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

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Abstract. Road traffic has significant impacts on air quality particularly in densely urbanized and populated areas where vehicle emissions are a major local source of ambient particulate matter. Engine type (i.e. fuel use) significantly impacts the

10   chemical characteristics of tailpipe emission and thus the distribution of engine types in traffic impacts measured ambient concentrations. This study provides an estimation of the contribution of vehicles powered by different fuels (gasoline, diesel, LPG) to carbonaceous submicron aerosol (PM_1) based on ambient aerosol mass spectrometer (AMS) and elemental carbon (EC) measurements and vehicle count data in an urban inner city environment in Hong Kong with the aim to gauge the importance of different engine types to particulate matter burdens in a typical urban street canyon. On an average per-vehicle-

15   basis, gasoline vehicles emitted 75% and 93% more organics than diesel and LPG vehicles respectively, while EC emissions from diesel vehicles were 45% higher than those from gasoline vehicles. LPG vehicles showed no appreciable contributions to EC and thus overall represented a small contributor to traffic-related primary ambient PM_1 despite their high abundance (~30%) in the traffic mix. Total carbonaceous particle mass contributions to ambient PM_1 from diesel engines were only marginally higher (~4%) than those from gasoline engines, which is likely an effect of recently introduced control strategies

20   targeted at commercial vehicles and buses. Overall, gasoline vehicles contributed 1.2 µg/m³ of EC and 1.1 µg/m³ of organics, LPG vehicles 0.6 µg/m³ of organics and diesel vehicles 2.0 µg/m³ of EC and 0.7 µg/m³ of organics to ambient carbonaceous PM_1.

1. Introduction

1.1. Particulate matter from motor vehicles in urban areas

Emissions from on-road motor vehicles comprise gas-phase species (CO, CO_2, NO_x, SO_2) and volatile organic compounds among them important precursors of secondary organic aerosol (SOA), as well primary particulate matter predominantly in the fine ($PM_{2.5}$) and ultrafine particle size range ($PM_{0.1}$) mostly as carbonaceous species which encompass primary organic aerosol (POA) components and elemental carbon (Giechaskiel et al., 2014). Increased acute occurrence and risk of chronic development of cardiovascular and pulmonary diseases are important epidemiological effects of particulate matter inhalation (Davidson et al., 2005;Pope, 2007;Valavanidis et al., 2008;World Health Organization, 2011). In urban areas with high population and building densities, proximity to vehicle emissions poses a significant public health risk and renders paramount importance to the characterization and quantification of vehicle emissions (Kumar et al., 2014;Uherek et al., 2010).

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

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

Investigations on the impacts of motor vehicle emissions on ambient air pollution and related effects such as health implications (Kheirbek et al., 2016;Zhang and Batterman, 2013;Levy et al., 2010) typically rely on combinations of source inventories with dispersion modelling and combined physical-chemical models with significant uncertainties arising from the complexity of the urban built environment (Kumar et al., 2011;Belcher et al., 2013), inventory currency and accuracy of emission factors (Simon et al., 2008;Fuzzi et al., 2015). The latter is critically affected by the design of dynamometer test cycles with discrepancies having been noted between type approval procedure derived emissions and on-road measured driving emissions, which underlines the continued need to evaluate and monitor contributions of traffic emissions to urban air pollution by direct ambient measurements (Harrison and Beddows, 2017). A dominant factor that affects both gas- and particle-phase species distribution is the engine type used (Alves et al., 2015;Franco et al., 2013;Jang et al., 2016;Kam et al., 2012;Karjalainen et al.,

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

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

1.2. Background of vehicle emission studies in Hong Kong

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

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

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

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

100   By contrast, seasonal variations of EC concentrations were weak. Higher EC concentrations were typically associated with rush hour traffic and confirm the local nature of EC and its relation to mainly on-road traffic (Lee et al., 2006;Cheng et al., 2010;Huang et al., 2014;Louie et al., 2005). While earlier studies (*samples dated pre-2005*) reported EC concentrations in the range of 11-25 µg C m^{-3} in PM$_{2.5}$ at the open roadside (Louie et al., 2005;Cheng et al., 2010;Lee et al., 2006), more recent measurements (*2012-2013*) indicate a significant reduction in overall roadside EC burdens with concentrations ~ 5 µg C m^{-3}

105 (Huang et al., 2014;Sun et al., 2016;Lee et al., 2015). Vehicle-related OC and organic matter (OM) concentrations at a roadside measurement site were estimated to ~2 µg C m^{-3} and ~7 µg m^{-3} respectively in 2012, accounting for one-third of total OC in PM$_{2.5}$ and one-quarter of total PM$_{2.5}$ (Huang et al., 2014).

    Contributions of traffic emissions to ambient gas-phase species were evaluated utilizing VOC canister samples collected in locations with different dominant-vehicle types in Hong Kong in 2003 (Ho et al., 2013). Propane, n- and i-butane accounted

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

    Vehicle emission factors were estimated by a mass-balance model with differential measurements near the entrance and exit

115 of a tunnel bore in 2003 and manual vehicle counts. Diesel vehicles were found to contribute substantially more to NO$_x$, PM$_{2.5,}$ and PAH than gasoline vehicles which dominated CO emissions. Diesel and LPG-fueled vehicles dominated total measured VOCs and total measured carbonyl compounds. EC emission factors of diesel vehicles (131 mg veh^{-1} km^{-1}) were ~40 times higher than those of non-diesel (i.e. gasoline and LPG-fueled) vehicles (3.2 mg veh^{-1} km^{-1}). For OC, the difference amounted to a smaller factor of ~8 with emission factors of 67.9 mg veh^{-1} km^{-1} and 8.5 mg veh^{-1} km^{-1} for diesel and non-diesel vehicles

120 respectively. In terms of fractional contribution, OC made up 51% of PM$_{2.5}$ in non-diesel emissions, while EC made up 51% of PM$_{2.5}$ in diesel emissions. Contributions of inorganic species including ions and trace metals in traffic emissions were minor amounting to only ~10% (Cheng et al., 2010;Research Centre of Environmental Technology and Management, 2005). While these emission factors compared fairly well with those from similar studies undertaken in the 1990s (Gertler et al.,

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

A mobile measurement platform with an array of on-board PM and gas monitors was deployed in early 2012 for a more up-to-date characterization of fuel-based emission factors of PM, NO_x, and butane by chasing vehicle plumes on major roadways and highways (Ning et al., 2012). The measurements focused on the evaluation of heavy duty diesel trucks (HDDV), franchised diesel buses (DB) and LPG fueled light buses (LB) with plumes sampled at cruise speed to mitigate biases from engine load-dependent emission variability. HDDVs were the highest emitters of BC (1.6 g kg^{-1} of fuel) followed by diesel buses (1.1 g kg^{-1} of fuel) and LPG light buses (0.1 g kg^{-1} of fuel). Analogously, average particle number emission factors were 3.1 x 10^{15} kg^{-1} of fuel, 3.1 x 10^{15} kg^{-1} of fuel and 0.9 x 10^{15} kg^{-1} of fuel for HDDVs, diesel buses and LPG light buses respectively.

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

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

2.   Methodology

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

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

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

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

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

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

3.   Results

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

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

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

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

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

Both HOA and EC exhibited significant reductions in overall mass concentrations on Sundays between 14% and 27%, compared to the rest of the week (Monday to Saturday) as shown in Figure 1d. Days with less than 75% (>18h) data availability were excluded from both time series in the analysis to obviate bias in the daily averaging from their strong diurnal patterns, as opposed to the original data presented by Lee at al. (2015) where the entire unabridged time series were used. Mean reductions in nominal mass concentrations were generally quite similar in the range of $0.8 - 1.0$ µg m^{-3} for EC and ~0.5 µg m^{-3} for HOA. The greater overall reduction in EC indicates a greater reduction in the number of diesel vehicles on Sundays, owing to reduced commercial traffic and lower bus frequency. Contributions of vehicle emissions to ambient organic aerosol have been derived from one-year long measurements at the same site based on analysis of semi-continuous ECOC measurements (Huang et al., 2014). The EC tracer method (Turpin and Huntzicker, 1991;Lim and Turpin, 2002) was employed which evaluates minimum OC/EC ratios, and OC and EC measurements during time periods of high primary source strength and low photochemical activities when OC is mainly of local origin and attributable to the dominant primary sources in the vicinity. A characteristic vehicle-related OC/EC value (OC/EC$_{vehicle}$) of 0.5 was derived from the 5% lowest summer OC/EC ratio data to approximate organic vehicular emissions. The reported monthly average primary organic matter concentrations from traffic in PM$_{2.5}$ (using an OM/OC conversion factor of 1.4) ranged between $2.7 - 3.4$ µg m^{-3} with little seasonal variation. To compare to our measurement base of PM$_1$, we use the previously mentioned average ratio of PM$_1$/PM$_{2.5}$ of 0.8 for conversion. The OC/EC approach would thus yield an approximate $2.2 - 2.7$ µg m^{-3} of traffic-related primary organics in PM$_1$ which compares well with the overall average concentration of traffic-related HOA (2.8 µg m^{-3}) covering the entire AMS measurement campaign (March – July 2013). However, we observed significantly different concentrations depending on the season with much higher concentrations (3.5 µg m^{-3}) in spring than in summer (2.0 µg m^{-3}). The lack of a clear seasonal variation in OC/EC derived traffic-related organics is due to the use of a fixed conversion factor directly dependent on ambient EC concentration which did not exhibit a strong seasonal behavior and thus could not capture the same trends observed from the AMS measurements. The derivation of the OC/EC$_{vehicle}$ ratio is subject to further uncertainty due to the substantial influence of cooking-related organics at the site (Lee et al., 2015;Sun et al., 2016), i.e. traffic emissions may not be the dominant source of "local" organics at the site (Table 2), as well as the influence of background SOA which is likely to persist even in time periods of low OC/EC ratios.

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

3.2. Traffic and vehicle flow characteristics

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

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

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

3.3. Estimation of engine-type contributions

3.3.1. Multiple Linear Regression of HOA and EC

Utilizing the detailed information on average daily traffic composition at the measurement site from the counting exercise, contributions of exhaust from different engine types to overall ambient HOA and EC concentrations can be evaluated. Multiple linear regression was carried out with the concentration time series of HOA and EC during the traffic counting period as the dependent variables, the pooled sum of counted diesel, gasoline and LPG vehicles as the independent variables, and a constrained zero intercept, assuming negligible transport of HOA and EC (*see 2. Methodology*).

Two factor solutions (diesel + gasoline), as well as three factor solutions (diesel + gasoline + LPG), were considered, as LPG vehicles are expected to contribute less to particle-phase and more to gas-phase emissions (Faiz et al., 1996). The key statistical output parameters (adjusted R^2, regression coefficients, p-values) of the multiple linear regression analysis for HOA and EC for two and three factor models are presented in Table S1 in the Supplement. For HOA, both two and three factor models yielded acceptable results with all resolved coefficients statistically significant at the 95% confidence level ($p<0.05$). Exclusion of LPG vehicles (two factor model) led to additional apportionment of HOA to both gasoline (+25%) and diesel (+18%) vehicles. Overall model performance improved only slightly when moving from the two factor solution ($R^2_{adj}=0.86$) to the three factor solution ($R^2_{adj}=0.90$). Comparative studies on particulate vehicle emissions have reported non-negligible particle

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

3.3.2.   Analysis of engine-type reconstructed carbonaceous components

The time series of measured and reconstructed HOA and EC concentrations based on the regression coefficients are depicted individually in Figure S5a and S5c in the Supplement and in combination in Figure 3 to represent the sum of motor vehicle related primary carbonaceous particulate compounds, not including possible additional SOA species formed through subsequent atmospheric processing of gas- and particle-phase species in tailpipe exhaust. Gasoline vehicles accounted for more than 45% of total HOA, while diesel and LPG-fueled vehicles were responsible for similar shares of 25-29% (Figure S5b in the Supplement). Over 60% of total EC in PM_1 was accounted for by diesel vehicles (Figure S5d in the Supplement). The sum of HOA and EC in PM_1 represent approximately the total submicron primary carbonaceous aerosol contributions from road traffic. Despite the limited time frame of the counting exercise, the MLR factors adequately resolved the overall trend in EC and HOA concentration (Figure 3a) and diurnal variation (Figure 3b). Relative residuals from the reconstructed sum of HOA and EC were ≤25% with overestimation (*Residuals < 0*) predominant in the later afternoon hours between 14:00 and 18:00, while significant underestimation (*Residuals > 0*) was observed mostly in the late evening hours between 19:00 and 0:00. The semi-volatile character of HOA would contribute to the observed reduced afternoon particle-phase concentration and conversely enhanced nighttime particle-phase concentrations. Averaged over the whole 72h period, measured and reconstructed carbonaceous submicron PM_1 differed by only 4% indicating that the overall reconstruction was not affected

considerably by these diurnal trends in residuals. Vehicle emission factors are typically based on the concentration of emitted species per unit of consumed fuel (derived from CO_2 measurements) or per driven distance. As concurrent CO_2 measurement data were not available, emission factors were not evaluated in this study. Instead, the relative emission behavior of different vehicle groups and their impacts on ambient PM concentrations were approximated by normalizing the resolved engine-specific carbonaceous primary aerosol concentrations by the total number of vehicles in each category (Figure 3b). On a per-vehicle basis (Figure 3b), gasoline vehicles emitted 75% more HOA than diesel vehicles and 93% more HOA than LPG vehicles, while EC emissions from diesel vehicles were 45% higher than those from gasoline vehicles. Therefore, on average gasoline and diesel vehicles emitted similar total amounts of primary carbonaceous PM_1 and each about three times more than LPG powered vehicles. In terms of ambient concentrations, gasoline vehicles accounted for 78% more ambient HOA than diesel vehicles and 56% more ambient HOA than LPG-fueled vehicles. As opposed diesel vehicles accounted for 62% more ambient EC than gasoline-powered vehicles. Each diesel and gasoline vehicles contributed 75-85% more carbonaceous PM_1 mass than LPG vehicles, which despite making up ~30% of total counted traffic accounted for less than 13% of traffic-related primary PM_1 (Figure 3e).

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

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

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

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

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

At the same time, various control schemes targeting diesel vehicle emissions have been introduced in recent years. In Hong Kong these included inter alia an incentive scheme in 2010 for the replacement of diesel commercial vehicles, the retrofitting of diesel particulate filters (DPF) on pre-Euro IV buses and subsequent replacement of older buses with Euro IV, V and VI standard models (Environment Bureau, 2013;Environmental Protection Department, 2013). $PM_{2.5}$ emission factors determined from dynamometer test runs of various vehicle types in Hong Kong were generally higher for diesel vehicles compared to gasoline and LPG vehicles (Chan et al., 2007), while we observed similar total particle mass emission on a per-vehicle basis for diesel and gasoline vehicles in this study. The dynamometer tests were conducted on relatively old vehicles (*newest date of manufacture: 2001*) and are unlikely to adequately reflect the ambient vehicle mix in our study, particularly with regard to the implementation of recent emission control programs. New engine technologies for diesel vehicles, such as DPFs, have been shown to greatly reduce both EC and OC emissions (Alves et al., 2015) thus leading to overall little total particle mass emissions (Li et al., 2014;Quiros et al., 2015) partially due to shifts in the particle size distributions towards a greater fraction of particles in the ultrafine mode (Giechaskiel et al., 2012). The Euro III, IV and V standards for trucks and buses were introduced in late 2000, late 2005 and late 2008 respectively. While the year of manufacture does not directly infer compliance to a specific emission standard, an approximation of the fraction of vehicles fulfilling a certain standard can be made by assuming that vehicles produced between 2001 and 2005, between 2006 and 2008 and between 2009 and 2013 very likely comply with the Euro III, IV and V standards respectively. In this case, it is assumed that emission standards were immediately or had already been adopted in vehicle models in the corresponding year of manufacture. Figure S6 in the Supplement depicts the number and proportion of vehicles of certain years of manufacture and their assumed Euro standard. For goods vehicles, 52% of counted vehicles were built between 2005 and 2013 (i.e. likely fulfilling Euro IV and V), while for buses the proportion

was slightly lower at 33%. With these two vehicle groups representing the bulk of diesel powered vehicles, an estimated 40% of diesel vehicles complied with Euro IV and Euro V standards during the time of our ambient measurements, further rationalizing the relatively low per-vehicle contribution of diesel vehicles to ambient exhaust-derived carbonaceous PM_1 in this study.

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

4. Conclusion

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

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

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

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Disclaimer

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

References

Aiken, A. C., Salcedo, D., Cubison, M. J., Huffman, J. A., DeCarlo, P. F., Ulbrich, I. M., Docherty, K. S., Sueper, D., Kimmel, J. R., Worsnop, D. R., Trimborn, A., Northway, M., Stone, E. A., Schauer, J. J., Volkamer, R. M., Fortner, E., de Foy, B., Wang, J., Laskin, A., Shutthanandan, V., Zheng, J., Zhang, R., Gaffney, J., Marley, N. A., Paredes-Miranda, G., Arnott, W. P., Molina, L. T., Sosa, G., and Jimenez, J. L.: Mexico City aerosol analysis during MILAGRO using high resolution aerosol mass spectrometry at the urban supersite (T0) - Part 1: Fine particle composition and organic source apportionment, Atmos. Chem. Phys., 9, 6633-6653, https://doi.org/10.5194/acp-9-6633-2009, 2009.

Alves, C. A., Lopes, D. J., Calvo, A. I., Evtyugina, M., Rocha, S., and Nunes, T.: Emissions from light-duty diesel and gasoline in-use vehicles measured on chassis dynamometer test cycles, Aerosol Air Qual. Res., 15, 99-116, https://doi.org/10.4209/aaqr.2014.01.0006, 2015.

Ban-Weiss, G. A., McLaughlin, J. P., Harley, R. A., Lunden, M. M., Kirchstetter, T. W., Kean, A. J., Strawa, A. W., Stevenson, E. D., and Kendall, G. R.: Long-term changes in emissions of nitrogen oxides and particulate matter from on-road gasoline and diesel vehicles, Atmos. Environ., 42, 220-232, https://doi.org/10.1016/j.atmosenv.2007.09.049, 2008.

Belcher, S., Coceal, O., Hunt, J., Carruthers, D., and Robins, A.: A review of urban dispersion modelling, ADMLC, UK, 2013.

Cappa, C. D., and Jimenez, J. L.: Quantitative estimates of the volatility of ambient organic aerosol, Atmos. Chem. Phys., 10, 5409-5424, https://doi.org/10.5194/acp-10-5409-2010, 2010.

Chan, T. L., Ning, Z., Wang, J. S., Cheung, C. S., Leung, C. W., and Hung, W. T.: Gaseous and Particle Emission Factors from the Selected On-Road Petrol/Gasoline, Diesel, and Liquefied Petroleum Gas Vehicles, Energy & Fuels, 21, 2710-2718, https://doi.org/10.1021/ef070172i, 2007.

Cheng, Y., Ho, K. F., Lee, S. C., and Law, S. W.: Seasonal and diurnal variations of PM1.0, PM2.5 and PM10 in the
480   roadside environment of hong kong, China Particuology, 4, 312-315, https://doi.org/10.1016/S1672-
2515(07)60281-4, 2006.

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-speciated
on-road PM2.5 motor vehicle emission factors in Hong Kong, Sci. Total Environ., 408, 1621-1627,
https://doi.org/10.1016/j.scitotenv.2009.11.061, 2010.

485   Cui, M., Chen, Y., Tian, C., Zhang, F., Yan, C., and Zheng, M.: Chemical composition of PM2.5 from two tunnels with
different vehicular fleet characteristics, Sci. Total Environ., 550, 123-132,
https://doi.org/10.1016/j.scitotenv.2016.01.077, 2016.

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

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

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

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.,
Jathar, S., Robinson, A. L., and Seinfeld, J. H.: Emission factor ratios, SOA mass yields, and the impact of
vehicular emissions on SOA formation, Atmos. Chem. Phys., 14, 2383-2397, https://doi.org/10.5194/acp-14-2383-
500   2014, 2014.

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

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

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

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

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

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

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

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

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

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., de Gouw, J., Goldstein, A. H., Harley, R. A., Jimenez, J. L., Prévôt, A. S. H., and Robinson, A. L.: Review of Urban Secondary Organic Aerosol Formation from Gasoline and Diesel Motor Vehicle Emissions, Environ. Sci. Technol., 51, 1074-1093, https://doi.org/10.1021/acs.est.6b04509, 2017.

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

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

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

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

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

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, Y. H., Jayne, J. T., Ng, N. L., and Worsnop, D. R.: Highly time-resolved chemical characterization of atmospheric

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

Huang, X. H. H., Bian, Q. J., Louie, P. K. K., and Yu, J. Z.: Contributions of vehicular carbonaceous aerosols to PM2.5 in a
         roadside environment in Hong Kong, Atmos. Chem. Phys., 14, 9279-9293, https://doi.org/10.5194/acp-14-9279-
         2014, 2014.

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

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

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

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.,
         Smith, K. A., Morris, J. W., and Davidovits, P.: Ambient aerosol sampling using the Aerodyne Aerosol Mass
560   Spectrometer, J. Geophys. Res. Atmos., 108, 8425, https://doi.org/10.1029/2001jd001213, 2003.

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

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

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

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

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

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

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

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

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

Lee, B. P., Li, Y. J., Yu, J. Z., Louie, P. K. K., and Chan, C. K.: Characteristics of submicron particulate matter at the urban roadside in downtown Hong Kong-Overview of 4 months of continuous high-resolution aerosol mass spectrometer measurements, J. Geophys. Res. Atmos.,, 120, 7040-7058, https://doi.org/10.1002/2015jd023311, 2015.

Lee, S. C., Cheng, Y., Ho, K. F., Cao, J. J., Louie, P. K. K., Chow, J. C., and Watson, J. G.: PM1.0 and PM2.5 Characteristics in the Roadside Environment of Hong Kong, Aerosol Sci. Technol., 40, 157-165, https://doi.org/10.1080/02786820500494544, 2006.

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

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

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

Louie, P. K., Chow, J. C., Chen, L. W., Watson, J. G., Leung, G., and Sin, D. W.: PM2.5 chemical composition in Hong Kong: urban and regional variations, Sci. Total Environ., 338, 267-281, https://doi.org/10.1016/j.scitotenv.2004.07.021, 2005.

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

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, S. H., and Blake, D. R.: Evaluation of the effectiveness of air pollution control measures in Hong Kong, Environ. Pollut., 220, Part A, 87-94, https://doi.org/10.1016/j.envpol.2016.09.025, 2017.

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

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

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

Ning, Z., Wubulihairen, M., and Yang, F. H.: PM, NOx and butane emissions from on-road vehicle fleets in Hong Kong and 620 their implications on emission control policy, Atmos. Environ., 61, 265-274, https://doi.org/10.1016/j.atmosenv.2012.07.047, 2012.

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

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

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

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.: Particle effective density and mass during steady-state operation of GDI, PFI, and diesel passenger cars, J. Aerosol Sci, 83, 39-54, https://doi.org/10.1016/j.jaerosci.2014.12.004, 2015.

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

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

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

Saliba, G., Saleh, R., Zhao, Y., Presto, A. A., Lambe, A. T., Frodin, B., Sardar, S., Maldonado, H., Maddox, C., May, A. A., Drozd, G. T., Goldstein, A. H., Russell, L. M., Hagen, F., and Robinson, A. L.: Comparison of Gasoline Direct-645 Injection (GDI) and Port Fuel Injection (PFI) Vehicle Emissions: Emission Certification Standards, Cold-Start,

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

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

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

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

660   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 Worsnop, D. R.: Highly time- and size-resolved characterization of submicron aerosol particles in Beijing using an Aerodyne Aerosol Mass Spectrometer, Atmos. Environ., 44, 131-140, https://doi.org/10.1016/j.atmosenv.2009.03.020, 2010.

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

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., Williams, L. R., and Demerjian, K. L.: Characterization of near-highway submicron aerosols in New York City
670   with a high-resolution aerosol mass spectrometer, Atmos. Chem. Phys., 12, 2215-2227, https://doi.org/10.5194/acp-12-2215-2012, 2012.

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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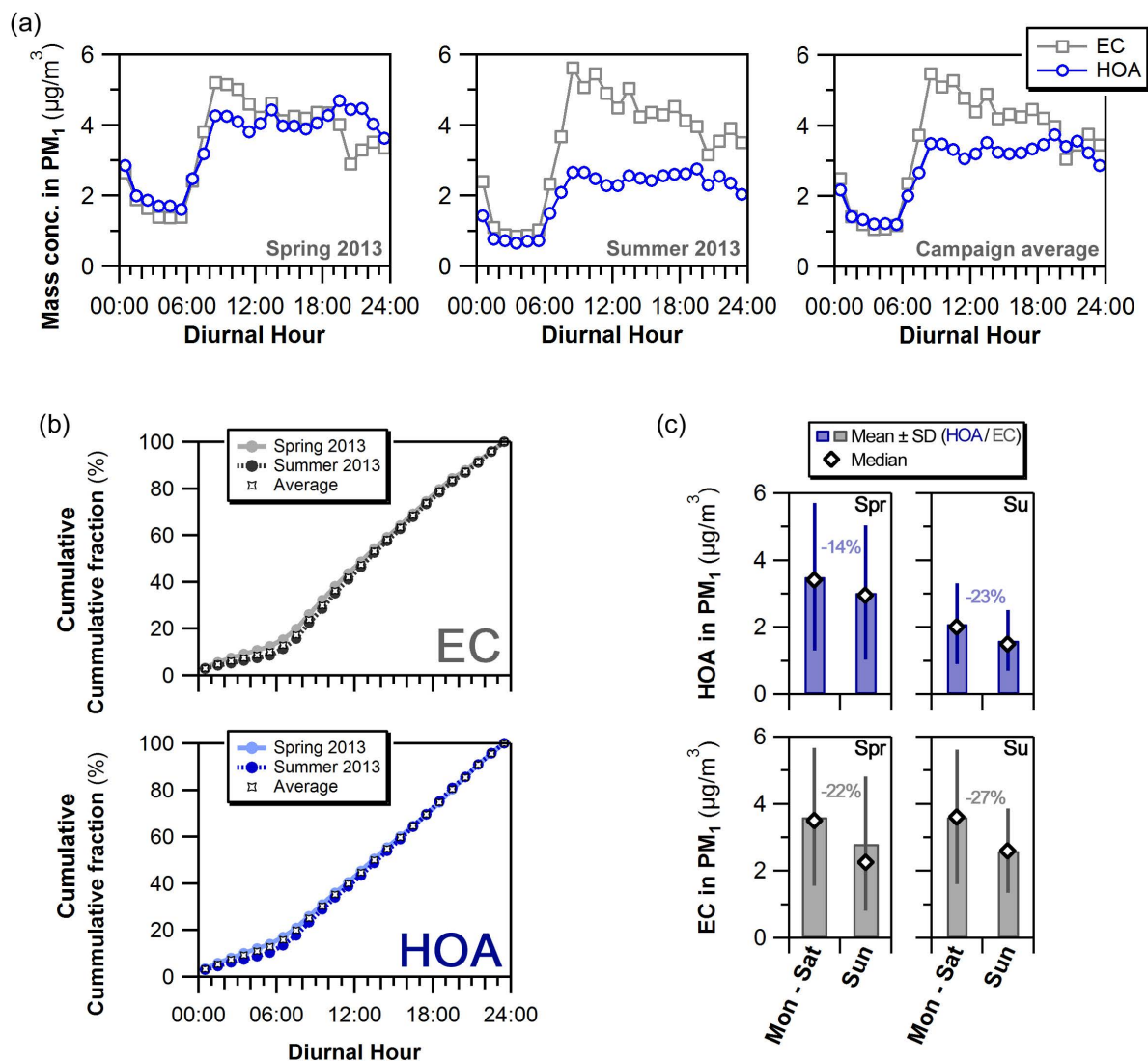
Table 1. Species concentrations from AMS and total PM$_{2.5}$ mass from TEOM in (a); EC, OC and organic aerosol constituent concentrations (AMS PMF analysis) in (b); average concentrations during the traffic counting period (May 28-31, 2013) and seasonal averages (Spring: Mar – May 2013, Summer: May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

(a)

µg/m³ Mean ± SD	Org (AMS)	SO4 (AMS)	NO3 (AMS)	NH4 (AMS)	Chl (AMS)	NR-PM$_1$ (AMS)	PM$_{2.5}$ (TEOM)
May 28-31	5.9±3.0	4.8±1.2	0.3± 0.1	1.5±0.3	<0.1	12.5±3.7	16.0±5.3
Spring	12.8±7.6	7.0±3.8	2.5±2.1	2.5±1.5	0.4±0.3	25.3±13.1	32.3±12.4
Summer	7.9±5.4	3.4±2.0	0.4±0.4	1.1±0.5	<0.1	12.7±3.6	17.3±7.5

(b)

µg/m³ Mean ± SD	EC (PM$_{2.5}$)	OC (PM$_{2.5}$)	EC (PM$_1$)	OC (PM$_1$)	HOA (PM$_1$)	COA[a] (PM$_1$)	SOA[b] (PM$_1$)
May 28-31	4.2±2.3	3.0±1.2	3.2±1.7	2.3±0.9	2.4±1.4	2.0±1.2	1.6±1.1
Spring	4.3±2.6	7.6±3.9	3.2±1.9	5.7±2.9	3.5±2.4	4.4±4.3	4.9±3.4
Summer	4.3±2.5	4.1±2.1	3.2±1.8	3.0±1.6	2.0±1.3	3.6±3.4	2.2±2.2

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

730 **Figure 1.**

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

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

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

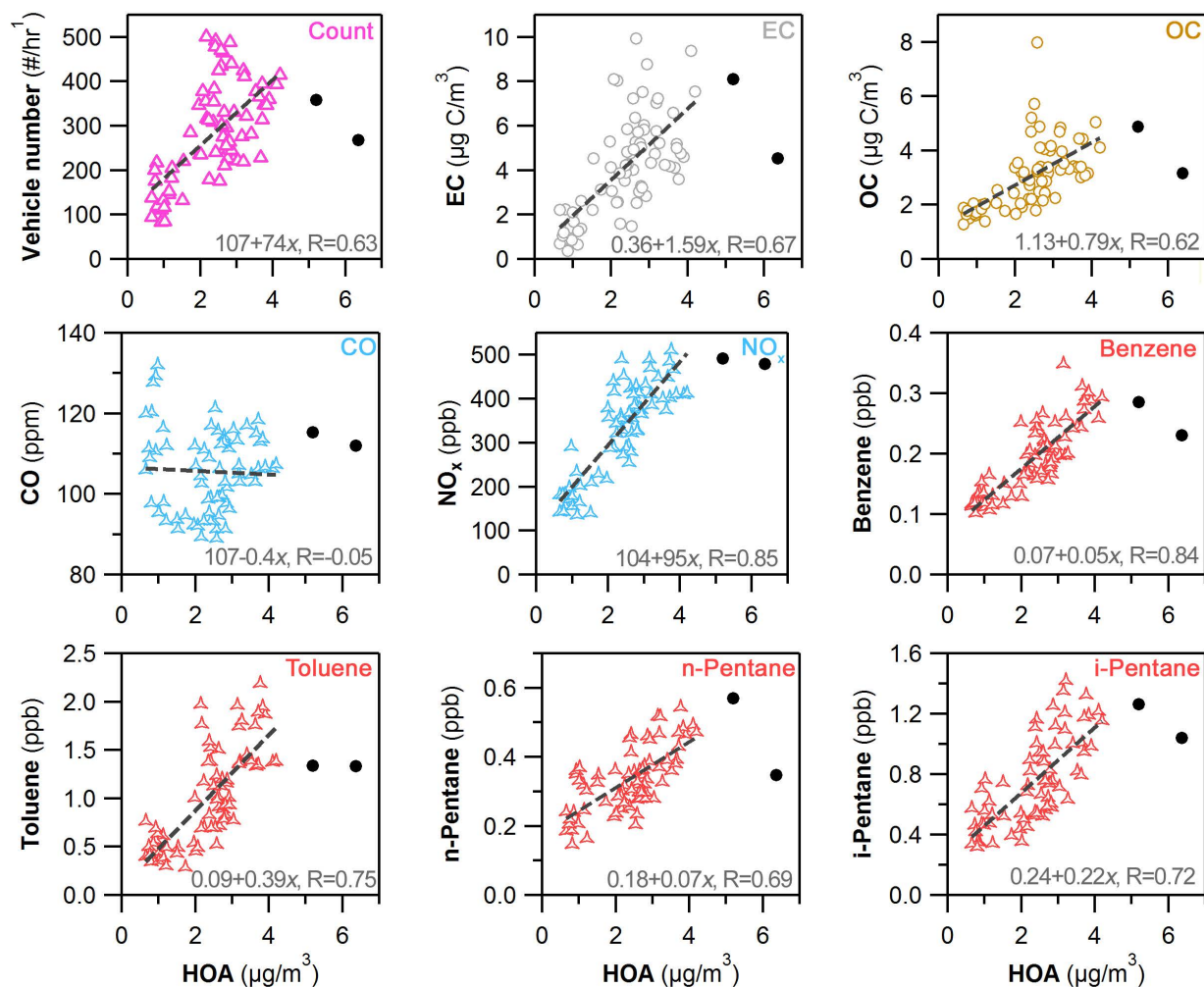



Figure 2.

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

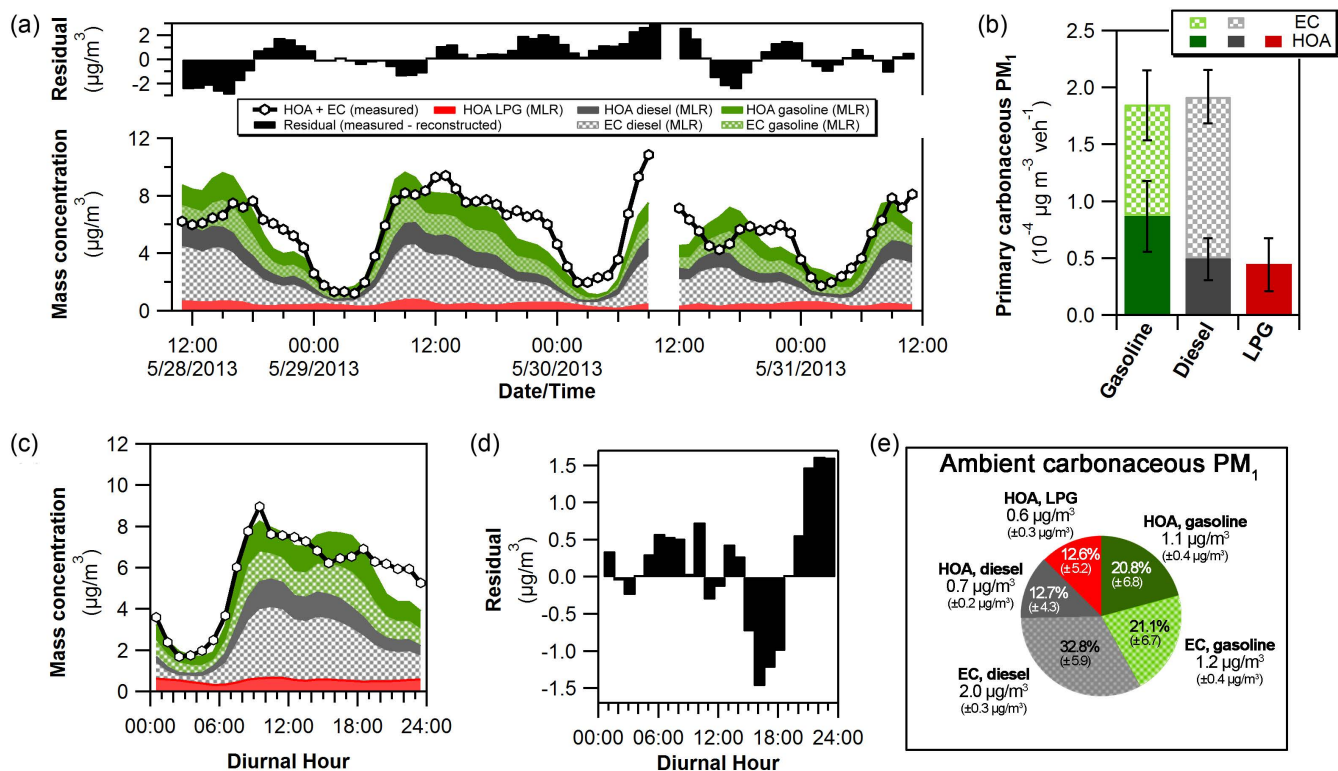



Figure 3.

745   (a) Time series of engine-type separated HOA and EC in PM$_1$, sum of actual measured HOA and EC in PM$_1$, as well as residuals as the difference between resolved HOA+EC and measured HOA+EC

(b) Contribution of engine-type separated HOA and EC to PM$_1$ normalized by the total number of vehicles per engine category; vehicle composition based on traffic count (28 – 31 May, 2013) and vehicle number scaled to average annual daily traffic (Transport Department, 2014), uncertainties from resolved fit parameters (MLR) included

750   (c) Diurnal variation of vehicle-type separated HOA and EC concentrations as well as sum of actual measured HOA and EC in PM$_1$

(d) Diurnal variation of residuals as the difference between resolved HOA+EC and measured HOA+EC in PM$_1$

(e) Average contribution of engine-type separated HOA and EC in total reconstructed HOA+EC in PM$_1$, uncertainties from resolved fit parameters (MLR) included