Atmospheric Environment 122 (2015) 876–884

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Emission factors of fine particles, carbonaceous aerosols and traces gases from road vehicles: Recent tests in an urban tunnel in the Pearl River Delta, China

Yanli Zhang [a], Xinming Wang [a, *], Guanghui Li [a, b], Weiqiang Yang [a, b], Zhonghui Huang [a, b], Zhou Zhang [a, b], Xinyu Huang [a, b], Wei Deng [a, b], Tengyu Liu [a, b], Zuzhao Huang [c], Zhanyi Zhang [d]

[a] Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China
[b] University of Chinese Academy of Sciences, Beijing 100049, China
[c] Guangzhou Environmental Monitoring Center, Guangzhou 510030, China
[d] Guangzhou Research Institute of Environmental Protection, Guangzhou 510620, China

HIGHLIGHTS

- Emission factors of air pollutants for road vehicles measured in an urban tunnel.
- Decadal changes of $PM_{2.5}$, OC and EC emission factors measured in the same tunnel.
- EFs of SO_2, NO_x, CO, CO_2 and NMHCs for road vehicles from tunnel tests.
- Diurnal variation of emission factors revealed changes with fleet compositions.

ARTICLE INFO

Article history:
Received 31 January 2015
Received in revised form
7 August 2015
Accepted 10 August 2015
Available online 11 August 2015

Keywords:
Emission factors (EFs)
Tunnel study
Motor vehicles
Fine particles
Carbonaceous aerosols
Trace gases

ABSTRACT

Motor vehicles contribute primarily and secondarily to air quality problems due to fine particle ($PM_{2.5}$) and ozone (O_3) pollution in China's megacities. Characterizing vehicle emission with the rapid change of vehicle numbers and fleet compositions is vital for both bottom-up emission survey and top–down source apportioning. To obtain emission factors (EFs) of $PM_{2.5}$, carbonaceous aerosols and trace gases for road vehicles, in urban Guangzhou we conducted a field campaign in 2014 in the Zhujiang Tunnel, a heavily burdened tunnel with about 40,000 motor vehicles passing through each of its two separated bores per day. $PM_{2.5}$ and volatile organic compounds (VOCs) were sampled for offline analysis while trace gases including SO_2, NO_x and CO were measured online and in situ. An eddy covariance system with an integrated 3-D sonic anemometer was also adopted to measure CO_2 and winds inside the tunnel. We recorded an average fleet composition of 61% light-duty gasoline vehicles (LDVs) + 12% heavy-duty diesel vehicles (HDVs) + 27% liquefied petroleum gas vehicles (LPGVs), and EFs of 82.7 ± 28.3, 19.3 ± 4.7 and 13.3 ± 3.3 mg veh^{-1} km^{-1}, respectively, for $PM_{2.5}$, organic carbon (OC) and elemental carbon (EC). These EFs were respectively 23.4%, 18.3% and 72.3% lower when compared to that measured in the same tunnel in 2004. EFs of $PM_{2.5}$, OC and EC were higher at night time (148 ± 126, 29 ± 24 and 21 ± 18 mg veh^{-1} km^{-1}, respectively) due to significantly elevated fractions of HDVs in the traffic fleets. An average ratio of OC to EC 1.45 from this tunnel study was much higher than that of ~0.5 in previous tunnel studies. The EFs of SO_2, NO_x, CO, CO_2 and NMHCs for road traffic were also obtained from our tunnel tests, and they were 20.7 ± 2.9, (1.29 ± 0.2)E+03, (3.10 ± 0.68)E+03, (3.90 ± 0.49)E+05, and 448 ± 39 mg veh^{-1} km^{-1}, respectively.

1. Introduction

Air pollution due to fine particles ($PM_{2.5}$) and ozone (O_3) has become tough problems of wide concern in China's megacities

* Corresponding author. State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China.
E-mail address: wangxm@gig.ac.cn (X. Wang).

http://dx.doi.org/10.1016/j.atmosenv.2015.08.024
1352-2310/

(Chan and Yao, 2008; Zhang et al., 2012a; Guo et al., 2014) due to impaired visibility (Che et al., 2007; Wu et al., 2007; Cao et al., 2012), adverse health endpoints (Xu et al., 2008; Tie et al., 2009; Huang et al., 2012; Chen et al., 2013b; Shang et al., 2013) and damage to ecological services (Krupa et al., 2001; Wang et al., 2007). Particularly in the urban areas, vehicle exhausts are convinced a major source of air pollutants leading to China's air quality headaches including $PM_{2.5}$ and O_3. Apart from emission of primary particulate matters (PM) (Kleindienst et al., 2002; Huang et al., 2014), motor vehicles can also emit a variety of trace gases such as VOCs and NO_x (Schauer et al., 1999; Kirchstetter et al., 1999; Hao et al., 2000; Imhof et al., 2006), which are important precursors to both secondary aerosols (Ng et al., 2007) and ozone (Derwent et al., 1996). As an example, in China's Pearl River Delta (PRD) region, a substantial fraction of aromatic hydrocarbons were sourced from vehicle exhausts (Liu et al., 2008b; Cheng et al., 2010a; Zhang et al., 2012b), and they were major anthropogenic precursors of both secondary organic aerosols (SOA) (Ding et al., 2012; Wang et al., 2013) and ambient ozone (Cheng et al., 2010b; Zhang et al., 2013b). Therefore vehicle emission control is an enduring task to combat air pollution in China in light of its rapidly increasing vehicle numbers.

The contribution of vehicle emission to $PM_{2.5}$ in China, however, is a debatable issue. Even in Beijing, the capital city of China, observation-based $PM_{2.5}$ source apportioning revealed quite large gaps for the percentages shared by vehicle exhausts (Song et al., 2007; Zhang et al., 2013a; Huang et al., 2014). On the other hand, bottom-up emission inventories still have pretty large uncertainties for vehicle emission. Zheng et al. (2009) demonstrated that vehicle emissions accounted for 67%, 36%, 39%, 36%, and 22% of CO, NO_x VOCs, $PM_{2.5}$ and PM_{10}, respectively, in the Pearl River Delta (PRD) region with 2006 as the base year, and they claimed uncertainties from −60% to +70% for both particle and gaseous pollutants from vehicle emissions in the emission inventory. A major reason for the uncertainties related to the top–down or bottom-up survey of vehicle emission is the lack of extensive characterization of vehicle emission, particularly the source profiles and emission factors, in China. As a matter of fact, the possession of civil vehicles numbers in China increased rapidly from about 27 million in 2004 to over 127 million in 2013 with an annual increasing rate of ~40% (National Bureau of Statistics of China (NBSC), 2014). This would inevitably induce great changes in China's on-road vehicles including the engines, oil consumption, energy efficiency and emission standards, and thereby result in fast changing emission factors and chemical compositions of air pollutants from vehicles. Under this situation, updating emission factors and source profiles for motor vehicles is in pressing need to keep pace with the fast changing motor vehicles, either in numbers or in compositions.

Emission factors (EFs) for motor vehicles can be measured by a variety of methods including chassis and engine dynamometer testing under controlled conditions in laboratories (Artelt et al., 1999; Nine et al., 1999), remote sensing method (Bishop and Stedman, 1996), on-road (chase) measurements (Shorter et al., 2005) and tunnel studies (Jamriska et al., 2004; Hueglin et al., 2006). Among them tunnel study has its advantages in obtaining absolute levels of emissions by capturing a cross-section of the on-road vehicle fleet and representing real-world operation conditions (Franco et al., 2013). In China, on-board measurements of emission factors for pollutants from vehicles has been extensively performed by using a portable emissions measurement system (PEMS), mostly in Beijing (Liu et al., 2009; Huo et al., 2011; Wang et al., 2011; Shen et al., 2014; Yao et al., 2015), Shanghai (Chen et al., 2007), Chongqing (Wang et al., 2012a) and Macao (Hu et al., 2012; Wang et al., 2014). Remote sensing measurements of EFs were previously conducted in Hangzhou in 2004 and 2005 (Guo et al., 2007), and Li et al. (2013) reported $PM_{2.5}$ emissions from light-duty vehicles measured on a chassis dynamometer. However, only a few tunnel studies about the emission factors for road vehicles were carried out in mainland China, all in the Pearl River Delta region except one measuring PM_{10}-bound polycyclic aromatic hydrocarbons in the Fu Gui-shan Tunnel in Nanjing (Chen et al., 2013a). He et al. (2006) chemically characterized $PM_{2.5}$ based on the 4 sets of particle samples collected from the Wutong Tunnel in the PRD's Shenzhen city, and Cheng et al. (2006; 2010c) and Ho et al. (2006, 2009) measured EFs of $PM_{2.5}$, VOCs and carbonyl compounds in the Shing Mun Tunnel in Hong Kong. Based on studies in the Zhujiang Tunnel in urban Guangzhou, the same one for this study, Wang et al. (2001) calculated EFs of PM_{10} and some gaseous pollutants from a campaign in 1999; Huang et al. (2006) and He et al. (2008) reported size distribution of EC and $PM_{2.5}$ emissions from a campaign in 2004. As chemical compositions and EFs of pollutants from vehicles would change quite a lot accompanying with the rapid change of vehicle numbers, fleet compositions, fuel quality and control strategies, these EFs need to be updated to reflect current vehicle emissions and to include more pollutant species. However, a very recent study in the Zhujiang Tunnel in 2013 only secured EFs of ammonia (Liu et al., 2014), $PM_{2.5}$ and major $PM_{2.5}$ components (Dai et al., 2015). Moreover, only two particulate samples were collected per day, this would mask peak or valley values as large diurnal variations of EFs were observed in many tunnel and on-board tests (Ammoura et al., 2014; Shen et al., 2014).

This study aimed to update EFs of $PM_{2.5}$ and carbonaceous aerosols (OC and EC) and to extend our measurements for obtaining EFs of gaseous pollutants including VOCs, NO_x, CO and SO_2 for road vehicles in the Pearl River Delta region based on our campaign in the Zhujiang Tunnel in June 2014. These results are not only the latest emission factors of gaseous and particulate pollutants for road vehicles in the PRD region, but also valid data to assess the effectiveness of vehicle emission controls in the recent decade when comparing them with that previously measured in the same tunnel.

2. Methods

2.1. Description of the Zhujiang Tunnel

The Zhujiang Tunnel (23.11°N, 113.23°E), located in the Liwan District in urban Guangzhou, is the first underwater tunnel crossing the Pearl River in the Pearl River Delta. It has two bores with two lanes for each direction; its total length is 1238.5 m with a 721 m flat underwater section (Fig. 1). We conducted our tests in this flat underwater section. The outlet sampling site is about 50 m to the end of this flat underwater section. Further information can be

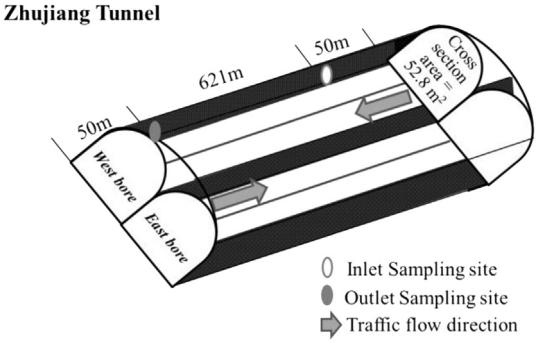

Fig. 1. Schematic diagram of the straight line segment of the Zhujiang Tunnel.

found elsewhere (Huang et al., 2006; He et al., 2008; Liu et al., 2014; Dai et al., 2015). The tunnel has a heavy traffic burden of ~40,000 vehicles per day. There is a 50 km/h speed limit in the tunnel. Normally the ventilation system supplies fresh air into the tunnel to lower air pollutants inside, however, the ventilation system and vent-pipes of the system were all closed during our tests in the tunnel to avoid dilution by ventilation so that accumulation of air pollutants inside the tunnel was solely attributed to vehicle emissions.

2.2. Field works

From June 25th to July 1st 2014, we set two monitoring stations at two ports, each 50 m from the inlet and outlet (Fig. 1), for simultaneously detecting NO_x, SO_2, CO and CO_2 and collecting filter-based $PM_{2.5}$ and canister-contained VOCs samples. At each station two filter-based $PM_{2.5}$ sampler were employed: one is a high-volume one (Tsich Environmental, Inc.) at a flow rate of 1.13 m^3 min^{-1} with the 8 × 10 inch Whatman quartz fiber filters, and another is a medium volume sampler (Model PQ-200, BGI Inc.) at a flow rate of 16.7 L min^{-1} with the Whatman 47 mm diameter PTFE filters. Each set of filter-based samples were collected in 4-hr intervals, namely 02:00–06:00, 06:00–10:00, 10:00–14:00, 14:00–18:00, 18:00–22:00, and 22:00–02:00. During the campaign altogether 28 pairs of filter-based $PM_{2.5}$ samples, including 56 Quartz-filter ones and 56 Teflon-filter ones, were obtained. Two sets of blank samples were also collected by loading filters into the sampler but without pulling air through. All samples were stored in a refrigerator (−20 °C) before chemical analysis.

VOCs samples were collected using pre-evacuated 2-L electropolished stainless steel canister. During the sampling a Model 910 Pressurized Canister Sampler (Xonteck, Inc.) was adopted to allow each canister to be filled in 60 min at a constant flow of 66.7 ml/min and with the ending air pressure of about 2 atm inside the canister. The samples were collected on two workdays (June 25th and 26th) and at weekends (June 28th and 29th) during intervals of 02:00–03:00, 07:00–08:00, 08:00–09:00, 09:00–10:00, 10:30–11:30, 14:00–15:00, 17:00–18:00, 18:00–19:00, and 19:00–20:00 in each day. We got one more pair during 02:00–03:00 on June 27th and another during 02:00–03:00 on June 30th. Totally 76 valid VOCs samples were collected in the tunnel. These samples were transported back to lab and get analyzed in a week.

Trace gases, including NO_x, SO_2, CO and CO_2, were continuously monitored. NO and NO_x were detected using a chemiluminescence NO–NO_2–NO_x analyzer (Thermo Electron Corporation, Model 42i); SO_2 was measured by a pulsed UV fluorescence (TEI, model 43S); CO was measured with a gas filter correlation, nondispersive infrared analyzer (API, Model 300). Detailed information about online monitoring of these gases can be found elsewhere (Zhang et al., 2010; Liu et al., 2014).

Carbon dioxide (CO_2), as well as wind speed, wind direction, and temperature, were measured in situ using the IRGASON eddy covariance system (Campbell, Inc.) with an integrated CO_2 and H_2O open-path gas analyzer and a 3-D Sonic Anemometer. The IRGASON gas analyzer is a non-dispersive mid-infrared absorption analyzer with a mercury cadmium telluride (MCT) detector in its lower arm to measure the decrease in radiation intensity due to absorption and then to infer concentrations using the Beer–Lambert Law. The accuracy is 1% and the precision is 0.2 mg/m^3 (0.15 μmol/mol) for CO_2. The 3-D Sonic Anemometer measures the 3-D wind speed u_x, u_y and u_z with accuracies < ±0.08 m s^{-1} for u_x and u_y and < ±4.0 cm s^{-1} for u_z. The accuracy of wind direction is within ±0.7° while horizontal wind at 1 m s^{-1}, and ambient temperature was measured with an accuracy of ±0.5 °C.

2.3. Traffic counts

A video camera was placed at the outlet to record the passing vehicles during the sampling periods. The videotapes were used to determine the vehicle counts and to classify the vehicles into 11 types, which including sedan cars, large passengers cars, mini-trucks (MTs), light-duty trucks (LDTs), medium-duty trucks (MDTs), heavy-duty trucks (HDTs), light-duty buses (LDBs), medium-duty buses (MDBs), heavy-duty buses (LDBs), taxis, and motorcycles according to the taxonomy in NBSC (2014).

2.4. Chemical analysis

Methods of $PM_{2.5}$ and OC/EC mass concentrations were described elsewhere (Wang et al., 2012b). The Teflon filters were used for the gravimetric analysis, and quartz filters were chosen for the OC/EC analysis. VOCs were analyzed using a Model 7100 Preconcentrator (Entech Instruments Inc., California, USA) coupled with an Agilent 5973N gas chromatography-mass selective detector/flame ionization detector (GC-MSD/FID, Agilent Technologies, USA). Detailed cryogenically concentration steps are described elsewhere (Zhang et al., 2013b).

2.5. Quality control and quality assurance

For collecting $PM_{2.5}$ in the tunnel, quartz filters were baked at 450 °C for 4 h to remove organic impurities prior to field sampling, wrapped in aluminum foils, and zipped in Teflon bags. Sampling flow was checked using an orifice calibration unit with a manometer. After sampling, the filters were again wrapped in aluminum foils, zipped in Teflon bags, and stored in a freezer at −20 °C until analysis. All the OC/EC data were corrected using the field blanks. All trace gas analyzers were zero-point adjusted with zero air twice a day and re-calibrated with authentic standards once a day.

For sampling VOCs, each canister before use was subjected to at least five evacuation/pressurization cycles using humidified zero air. To check if there was any contamination in the canisters, each canister was again pressurized with humidified zero air at the end of the evacuation/pressurization cycles and stored in the laboratory for at least 24 h, and then analyzed the same way as air samples. A canister was certificated as clean if all target compounds were not detected or were below method detection limits (MDLs). After leak checking and cleansing, the canisters were re-evacuated to <0.05 mm Hg and remained in this condition until field use. After lab analysis, target VOCs were identified based on their retention times and mass spectra, and were quantified by an external calibration method. Field blank canisters were refilled with zero air in the lab and then analyzed the same way as the ambient air samples after at least 24 h storage. All target compounds in the field blank samples were below their MDLs.

2.6. Emission factor

Average emission factors for vehicles traveling through the tunnel during a time interval T were calculated the same as that in previous studies (Pierson and Brachaczek, 1983; Pierson et al., 1996).

$$EF_{\text{fleet}} = \frac{\Delta C_p \times V_{\text{air}} \times T \times A}{N \times l} \quad (1)$$

Where $\boldsymbol{EF}_{fleet}$ (mg veh^{-1} km^{-1}) is the mean emission factor per vehicle for a given species expressed as emitted mass per kilometer, $\boldsymbol{\Delta C}_p$ (mg/m^3) is the concentration difference between the measurements at the tunnel exit and its entrance, $\boldsymbol{V}_{air}$ (m/s) is the air

velocity parrell to the tunnel sensed by the 3-D Sonic Anemometer, Air velocity varied from 2.75 to 4.00 m/s with an average of 3.44 m/s during the sampling period. ***A*** is the tunnel cross section area in m^2 ($A = 52.8\ m^2$), ***N*** is the total traffic number passing the tunnel during the time interval ***T***(s) (1 h in this study), and ***l*** is the length of the tunnel between the two monitoring stations in km, The length of the tunnel between the two stations is 621 m within the 721 m flat underwater section. As there is a 50 km/h speed limit in the tunnel, the traffic speed range is 20–47 km/h during our sampling period.

3. Results and discussion

3.1. Traffic fleets passing through the tunnel bore

In the tunnel bore where we conducted our tests, 39,164–43,306 motor vehicles passed through per day. The traffic density varied between 754 and 2554 vehicles per hour. We further categorized motor vehicles into three fuel-types, namely heavy-duty diesel vehicles (HDVs) (which including heavy-duty trucks and large passenger cars), light-duty gasoline vehicles (LDVs) (including sedan cars, light-duty trucks, and motorcycles), and liquefied petroleum gas vehicles (LPGVs) (including buses and taxies). Variations of vehicle numbers and the percentages shared by LDVs, HDVs, and LPGVs on a weekend day and a workday were shown in Fig. 2. In average, LDVs, HDVs and LPGVs shared fractions of 61%, 12%, and 27%, respectively. Percentages of LDVs ranged 29–84% and 35–77%, respectively, on workdays and weekends. On weekends, the average fraction of LDVs in the daytime (08:00 am – 20:00 pm) was 73 ± 7%, higher than that of 53 ± 24% in the night time (20:00 pm – the next day 08:00 am). LDVs showed a diurnal pattern: increasing rapidly from 06:00 am to ~10:30 am, maintaining at this high platform levels to ~22:30 am, and then dropping to a minimum at about 02:00 am in the next day. HDVs and LPGVs, on the contrary, shared higher fractions in the night time than in the daytime. While fractions of HDVs and LPGVs were quite stable during daytime with an average of 9% and 20%, respectively, they reached as high as 24% and 60%, respectively, at about 05:30–06:00 am and 01:30–02:00 am. On weekdays, a distinct difference was that in the early morning vehicle numbers increased more rapidly and reached the high platform levels about 2 h earlier when compared to the diurnal pattern in weekends.

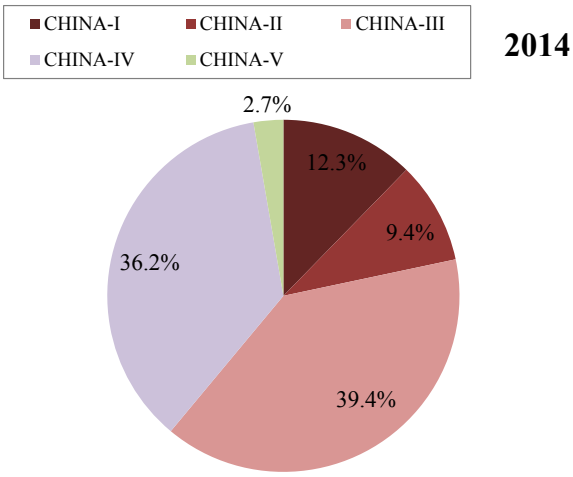

Fig. 3. Compositions of vehicles in 2014 in Guangzhou by emission standards.

3.2. Concentrations and emission factors (EFs)

Table 1 showed measured inlet/outlet concentrations and calculated emission factors, as well as their ranges and 95% confidence interval (95% C.I.) for $PM_{2.5}$, OC, EC, SO_2, NO_x, CO, CO_2, and total NMHCs. Total NMHCs in this study refers to the sum of sixty-two C_2–C_{11} hydrocarbons. The CO_2 outlet-inlet concentration gaps in the Zhujiang Tunnel were about 2 times of that in the Shing Mun Tunnel in Hong Kong (Ho et al., 2009), largely due to heavier traffic burdens. The average inlet concentration of OC and EC were 20.8 ± 2.5 μg/m^3 and 6.2 ± 1.5 μg/m^3, respectively, with an average OC/EC ratio of 3.8 ± 0.4; while that at the outlet were 47.8 ± 5.5 and 23.7 ± 2.7 μg/m^3, respectively, with an average OC/EC ratio of 2.1 ± 0.2.

As listed in Table 1, the emission factors for the traffic fleets in the Zhujiang Tunnel were 82.7 ± 28.3, 19.3 ± 4.7, 13.3 ± 3.3, 20.7 ± 2.9, (1.29 ± 0.2)E+03, (3.10 ± 0.68)E+03, (3.90 ± 0.49)E+05,

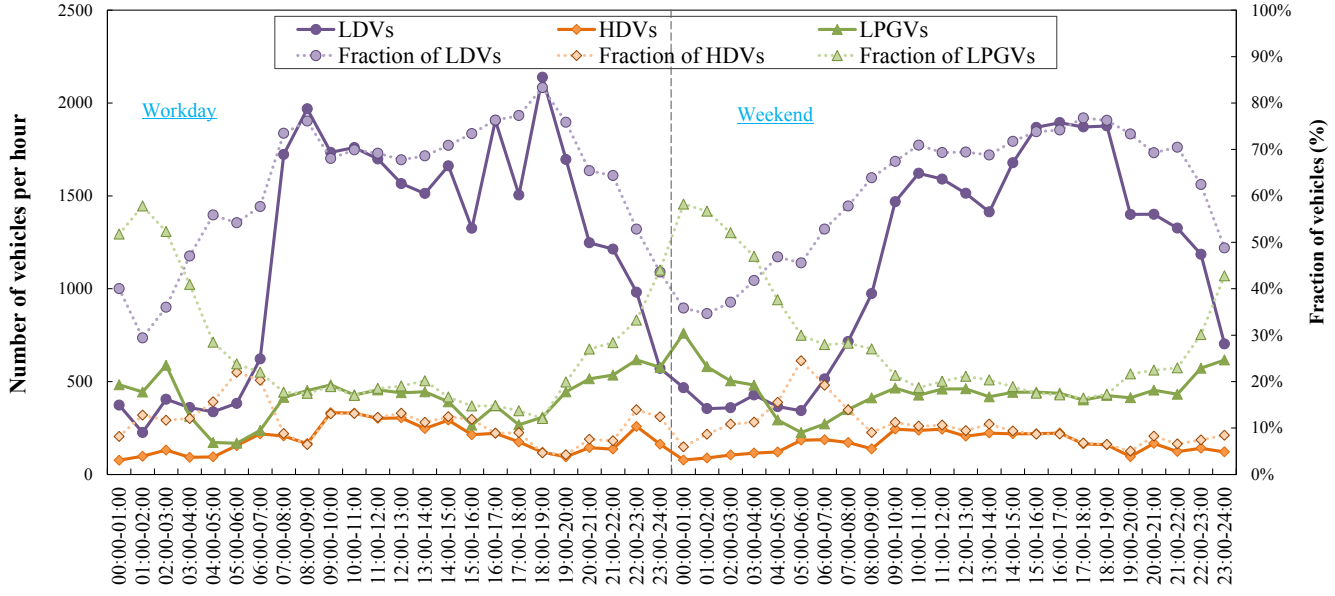

Fig. 2. Diurnal variations of three categories of vehicles on workday and weekend.

Table 1
Average concentrations (μg/m^3) and emission factors (mg veh^{-1} km^{-1}) of target compounds in Zhujiang Tunnel.

Species	Inlet concentration		Outlet concentration		Emission factors		
	Mean	95% C.I.	Mean	95% C.I.	Mean	95% C.I.	Range
$PM_{2.5}$	101	14.7	204	35.8	82.7	28.3	4.4–310
OC	20.8	2.5	47.8	5.5	19.3	4.7	1.6–58
EC	6.2	1.5	23.7	2.7	13.3	3.3	0.3–45
SO_2	17.5	0.8	43.0	1.1	20.7	2.9	11–39
NO_x	0.58E+03	0.03E+03	2.45E+03	0.01E+03	1.29E+03	0.20E+03	(0.43–2.26)E+03
CO	1.40E+03	0.15E+03	5.35E+03	0.02E+03	3.10E+03	0.68E+03	(1.60–12.4)E+03
CO_2	8.3E+05	0.07E+05	1.30E+06	0.02E+06	3.90E+05	0.49E+05	(0.78–7.2)E+05
TNMHCs	133	14	534	70	448	39	272–727

and 448 ± 39 mg veh^{-1} km^{-1} for $PM_{2.5}$, OC, EC, SO_2, NO_x, CO, CO_2 and NMHCs, respectively. OC and EC were found to account for 23.6% and 16.7% of the emitted $PM_{2.5}$ mass with an average OC/EC of 1.45. This was different from higher emission factors of EC than OC in previous studies, such as that with OC/EC ratio of 0.30 in the Kaisernuhlen Tunnel in Austria (Handler et al., 2008), 0.54 in the Shing Mun Tunnel in Hong Kong (Cheng et al., 2010c), and 0.49 in the same Zhujiang Tunnel in 2004 (He et al., 2008).

3.3. Comparison of emission factors with that from other tunnel studies

The emission factors of $PM_{2.5}$, OC and EC in this study were compared with that from other tunnel studies over the world (Table 2). EF of $PM_{2.5}$ (82.7 mg veh^{-1} km^{-1}) in this study was lower than that of 131 mg veh^{-1} km^{-1} in an urban tunnel in Hong Kong (Cheng et al., 2010c) with traffic fleet composition of 41% gasoline + 50% Diesel + 9% LPG. It also showed a decreasing trend when compared to that of 110 mg veh^{-1} km^{-1} in the same tunnel in 2004 with fleet composition of 80% LDV + 20% HDV (He et al., 2008) or that of 92.4 mg veh^{-1} km^{-1} in 2013 with fleet composition of 60% LDV + 14% HDV + 26% LPGV (Dai et al., 2015). However, this EF for $PM_{2.5}$ was much higher than that reported about a decade ago in the USA or Europe, such as 52 mg veh^{-1} km^{-1} in the Sepulveda Tunnel in Los Angeles with 97.4% LDV + 2.6% HDV (Gillies et al., 2001), 46.8 mg veh^{-1} km^{-1}in the Kaisernuhlen Tunnel in Vienna (Laschober et al., 2004), 49 mg veh^{-1} km^{-1} in the Kingsway Tunnel in the United Kingdom (Imhof et al., 2006), and 67 mg veh^{-1} km^{-1} in the Söderleds Tunnel in Stockholm with 90% LDV + 5% HDV + 5% Diesel (Kristensson et al., 2004). Apart from emission control technologies, fleet composition is also an important factor determining the EFs. Typically higher EFs of $PM_{2.5}$, 3–5 times that from this study, were observed for HDVs or Diesel vehicles (Allen et al., 2001; Cheng et al., 2010c; Jamriska et al., 2004; Imhof et al., 2006). Contrarily, in many tunnels dominated by LDVs or gasoline vehicles, EFs of $PM_{2.5}$ were much lower: the maximum of 53.4 mg veh^{-1} km^{-1} was reported in Milwaukee (Lough et al., 2005) and as low as 5.5 mg veh^{-1} km^{-1} was reported in San Francisco (Allen et al., 2001), and quite a lot were fell between them (Table 2; Lough et al., 2005; Imhof et al., 2006; Handler et al., 2008; Chiang and Huang, 2009; Cheng et al., 2010c; Mancilla and Mendoza, 2012). In this study the traffic fleet in the Zhujiang Tunnel has a much higher fractions of HDV, so the EFs of $PM_{2.5}$ were between that in HDV-dominated tunnels and LDV-dominated ones.

The OC emission factor of 19.3 mg veh^{-1} km^{-1} in this study were lower than that of 35.7 mg veh^{-1} km^{-1} for total fleet in the Shing

Table 2
Comparison of emission factors of $PM_{2.5}$, OC and EC with other studies.

References	Tunnel	Cities/Country	Test year	Fuel type	Emission factors (mg veh^{-1} km^{-1})		
					$PM_{2.5}$	OC	EC
LDV dominance							
(Mancilla and Mendoza, 2012)	Monterrey tunnel	Mexico	2009	97% Gasoline	22.8	12.6	5.7
(Allen et al., 2001)	Caldecott tunnel	San Franciso	1997	LDV	5.5	–	–
(Kristensson et al., 2004)	Sweden	Stockholm	1998–1999	90%LDV + 5%HDV+5%Diesel	67.0	–	–
(Cheng et al., 2010c)	Shing Mun Tunnel	Hong Kong, China	2003	Gasoline + LPG	16.6	8.5	3.2
(Lough et al., 2005)	Kilborn Tunnel	Milwaukee, USA	2000	>97%LDV	24.1	6.4	6.9
(Lough et al., 2005)	Howell Tunnel	Milwaukee, USA	2000	>90%LDV	53.4	12.9	6.6
			2000–2001	>90%LDV	35.9	8.4	3.3
(Handler et al., 2008)	Kaisernuhlen tunnel	Vienna, Austria	2005	mixed LDV + 9.6%HDV	26.0	5.4	17.8
(Imhof et al., 2006)	Kingsway Tunnel	United Kingdom	2003	LDV	19.0	–	–
(Chiang and Huang, 2009)	Chung-Liao tunnel	Taiwan	Unknown	>90%LDV	38.0	15.1	4.7
HDV dominance							
(Gillies et al., 2001)	Sepulveda Tunnel	Los Angeles	1996	97.4%HDV + 2.6%LDV	52.0	–	–
(Allen et al., 2001)	Caldecott tunnel	San Franciso	1997	HDV	430	–	–
(Cheng et al., 2010c)	Shing Mun Tunnel	Hong Kong, China	2003	Diesel	257	67.9	131.0
(Jamriska et al., 2004)	Woolloongabba Tunnel	Brisbane City, Australia	Unknown	Diesel bus	267	–	–
(Imhof et al., 2006)	Kingsway Tunnel	United Kingdom	2003	HDV	381	–	–
Total fleet							
(Laschober et al., 2004)	Kaisernuhlen tunnel	Vienna, Austria	2002	Total fleet	46.8	–	27.9
(Cheng et al., 2010c)	Shing Mun Tunnel	Hong Kong, China	2003	41%Gasoline + 50%Diesel+9%LPG	131	35.7	65.8
(He et al., 2008)	Zhujiang Tunnel	Guangzhou, China	2004	80%LD + 20%HD	110	24.3	49.6
(Dai et al., 2015)	Zhujiang Tunnel	Guangzhou, China	2013	60%LDV + 14%HDV + 26%LPGV	92.4	16.7	16.4
This study	**Zhujiang Tunnel**	**Guangzhou, China**	**2014**	**61%LDV + 12%HDV + 27%LPGV**	**82.7**	**19.3**	**13.3**

Mun Tunnel in Hong Kong (Cheng et al., 2010c) and 24.3 mg veh^{-1} km^{-1} in the Zhujiang Tunnel in 2004 (He et al., 2008), but accordingly higher than that ranging 5.40–15.1 mg veh^{-1} km^{-1} in LDV-dominated tunnels (Mancilla and Mendoza, 2012; Cheng et al., 2010c; Lough et al., 2005; Handler et al., 2008; Chiang and Huang, 2009), and near that of 16.7 mg veh^{-1} km^{-1} tested in 2013 in the Zhujiang Tunnel (Dai et al., 2015). Similarly, the EC emission factor of 13.3 mg veh^{-1} km^{-1} in this study were much lower when compared to that of 65.8 mg veh^{-1} km^{-1} for total fleet in Hong Kong (Cheng et al., 2010c) and 49.6 mg veh^{-1} km^{-1} in the Zhujiang Tunnel in 2004 (He et al., 2008). It was also about 20% lower than that of 16.4 mg veh^{-1} km^{-1} in the Zhujiang Tunnel in 2013 (Dai et al., 2015). However, this value was much higher when compared to 3.2–6.9 mg veh^{-1} km^{-1} reported in other LDV-dominated tunnels (Mancilla and Mendoza, 2012; Cheng et al., 2010c; Lough et al., 2005; Chiang and Huang, 2009), except an LDV-dominated tunnel in Vienna, where EF of EC reached 17.8 mg veh^{-1} km^{-1} (Handler et al., 2008).

A comparison of EFs for gaseous pollutants is shown in Table 3. Only a study in the Tauern Tunnel revealed much higher emission factor of 3131 mg veh^{-1} km^{-1} for NO_x (Schmid et al., 2001) than that of 1286 ± 204 mg veh^{-1} km^{-1} in this study. Studies in the Söderleds Tunnel (1360 mg veh^{-1} km^{-1}; Kristensson et al., 2004) and in the Gubrist Tunnel (1050 mg veh^{-1} km^{-1}; Legreid et al., 2007) obtained comparable high EFs for NO_x. Much lower EFs for NO_x were observed in gasoline vehicle tunnels, such as 145–331 mg veh^{-1} km^{-1} in the Hsuehshan Tunnel (Chang et al., 2009) and 110–130 mg veh^{-1} km^{-1} in the Loma Larga Tunnel (Mancilla et al., 2012; Araizaga et al., 2013). These large differences suggest that HDVs contribute more to NO_x than LDVs. Other studies reported moderate EFs for NO_x within 263–900 mg veh^{-1} km^{-1} (Hwa et al., 2002; Cheng et al., 2006; Chiang et al., 2007; Ameur-Bouddabbous et al., 2012). EFs for NMHCs averaged 448 ± 38 mg veh^{-1} km^{-1} in this study, quite similar to that of 440 mg veh^{-1} km^{-1} in the Taipei Tunnel (Hwa et al., 2002) and 460 mg veh^{-1} km^{-1} in the Chuang-Liao tunnel (Chiang et al., 2007), or 460 mg veh^{-1} km^{-1} in the Gubrist Tunnel (Legreid et al., 2007), but higher than 115 mg veh^{-1} km^{-1} in the Shing Mun Tunnel (Ho et al., 2009) and 96–121 mg veh^{-1} km^{-1} in the Hsuehshan Tunnel (Chang et al., 2009). However, all the above mentioned EFs for NMHCs are quite low when compared to that from 1160 ± 50 (Mancilla et al., 2012) to 1540 ± 80 (Araizaga et al., 2013) reported in the Loma Large Tunnel.

EFs of SO_2 averaged 21 ± 3 mg veh^{-1} km^{-1}, comparable with that of 20 mg veh^{-1} km^{-1} in Taiwan (Chang et al., 2009), but much higher than that ranging 3.0–11.6 mg veh^{-1} km^{-1} from other surveys (Chang et al., 2009; Ameur-Bouddabbous et al., 2012). The relatively higher EFs for SO_2 in this study might be a reflection of higher sulfur contents in our fuels (Liu et al., 2008a).

The EFs of CO and CO_2 in different tunnels were also listed in Table 3. The EFs for CO was unusually high (11800 mg veh^{-1} km^{-1}) in the Loma Larga Tunnel (Chang et al., 2009), 2.2–6.4 times of those from other studies. EFs of CO (3096 ± 680 mg veh^{-1} km^{-1}) in this study also quite approximated that of 3640 mg veh^{-1} km^{-1} in the Taipai Tunnel (Hwa et al., 2002). They were within the 5300 mg veh^{-1} km^{-1} in the Söderleds Tunnel (Kristensson et al., 2004) and 1845–1900 mg veh^{-1} km^{-1} in the Shing Mun Tunnel (Cheng et al., 2006; Ho et al., 2009). The CO_2 emission factor, as an indicator of energy efficiency, was 3.9E + 05 mg veh^{-1} km^{-1} from this study. It was nearly 30% higher than that of 3.10E + 05 mg veh^{-1} km^{-1} in the Shing Mun Tunnel (Ho et al., 2009) and more than double that of 1.70E + 05 mg veh^{-1} km^{-1} in the Loma Large Tunnel (Mancilla et al., 2012). The mole fraction ratios of $\Delta CO/\Delta CO_2$ (ppb ppm^{-1}) could be applied to differentiate the gasoline vehicles from diesel vehicles (Ammoura et al., 2014) as gasoline vehicles are known to emit much more CO than diesel vehicles. The average $\Delta CO/\Delta CO_2$ of 11.1 in this study was much higher than that of 5.68–8.44 in the Guy Môquet Tunnel in Paris, indicating higher percentage of LDVs in traffic fleets passing through the Zhujiang Tunnel than that through the Guy Môquet Tunnel.

3.4. Decadal variations of EFs for $PM_{2.5}$, OC and EC measured in the Zhujiang Tunnel

With the motor vehicle numbers increasing from 0.68 million in 2004 to 2.4 million in 2014 (Guangzhou Statistical Yearbook, 2015), measured EFs of $PM_{2.5}$, OC and EC for total fleet in the same Zhujiang Tunnel recorded a decrease of 23.4%, 18.3%, and 72.3%, respectively, in 2014 when compared to those in 2004 (He et al., 2008) (Fig. 4). This change is largely attributed to changing fleet compositions, and also to upgraded vehicle emission standards and fuel quality. In 2004 motor vehicle fleets passing through the tunnel were 80% gasoline vehicles and 20% diesel vehicles (He et al., 2008), while in 2014 during our campaign the compositions had changed to be 61% of gasoline vehicles, 12% diesel vehicles and 27% of LPG vehicles. A drastic change beneficial to lowering EFs of $PM_{2.5}$ from automobiles was that 80% buses and 100% taxies were replaced as LPG-driven ones as a measure to reduce particulate emission from public transportation sector.

Table 3
Emission factors of gaseous pollutants comparison with other recent studies.

References	Tunnel	Fuel type	Emission factors (mg veh^{-1} km^{-1})				
			NO_x	TNMHCs	SO_2	CO	CO_2
(Schmid et al., 2001)	Tauern Tunnel 2001	Total fleet	3131 ± 383	–	–	–	–
(Hwa et al., 2002)	Tapei Tunnel 2002	Total fleet	900 ± 180	440 ± 60	–	3640 ± 260	–
(Kristensson et al., 2004)	Söderleds Tunnel Winter 98/99	Total fleet	1360 ± 30	–	–	5300	–
(Cheng et al., 2006)	Shing Mun Tunnel, 2003/2004	Total fleet	878 ± 308	–	–	1845 ± 434	–
(Chiang et al., 2007)	Chung-Liao Tunnel	Total fleet	730 ± 150	460 ± 170	20 ± 10	–	–
(Legreid et al., 2007)	Gubrist Tunnel 2004	Total fleet	1050 ± 90	460 ± 40	–	–	–
(Ho et al., 2009)	Shing Mun Tunnel 2004	Total fleet	–	115 ± 26	–	1900 ± 380	3.10E+05
(Chang et al., 2009)	Hsuehshan Tunnel	LDV	145 ± 67	96 ± 65	3 ± 2	–	–
	Hsuehshan Tunnel	LDV	331 ± 166	121 ± 63	6 ± 3	–	–
(Mancilla et al., 2012)	Loma Larga Tunnel (LLT)	>97% gasoline	130 ± 100	1540 ± 80	–	11800	1.70E+05
(Ameur-Bouddabbous et al., 2012)	Grand Mare Tunnel Summer 2007	Total fleet	263 ± 115 (NO_2)	–	3.2 ± 2.1	–	–
	Grand Mare Tunnel Winter 2009	Total fleet	589 ± 289 (NO_2)	–	11.6 ± 8.4	–	–
(Araizaga et al., 2013)	Loma Larga Tunnel 2009	LDV	110 ± 70	1160 ± 50	–	–	–
This study	**Zhujiang Tunnel 2014**	**Total fleet**	**1286 ± 204**	**448 ± 38**	**21 ± 3**	**3096 ± 680**	**3.9E+05**

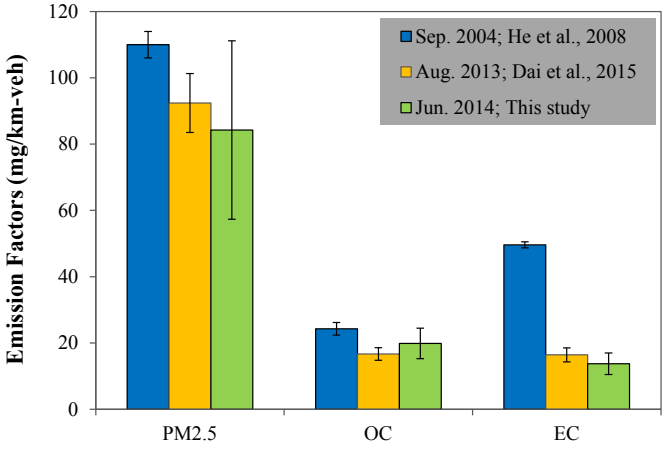

Fig. 4. Comparison of emission factors for $PM_{2.5}$, OC and EC based on tests in the Zhujiang tunnel in 2004, 2013 and 2014.

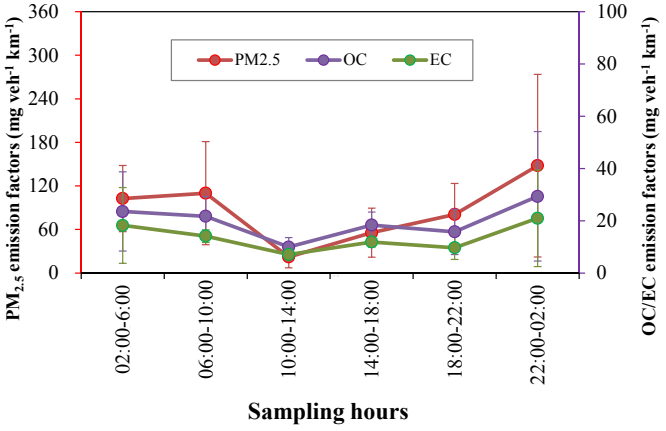

Fig. 5. Diurnal variations of emission factors of $PM_{2.5}$, OC, and EC.

Tightening motor vehicle emission standards is another important factor. For gasoline vehicles, emission standards were upgraded from CHINA-II (or Euro-2) in 2004 to CHINA-III (or Euro-3) in 2007, then to CHINA-IV (or Euro-4) in 2010, and recently further to CHINA-V (or Euro-5) in 2014. The PM emission limit was lowered from 0.10 to 0.20 g/km for direct injection compression ignition engine and 0.08–0.17 g/km for indirect injection compression ignition engine in CHINA-II (GB18352.2–2001) to 0.0045 g/km for all types of engines in CHINA-V (GB 18352.5-2013). Meanwhile, diesel emission standards were upgraded from CHINA-II in 2003 to CHINA-V in 2012. The PM emission limits were constrained from 0.15 g/(kW h) in CHINA-II (GB-17691-2001) to 0.02 g/(kW h) in CHINA-IV and CHINA-V (GB-17691-2005). Accompanying with the upgrade of emission standards, quality of gasoline and diesel oils was also upgraded. For example, sulfur contents was lowered from 1000 ppm in 1999 (GB 17930-1999) to 10 ppm in 2013 (GB 17930-2013) for gasoline. In 2004 vehicles were CHINA-I or CHINA-II ones. In 2014, however, as showed in Fig. 3, CHINA-III and CHINA-IV vehicles became dominated with shares of 39.4% and 36.2%, respectively (NBSC, 2014), and the less green CHINA-I and CHINA-II vehicles only accounted for 21.7% of the total motor vehicles. As tested in Beijing by Shen et al. (2014), EFs of $PM_{2.5}$ for LGVs could decline by 99% from Euro-0 to Euro-4. Therefore, traffic fleets with increasing portion of cleaner automobiles would result in lowered EFs of $PM_{2.5}$ and the carbonaceous aerosols as well.

It is worth noting that the design of sampling campaign in the tunnel would also influence EFs as factors like fleet composition and driving speed would change during a day (Grieshop et al., 2006; Mancilla and Mendoza, 2012; Shen et al., 2014). As showed in Fig. 5, EFs of $PM_{2.5}$, OC and EC showed similar diurnal variations; they were the highest at 22:00–02:00, which were 148 ± 126, 29 ± 24 and 21 ± 18 mg veh^{-1} km^{-1}, and the lowest at 10:00–14:00, which were 22 ± 15, 10 ± 4, and 7 ± 2 mg veh^{-1} km^{-1} for $PM_{2.5}$, OC and EC, respectively. The average EFs of $PM_{2.5}$, OC and EC at night time was observed to be 1.7, 1.4 and 1.5 times of that at daytime due largely to a much higher fraction of HDVs, which had much higher EFs for $PM_{2.5}$, OC and EC than LDVs or LPGVs (Ban-Weiss, 2008). This was also consistent with the very recent study by Shen et al. (2014), which demonstrated that the contribution of diesel vehicles to $PM_{2.5}$ became more important at night time than that at the daytime. Our sampling periods inside the tunnel not only covered full days but have more samples during each day when compared to previous campaigns, thus would get EFs more representative of road traffic fleets.

4. Conclusions

From tests of road vehicle emissions in the Zhujiang Tunnel in urban Guangzhou, we updated EFs of $PM_{2.5}$ and carbonaceous aerosols, and obtained EFs of trace gases including NO_x, CO, SO_2, CO_2 and total NMHCs. One distinction of this study is that we used an eddy covariance system with an integrated CO_2 open-path analyzer and a 3-D Sonic Anemometer for more accurate and precise CO_2 and wind measurements inside the tunnel. We found that EF values per kilometer per vehicle in 2014 for $PM_{2.5}$, OC and EC from this study were only −23.4%, −18.3%, and −72.3% lower when compared to that previously measured in 2004 in the same tunnel. As motor vehicle numbers increased more than doubled in the decade, total vehicle emission of $PM_{2.5}$ would be still increasing although vehicle emission standards have been upgraded to Euro-V. Our extensive measurements of diurnal variations of EFs for $PM_{2.5}$, OC and EC revealed higher values at night time due to higher fractions of DVs.

Acknowledgments

This study was funded by Strategic Priority Research Program of the Chinese Academy of Sciences (Grant No. XDB05010200), Natural Science Foundation of China (Project No. 41025012/41121063), and the Ministry of Science and Technology of China (2012IM030700).

References

Allen, J.O., Mayo, P.R., Hughes, L.S., Salmon, L.G., Cass, G.R., 2001. Emissions of size-segregated aerosols from on-road vehicles in the Caldecott Tunnel. Environ. Sci. Technol. 35 (21), 4189–4197.

Ameur-Bouddabbous, I., Kasperek, J., Barbier, A., Harel, F., Hannoyer, B., 2012. Transverse approach between real world concentrations of SO_2, NO_2, BTEX, aldehyde emissions and corrosion in the Grand Mare tunnel. J. Environ. Sci.-China 24 (7), 1240–1250.

Ammoura, L., Xueref-Remy, I., Gros, V., Baudic, A., Bonsang, B., Petit, J.E., Perrussel, O., Bonnaire, N., Sciare, J., Chevallier, E., 2014. Atmospheric measurements of ratios between CO_2 and co-emitted species from traffic: a tunnel study in the Paris megacity. Atmos. Chem. Phys. 14 (23), 12871–12882.

Araizaga, A.E., Mancilla, Y., Mendoza, A., 2013. Volatile organic compound emissions from light-duty vehicles in Monterrey, Mexico: a tunnel study. Int. J. Environ. Res. 7 (2), 277–292.

Artelt, S., Kock, H., König, H., Levsen, K., Rosner, G., 1999. Engine dynamometer experiments: platinum emissions from differently aged three-way catalytic converters. Atmos. Environ. 33, 3559–3567.

Ban-Weiss, G., et al., 2008. Long-term changes in emissions of nitrogen oxides and particulate matter from on-road gasoline and diesel vehicles. Atmos. Environ. 42 (2), 220–232.

Bishop, G.A., Stedman, D.H., 1996. Measuring the emissions of passing cars. Acc. Chem. Res. 29, 489–495.

Cao, J.J., Wang, Q.Y., Chow, J.C., Watson, J.G., Tie, X.X., Shen, Z.X., Wang, P., An, Z.S., 2012. Impacts of aerosol compositions on visibility impairment in Xi'an, China. Atmos. Environ. 59, 559–566.
Chan, C.K., Yao, X., 2008. Air pollution in mega cities in China. Atmos. Environ. 42 (1), 1–42.
Chang, S.C., Lin, T.H., Lee, C.T., 2009. On-road emission factors from light-duty vehicles measured in Hsuehshan Tunnel (12.9 km), the longest tunnel in Asia. Environ. Monit. Assess. 153 (1–4), 187–200.
Che, H., Zhang, X., Li, Y., Zhou, Z., Qu, J.J., 2007. Horizontal visibility trends in China 1981–2005. Geophys. Res. Lett. 34 (24).
Chen, C., Huang, C., Jing, Q., Wang, H., Pan, H., Li, L., Zhao, J., Dai, Y., Huang, H., Schipper, L., Streets, D.G., 2007. On-road emission characteristics of heavy-duty diesel vehicles in Shanghai. Atmos. Environ. 41 (26), 5334–5344.
Chen, F., Hu, W., Zhong, Q., 2013a. Emissions of particle-phase polycyclic aromatic hydrocarbons (PAHs) in the Fu Gui-shan Tunnel of Nanjing, China. Atmos. Res. 124, 53–60.
Chen, Z., Wang, J.-N., Ma, G.-X., Zhang, Y.-S., 2013b. China tackles the health effects of air pollution. Lancet 382 (9909), 1959–1960.
Cheng, Y., Lee, S.C., Ho, K.F., Louie, P.K.K., 2006. On-road particulate matter ($PM_{2.5}$) and gaseous emissions in the Shing Mun Tunnel, Hong Kong. Atmos. Environ. 40 (23), 4235–4245.
Cheng, H.R., Guo, H., Saunders, S.M., Lam, S.H.M., Jiang, F., Wang, X.M., Wang, T.J., 2010a. Assessing photochemical ozone formation in the Pearl River Delta using a photochemical trajectory model. Atmos. Environ. 44, 4199–4208. http://dx.doi.org/10.1016/j.atmosenv, 2010. 07.019.
Cheng, H., Guo, H., Wang, X., Saunders, S.M., Lam, S.H.M., Jiang, F., Wang, T., Ding, A., Lee, S., Ho, K.F., 2010b. On the relationship between ozone and its precursors in the Pearl River Delta: application of an observation-based model (OBM). Environ. Sci. Pollut. Res. 17, 547–560.
Cheng, Y., Lee, S.C., Ho, K.F., Chow, J.C., Watson, J.G., Louie, P.K.K., Cao, J.J., Hai, X., 2010c. Chemically speciated on-road $PM_{2.5}$ motor vehicle emission factors in Hong Kong. Sci. Total Environ. 408 (7), 1621–1627.
Chiang, H.-L., Huang, Y.-S., 2009. Particulate matter emissions from on-road vehicles in a freeway tunnel study. Atmos. Environ. 43 (26), 4014–4022.
Chiang, H.-L., Hwu, C.-S., Chen, S.-Y., Wu, M.-C., Ma, S.-Y., Huang, Y.-S., 2007. Emission factors and characteristics of criteria pollutants and volatile organic compounds (VOCs) in a freeway tunnel study. Sci. Total Environ. 381 (1–3), 200–211.
Dai, S., Bi, X., Chan, L.Y., He, J., Wang, B., Wang, X., Peng, P., Sheng, G., Fu, J., 2015. Atmos. Chem. Phys. 15 (6), 3097–3108.
Derwent, R.G., Jenkin, M.E., Saunder, S.M., 1996. Photochemical ozone creation potentials for a large number of reactive hydrocarbons under European conditions. Atmos. Environ. 30, 181–199.
Ding, X., Wang, X.M., Gao, B., Fu, X.X., He, Q.F., Zhao, X.Y., Yu, J.Z., Zheng, M., 2012. "Tracer-based estimation of secondary organic carbon in the Pearl River Delta, south China." J. Geophys. Res.-Atmos. 117.
Franco, V., Kousoulidou, M., Muntean, M., Ntziachristos, L., Hausberger, S., Dilara, P., 2013. Road vehicle emission factors development: a review. Atmos. Environ. 70, 84–97.
Gillies, J.A., Gertler, A.W., Sagebiel, J.C., Dippel, W.A., 2001. On-road particulate matter ($PM_{2.5}$ and PM_{10}) emissions in the Sepulveda Tunnel, Los Angeles, California. Environ. Sci. Technol. 35 (6), 1054–1063.
Grieshop, A.P., Lipsky, E.M., Pekney, N.J., Takahama, S., Robinson, A.L., 2006. Fine particle emission factors from vehicles in a highway tunnel: effects of fleet composition and season. Atmos. Environ. 40, S287–S298.
Guangzhou Statistical Yearbook, 2015. China Statistics Press, Beijing, p. 2014.
Guo, H., Zhang, Q.Y., Shi, Y., Wang, D.H., 2007. Evaluation of the International Vehicle Emission (IVE) model with on-road remote sensing measurements. J. Environ. Sci.-China 19 (7), 818–826.
Guo, S., Hu, M., Zamora, M.L., Peng, J., Shang, D., Zheng, J., Du, Z., Wu, Z., Shao, M., Zeng, L., Molina, M.J., Zhang, R., 2014. Elucidating severe urban haze formation in China. Proc. Natl. Acad. Sci. U. S. A. 111 (49), 17373–17378.
Handler, M., Puls, C., Zbiral, J., Marr, I., Puxbaum, H., Limbeck, A., 2008. Size and composition of particulate emissions from motor vehicles in the Kaisermuhlen-Tunnel, Vienna. Atmos. Environ. 42 (9), 2173–2186.
Hao, J.M., He, D.Q., Wu, Y., Fu, L., He, K.B., 2000. A study of the emission and concentration distribution of vehicular pollutants in the urban area of Beijing. Atmos. Environ. 34 (3), 453–465.
He, L.Y., Hu, M., Huang, X.F., Zhang, Y.H., Yu, B.D., Liu, D.Q., 2006. Chemical characterization of fine particles from on-road vehicles in the Wutong tunnel in Shenzhen, China. Chemosphere 62 (10), 1565–1573.
He, L.Y., Hu, M., Zhang, Y.-H., Huang, X.-F., Yao, T.-T., 2008. Fine particle emissions from on-road vehicles in the Zhujiang Tunnel, China. Environ. Sci. Technol. 42 (12), 4461–4466.
Ho, K.F., Lee, S.C., Tsai, W.Y., 2006. Carbonyl compounds in the roadside environment of Hong Kong. J. Hazard. Mater. 133 (1–3), 24–29.
Ho, K.F., Lee, S.C., Ho, W.K., Blake, D.R., Cheng, Y., Li, Y.S., Ho, S.S.H., Fung, K., Louie, P.K.K., Park, D., 2009. Vehicular emission of volatile organic compounds (VOCs) from a tunnel study in Hong Kong. Atmos. Chem. Phys. 9 (19), 7491–7504.
Hu, J.N., Wu, Y., Wang, Z.S., Li, Z.H., Zhou, Y., Wang, H.T., Bao, X.F., Hao, J.M., 2012. Real-world fuel efficiency and exhaust emissions of light-duty diesel vehicles and their correlation with road conditions. J. Environ. Sci.-China 24 (5), 865–874.
Huang, X.F., Yu, J.Z., He, L.Y., Hu, M., 2006. Size distribution characteristics of elemental carbon emitted from Chinese vehicles: results of a tunnel study and atmospheric implications. Environ. Sci. Technol. 40 (17), 5355–5360.
Huang, W., Cao, J., Tao, Y., Dai, L., Lu, S.-E., Hou, B., Wang, Z., Zhu, T., 2012. Seasonal variation of chemical species associated with short-term mortality effects of $PM_{2.5}$ in Xi'an, a central city in China. Am. J. Epidemiol. 175 (6), 556–566.
Huang, R.-J., Zhang, Y., Bozzetti, C., Ho, K.-F., Cao, J.-J., Han, Y., Daellenbach, K.R., Slowik, J.G., Platt, S.M., Canonaco, F., Zotter, P., Wolf, R., Pieber, S.M., Bruns, E.A., Crippa, M., Ciarelli, G., Piazzalunga, A., Schwikowski, M., Abbaszade, G., Schnelle-Kreis, J., Zimmermann, R., An, Z., Szidat, S., Baltensperger, U., El Haddad, I., Prevot, A.S.H., 2014. High secondary aerosol contribution to particulate pollution during haze events in China. Nature 514 (7521), 218–222.
Hueglin, C., Buchmann, B., Weber, R., 2006. Long-term observation of real-world road traffic emission factors on a motorway in Switzerland. Atmos. Environ. 40, 3696–3709.
Huo, H., Zhang, Q., He, K., Yao, Z., Wang, X., Zheng, B., Streets, D.G., Wang, Q., Ding, Y., 2011. Modeling vehicle emissions in different types of Chinese cities: importance of vehicle fleet and local features. Environ. Pollut. 159 (10), 2954–2960.
Hwa, M.Y., Hsieh, C.C., Wu, T.C., Chang, L.F.W., 2002. Real-world vehicle emissions and VOCs profile in the Taipei tunnel located at Taiwan Taipei area. Atmos. Environ. 36 (12), 1993–2002.
Imhof, D., Weingartner, E., Prevot, A.S.H., Ordonez, C., Kurtenbach, R., Wiesen, P., Rodler, J., Sturm, P., McCrae, I., Ekstrom, M., Baltensperger, U., 2006. Aerosol and NO_x emission factors and submicron particle number size distributions in two road tunnels with different traffic regimes. Atmos. Chem. Phys. 6, 2215–2230.
Jamriska, M., Morawska, L., Thomas, S., He, C., 2004. Diesel bus emissions measured in a tunnel study. Environ. Sci. Technol. 38, 6701–6709.
Kirchstetter, T.W., Harley, R.A., Kreisberg, N.M., Stolzenburg, M.R., Hering, S.V., 1999. On-road measurement of fine particle and nitrogen oxide emissions from light- and heavy-duty motor vehicles. Atmos. Environ. 33 (18), 2955–2968.
Kleindienst, T.E., Corse, E.W., Li, W., McIver, C.D., Conver, T.S., Edney, E.O., Driscoll, D.J., Speer, R.E., Weathers, W.S., Tejada, S.B., 2002. Secondary organic aerosol formation from the irradiation of simulated automobile exhaust. J. Air & Waste Manag. Assoc. 52 (3), 259–272.
Kristensson, A., Johansson, C., Westerholm, R., Swietlicki, E., Gidhagen, L., Wideqvist, U., Vesely, V., 2004. Real-world traffic emission factors of gases and particles measured in a road tunnel in Stockholm, Sweden. Atmos. Environ. 38 (5), 657–673.
Krupa, S., McGrath, M.T., Andersen, C.P., Booker, F.L., Burkey, K.O., Chappelka, A.H., Chevone, B.I., Pell, E.J., Zilinskas, B.A., 2001. Ambient ozone and plant health. Plant Dis. 85, 4–12.
Laschober, C., Limbeck, A., Rendl, J., Puxbaum, H., 2004. Particulate emissions from on-road vehicles in the Kaisermuhlen-tunnel (Vienna, Austria). Atmos. Environ. 38 (14), 2187–2195.
Legreid, G., Reimann, S., Steinbacher, M., Staehelin, J., Young, D., Stemmler, K., 2007. Measurements of OVOCs and NMHCs in a Swiss highway tunnel for estimation of road transport emissions. Environ. Sci. Technol. 41 (20), 7060–7066.
Li, T., Chen, X., Yan, Z., 2013. Comparison of fine particles emissions of light-duty gasoline vehicles from chassis dynamometer tests and on-road measurements. Atmos. Environ. 68, 82–91.
Liu, H.A., He, K.B., He, D.Q., Fu, L.X., Zhou, Y., Walsh, M.P., Blumberg, K.O., 2008a. Analysis of the impacts of fuel sulfur on vehicle emissions in China. Fuel 87 (13–14), 3147–3154.
Liu, Y., Shao, M., Lu, S., Chang, C.C., Wang, J.L., Fu, L.L., 2008b. Source apportionment of ambient volatile organic compounds in the Pearl River Delta, China: part II. Atmos. Environ. 42, 6261–6274.
Liu, H., He, K., Lents, J.M., Wang, Q., Tolvett, S., 2009. Characteristics of diesel truck emission in China based on portable emissions measurement systems. Environ. Sci. Technol. 43 (24), 9507–9511.
Liu, T., Wang, X., Wang, B., Ding, X., Deng, W., Lu, S., Zhang, Y., 2014. Emission factor of ammonia (NH_3) from on-road vehicles in China: tunnel tests in urban Guangzhou. Environ. Res. Lett. 9 (6).
Lough, G.C., Schauer, J.J., Lonneman, W.A., Allen, M.K., 2005. Summer and winter nonmethane hydrocarbon emissions from on-road motor vehicles in the Midwestern United States. J. Air & Waste Manag. Assoc. 55 (5), 629–646.
Mancilla, Y., Mendoza, A., 2012. A tunnel study to characterize $PM_{2.5}$ emissions from gasoline-powered vehicles in Monterrey, Mexico. Atmos. Environ. 59, 449–460.
Mancilla, Y., Araizaga, A.E., Mendoza, A., 2012. A tunnel study to estimate emission factors from mobile sources in Monterrey, Mexico. J. Air & Waste Manag. Assoc. 62 (12), 1431–1442.
National Bureau of Statistics of China (NBSC), 2014. China Statistical Yearbook. China Statistics Press, Beijing.
Ng, N.L., Kroll, J.H., Chan, A.W.H., Chhabra, P.S., Flagan, R.C., Seinfeld, J.H., 2007. Secondary organic aerosol formation from m-xylene, toluene, and benzene. Atmos. Chem. Phys. 7, 3909–3922.
Nine, R.D., Clark, N., Daley, J.J., Atkinson, C., 1999. Development of a heavy-duty chassis dynamometer driving route. Proc. Inst. Mech. Eng. Part D J. Automob. Eng. 213, 561–574.
Pierson, W.R., Brachaczek, W.W., 1983. EMISSIONS of ammonia and amines from vehicles on the road. Environ. Sci. Technol. 17 (12), 757–760.
Pierson, W.R., Gertler, A.W., Robinson, N.F., Sagebiel, J.C., Zielinska, B., Bishop, G.A., Stedman, D.H., Zweidinger, R.B., Ray, W.D., 1996. Real-world automotive emissions – summary of studies in the Fort McHenry and Tuscarora Mountain Tunnels. Atmos. Environ. 30 (12), 2233–2256.
Schauer, J.J., Kleeman, M.J., Cass, G.R., Simoneit, B.R.T., 1999. Measurement of

emissions from air pollution sources. 2. C-1 through C-30 organic compounds from medium duty diesel trucks. Environ. Sci. Technol. 33 (10), 1578–1587.
Schmid, H., Pucher, E., Ellinger, R., Biebl, P., Puxbaum, H., 2001. Decadal reductions of traffic emissions on a transit route in Austria – results of the Tauerntunnel experiment 1997. Atmos. Environ. 35 (21), 3585–3593.
Shang, Y., Sun, Z.W., Cao, J.J., Wang, X.M., Zhong, L.J., Bi, X.H., Li, H., Liu, W.X., Zhu, T., Huang, W., 2013. Systematic review of Chinese studies of short-term exposure to air pollution and daily mortality. Environ. Int. 54, 100–111.
Shen, X., Yao, Z., Huo, H., He, K., Zhang, Y., Liu, H., Ye, Y., 2014. $PM_{2.5}$ emissions from light-duty gasoline vehicles in Beijing, China. Sci. Total Environ. 487, 521–527.
Shorter, J.H., Herndon, S., Zahniser, M.S., Nelson, D.D., Wormhoudt, J., Demerjian, K.L., Kolb, C.E., 2005. Real-time measurements of nitrogen oxide emissions from in-use New York City transit buses using a chase vehicle. Environ. Sci. Technol. 39, 7991–8000.
Song, Y., Tang, X., Xie, S., Zhang, Y., Wei, Y., Zhang, M., Zeng, L., Lu, S., 2007. Source apportionment of $PM_{2.5}$ in Beijing in 2004. J. Hazard. Mater. 146 (1–2), 124–130.
Tie, X., Wu, D., Brasseur, G., 2009. Lung cancer mortality and exposure to atmospheric aerosol particles in Guangzhou, China. Atmos. Environ. 43 (14), 2375–2377.
Wang, B., Zhang, Y., Zhu, C., Yu, K., Chan, L., Chan, Z., 2001. A study on city motor vehicle emission factors by tunnel test. Huan Jing Ke Xue 22 (2), 55–59 (in Chinese).
Wang, X.K., Manning, W.J., Feng, Z.W., Zhu, Y.G., 2007. Ground-level ozone in China: distribution and effects on crop yields. Environ. Pollut. 147, 394–400.
Wang, X., Westerdahl, D., Wu, Y., Pan, X., Zhang, K.M., 2011. On-road emission factor distributions of individual diesel vehicles in and around Beijing, China. Atmos. Environ. 45 (2), 503–513.
Wang, X., Westerdahl, D., Hu, J.N., Wu, Y., Yin, H., Pan, X.C., Zhang, K.M., 2012a. On-road diesel vehicle emission factors for nitrogen oxides and black carbon in two Chinese cities. Atmos. Environ. 46, 45–55.
Wang, X.M., Ding, X., Fu, X., He, Q., Wang, S., Bernard, F., Zhao, X., Wu, D., 2012b. Aerosol scattering coefficients and major chemical compositions of fine particles observed at a rural site hit the central Pearl River Delta, South China. J. Environ. Sci.-China 24 (1), 72–77.
Wang, S.Y., Wu, D.W., Wang, X.M., Fung, J.C.H., Yu, J.Z., 2013. Relative contributions of secondary organic aerosol formation from toluene, xylenes, isoprene, and monoterpenes in Hong Kong and Guangzhou in the Pearl River Delta, China: an emission-based box modeling study. J. Geophys. Res.-Atmos. 118 (2), 507–519.
Wang, Z., Wu, Y., Zhou, Y., Li, Z., Wang, Y., Zhang, S., Hao, J., 2014. Real-world emissions of gasoline passenger cars in Macao and their correlation with driving conditions. Int. J. Environ. Sci. Technol. 11 (4), 1135–1146.
Wu, D., Bi, X., Deng, X., Li, F., Tan, H., Liao, G., Huang, J., 2007. Effect of atmospheric haze on the deterioration of visibility over the Pearl River Delta. Acta Meteorol. Sin. 21 (2), 215–223.
Xu, H., Wang, X., Poesch, U., Feng, S., Wu, D., Yang, L., Li, S., Song, W., Sheng, G., Fu, J., 2008. Genotoxicity of total and fractionated extractable organic matter in fine air particulate matter from urban Guangzhou: comparison between haze and nonhaze episodes. Environ. Toxicol. Chem. 27 (1), 206–212.
Yao, Z.L., Shen, X., Ye, Y., Cao, X., Jiang, X., Zhang, Y., He, K., 2015. On-road emission characteristics of VOCs from diesel trucks in Beijing, China. Atmos. Environ. 103, 87–93.
Zhang, Q., He, K., Huo, H., 2012. Cleaning China's air. Nature 484 (7393), 161–162.
Zhang, R., Jing, J., Tao, J., Hsu, S.C., Wang, G., Cao, J., Lee, C.S.L., Zhu, L., Chen, Z., Zhao, Y., Shen, Z., 2013a. Chemical characterization and source apportionment of $PM_{2.5}$ in Beijing: seasonal perspective. Atmos. Chem. Phys. 13 (14), 7053–7074.
Zhang, Y.L., Guo, H., Wang, X.M., Simpson, I.J., Barletta, B., Blake, D.R., Meinardi, S., Rowland, F.S., Cheng, H.R., Saunders, S.M., Lam, S.H.M., 2010. Emission patterns and spatiotemporal variations of halocarbons in the Pearl River Delta region, southern China. J. Geophys. Res.-Atmos. 115.
Zhang, Y., Wang, X., Blake, D.R., Li, L., Zhang, Z., Wang, S., Guo, H., Lee, F.S.C., Gao, B., Chan, L., Wu, D., Rowland, F.S., 2012b. Aromatic hydrocarbons as ozone precursors before and after outbreak of the 2008 financial crisis in the Pearl River Delta region, south China. J. Geophys. Res.-Atmos. 117.
Zhang, Y., Wang, X., Barletta, B., Simpson, I.J., Blake, D.R., Fu, X., Zhang, Z., He, Q., Liu, T., Zhao, X., Ding, X., 2013b. Source attributions of hazardous aromatic hydrocarbons in urban, suburban and rural areas in the Pearl River Delta (PRD) region. J. Hazard. Mater. 250, 403–411.
Zheng, J., Zhang, L., Che, W., Zheng, Z., Yin, S., 2009. A highly resolved temporal and spatial air pollutant emission inventory for the Pearl River Delta region, China and its uncertainty assessment. Atmos. Environ. 43 (32), 5112–5122.