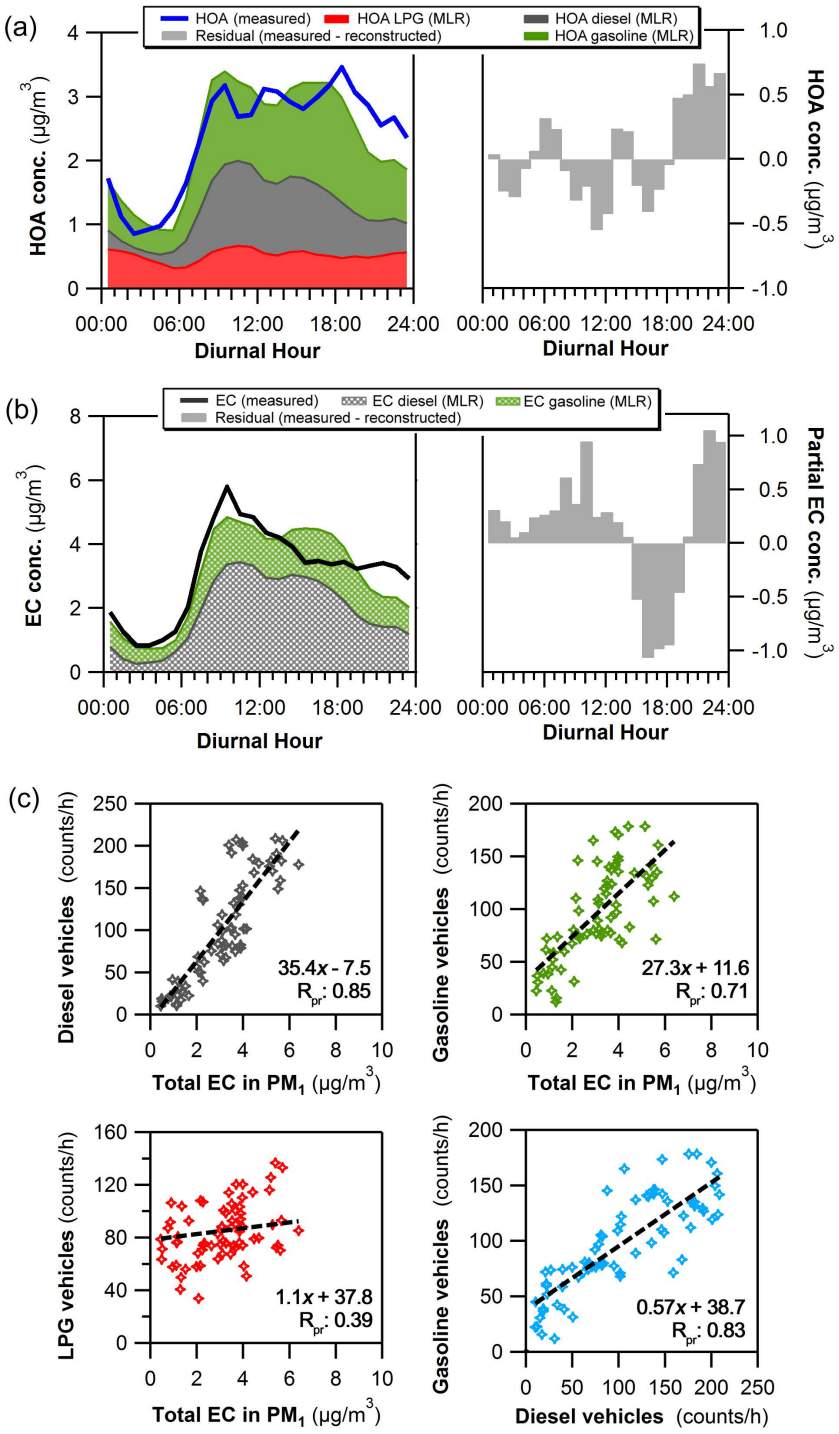

Figure S4

(a) Diurnal variations of engine-type resolved HOA concentrations and actual measured HOA *(left)*, and diurnal variation of HOA residual as the difference between actual and reconstructed HOA *(right)*

(b) Diurnal variations of engine -type resolved EC concentration and actual measured EC in PM_1 *(left)*, and diurnal variation of EC residual as the difference between actual and reconstructed EC in PM_1 *(right)*

(c) Scatter plots of vehicle counts (by engine type) against total EC in PM_1 *(grey, green, red)*, and gasoline against diesel vehicle count *(blue)*

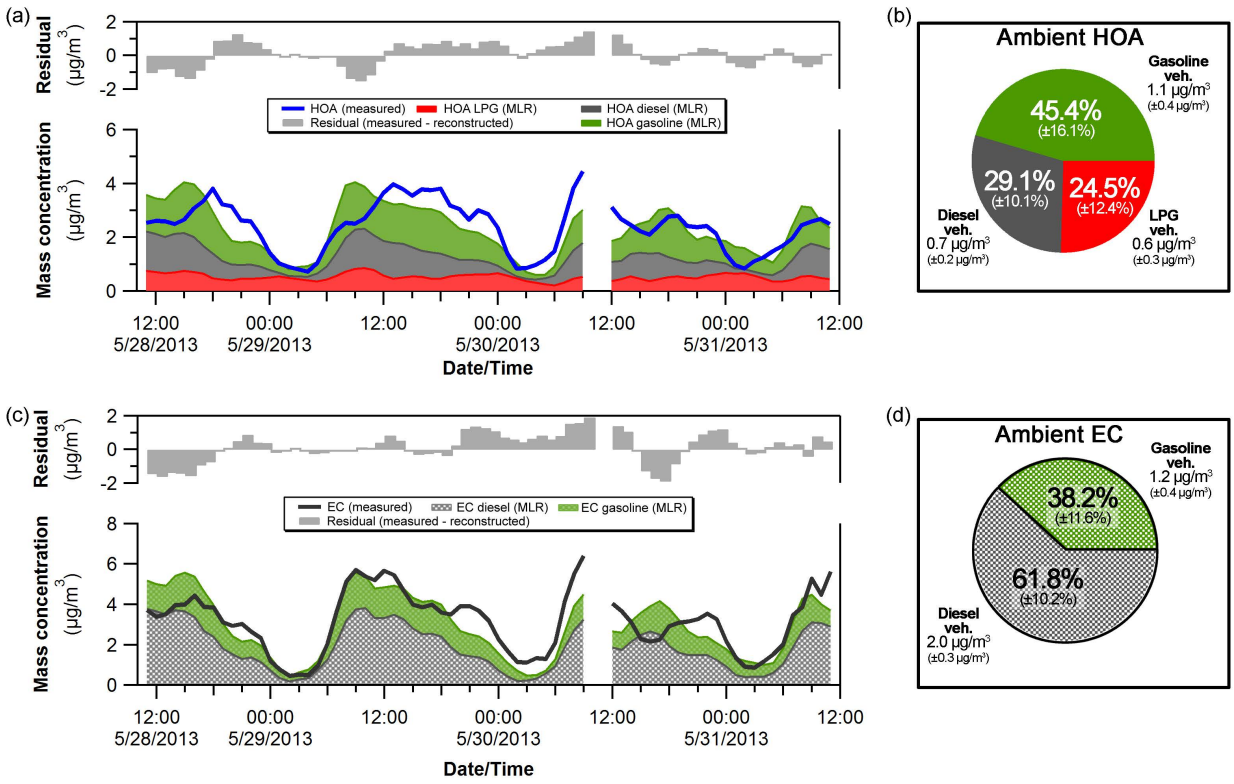

Figure S5

(a) Time series of engine-type resolved HOA, actual measured HOA, as well as residuals as the difference between resolved HOA and measured HOA in PM_1

(b) Average contribution of engine-type resolved HOA in total reconstructed HOA in PM_1

(c) Time series of engine-type separated EC, actual measured EC, as well as residuals as the difference between resolved EC and measured EC in in PM_1

(d) Average contribution of engine-type separated EC in total reconstructed EC in PM_1

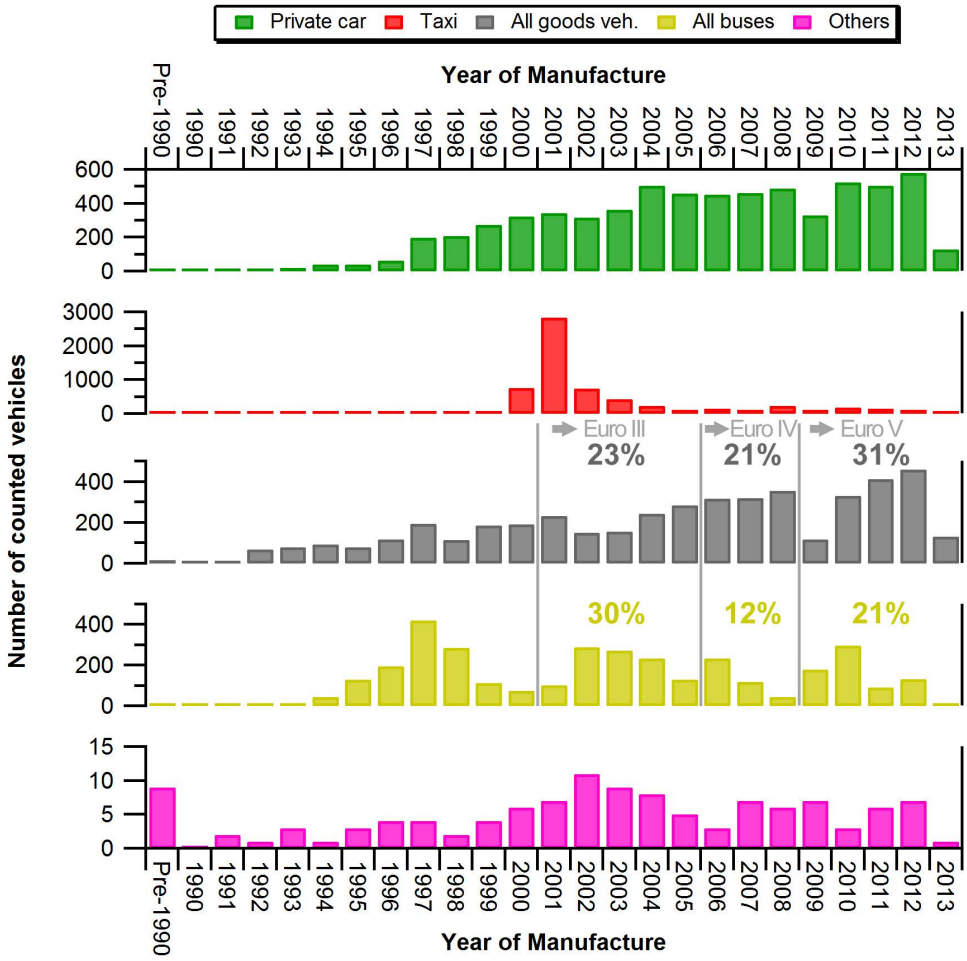

Figure S6

Year of manufacture of vehicles identified at the measurement site between 28 May 2013 and 31 May 2013. Vertical gray lines indicate the year of introduction of respective Euro emission standards for heavy diesel vehicles and percentage numbers indicate the fraction of vehicles within each bin.

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Lee, B. P., Li, Y. J., Yu, J. Z., Louie, P. K. K., and Chan, C. K.: Characteristics of submicron particulate matter at the urban roadside in downtown Hong Kong—Overview of 4 months of continuous high-resolution aerosol mass spectrometer measurements, Journal of Geophysical Research: Atmospheres, 120, 7040-7058, 10.1002/2015JD023311, 2015.

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