

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 treated, 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 et 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 PM₁ 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 $2.7 - 3.4 \mu\text{g m}^{-3}$ 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 $2.2 - 2.7 \mu\text{g m}^{-3}$ of traffic-related primary organics in PM_1 which compares well with the overall average concentration of traffic-related HOA ($2.8 \mu\text{g m}^{-3}$) covering the entire AMS measurement campaign (March – July 2013). However, we observed significantly different concentrations depending on the season with much higher concentrations ($3.5 \mu\text{g m}^{-3}$) in spring than in summer ($2.0 \mu\text{g m}^{-3}$). 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 (SO_4 , NO_3 , NH_4 , 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)	$\mu\text{g}/\text{m}^3$ Mean \pm SD	Org (AMS)	SO ₄ (AMS)	NO ₃ (AMS)	NH ₄ (AMS)	Chl (AMS)	NR-PM ₁ (AMS)	PM _{2.5} (TEOM)
May 28-31		5.9 \pm 3.0	4.8 \pm 1.2	0.3 \pm 0.1	1.5 \pm 0.3	0.1 \pm 0.1	12.5 \pm 3.7	16.0 \pm 5.3
Spring		12.8 \pm 7.6	7.0 \pm 3.8	2.5 \pm 2.1	2.5 \pm 1.5	0.4 \pm 0.3	25.3 \pm 13.1	32.3 \pm 12.4
Summer		7.9 \pm 5.4	3.4 \pm 2.0	0.4 \pm 0.4	1.1 \pm 0.5	0.1 \pm 0.1	12.7 \pm 3.6	17.3 \pm 7.5

(b)	$\mu\text{g}/\text{m}^3$ Mean \pm 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 \pm 2.3	3.0 \pm 1.2	3.2 \pm 1.7	2.3 \pm 0.9	2.4 \pm 1.4	2.0 \pm 1.2	1.6 \pm 1.1
Spring		4.3 \pm 2.6	7.6 \pm 3.9	3.2 \pm 1.9	5.7 \pm 2.9	3.5 \pm 2.4	4.4 \pm 4.3	4.9 \pm 3.4
Summer		4.3 \pm 2.5	4.1 \pm 2.1	3.2 \pm 1.8	3.0 \pm 1.6	2.0 \pm 1.3	3.6 \pm 3.4	2.2 \pm 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 PM₁ despite their high abundance in the traffic mix (~30%). Total carbonaceous particle mass contributions to ambient PM₁ 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 $\mu\text{g}/\text{m}^3$ of EC and 1.1 $\mu\text{g}/\text{m}^3$ of organics, LPG vehicles 0.6 $\mu\text{g}/\text{m}^3$ of organics and diesel vehicles 2.0 $\mu\text{g}/\text{m}^3$ of EC and 0.7 $\mu\text{g}/\text{m}^3$ of organics to ambient carbonaceous PM₁. [...]

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, CO₂ 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 $1.7 \mu\text{g}/\text{m}^3$) 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.

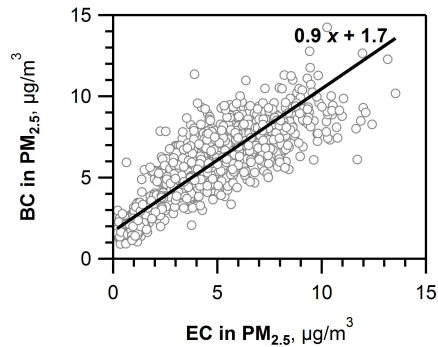


Figure R1. Scatter plot of hourly BC (aethalometer) and hourly EC (ECOC analyzer) measurements (*open circles*) with orthogonal distance regression (*solid line*) between June 1 and July 19, 2013 at the Mong Kok site

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 PM₁ were estimated by converting the raw PM_{2.5} concentrations using a PM₁ to PM_{2.5} ratio of 0.8 representing the overall average of PM₁ 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 CO₂ measurements) or per driven distance. As concurrent CO₂ 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 PM_{2.5}, 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 partitioning/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, CO₂, NO_x, SO₂) 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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