

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

4 Berto Paul Lee¹, Peter Kwok Keung Louie², Connie Luk², and Chak Keung Chan^{1*}

5 ¹School of Energy and Environment, City University of Hong Kong, Hong Kong, China

6 ²Environmental Protection Department, HKSAR Government, Wan Chai, Hong Kong, China

7 *Correspondence to:* Chak K. Chan (chak.k.chan@cityu.edu.hk)

8 **Abstract.** Road traffic has significant impacts on air quality particularly in densely urbanized and populated areas where
9 vehicle emissions are a major local source of ambient particulate matter. Engine type (i.e. fuel use) significantly impacts the
10 chemical characteristics of tailpipe emission and thus the distribution of engine types in traffic impacts measured ambient
11 concentrations. This study provides an estimation of the contribution of vehicles powered by different fuels (gasoline, diesel,
12 LPG) to carbonaceous submicron aerosol (PM₁) based on ambient aerosol mass spectrometer (AMS) and elemental carbon
13 (EC) measurements and vehicle count data in an urban inner city environment in Hong Kong with the aim to gauge the
14 importance of different engine types to particulate matter burdens in a typical urban street canyon. On an average per-vehicle-
15 basis, gasoline vehicles emitted 75% and 93% more organics than diesel and LPG vehicles respectively, while EC emissions
16 from diesel vehicles were 45% higher than those from gasoline vehicles. LPG vehicles showed no appreciable contributions
17 to EC and thus overall represented a small contributor to traffic-related primary ambient PM₁ despite their high abundance
18 (~30%) in the traffic mix. Total carbonaceous particle mass contributions to ambient PM₁ from diesel engines were only
19 marginally higher (~4%) than those from gasoline engines, which is likely an effect of recently introduced control strategies
20 targeted at commercial vehicles and buses. Overall, gasoline vehicles contributed 1.2 µg/m³ of EC and 1.1 µg/m³ of organics,
21 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
22 PM₁.

23
24

25 1. Introduction

26 1.1. Particulate matter from motor vehicles in urban areas

27 Emissions from on-road motor vehicles comprise gas-phase species (CO, CO₂, NO_x, SO₂) and volatile organic compounds
28 among them important precursors of secondary organic aerosol (SOA), as well primary particulate matter predominantly in
29 the fine (PM_{2.5}) and ultrafine particle size range (PM_{0.1}) mostly as carbonaceous species which encompass primary organic
30 aerosol (POA) components and elemental carbon (Giechaskiel et al., 2014). Increased acute occurrence and risk of chronic
31 development of cardiovascular and pulmonary diseases are important epidemiological effects of particulate matter inhalation
32 (Davidson et al., 2005;Pope, 2007;Valavanidis et al., 2008;World Health Organization, 2011). In urban areas with high
33 population and building densities, proximity to vehicle emissions poses a significant public health risk and renders paramount
34 importance to the characterization and quantification of vehicle emissions (Kumar et al., 2014;Uherek et al., 2010).

35 Their contribution to total ambient concentrations however remains elusive with considerable variability in measured emission
36 rates and species composition between as well as within vehicle classes from exhaust measurements in laboratory settings,
37 vehicle chase or portable emission measurement systems (PEMS) studies (Franco et al., 2013;Kwak et al., 2014;Karjalainen
38 et al., 2014;Giechaskiel et al., 2014;Alves et al., 2015) . These differences have been attributed to numerous influencing factors
39 such as vehicle age, fuel use, operational parameters, environmental conditions and the subsequent introduction of more
40 advanced engine technology (e.g. gasoline direct injection / GDI) and exhaust after-treatment (e.g. diesel particulate filters /
41 DPF) (May et al., 2014). Apart from combustion-related compounds, species originating from lubricating oils have been
42 identified as major components in primary gasoline and diesel exhaust and may be responsible for more than half of the total
43 in-use POA emissions (Worton et al., 2014).

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

49 Investigations on the impacts of motor vehicle emissions on ambient air pollution and related effects such as health implications
50 (Kheirbek et al., 2016;Zhang and Batterman, 2013;Levy et al., 2010) typically rely on combinations of source inventories with
51 dispersion modelling and combined physical-chemical models with significant uncertainties arising from the complexity of
52 the urban built environment (Kumar et al., 2011;Belcher et al., 2013), inventory currency and accuracy of emission factors
53 (Simon et al., 2008;Fuzzi et al., 2015). The latter is critically affected by the design of dynamometer test cycles with
54 discrepancies having been noted between type approval procedure derived emissions and on-road measured driving emissions,
55 which underlines the continued need to evaluate and monitor contributions of traffic emissions to urban air pollution by direct
56 ambient measurements (Harrison and Beddows, 2017). A dominant factor that affects both gas- and particle-phase species
57 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.,

58 2014;Kwak et al., 2014), i.e. the distribution of engine types in the traffic mix will have significant influence on ambient
59 concentrations. In the US gasoline fuel use in light duty vehicles is wide spread contributing to a greater dominance of gasoline-
60 related VOC and particle-phase organic emissions, whereas in Europe diesel-vehicle use prevails leading to higher ambient
61 burdens of black carbon (BC) (Zotter et al., 2014;Gentner et al., 2017). Evaluating the contribution of different vehicle groups
62 to exhaust related species in ambient measurements is therefore of vital interest to gauge their current importance to urban air
63 pollution and to acquire a baseline for assessing the efficacy of control policies and future effects of traffic mix and vehicle
64 technology changes.

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

74 **1.2. Background of vehicle emission studies in Hong Kong**

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

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

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

95 Generally, low overall organic carbon (OC) to elemental carbon (EC) ratios (0.6-0.8) which are typical for locations in direct
96 proximity to primary combustion sources were observed. In comparison, samples from urban rooftop sites exhibited lower
97 contributions of carbonaceous constituents (<50%) and were impacted more by oxidized secondary organic species with
98 correspondingly higher overall OC/EC ratios ~ 1.9 (Louie et al., 2005; Cheng et al., 2010; Lee et al., 2006). Impacts of long-
99 range transport of continental aerosol were evident in both higher total PM and higher OC concentrations in the winter months.

100 By contrast, seasonal variations of EC concentrations were weak. Higher EC concentrations were typically associated with
101 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.,
102 2010; Huang et al., 2014; Louie et al., 2005). While earlier studies (*samples dated pre-2005*) reported EC concentrations in the
103 range of 11-25 $\mu\text{g C m}^{-3}$ in $\text{PM}_{2.5}$ at the open roadside (Louie et al., 2005; Cheng et al., 2010; Lee et al., 2006), more recent
104 measurements (2012-2013) indicate a significant reduction in overall roadside EC burdens with concentrations $\sim 5 \mu\text{g C m}^{-3}$
105 (Huang et al., 2014; Sun et al., 2016; Lee et al., 2015). Vehicle-related OC and organic matter (OM) concentrations at a roadside
106 measurement site were estimated to $\sim 2 \mu\text{g C m}^{-3}$ and $\sim 7 \mu\text{g m}^{-3}$ respectively in 2012, accounting for one-third of total OC in
107 $\text{PM}_{2.5}$ and one-quarter of total $\text{PM}_{2.5}$ (Huang et al., 2014).

108 Contributions of traffic emissions to ambient gas-phase species were evaluated utilizing VOC canister samples collected in
109 locations with different dominant-vehicle types in Hong Kong in 2003 (Ho et al., 2013). Propane, n- and i-butane accounted
110 for over 80% of identified species in the LPG vehicle dominated samples, while toluene was the most abundant species in the
111 gasoline vehicle dominated samples. Ethene and ethyne were enriched in diesel exhaust dominated samples. Contributions of
112 vehicle-related VOCs to ozone formation were evaluated from the maximum incremental reactivity, with toluene (15-17%)
113 and propene (7-8%) accounting for the largest shares (Ho et al., 2013).

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

124 2001;Lowenthal et al., 1994;Kirchstetter et al., 1999) in other parts of the world, they are unlikely to reflect the properties of
125 contemporary mobile road traffic emissions in Hong Kong with substantial changes in vehicle fleet composition and the
126 emergence of more advanced engine and emission control technologies over the last 15 years.

127 A mobile measurement platform with an array of on-board PM and gas monitors was deployed in early 2012 for a more up-
128 to-date characterization of fuel-based emission factors of PM, NO_x, and butane by chasing vehicle plumes on major roadways
129 and highways (Ning et al., 2012). The measurements focused on the evaluation of heavy duty diesel trucks (HDDV), franchised
130 diesel buses (DB) and LPG fueled light buses (LB) with plumes sampled at cruise speed to mitigate biases from engine load-
131 dependent emission variability. HDDVs were the highest emitters of BC (1.6 g kg⁻¹ of fuel) followed by diesel buses (1.1 g
132 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 10¹⁵
133 kg⁻¹ of fuel, 3.1 x 10¹⁵ kg⁻¹ of fuel and 0.9 x 10¹⁵ kg⁻¹ of fuel for HDDVs, diesel buses and LPG light buses respectively.

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

141 Despite road traffic representing a major local source of ambient particulate matter in Hong Kong, characterization studies on
142 vehicle emissions in Hong Kong remain sparse. While most previous studies evaluated traffic exhaust contributions in confined
143 environments (tunnels), focal locations (e.g. bus terminus, taxi stand) or by plume chase, this work provides a quantitative
144 estimation of vehicle exhaust related carbonaceous aerosol contributions and their relationship to traffic flow characteristics
145 from ambient measurements in a more complex urban environment. We aim to complement previous studies which only
146 estimated overall traffic-related aerosol contributions from online and offline ECOC measurements at different time resolutions
147 (from hourly to 24h samples) (Louie et al., 2005;Huang et al., 2014) and online mass spectrometric methods (Sun et al.,
148 2016;Lee et al., 2015) at the same location by combining resolved traffic-related organic aerosol concentrations from factor
149 analysis of aerosol mass spectrometer (AMS) data, online EC measurements and traffic count data to gain an improved
150 understanding of the influence of traffic composition on traffic-related submicron carbonaceous aerosol.

151 2. Methodology

152 Sampling of ambient particulate matter by a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS,
153 Aerodyne Inc.) took place in spring 2013 (7 March 2013 to 15 May 2013) and summer 2013 (16 May 2013 to 19 July 2013).
154 Measurements were undertaken at a ground-level site on a pedestrian island at a road junction in the Mong Kok (MK) district
155 of the densely populated and built-up Kowloon peninsula. Additional data such as meteorological data (wind, temperature,
156 relative humidity, solar irradiation), concentrations of volatile organic compounds (VOCs), as well as trace gases (NO_x, SO₂,
157 and O₃) were available from the adjacent air quality monitoring site (AQMS) operated by the Environmental Protection
158 Department (EPD) of the HKSAR Government on the same road divider island. Details on employed instrument model and
159 sampling methodologies are available in other dedicated publications (Lee et al., 2015; Environmental Protection Department,
160 2016; Sun et al., 2016). Elemental and organic carbon (EC, OC) were measured (semi-continuous thermo-optical ECOC
161 analyzer RT-3131, Sunset Inc., USA) at the AQMS at 1h time resolution and at similar sampling height (~2m) employing a
162 modified National Institute for Occupational Safety and Health (NIOSH) method 5040 protocol which has been described in
163 a previous study (Huang et al., 2014). Concentrations of EC and OC in PM₁ were estimated by converting the raw PM_{2.5}
164 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
165 sampling studies (0.75-0.85) in Hong Kong (Cheng et al., 2006; Lee et al., 2006).

166 AMS data were treated according to general AMS data treatment principles (DeCarlo et al., 2006; Jimenez et al., 2003) with
167 standard software packages (SQUIRREL v1.53G, PIKA v1.12G). Analysis of the unit-mass resolution mass spectra yielded
168 non-refractory submicron particle species concentrations of major inorganic constituents (SO₄, NO₃, NH₄, Chl) and total
169 organics at a base time resolution of 10 min. Positive Matrix Factorization (PMF) was used to deconvolute high-resolution
170 organic mass spectra acquired at 10 min time resolution following recommended PMF guidelines for AMS data (Zhang et al.,
171 2011) with the AMS PMF analysis toolkit (Ulbrich et al., 2009). Six organic aerosol (OA) factors were identified encompassing
172 three secondary organic aerosol (SOA) and three primary organic aerosol (POA) factors of which one was attributed to traffic
173 emissions (hydrocarbon-like organic aerosol, HOA (Zhang et al., 2005)) and two to cooking activities. OA factor
174 concentrations were established using the fractional factor contributions from the high-resolution PMF analysis and the total
175 submicron organics concentrations from the unit-mass resolution analysis. Both unit- and high-resolution mass spectral derived
176 data were averaged to 1h time resolution to match ECOC measurements and traffic count data (*see below*), assessed from 28
177 May to 31 May, 2013. Average species concentrations measured during this time as well as the seasonal averages (spring,
178 summer) from the whole measurement campaign are summarized in Table 1. Further specific details on instrument parameters,
179 data analysis and an overview of the general characteristics of submicron particulate matter from the same campaign discussing
180 the whole measurement period can be found in a previous publication (Lee et al., 2015).

181 Details on traffic composition are usually obtained by concurrent video image recordings and subsequent manual assessment
182 of vehicle numbers and identification of vehicle types with classifications e.g. by vehicle function (Wang et al., 2016) or
183 broader groups such as fuel types (Cui et al., 2016). In this work, vehicle data were obtained from the Hong Kong government,

184 which conducted a counting exercise during the sampling period over three weekdays (noon 28 May 2013 to noon 31 May
185 2013, Tue to Fri) using automated license plate recognition (ALPR) with four infrared cameras on-site. Traffic along Lai Chi
186 Kok road was monitored with three cameras monitoring all three lanes in one direction (out- or inbound) and the remaining
187 camera monitoring one lane of the opposite direction. Camera positions and the monitored single lane were swapped in 2-3h
188 intervals. Non-personalized data on registered vehicles were obtained, including license class (*vehicle type*), year of
189 manufacture (*vehicle age*) and engine type (*gasoline, diesel, LPG, others*), thus providing a direct linkage to actual vehicle
190 inventory data and circumventing the need for manual assessments. The obtained data were pooled by engine type (i.e. type
191 of fuel used) to construct an hourly time series of diesel, gasoline and LPG powered vehicles.

192 Measured HOA and EC during the traffic counting period were decomposed by multiple linear regression (MLR) based on the
193 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
194 diesel, gasoline and LPG powered vehicles representing the independent regression variables. To reduce the skewing impact
195 of scattered spikes in vehicle count number, HOA and EC concentrations, the time series were subjected to three-point box
196 smoothing prior to regression analysis. Multiple linear regression was performed for two and three factor solutions and with a
197 constrained zero intercept. A non-zero intercept may not be physically meaningful as it requires the assumption of a constant
198 background level of HOA and EC regardless of changes in surface wind, air mass influence or diurnal changes in background
199 source strength. Contrarily, traffic is largely homogenous in the vicinity of the sampling site (dense inner city traffic) and other
200 possible combustion sources were sufficiently removed (i.e. shipping with ~2km of straight-line distance to the nearest
201 coastline) and shielded by complex and tall urban geometry from the ground-level sampling site, i.e. impacts of transport on
202 measured traffic-related carbonaceous constituents can be assumed to be of minor influence. MLR solutions were assessed per
203 statistical significance of resolved regression coefficients, adjusted R^2 of the regression and the distribution of residuals.
204 Further details are discussed in the corresponding discussion section 3.3.1.

205 **3. Results**

206 **3.1. Chemical and temporal characteristics of traffic-related carbonaceous species**

207 Organic aerosol components were resolved by PMF from the organic part of the measured high-resolution mass spectra
208 acquired during the measurement campaign and scaled to the measured total organic species concentrations based on the unit-
209 mass resolution acquisition mode (Lee et al., 2015). Hydrocarbon-like organic aerosol (HOA) was resolved and identified as
210 the traffic exhaust related factor, exhibiting chemical characteristics typically associated with freshly emitted primary organic
211 aerosol from fossil fuel combustion (Zhang et al., 2005) and with appreciable correlations to other traffic-related species such
212 as NO_x or pentanes (Lee et al., 2015). Its mass spectrum (Fig. S1a in the Supplement) is dominated by ions of saturated
213 hydrocarbons ($\text{C}_n\text{H}_{2n+1}$ series) and its elemental composition is largely void of oxygenated components with correspondingly
214 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

215 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.,
216 2010;Aiken et al., 2009;Setyan et al., 2012).

217 The highest fractions of HOA among total organic particulate matter in PM₁ were observed with southeasterly and
218 northwesterly surface winds following the street canyon axis along Lai Chi Kok Road (Fig. S2a in the Supplement). Average
219 HOA mass concentrations varied between spring (3.5 $\mu\text{g m}^{-3}$) and summer (2.0 $\mu\text{g m}^{-3}$), with an overall campaign average of
220 2.8 $\mu\text{g m}^{-3}$. Diurnal variations of HOA were substantial, with mass concentrations rising sharply in the early morning (06:00),
221 increasing almost linearly during the morning rush hour at rates between 0.6 and 0.8 $\mu\text{g m}^{-3} \text{ h}^{-1}$ and leveling off beyond 09:00
222 (Fig. 1a). 90% of the daily HOA was accumulated between the start of the rush hour and midnight. Minimum HOA
223 concentrations in the low traffic period (00:00 to 06:00) amounted to $\sim 1.7 \mu\text{g m}^{-3}$ in spring and $\sim 0.7 \mu\text{g m}^{-3}$ in summer. Average
224 post rush-hour concentrations (09:00 to 00:00) were 4.1 $\mu\text{g m}^{-3}$ and 2.6 $\mu\text{g m}^{-3}$ in spring and summer respectively.

225 Elemental carbon (EC) and organic carbon (OC) in PM_{2.5} were measured semi-continuously at the adjacent AQMS following
226 Huang et al. (Huang et al., 2014) and converted to PM₁ as described in the methodology section. In the following, we refer to
227 the PM₁ concentrations for discussion. Diurnal variations of EC were similar to those of HOA, however, nominal
228 concentrations were not affected by the seasonal transition. This led to significantly different EC/HOA ratios of 0.86 in spring
229 and 1.78 in summer (Fig. S1b in the Supplement). As the measurement location falls within the Kowloon urban area, which is
230 characterized by uniform annual traffic flows (Transport Department, 2014), the observed decrease in HOA concentrations
231 was more likely due to significant amounts of HOA being semi-volatile and partitioning in the gas-phase portion of the exhaust
232 emissions in summer than changes in traffic flows. This is also consistent with the trend in relative diurnal variation of HOA
233 (depicted cumulatively in Fig. 1b) which remained constant across seasons despite the seasonality in nominal concentrations,
234 and the overall negative correlation of HOA mass concentration and temperature (Fig. S2b in the Supplement). The semi-
235 volatility of HOA in this study is inferred from the observed trend in ambient mass concentrations and affirms more recent
236 studies on the partitioning behavior of POA components. Evaporation of semi-volatile components upon dilution decreases
237 ambient POA concentrations considerably and is expected to be temperature-dependent (Robinson et al., 2007), as is the case
238 in this study. Measurements of thermally denuded particles of primary origin by TD-AMS indicate that POA components can
239 exhibit properties of semi-volatility on a scale similar to SOA in urban environments (Huffman et al., 2009a;Huffman et al.,
240 2009b), and more recent studies suggest that HOA possesses a relatively wide volatility distribution with up to 50-60% of its
241 mass consisting of semi-volatile organic compounds (Cappa and Jimenez, 2010;Paciga et al., 2016). As noted earlier, in this
242 study the seasonal decrease in submicron particle-phase organic mass concentrations (HOA) by 40% was associated with a
243 substantial increase in mean temperature by 7°C between spring (23°C \pm 3°C) and summer (30°C \pm 2°C).

244 Both HOA and EC exhibited significant reductions in overall mass concentrations on Sundays between 14% and 27%,
245 compared to the rest of the week (Monday to Saturday) as shown in Fig. 1d. Days with less than 75% (>18h) data availability
246 were excluded from both time series in the analysis to obviate bias in the daily averaging from their strong diurnal patterns, as
247 opposed to the original data presented by Lee et al. (2015) where the entire unabridged time series were used. Mean reductions
248 in nominal mass concentrations were generally quite similar in the range of 0.8 – 1.0 $\mu\text{g m}^{-3}$ for EC and $\sim 0.5 \mu\text{g m}^{-3}$ for HOA.

249 The greater overall reduction in EC indicates a greater reduction in the number of diesel vehicles on Sundays, owing to reduced
250 commercial traffic and lower bus frequency. Contributions of vehicle emissions to ambient organic aerosol have been derived
251 from one-year long measurements at the same site based on analysis of semi-continuous ECOC measurements (Huang et al.,
252 2014). The EC tracer method (Turpin and Huntzicker, 1991; Lim and Turpin, 2002) was employed which evaluates minimum
253 OC/EC ratios, and OC and EC measurements during time periods of high primary source strength and low photochemical
254 activities when OC is mainly of local origin and attributable to the dominant primary sources in the vicinity. A characteristic
255 vehicle-related OC/EC value (OC/EC_{vehicle}) of 0.5 was derived from the 5% lowest summer OC/EC ratio data to approximate
256 organic vehicular emissions. The reported monthly average primary organic matter concentrations from traffic in $PM_{2.5}$ (using
257 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
258 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
259 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
260 with the overall average concentration of traffic-related HOA ($2.8 \mu\text{g m}^{-3}$) covering the entire AMS measurement campaign
261 (March – July 2013). However, we observed significantly different concentrations depending on the season with much higher
262 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
263 traffic-related organics is due to the use of a fixed conversion factor directly dependent on ambient EC concentration which
264 did not exhibit a strong seasonal behavior and thus could not capture the same trends observed from the AMS measurements.
265 The derivation of the OC/EC_{vehicle} ratio is subject to further uncertainty due to the substantial influence of cooking-related
266 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
267 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
268 ratios.

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

273 **3.2. Traffic and vehicle flow characteristics**

274 During the three-day counting exercise, a total of 21,048 vehicles were registered (~7016 vehicles per day). This accounts for
275 approximately 18% of the annual average daily traffic (A.A.D.T.) of 38,220 vehicles estimated in the 2013 Annual Traffic
276 Census (Transport Department, 2014). The diurnal variation of the counted vehicle number, resolved by broad vehicle classes,
277 and their varying contribution to the vehicle mix during the day are depicted in Fig. S2c in the Supplement. The maximum
278 hourly vehicle number occurred towards the end of the rush-hour (~9:00) for most vehicle classes (except “*other vehicles*”,
279 which mostly comprised motorcycles). For the evening rush-hour (17:00-19:00), only the private car category exhibited a peak
280 in vehicle number. Overall, the post rush-hour total vehicle count remained stable throughout the day, and only fell off after

281 19:00, largely due to a strong decrease in the number of goods and private vehicles. The variation in the fractional contribution
282 of different vehicles types over the day resembles that assessed by the Transport Department (Transport Department, 2014) at
283 the closest core traffic counting station at Nathan Road (Fig. S2c in the Supplement) and affirms that the measured traffic
284 composition during the 72h counting period at Mong Kok was largely representative. At the Mong Kok site, smaller vehicles
285 (*cars and taxis*) accounted for about 60% of total registered vehicles, while heavier vehicles (*buses, vans, trucks*) made up
286 most of the remainder.

287 Total HOA concentrations showed good correlations ($R_{pr}>0.6$) with most of the measured vehicle-related VOCs, NO_x and EC
288 concentrations as well as the total number of passing vehicles during the counting period (Fig. 2). As with the overall campaign
289 data, CO did not correlate with either HOA or EC, indicating the impact of significant non-traffic-related CO sources in the
290 Mong Kok area. Among the counted buses, light buses and goods vehicle, over 96% were running on diesel engines, while
291 99.9% of counted taxis utilized LPG engines. Private vehicles were almost exclusively powered by gasoline engines (99.4%)
292 due to government restrictions on the import and use of diesel-powered private vehicles at the time of this study.

293 3.3. Estimation of engine-type contributions

294 3.3.1. Multiple Linear Regression of HOA and EC

295 Utilizing the detailed information on average daily traffic composition at the measurement site from the counting exercise,
296 contributions of exhaust from different engine types to overall ambient HOA and EC concentrations can be evaluated. Multiple
297 linear regression was carried out with the concentration time series of HOA and EC during the traffic counting period as the
298 dependent variables, the pooled sum of counted diesel, gasoline and LPG vehicles as the independent variables, and a
299 constrained zero intercept, assuming negligible transport of HOA and EC (*see Methodology section*).

300 Two factor solutions (diesel + gasoline), as well as three factor solutions (diesel + gasoline + LPG), were considered, as LPG
301 vehicles are expected to contribute less to particle-phase and more to gas-phase emissions (Faiz et al., 1996). The key statistical
302 output parameters (adjusted R², regression coefficients, p-values) of the multiple linear regression analysis for HOA and EC
303 for two and three factor models are presented in Table S1 in the Supplement. For HOA, both two and three factor models
304 yielded acceptable results with all resolved coefficients statistically significant at the 95% confidence level ($p<0.05$). Exclusion
305 of LPG vehicles (two factor model) led to additional apportionment of HOA to both gasoline (+25%) and diesel (+18%)
306 vehicles. Overall model performance improved only slightly when moving from the two factor solution ($R^2_{adj}=0.86$) to the
307 three factor solution ($R^2_{adj}=0.90$). Comparative studies on particulate vehicle emissions have reported non-negligible particle
308 mass contributions of LPG vehicles. Estimated particle mass emission factors from LPG passenger vehicles were similar (Chan
309 et al., 2007) or ~30% lower (Ristovski et al., 2005) compared to gasoline powered models in chassis dynamometer runs. In
310 on-road environments, absolute particle number concentrations in bus exhaust plumes were lower in samples from CNG and
311 LPG fueled buses compared to diesel and DME (dimethyl ether) fueled buses, but still 10 times higher than typical ambient
312 background particle concentrations (Kwak et al., 2014). Evaluating HOA concentrations measured between 0:00 and 5:00

313 when LPG vehicles dominated the total vehicle population (45-60% of total counted vehicles) in our study shows a clear
314 positive relationship between ambient HOA and LPG vehicle count (Fig. S3a in the Supplement). The two factor MLR model
315 tends to underestimate measured HOA mass concentrations with the distribution of absolute residuals (*difference of*
316 *reconstructed and measured HOA mass concentration*) biased to negative values whereas the three factor model yielded more
317 normally distributed residuals (Fig. S3b, c in the Supplement). We thus consider the three factor solution including LPG
318 vehicles as a more appropriate representation for the deconvolution of the ambient HOA mix from our measurements. For EC,
319 a three factor representation could not resolve variable coefficients that were all statistically significant at the 95% confidence
320 level yielding $p > 0.05$ for the gasoline vehicle factor. While both diesel and gasoline vehicle number correlated appreciably
321 with EC (Fig. S4c in the Supplement), the number of LPG vehicles lacked a corresponding relation and stayed almost constant
322 over the range of measured EC concentrations, rendering the two factor model the more appropriate representation in
323 deconvoluting measured ambient EC.

324 As discussed in the methodology section, three point box smoothing was applied to the vehicle counts and carbonaceous PM₁
325 species concentrations to reduce biasing of resolved regression parameters induced from spikes in the time series (Figure S5
326 in the Supplement). Comparing MLR results from the smoothed and raw time series data (Table S2 in the Supplement), impacts
327 on the variable coefficients and their variability (standard deviations) were minor and factor statistical significance was not
328 detrimentally affected. The smoothed data set overall yielded better results with higher adjusted R² as well as lower average
329 and summed residual values. The smoothing process thus did not incur substantial additional uncertainties in the deconvolution
330 of HOA and EC.

331 3.3.2. Analysis of engine-type reconstructed carbonaceous components

332 The time series of measured and reconstructed HOA and EC concentrations based on the regression coefficients are depicted
333 individually in Fig. S6a and S6c in the Supplement and in combination in Fig. 3 to represent the sum of motor vehicle related
334 primary carbonaceous particulate compounds, not including possible additional SOA species formed through subsequent
335 atmospheric processing of gas- and particle-phase species in tailpipe exhaust. Gasoline vehicles accounted for more than 45%
336 of total HOA, while diesel and LPG-fueled vehicles were responsible for similar shares of 25-29% (Fig. S6b in the
337 Supplement). Over 60% of total EC in PM₁ was accounted for by diesel vehicles (Fig. S6d in the Supplement). The sum of
338 HOA and EC in PM₁ represent approximately the total submicron primary carbonaceous aerosol contributions from road
339 traffic. Despite the limited time frame of the counting exercise, the MLR factors adequately resolved the overall trend in EC
340 and HOA concentration (Fig. 3a) and diurnal variation (Fig. 3b). Relative residuals from the reconstructed sum of HOA and
341 EC were $\leq 25\%$ with overestimation (*Residuals* < 0) predominant in the later afternoon hours between 14:00 and 18:00, while
342 significant underestimation (*Residuals* > 0) was observed mostly in the late evening hours between 19:00 and 0:00. The semi-
343 volatile character of HOA would contribute to the observed reduced afternoon particle-phase concentration and conversely
344 enhanced nighttime particle-phase concentrations. Averaged over the whole 72h period, measured and reconstructed

345 carbonaceous submicron PM_1 differed by only 4% indicating that the overall reconstruction was not affected considerably by
346 these diurnal trends in residuals. Vehicle emission factors are typically based on the concentration of emitted species per unit
347 of consumed fuel (derived from CO_2 measurements) or per driven distance. As concurrent CO_2 measurement data were not
348 available, emission factors were not evaluated in this study. Instead, the relative emission behavior of different vehicle groups
349 and their impacts on ambient PM concentrations were approximated by normalizing the resolved engine-specific carbonaceous
350 primary aerosol concentrations by the total number of vehicles in each category (Fig. 3b). On a per-vehicle basis (Fig. 3b),
351 gasoline vehicles emitted 75% more HOA than diesel vehicles and 93% more HOA than LPG vehicles, while EC emissions
352 from diesel vehicles were 45% higher than those from gasoline vehicles. Therefore, on average gasoline and diesel vehicles
353 emitted similar total amounts of primary carbonaceous PM_1 and each about three times more than LPG powered vehicles. In
354 terms of ambient concentrations, gasoline vehicles accounted for 78% more ambient HOA than diesel vehicles and 56% more
355 ambient HOA than LPG-fueled vehicles. As opposed diesel vehicles accounted for 62% more ambient EC than gasoline-
356 powered vehicles. Each diesel and gasoline vehicles contributed 75-85% more carbonaceous PM_1 mass than LPG vehicles,
357 which despite making up ~30% of total counted traffic accounted for less than 13% of traffic-related primary PM_1 (Fig. 3e).
358 It must be noted that these data represent the total average of the encountered diesel, gasoline and LPG vehicle fleet,
359 notwithstanding that each of these broad classes is inhomogeneous consisting of a wide variety of vehicles of different size,
360 engine power and age, and thus different individual emission characteristics.

361 As the fractional contributions of diesel and gasoline vehicles to ambient HOA and EC varied substantially, i.e. diesel vehicles
362 emitted almost three times as much EC as HOA ($EC/HOA=2.85$) and gasoline vehicles contributed similar mass concentrations
363 to both HOA and EC ($EC/HOA=1.02$). This caused significant diurnal variations in the EC/HOA ratio (Fig. S1c in the
364 Supplement) with low values in the nighttime hours, where LPG- and gasoline-fueled vehicles were more dominant and highest
365 values occurring at the tail of the morning rush-hour where fractions of goods vehicles and buses among overall traffic were
366 highest. Previous measurements of vehicle emissions in a highway road tunnel in the San Francisco Bay Area (Dallmann et
367 al., 2014) employing a soot-particle aerosol mass spectrometer (SP-AMS) yielded a similar characteristic ratio of black carbon
368 to organic aerosol ($BC/OA\sim 2.6$) for diesel truck plumes and a smaller ratio of $BC/OA<0.1$ for gasoline vehicle plumes, while
369 filter samples at the same location from light duty vehicles location exhibited BC/OA ratios of ~0.7 for light-duty vehicles and
370 ~2.1 for medium and heavy diesel trucks (Ban-Weiss et al., 2008). Dynamometer studies mostly reported EC/OC values for
371 gasoline powered vehicles < 1 (Alves et al., 2015;Geller et al., 2006) indicating overall a greater importance of organics in
372 particulate emissions.

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

379 2010). The observed seasonal variability of particle-phase organic concentrations due to partitioning of semi-volatile
380 components into the gas-phase would also decrease particle-phase HOA concentrations in warmer seasons, thus elevating the
381 observed ambient EC/HOA ratio with our measurements taking place in summer. While this would affect all engine types, the
382 extent may vary with the distribution of semi-volatile species in the respective tailpipe emissions of different vehicle groups.
383 Due to the characteristics of the sampling site (inner-city urban road traffic and proximity to both a road junction and pedestrian
384 crossing) sampled emissions include a considerable fraction of variable and higher engine loads during the acceleration phase
385 whereas road tunnel environments are characterized by largely constant engine loads and traveling speeds. As gasoline vehicles
386 are typically not equipped with particle filters, they have been shown to emit significant amounts of EC under unstable engine
387 loads (Karjalainen et al., 2014).

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

394 At the same time, various control schemes targeting diesel vehicle emissions have been introduced in recent years. In Hong
395 Kong these included inter alia an incentive scheme in 2010 for the replacement of diesel commercial vehicles, the retrofitting
396 of diesel particulate filters (DPF) on pre-Euro IV buses and subsequent replacement of older buses with Euro IV, V and VI
397 standard models (Environment Bureau, 2013; Environmental Protection Department, 2013). $PM_{2.5}$ emission factors determined
398 from dynamometer test runs of various vehicle types in Hong Kong were generally higher for diesel vehicles compared to
399 gasoline and LPG vehicles (Chan et al., 2007), while we observed similar total particle mass emission on a per-vehicle basis
400 for diesel and gasoline vehicles in this study. The dynamometer tests were conducted on relatively old vehicles (*newest date
401 of manufacture: 2001*) and are unlikely to adequately reflect the ambient vehicle mix in our study, particularly with regard to
402 the implementation of recent emission control programs. New engine technologies for diesel vehicles, such as DPFs, have
403 been shown to greatly reduce both EC and OC emissions (Alves et al., 2015) thus leading to overall little total particle mass
404 emissions (Li et al., 2014; Quiros et al., 2015) partially due to shifts in the particle size distributions towards a greater fraction
405 of particles in the ultrafine mode (Giechaskiel et al., 2012). The Euro III, IV and V standards for trucks and buses were
406 introduced in late 2000, late 2005 and late 2008 respectively. While the year of manufacture does not directly infer compliance
407 to a specific emission standard, an approximation of the fraction of vehicles fulfilling a certain standard can be made by
408 assuming that vehicles produced between 2001 and 2005, between 2006 and 2008 and between 2009 and 2013 very likely
409 comply with the Euro III, IV and V standards respectively. In this case, it is assumed that emission standards were immediately
410 or had already been adopted in vehicle models in the corresponding year of manufacture. Figure S7 in the Supplement depicts
411 the number and proportion of vehicles of certain years of manufacture and their assumed Euro standard. For goods vehicles,
412 52% of counted vehicles were built between 2005 and 2013 (i.e. likely fulfilling Euro IV and V), while for buses the proportion

413 was slightly lower at 33%. With these two vehicle groups representing the bulk of diesel powered vehicles, an estimated 40%
414 of diesel vehicles complied with Euro IV and Euro V standards during the time of our ambient measurements, further
415 rationalizing the relatively low per-vehicle contribution of diesel vehicles to ambient exhaust-derived carbonaceous PM₁ in
416 this study.

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

420 **4. Conclusion**

421 Primary submicron carbonaceous aerosol was measured by combined HR-ToF-AMS and ECOC measurements, supplemented
422 by various VOCs and other gas-phase species, at an inner-city urban roadside sampling site in the Mong Kok District in Hong
423 Kong. Organic species concentrations (HOA) were of similar magnitude as traffic-related organic matter estimated previously,
424 but with strong seasonal dependency with on average 40% lower concentrations in summer (2.0 µg/m³) compared to spring
425 (3.5 µg/m³) likely due to greater fractions of semi-volatile species remaining in the gas-phase in the warmer season. EC
426 concentrations remained at similar levels throughout the sampling period, leading to significantly different average EC/HOA
427 ratios in spring (0.86) and summer (1.78). Sunday concentrations of both HOA and EC were 14-27% lower than during the
428 rest of the week (Mon-Sat) reflecting significant reductions in traffic volume on Sundays. Larger relative reductions in EC
429 compared to HOA indicate a greater reduction in the number of diesel vehicles, e.g. commercial trucks and buses.

430 Detailed vehicle counting data acquired during a 72h counting exercise in late May 2013 were utilized to decompose HOA
431 and EC concentrations into vehicle-type factors (gasoline-, diesel- and LPG-powered vehicles) by multiple linear regression
432 analysis. Gasoline vehicles contributed similar amounts of EC and HOA, while diesel vehicles emitted predominantly EC and
433 to a lesser extent HOA. On an average per-vehicle basis, contributions of diesel and gasoline vehicles to carbonaceous PM₁
434 were similar, contrary to previous studies which attributed higher particulate matter emissions to diesel vehicles. This clear
435 reduction is likely due to recent control strategies targeted at commercial vehicles and buses, which represent the bulk of diesel
436 powered vehicles at the measurement site. This becomes especially important in view of the growing number of higher Euro
437 standard diesel vehicles which, while effectively reducing overall emitted particle mass may emit similar number
438 concentrations of particles with particle sizes having been shown to shift more into the ultrafine region (Tartakovsky et al.,
439 2015). The number of LPG-vehicles did not exhibit a significant correlation with EC concentrations and was thus considered
440 to be a negligible EC source. Its contributions to ambient primary traffic-related PM₁ were small (13%) despite representing
441 about one-third of total counted vehicles.

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

445 **Acknowledgments**

446 This work was supported by the Environment and Conservation Fund of Hong Kong (project number ECWW09EG04). Chak
447 K. Chan gratefully acknowledges the startup fund of the City University of Hong Kong.

448 **Disclaimer**

449 The opinions expressed in this paper are those of the author and do not necessarily reflect the views or policies of the
450 Government of the Hong Kong Special Administrative Region, nor does mention of trade names or commercial products
451 constitute an endorsement or recommendation of their use.

452 **References**

- 453 Aiken, A. C., Salcedo, D., Cubison, M. J., Huffman, J. A., DeCarlo, P. F., Ulbrich, I. M., Docherty, K. S., Sueper, D.,
454 Kimmel, J. R., Worsnop, D. R., Trimborn, A., Northway, M., Stone, E. A., Schauer, J. J., Volkamer, R. M., Fortner,
455 E., de Foy, B., Wang, J., Laskin, A., Shutthanandan, V., Zheng, J., Zhang, R., Gaffney, J., Marley, N. A., Paredes-
456 Miranda, G., Arnott, W. P., Molina, L. T., Sosa, G., and Jimenez, J. L.: Mexico City aerosol analysis during
457 MILAGRO using high resolution aerosol mass spectrometry at the urban supersite (T0) - Part 1: Fine particle
458 composition and organic source apportionment, *Atmos. Chem. Phys.*, 9, 6633-6653, [https://doi.org/10.5194/acp-9-](https://doi.org/10.5194/acp-9-6633-2009)
459 [6633-2009](https://doi.org/10.5194/acp-9-6633-2009), 2009.
- 460 Alves, C. A., Lopes, D. J., Calvo, A. I., Evtugina, M., Rocha, S., and Nunes, T.: Emissions from light-duty diesel and
461 gasoline in-use vehicles measured on chassis dynamometer test cycles, *Aerosol Air Qual. Res.*, 15, 99-116,
462 <https://doi.org/10.4209/aaqr.2014.01.0006>, 2015.
- 463 Ban-Weiss, G. A., McLaughlin, J. P., Harley, R. A., Lunden, M. M., Kirchstetter, T. W., Kean, A. J., Strawa, A. W.,
464 Stevenson, E. D., and Kendall, G. R.: Long-term changes in emissions of nitrogen oxides and particulate matter
465 from on-road gasoline and diesel vehicles, *Atmos. Environ.*, 42, 220-232,
466 <https://doi.org/10.1016/j.atmosenv.2007.09.049>, 2008.
- 467 Belcher, S., Coceal, O., Hunt, J., Carruthers, D., and Robins, A.: A review of urban dispersion modelling, ADMLC, UK,
468 2013.
- 469 Cappa, C. D., and Jimenez, J. L.: Quantitative estimates of the volatility of ambient organic aerosol, *Atmos. Chem. Phys.*,
470 10, 5409-5424, <https://doi.org/10.5194/acp-10-5409-2010>, 2010.
- 471 Chan, T. L., Ning, Z., Wang, J. S., Cheung, C. S., Leung, C. W., and Hung, W. T.: Gaseous and Particle Emission Factors
472 from the Selected On-Road Petrol/Gasoline, Diesel, and Liquefied Petroleum Gas Vehicles, *Energy & Fuels*, 21,
473 2710-2718, <https://doi.org/10.1021/ef070172i>, 2007.

474 Cheng, Y., Ho, K. F., Lee, S. C., and Law, S. W.: Seasonal and diurnal variations of PM_{1.0}, PM_{2.5} and PM₁₀ in the
475 roadside environment of hong kong, China Particuology, 4, 312-315, [https://doi.org/10.1016/S1672-](https://doi.org/10.1016/S1672-2515(07)60281-4)
476 2515(07)60281-4, 2006.

477 Cheng, Y., Lee, S. C., Ho, K. F., Chow, J. C., Watson, J. G., Louie, P. K. K., Cao, J. J., and Hai, X.: Chemically-specified
478 on-road PM_{2.5} motor vehicle emission factors in Hong Kong, Sci. Total Environ., 408, 1621-1627,
479 <https://doi.org/10.1016/j.scitotenv.2009.11.061>, 2010.

480 Cui, M., Chen, Y., Tian, C., Zhang, F., Yan, C., and Zheng, M.: Chemical composition of PM_{2.5} from two tunnels with
481 different vehicular fleet characteristics, Sci. Total Environ., 550, 123-132,
482 <https://doi.org/10.1016/j.scitotenv.2016.01.077>, 2016.

483 Dallmann, T. R., Onasch, T. B., Kirchstetter, T. W., Worton, D. R., Fortner, E. C., Herndon, S. C., Wood, E. C., Franklin, J.
484 P., Worsnop, D. R., Goldstein, A. H., and Harley, R. A.: Characterization of particulate matter emissions from on-
485 road gasoline and diesel vehicles using a soot particle aerosol mass spectrometer, Atmos. Chem. Phys., 14, 7585-
486 7599, <https://doi.org/10.5194/acp-14-7585-2014>, 2014.

487 Davidson, C. I., Phalen, R. F., and Solomon, P. A.: Airborne particulate matter and human health: A review, Aerosol Sci.
488 Technol., 39, 737-749, <https://doi.org/10.1080/02786820500191348>, 2005.

489 DeCarlo, P. F., Kimmel, J. R., Trimborn, A., Northway, M. J., Jayne, J. T., Aiken, A. C., Gonin, M., Fuhrer, K., Horvath, T.,
490 Docherty, K. S., Worsnop, D. R., and Jimenez, J. L.: Field-deployable, high-resolution, time-of-flight aerosol mass
491 spectrometer, Anal. Chem., 78, 8281-8289, <https://doi.org/10.1021/ac061249n>, 2006.

492 Ensberg, J. J., Hayes, P. L., Jimenez, J. L., Gilman, J. B., Kuster, W. C., de Gouw, J. A., Holloway, J. S., Gordon, T. D.,
493 Jathar, S., Robinson, A. L., and Seinfeld, J. H.: Emission factor ratios, SOA mass yields, and the impact of
494 vehicular emissions on SOA formation, Atmos. Chem. Phys., 14, 2383-2397, [https://doi.org/10.5194/acp-14-2383-](https://doi.org/10.5194/acp-14-2383-2014)
495 2014, 2014.

496 Environment Bureau: A Clean Air Plan For Hong Kong, HKSAR Government,, Hong Kong, 2013.

497 Environmental Protection Department: Cleaning the Air at Street Level:
498 http://www.epd.gov.hk/epd/english/environmentinhk/air/prob_solutions/cleaning_air_atroad.html, access: 1/2/2014,
499 2013.

500 Environmental Protection Department: Air Quality Monitoring Stations: [http://www.aqhi.gov.hk/en/monitoring-network/air-](http://www.aqhi.gov.hk/en/monitoring-network/air-quality-monitoring-stations.html)
501 [quality-monitoring-stations.html](http://www.aqhi.gov.hk/en/monitoring-network/air-quality-monitoring-stations.html), access: 27-2-2016, 2016.

502 Faiz, A., Weaver, C. S., and Walsh, M. P.: Air pollution from motor vehicles : standards and technologies for controlling
503 emissions, edited by: Weaver, C. S., and Walsh, M. P., World Bank, Washington, D.C, 1996.

504 Franco, V., Kousoulidou, M., Muntean, M., Ntziachristos, L., Hausberger, S., and Dilara, P.: Road vehicle emission factors
505 development: A review, Atmos. Environ., 70, 84-97, <https://doi.org/10.1016/j.atmosenv.2013.01.006>, 2013.

506 Fushimi, A., Kondo, Y., Kobayashi, S., Fujitani, Y., Saitoh, K., Takami, A., and Tanabe, K.: Chemical composition and
507 source of fine and nanoparticles from recent direct injection gasoline passenger cars: Effects of fuel and ambient
508 temperature, *Atmos. Environ.*, 124, 77-84, <https://doi.org/10.1016/j.atmosenv.2015.11.017>, 2016.

509 Fuzzi, S., Baltensperger, U., Carslaw, K., Decesari, S., Denier van der Gon, H., Facchini, M. C., Fowler, D., Koren, I.,
510 Langford, B., Lohmann, U., Nemitz, E., Pandis, S., Riipinen, I., Rudich, Y., Schaap, M., Slowik, J. G., Spracklen,
511 D. V., Vignati, E., Wild, M., Williams, M., and Gilardoni, S.: Particulate matter, air quality and climate: lessons
512 learned and future needs, *Atmos. Chem. Phys.*, 15, 8217-8299, <https://doi.org/10.5194/acp-15-8217-2015>, 2015.

513 Geller, M. D., Ntziachristos, L., Mamakos, A., Samaras, Z., Schmitz, D. A., Froines, J. R., and Sioutas, C.: Physicochemical
514 and redox characteristics of particulate matter (PM) emitted from gasoline and diesel passenger cars, *Atmos.*
515 *Environ.*, 40, 6988-7004, <https://doi.org/10.1016/j.atmosenv.2006.06.018>, 2006.

516 Gentner, D. R., Isaacman, G., Worton, D. R., Chan, A. W. H., Dallmann, T. R., Davis, L., Liu, S., Day, D. A., Russell, L.
517 M., Wilson, K. R., Weber, R., Guha, A., Harley, R. A., and Goldstein, A. H.: Elucidating secondary organic aerosol
518 from diesel and gasoline vehicles through detailed characterization of organic carbon emissions, *Proceedings of the*
519 *National Academy of Sciences*, 109, 18318-18323, <https://doi.org/10.1073/pnas.1212272109>, 2012.

520 Gentner, D. R., Jathar, S. H., Gordon, T. D., Bahreini, R., Day, D. A., El Haddad, I., Hayes, P. L., Pieber, S. M., Platt, S. M.,
521 de Gouw, J., Goldstein, A. H., Harley, R. A., Jimenez, J. L., Prévôt, A. S. H., and Robinson, A. L.: Review of
522 Urban Secondary Organic Aerosol Formation from Gasoline and Diesel Motor Vehicle Emissions, *Environ. Sci.*
523 *Technol.*, 51, 1074-1093, <https://doi.org/10.1021/acs.est.6b04509>, 2017.

524 Gertler, A. W., Abu-Allaban, M., Coulombe, W., Gillies, J. A., Pierson, W. R., Rogers, C. F., Sagebiel, J. C., Tarnay, L., and
525 Cahill, T. A.: Measurements of mobile source particulate emissions in a highway tunnel, *Int. J. Veh. Des.*, 27, 86-
526 93, <https://doi.org/10.1504/ijvd.2001.001954>, 2001.

527 Giechaskiel, B., Mamakos, A., Andersson, J., Dilara, P., Martini, G., Schindler, W., and Bergmann, A.: Measurement of
528 Automotive Nonvolatile Particle Number Emissions within the European Legislative Framework: A Review,
529 *Aerosol Sci. Technol.*, 46, 719-749, <https://doi.org/10.1080/02786826.2012.661103>, 2012.

530 Giechaskiel, B., Maricq, M., Ntziachristos, L., Dardiotis, C., Wang, X., Axmann, H., Bergmann, A., and Schindler, W.:
531 Review of motor vehicle particulate emissions sampling and measurement: From smoke and filter mass to particle
532 number, *J. Aerosol Sci.*, 67, 48-86, <https://doi.org/10.1016/j.jaerosci.2013.09.003>, 2014.

533 Harrison, R. M., and Beddows, D. C.: Efficacy of Recent Emissions Controls on Road Vehicles in Europe and Implications
534 for Public Health, *Scientific Reports*, 7, 1152, <https://doi.org/10.1038/s41598-017-01135-2>, 2017.

535 Ho, K. F., Ho, S. H. S., Lee, S. C., Louie, P. K. K., Cao, J., and Deng, W.: Volatile organic compounds in roadside
536 environment of Hong Kong, *Aerosol Air Qual. Res.*, 13, 1331-1347, 2013.

537 Huang, X. F., He, L. Y., Hu, M., Canagaratna, M. R., Sun, Y., Zhang, Q., Zhu, T., Xue, L., Zeng, L. W., Liu, X. G., Zhang,
538 Y. H., Jayne, J. T., Ng, N. L., and Worsnop, D. R.: Highly time-resolved chemical characterization of atmospheric

539 submicron particles during 2008 Beijing Olympic Games using an Aerodyne High-Resolution Aerosol Mass
540 Spectrometer, *Atmos. Chem. Phys.*, 10, 8933-8945, <https://doi.org/10.5194/acp-10-8933-2010>, 2010.

541 Huang, X. H. H., Bian, Q. J., Louie, P. K. K., and Yu, J. Z.: Contributions of vehicular carbonaceous aerosols to PM_{2.5} in a
542 roadside environment in Hong Kong, *Atmos. Chem. Phys.*, 14, 9279-9293, [https://doi.org/10.5194/acp-14-9279-](https://doi.org/10.5194/acp-14-9279-2014)
543 2014, 2014.

544 Huffman, J. A., Docherty, K. S., Aiken, A. C., Cubison, M. J., Ulbrich, I. M., DeCarlo, P. F., Sueper, D., Jayne, J. T.,
545 Worsnop, D. R., Ziemann, P. J., and Jimenez, J. L.: Chemically-resolved aerosol volatility measurements from two
546 megacity field studies, *Atmos. Chem. Phys.*, 9, 7161-7182, 2009a.

547 Huffman, J. A., Docherty, K. S., Mohr, C., Cubison, M. J., Ulbrich, I. M., Ziemann, P. J., Onasch, T. B., and Jimenez, J. L.:
548 Chemically-Resolved Volatility Measurements of Organic Aerosol from Different Sources, *Environ. Sci. Technol.*,
549 43, 5351-5357, <https://doi.org/10.1021/es803539d>, 2009b.

550 Jang, J., Lee, Y., Kwon, O., Woo, Y., Cho, C., Kim, G., Pyo, Y., and Lee, M.: Exhaust Gas Emission and Particulate Matter
551 (PM) from Gasoline, LPG and Diesel Vehicle Using Different Engine Oil, *Transactions of the Korean Society of*
552 *Automotive Engineers*, 24, 144-151, <https://doi.org/10.7467/KSAE.2016.24.2.144>, 2016.

553 Jimenez, J. L., Jayne, J. T., Shi, Q., Kolb, C. E., Worsnop, D. R., Yourshaw, I., Seinfeld, J. H., Flagan, R. C., Zhang, X. F.,
554 Smith, K. A., Morris, J. W., and Davidovits, P.: Ambient aerosol sampling using the Aerodyne Aerosol Mass
555 Spectrometer, *J. Geophys. Res. Atmos.*, 108, 8425, <https://doi.org/10.1029/2001jd001213>, 2003.

556 Kam, W., Liacos, J. W., Schauer, J. J., Delfino, R. J., and Sioutas, C.: On-road emission factors of PM pollutants for light-
557 duty vehicles (LDVs) based on urban street driving conditions, *Atmos. Environ.*, 61, 378-386,
558 <https://doi.org/10.1016/j.atmosenv.2012.07.072>, 2012.

559 Karjalainen, P., Pirjola, L., Heikkilä, J., Lähde, T., Tzamkiozis, T., Ntziachristos, L., Keskinen, J., and Rönkkö, T.: Exhaust
560 particles of modern gasoline vehicles: A laboratory and an on-road study, *Atmos. Environ.*, 97, 262-270,
561 <https://doi.org/10.1016/j.atmosenv.2014.08.025>, 2014.

562 Kheirbek, I., Haney, J., Douglas, S., Ito, K., and Matte, T.: The contribution of motor vehicle emissions to ambient fine
563 particulate matter public health impacts in New York City: a health burden assessment, *Environ. Health*, 15, 89,
564 <https://doi.org/10.1186/s12940-016-0172-6>, 2016.

565 Kirchstetter, T. W., Harley, R. A., Kreisberg, N. M., Stolzenburg, M. R., and Hering, S. V.: On-road measurement of fine
566 particle and nitrogen oxide emissions from light- and heavy-duty motor vehicles, *Atmos. Environ.*, 33, 2955-2968,
567 [https://doi.org/10.1016/S1352-2310\(99\)00089-8](https://doi.org/10.1016/S1352-2310(99)00089-8), 1999.

568 Kumar, P., Ketzler, M., Vardoulakis, S., Pirjola, L., and Britter, R.: Dynamics and dispersion modelling of nanoparticles from
569 road traffic in the urban atmospheric environment—A review, *J. Aerosol Sci*, 42, 580-603,
570 <https://doi.org/10.1016/j.jaerosci.2011.06.001>, 2011.

571 Kumar, P., Morawska, L., Birmili, W., Paasonen, P., Hu, M., Kulmala, M., Harrison, R. M., Norford, L., and Britter, R.:
572 Ultrafine particles in cities, *Environ. Int.*, 66, 1-10, <https://doi.org/10.1016/j.envint.2014.01.013>, 2014.

573 Kwak, J. H., Kim, H. S., Lee, J. H., and Lee, S. H.: On-road chasing measurement of exhaust particle emissions from diesel,
574 CNG, LPG, and DME-fueled vehicles using a mobile emission laboratory, *International Journal of Automotive*
575 *Technology*, 15, 543-551, <https://doi.org/10.1007/s12239-014-0057-z>, 2014.

576 Lanz, V. A., Alfarrá, M. R., Baltensperger, U., Buchmann, B., Hueglin, C., and Prevot, A. S. H.: Source apportionment of
577 submicron organic aerosols at an urban site by factor analytical modelling of aerosol mass spectra, *Atmos. Chem.*
578 *Phys.*, 7, 1503-1522, 2007.

579 Lau, J., Hung, W. T., and Cheung, C. S.: Observation of increases in emission from modern vehicles over time in Hong
580 Kong using remote sensing, *Environ. Pollut.*, 163, 14-23, <https://doi.org/10.1016/j.envpol.2011.12.021>, 2012.

581 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
582 roadside in downtown Hong Kong-Overview of 4 months of continuous high-resolution aerosol mass spectrometer
583 measurements, *J. Geophys. Res. Atmos.*, 120, 7040-7058, <https://doi.org/10.1002/2015jd023311>, 2015.

584 Lee, S. C., Cheng, Y., Ho, K. F., Cao, J. J., Louie, P. K. K., Chow, J. C., and Watson, J. G.: PM_{1.0} and PM_{2.5}
585 Characteristics in the Roadside Environment of Hong Kong, *Aerosol Sci. Technol.*, 40, 157-165,
586 <https://doi.org/10.1080/02786820500494544>, 2006.

587 Levy, J. I., Buonocore, J. J., and von Stackelberg, K.: Evaluation of the public health impacts of traffic congestion: a health
588 risk assessment, *Environ. Health*, 9, 65, <https://doi.org/10.1186/1476-069x-9-65>, 2010.

589 Li, Y., Xue, J., Johnson, K., Durbin, T., Villela, M., Pham, L., Hosseini, S., Zheng, Z., Short, D., Karavalakis, G., Asa-
590 Awuku, A., Jung, H., Wang, X., Quiros, D., Hu, S., Huai, T., and Ayala, A.: Determination of Suspended Exhaust
591 PM Mass for Light-Duty Vehicles, 2014.

592 Lim, H. J., and Turpin, B. J.: Origins of primary and secondary organic aerosol in Atlanta: Results' of time-resolved
593 measurements during the Atlanta supersite experiment, *Environ. Sci. Technol.*, 36, 4489-4496,
594 <https://doi.org/10.1021/es0206487>, 2002.

595 Louie, P. K., Chow, J. C., Chen, L. W., Watson, J. G., Leung, G., and Sin, D. W.: PM_{2.5} chemical composition in Hong
596 Kong: urban and regional variations, *Sci. Total Environ.*, 338, 267-281,
597 <https://doi.org/10.1016/j.scitotenv.2004.07.021>, 2005.

598 Lowenthal, D. H., Zielinska, B., Chow, J. C., Watson, J. G., Gautam, M., Ferguson, D. H., Neuroth, G. R., and Stevens, K.
599 D.: Characterization of Heavy-Duty Diesel Vehicle Emissions, *Atmos. Environ.*, 28, 731-743,
600 [https://doi.org/10.1016/1352-2310\(94\)90050-7](https://doi.org/10.1016/1352-2310(94)90050-7), 1994.

601 Lyu, X. P., Zeng, L. W., Guo, H., Simpson, I. J., Ling, Z. H., Wang, Y., Murray, F., Louie, P. K. K., Saunders, S. M., Lam,
602 S. H., and Blake, D. R.: Evaluation of the effectiveness of air pollution control measures in Hong Kong, *Environ.*
603 *Pollut.*, 220, Part A, 87-94, <https://doi.org/10.1016/j.envpol.2016.09.025>, 2017.

604 May, A. A., Nguyen, N. T., Presto, A. A., Gordon, T. D., Lipsky, E. M., Karve, M., Gutierrez, A., Robertson, W. H., Zhang,
605 M., Brandow, C., Chang, O., Chen, S. Y., Cicero-Fernandez, P., Dinkins, L., Fuentes, M., Huang, S. M., Ling, R.,
606 Long, J., Maddox, C., Massetti, J., McCauley, E., Miguel, A., Na, K., Ong, R., Pang, Y. B., Rieger, P., Sax, T.,

607 Truong, T., Vo, T., Chattopadhyay, S., Maldonado, H., Maricq, M. M., and Robinson, A. L.: Gas- and particle-
608 phase primary emissions from in-use, on-road gasoline and diesel vehicles, *Atmos. Environ.*, 88, 247-260,
609 <https://doi.org/10.1016/j.atmosenv.2014.01.046>, 2014.

610 Mohr, C., DeCarlo, P. F., Heringa, M. F., Chirico, R., Slowik, J. G., Richter, R., Reche, C., Alastuey, A., Querol, X., Seco,
611 R., Penuelas, J., Jimenez, J. L., Crippa, M., Zimmermann, R., Baltensperger, U., and Prevot, A. S. H.: Identification
612 and quantification of organic aerosol from cooking and other sources in Barcelona using aerosol mass spectrometer
613 data, *Atmos. Chem. Phys.*, 12, 1649-1665, <https://doi.org/10.5194/acp-12-1649-2012>, 2012.

614 Ning, Z., Wubulihairan, M., and Yang, F. H.: PM, NO_x and butane emissions from on-road vehicle fleets in Hong Kong and
615 their implications on emission control policy, *Atmos. Environ.*, 61, 265-274,
616 <https://doi.org/10.1016/j.atmosenv.2012.07.047>, 2012.

617 Paciga, A., Karnezi, E., Kostenidou, E., Hildebrandt, L., Psychoudaki, M., Engelhart, G. J., Lee, B. H., Crippa, M., Prevot, A.
618 S. H., Baltensperger, U., and Pandis, S. N.: Volatility of organic aerosol and its components in the megacity of
619 Paris, *Atmos. Chem. Phys.*, 16, 2013-2023, <https://doi.org/10.5194/acp-16-2013-2016>, 2016.

620 Platt, S. M., El Haddad, I., Zardini, A. A., Clairotte, M., Astorga, C., Wolf, R., Slowik, J. G., Temime-Roussel, B.,
621 Marchand, N., Jezek, I., Drinovec, L., Mocnik, G., Mohler, O., Richter, R., Barmet, P., Bianchi, F., Baltensperger,
622 U., and Prevot, A. S. H.: Secondary organic aerosol formation from gasoline vehicle emissions in a new mobile
623 environmental reaction chamber, *Atmos. Chem. Phys.*, 13, 9141-9158, <https://doi.org/10.5194/acp-13-9141-2013>,
624 2013.

625 Pope, C. A., 3rd: Mortality effects of longer term exposures to fine particulate air pollution: review of recent epidemiological
626 evidence, *Inhal Toxicol*, 19 Suppl 1, 33-38, <https://doi.org/10.1080/08958370701492961>, 2007.

627 Quiros, D. C., Hu, S. H., Hu, S. S., Lee, E. S., Sardar, S., Wang, X. L., Olfert, J. S., Jung, H. J. S., Zhu, Y. F., and Huai, T.:
628 Particle effective density and mass during steady-state operation of GDI, PFI, and diesel passenger cars, *J. Aerosol*
629 *Sci*, 83, 39-54, <https://doi.org/10.1016/j.jaerosci.2014.12.004>, 2015.

630 Research Centre of Environmental Technology and Management, H. K. P. U.: Determination of Suspended Particulate &
631 VOC Emission Profiles for Vehicular Sources in Hong Kong, Hong Kong Polytechnic University 2005.

632 Ristovski, Z. D., Jayaratne, E. R., Morawska, L., Ayoko, G. A., and Lim, M.: Particle and carbon dioxide emissions from
633 passenger vehicles operating on unleaded petrol and LPG fuel, *Sci. Total Environ.*, 345, 93-98,
634 <https://doi.org/10.1016/j.scitotenv.2004.10.021>, 2005.

635 Robinson, A. L., Donahue, N. M., Shrivastava, M. K., Weitkamp, E. A., Sage, A. M., Grieshop, A. P., Lane, T. E., Pierce, J.
636 R., and Pandis, S. N.: Rethinking organic aerosols: semivolatile emissions and photochemical aging, *Science*, 315,
637 1259-1262, <https://doi.org/10.1126/science.1133061>, 2007.

638 Saliba, G., Saleh, R., Zhao, Y., Presto, A. A., Lambe, A. T., Frodin, B., Sardar, S., Maldonado, H., Maddox, C., May, A. A.,
639 Drozd, G. T., Goldstein, A. H., Russell, L. M., Hagen, F., and Robinson, A. L.: Comparison of Gasoline Direct-
640 Injection (GDI) and Port Fuel Injection (PFI) Vehicle Emissions: Emission Certification Standards, Cold-Start,

641 Secondary Organic Aerosol Formation Potential, and Potential Climate Impacts, *Environ. Sci. Technol.*, 51, 6542-
642 6552, <https://doi.org/10.1021/acs.est.6b06509>, 2017.

643 Setyan, A., Zhang, Q., Merkel, M., Knighton, W. B., Sun, Y., Song, C., Shilling, J. E., Onasch, T. B., Herndon, S. C.,
644 Worsnop, D. R., Fast, J. D., Zaveri, R. A., Berg, L. K., Wiedensohler, A., Flowers, B. A., Dubey, M. K., and
645 Subramanian, R.: Characterization of submicron particles influenced by mixed biogenic and anthropogenic
646 emissions using high-resolution aerosol mass spectrometry: results from CARES, *Atmos. Chem. Phys.*, 12, 8131-
647 8156, <https://doi.org/10.5194/acp-12-8131-2012>, 2012.

648 Simon, H., Allen, D. T., and Wittig, A. E.: Fine Particulate Matter Emissions Inventories: Comparisons of Emissions
649 Estimates with Observations from Recent Field Programs, *J. Air Waste Manage. Assoc.*, 58, 320-343,
650 <https://doi.org/10.3155/1047-3289.58.2.320>, 2008.

651 Sun, C., Lee, B. P., Huang, D., Li, Y. J., Schurman, M. I., Louie, P. K. K., Luk, C., and Chan, C. K.: Continuous
652 measurements at the urban roadside in an Asian megacity by Aerosol Chemical Speciation Monitor (ACSM):
653 particulate matter characteristics during fall and winter seasons in Hong Kong, *Atmos. Chem. Phys.*, 16, 1713-1728,
654 <https://doi.org/10.5194/acp-16-1713-2016>, 2016.

655 Sun, J. Y., Zhang, Q., Canagaratna, M. R., Zhang, Y. M., Ng, N. L., Sun, Y. L., Jayne, J. T., Zhang, X. C., Zhang, X. Y., and
656 Worsnop, D. R.: Highly time- and size-resolved characterization of submicron aerosol particles in Beijing using an
657 Aerodyne Aerosol Mass Spectrometer, *Atmos. Environ.*, 44, 131-140,
658 <https://doi.org/10.1016/j.atmosenv.2009.03.020>, 2010.

659 Sun, Y. L., Zhang, Q., Schwab, J. J., Demerjian, K. L., Chen, W. N., Bae, M. S., Hung, H. M., Hogrefe, O., Frank, B.,
660 Rattigan, O. V., and Lin, Y. C.: Characterization of the sources and processes of organic and inorganic aerosols in
661 New York city with a high-resolution time-of-flight aerosol mass spectrometer, *Atmos. Chem. Phys.*, 11, 1581-
662 1602, <https://doi.org/10.5194/acp-11-1581-2011>, 2011.

663 Sun, Y. L., Zhang, Q., Schwab, J. J., Chen, W. N., Bae, M. S., Hung, H. M., Lin, Y. C., Ng, N. L., Jayne, J., Massoli, P.,
664 Williams, L. R., and Demerjian, K. L.: Characterization of near-highway submicron aerosols in New York City
665 with a high-resolution aerosol mass spectrometer, *Atmos. Chem. Phys.*, 12, 2215-2227, <https://doi.org/10.5194/acp-12-2215-2012>, 2012.

667 Tartakovsky, L., Baibikov, V., Comte, P., Czerwinski, J., Mayer, A., Veinblat, M., and Zimmerli, Y.: Ultrafine particle
668 emissions by in-use diesel buses of various generations at low-load regimes, *Atmos. Environ.*, 107, 273-280,
669 <https://doi.org/10.1016/j.atmosenv.2015.02.052>, 2015.

670 Transport Department: The Annual Traffic Census 2013, HKSAR Government, Hong Kong, 2014.

671 Turpin, B. J., and Huntzicker, J. J.: Secondary formation of organic aerosol in the Los Angeles basin: A descriptive analysis
672 of organic and elemental carbon concentrations, *Atmos. Environ.*, 25, 207-215, [https://doi.org/10.1016/0960-1686\(91\)90291-E](https://doi.org/10.1016/0960-1686(91)90291-E), 1991.

673

674 Uhrek, E., Halenka, T., Borken-Kleefeld, J., Balkanski, Y., Berntsen, T., Borrego, C., Gauss, M., Hoor, P., Juda-Rezler, K.,
675 Lelieveld, J., Melas, D., Rypdal, K., and Schmid, S.: Transport impacts on atmosphere and climate: Land transport,
676 *Atmos. Environ.*, 44, 4772-4816, <https://doi.org/10.1016/j.atmosenv.2010.01.002>, 2010.

677 Ulbrich, I. M., Canagaratna, M. R., Zhang, Q., Worsnop, D. R., and Jimenez, J. L.: Interpretation of organic components
678 from Positive Matrix Factorization of aerosol mass spectrometric data, *Atmos. Chem. Phys.*, 9, 2891-2918, 2009.

679 Valavanidis, A., Fiotakis, K., and Vlachogianni, T.: Airborne Particulate Matter and Human Health: Toxicological
680 Assessment and Importance of Size and Composition of Particles for Oxidative Damage and Carcinogenic
681 Mechanisms, *Journal of Environmental Science and Health, Part C*, 26, 339-362,
682 <https://doi.org/10.1080/10590500802494538>, 2008.

683 Wang, A., Fallah-Shorshani, M., Xu, J., and Hatzopoulou, M.: Characterizing near-road air pollution using local-scale
684 emission and dispersion models and validation against in-situ measurements, *Atmos. Environ.*, 142, 452-464,
685 <https://doi.org/10.1016/j.atmosenv.2016.08.020>, 2016.

686 Williams, L. R., Gonzalez, L. A., Peck, J., Trimborn, D., McInnis, J., Farrar, M. R., Moore, K. D., Jayne, J. T., Robinson, W.
687 A., Lewis, D. K., Onasch, T. B., Canagaratna, M. R., Trimborn, A., Timko, M. T., Magoon, G., Deng, R., Tang, D.,
688 Blanco, E. D. L. R., Prevot, A. S. H., Smith, K. A., and Worsnop, D. R.: Characterization of an aerodynamic lens
689 for transmitting particles greater than 1 micrometer in diameter into the Aerodyne aerosol mass spectrometer,
690 *Atmos. Meas. Tech.*, 6, 3271-3280, <https://doi.org/10.5194/amt-6-3271-2013>, 2013.

691 World Health Organization: Air Quality and Health, Fact Sheet N°313:
692 <http://www.who.int/mediacentre/factsheets/fs313/en/index.html>, access: 30 May, 2011.

693 Worton, D. R., Isaacman, G., Gentner, D. R., Dallmann, T. R., Chan, A. W. H., Ruehl, C., Kirchstetter, T. W., Wilson, K. R.,
694 Harley, R. A., and Goldstein, A. H.: Lubricating Oil Dominates Primary Organic Aerosol Emissions from Motor
695 Vehicles, *Environ. Sci. Technol.*, 48, 3698-3706, <https://doi.org/10.1021/es405375j>, 2014.

696 Yuan, Z. B., Yadav, V., Turner, J. R., Louie, P. K. K., and Lau, A. K. H.: Long-term trends of ambient particulate matter
697 emission source contributions and the accountability of control strategies in Hong Kong over 1998-2008, *Atmos.*
698 *Environ.*, 76, 21-31, <https://doi.org/10.1016/j.atmosenv.2012.09.026>, 2013.

699 Zhang, K., and Batterman, S.: Air pollution and health risks due to vehicle traffic, *Sci. Total Environ.*, 450, 307-316,
700 <https://doi.org/10.1016/j.scitotenv.2013.01.074>, 2013.

701 Zhang, Q., Alfarra, M. R., Worsnop, D. R., Allan, J. D., Coe, H., Canagaratna, M. R., and Jimenez, J. L.: Deconvolution and
702 quantification of hydrocarbon-like and oxygenated organic aerosols based on aerosol mass spectrometry, *Environ.*
703 *Sci. Technol.*, 39, 4938-4952, 2005.

704 Zhang, Q., Jimenez, J. L., Canagaratna, M. R., Ulbrich, I. M., Ng, N. L., Worsnop, D. R., and Sun, Y. L.: Understanding
705 atmospheric organic aerosols via factor analysis of aerosol mass spectrometry: a review, *Anal. Bioanal. Chem.*, 401,
706 3045-3067, <https://doi.org/10.1007/s00216-011-5355-y>, 2011.

707 Zhao, Y., Nguyen, N. T., Presto, A. A., Hennigan, C. J., May, A. A., and Robinson, A. L.: Intermediate Volatility Organic
708 Compound Emissions from On-Road Diesel Vehicles: Chemical Composition, Emission Factors, and Estimated
709 Secondary Organic Aerosol Production, *Environ. Sci. Technol.*, 49, 11516-11526,
710 <https://doi.org/10.1021/acs.est.5b02841>, 2015.

711 Zotter, P., El-Haddad, I., Zhang, Y., Hayes, P. L., Zhang, X., Lin, Y.-H., Wacker, L., Schnelle-Kreis, J., Abbaszade, G.,
712 Zimmermann, R., Surratt, J. D., Weber, R., Jimenez, J. L., Szidat, S., Baltensperger, U., and Prévôt, A. S. H.:
713 Diurnal cycle of fossil and nonfossil carbon using radiocarbon analyses during CalNex, *J. Geophys. Res. Atmos.*,
714 119, 6818-6835, <https://doi.org/10.1002/2013JD021114>, 2014.

715

716

718 **Table 1.** Species concentrations from AMS and total PM_{2.5} mass from TEOM in (a); EC, OC and organic aerosol constituent
 719 concentrations (AMS PMF analysis) in (b); average concentrations during the traffic counting period (May 28-31, 2013)
 720 and seasonal averages (Spring: Mar – May 2013, Summer: May – Jul 2013) based on Lee et al. (2015) at the Mong Kok
 721 site
 722

(a)

$\mu\text{g}/\text{m}^3$ Mean \pm SD	Org (AMS)	SO4 (AMS)	NO3 (AMS)	NH4 (AMS)	Chl (AMS)	NR-PM₁ (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	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	12.7 \pm 3.6	17.3 \pm 7.5

723

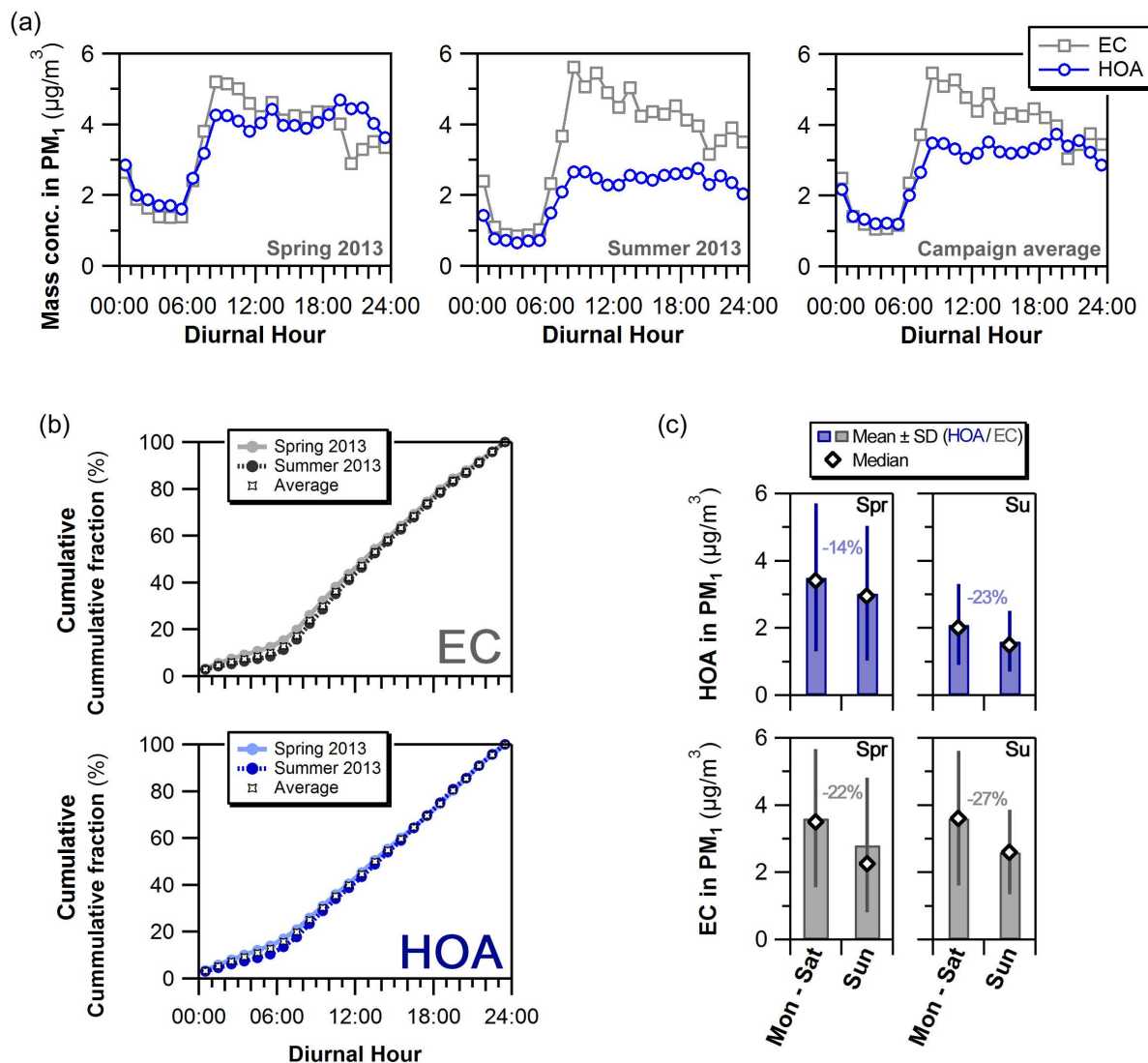
(b)

$\mu\text{g}/\text{m}^3$ 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

724



726

727 **Figure 1.**

728 (a) Diurnal variation of HOA and EC mass concentrations in spring 2013 (7 Mar 2013 to 15 May 2013), summer 2013 (16

729 May 2013 to 19 Jul 2013) and averaged over the entire sampling campaign (7 Mar 2013 to 19 Jul 2013)

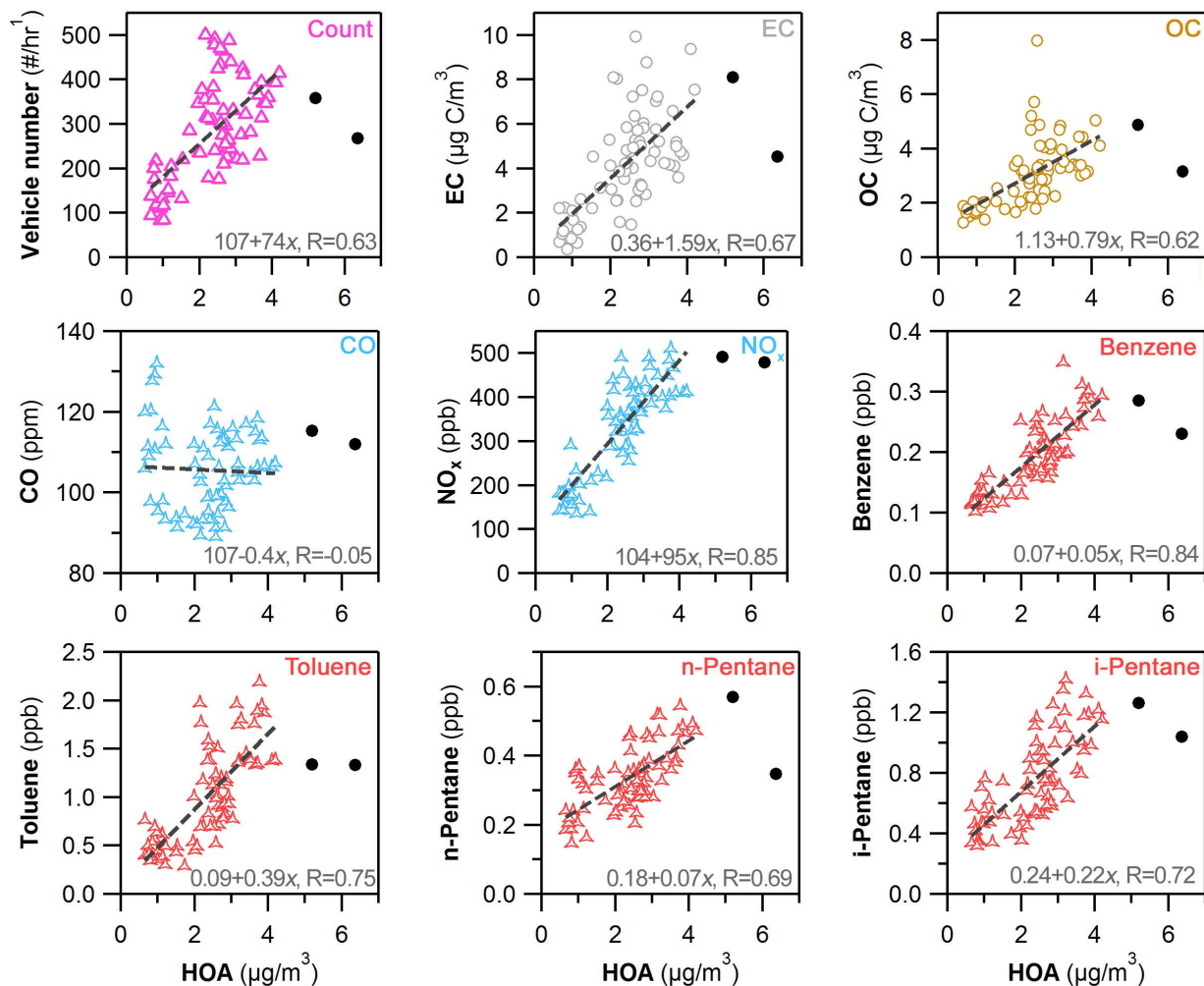
730 (b) Diurnal variation of the cumulative fraction of total daily HOA and EC mass concentrations in spring 2013, summer

731 2013 and averaged over the entire sampling campaign

732 (c) Mean HOA and EC concentrations (*bar with standard deviations, open marker depicts median*) from Monday through

733 Saturday and on Sunday in spring 2013 (Spr) and summer 2013 (Su); days with >25% (>6h) of missing data were

734 excluded



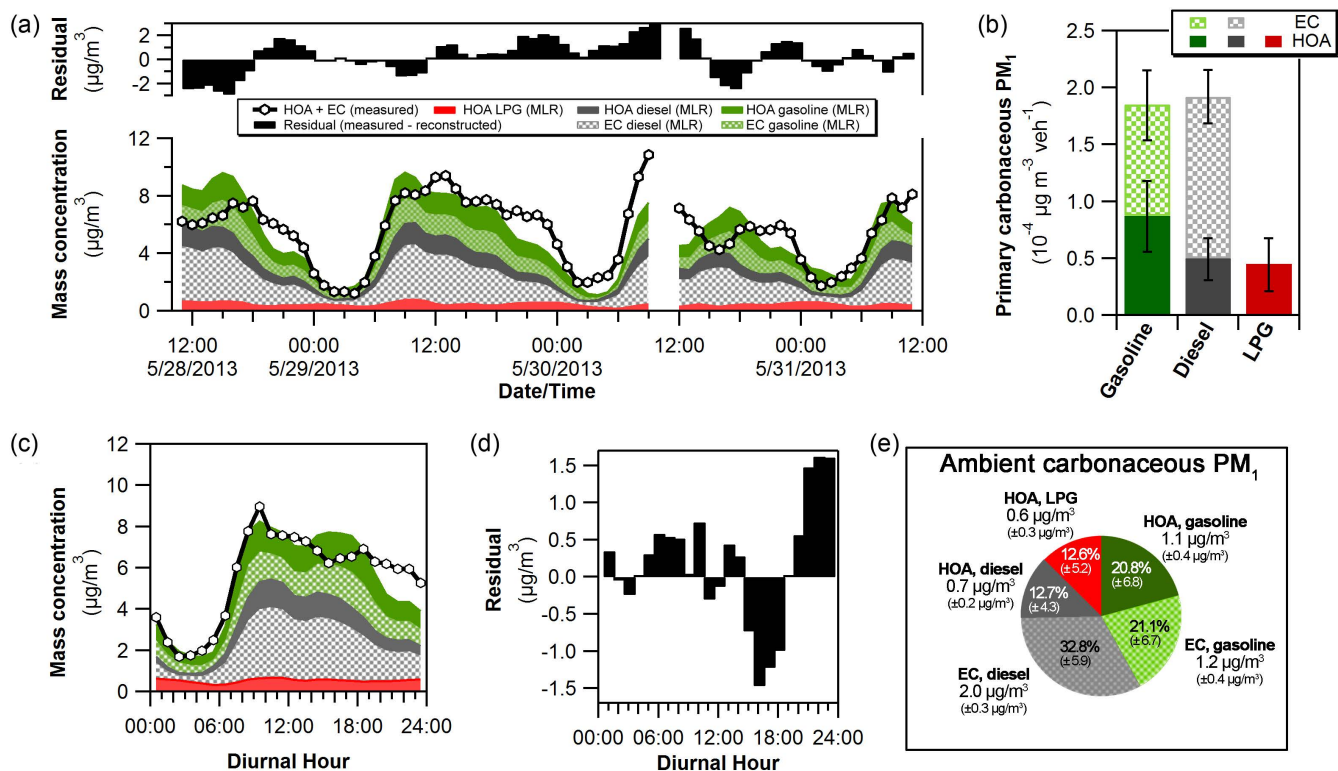
735

736 **Figure 2.**

737 Scatter plot of vehicle count, mass concentration of EC, OC and gas-phase concentrations of various VOCs, NO_x, and CO

738 against HOA during 28-31 May 2013, linear least squares fits with equations and Pearson's R values in grey, black dots

739 signify outlier values excluded from the line fits



740

741 **Figure 3.**

742 (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
 743 residuals as the difference between resolved HOA+EC and measured HOA+EC

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

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

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

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