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Heavy-duty diesel vehicles dominate vehicle emissions in a tunnel study in northern China

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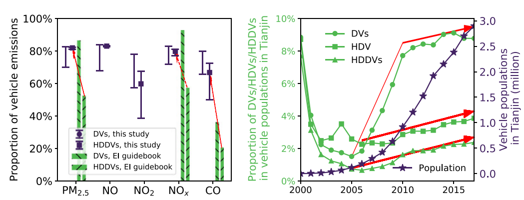
HIGHLIGHTS

- Real-world emission factors for heavy-duty-diesel vehicles (HDDVs) were measured.
- Emission inventory for HDDVs and non-HDDVs were established.
- HDDVs and diesel vehicles are major sources of vehicle emissions.
- The contribution of HDDVs to fleet emissions was underestimated.

GRAPHICAL ABSTRACT



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ABSTRACT

The relative importance of contributions of gasoline vehicles (GVs) and diesel vehicles (DVs), heavy-duty diesel vehicles (HDDVs) and non-HDDVs to on-road vehicle emissions remains unclear. Vehicle emission factors (EFs), including fine particulate matter ($PM_{2.5}$), NO-NO_2-NO_x, and carbon monoxide (CO), were measured (August 4-18, 2017) in an urban tunnel in Tianjin, northern China. The average EFs (mg km^{-1} veh^{-1}) of the fleet were as follows: 9.21 (95% confidence interval: 1.60, 23.07) for $PM_{2.5}$, 62.08 (21.21, 138.25) for NO, 20.42 (0.79, 45.48) for NO_2, 83.72 (26.29, 162.87) for NO_x, and 284.54 (18.22, 564.67) for CO. The fleet-average EFs exhibited diurnal variations, due to diurnal variations in the proportion of HDDVs in the fleet, though the hourly proportion of HDDVs never exceeded 10% during the study period. The reconstructed average EFs for on-road vehicle emissions of $PM_{2.5}$, NO, NO_2, and NO_x, and CO were approximately 2.2, 1.7, 1.5, 2.0, and 1.6 times as much as those in the tunnel, respectively, due to the higher HDDV fractions in the whole city than those in the tunnel. The EFs of $PM_{2.5}$, NO, NO_2, and NO_x, and CO from each HDDV were approximately 75, 81, 24, 65, and 33 times of those from each non-HDDV, respectively. HDDVs were responsible for approximately 81.92%, 83.02%, 59.79%, 79.79%, and 66.77% of the total $PM_{2.5}$, NO, NO_2, and NO_x, and CO emissions from on-road vehicles in Tianjin, respectively. DVs, especially HDDVs, are major sources of on-road $PM_{2.5}$, NO-NO_2-NO_x, and CO emissions in northern China. The contribution of HDDVs to fleet emissions calculated by the EFs from Chinese 'on-road vehicle emission inventory guidebook' were underestimated, as compared to our results. The EFs from on-road vehicles should be updated due to the rapid progression of vehicle technology combined with emission standards in China. The management and control of HDDV emissions have become urgent to reduction of on-road vehicle emissions.

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1. Introduction

Owing to rapid industrialization and urbanization, the severe and persistent air pollution in China has become an immense burden on healthcare and the economy (Song et al., 2017a,b). National air quality standards and emission control strategies are having a positive effect on air quality in China (He et al., 2017a,b). Local emission controls have great contribution to urban air quality (He et al., 2017c, 2016; Jing et al., 2016). Air pollution is becoming increasingly attributable to the growing and outdated vehicle fleet (mainly diesel vehicles) due to the refinement of industrial emission management, although total vehicle emissions have peaked and are now decreasing despite the increasing vehicle fleet size (Jiang et al., 2017; Wu et al., 2017). A growing body of evidence has emerged that specifically links exposure to traffic-related air pollution with increased health risks of the population (Sinharay et al., 2017; Cepeda et al., 2017; Chen et al., 2017; Hoek et al., 2002). On-road vehicles emit pollutants that damage health, including particulate matter (PM), specifically fine PM smaller than 2.5 microns (μm) in diameter ($PM_{2.5}$) that constitutes 90% of the PM emitted by on-road vehicles in China; and smog precursors, such as nitrogen oxides (NO_x=NO + NO_2), carbon monoxide (CO), and volatile organic compounds (VOCs), that contribute to the secondary formation of PM and ozone (O_3) (Jiang et al., 2017; Wu et al., 2016, 2017; Guo et al., 2014). However, it is difficult to evaluate the contribution of vehicle emissions to air pollution due to a lack of real-world vehicle emission data in China, despite vehicle emissions being important sources of health-damaging air pollutants. Accurately qualifying real-world vehicle emissions is essential to further our understanding of their impacts on urban air quality and public health (Li et al., 2017; Zhang et al., 2017b; Jing et al., 2016; He et al., 2016).

The estimation of real-world vehicle emissions requires a large amount of data, such as emission factors (EFs), traffic activity, fleet composition, and road information. Average EFs could be determined through four approaches, including emission models (such as Motor Vehicle Emission Simulator: MOVES, a European Road Transport Emission Inventory Model: COPERT, the International Vehicle Emissions model: IVE, and Mobile Source Emissions Factor: MOBILE), a chassis dynamometer, tunnel testing, and on-board testing. With the implementation of strict vehicle exhaust emission control strategies and the promotion of alternative-fuel vehicles worldwide, exhaust emission will markedly decline over the next few decades. However, emissions from non-exhaust are not well regulated. The contributions of non-exhaust emissions (including road abrasion, tire/brake wear, and road dust re-suspension) have become increasingly important in fleet emissions (Zhang et al., 2018a; Harrison et al., 2012; Thorpe and Harrison, 2008). Tunnel test might be the most idealized measure to estimate primary vehicle emissions including both exhaust and non-exhaust emissions without oxidation degradation by ultraviolet (UV) light. In addition, tunnel testing is a common method of obtaining average PM, NO_x, CO, and VOCs vehicle EFs for a typical urban fleet under actual road conditions (Zhang et al., 2018b; Lawrence et al., 2016; Cui et al., 2016; Dallmann et al., 2013; Hung-Lung et al., 2007; Jamriska et al., 2004; Hwa et al., 2002). Previous studies in China have focused on the average EFs of light-duty passenger vehicles (LDPVs, gasoline dominance) (Huang et al., 2017; Zhang et al., 2015; Shen et al., 2014) as they constitute the largest proportion of the Chinese vehicle fleet, accounting for 86.0% of the total fleet in 2016, based on 'China Vehicle Environmental Management Annual Report (2017)' (CVEMAR). However, on-road vehicles span a wide range of sizes, from light-duty vehicles (LDV), which are mostly fueled by gasoline in China, to heavy-duty vehicles (HDV), which are often diesel-powered. According to CVEMAR, diesel vehicles (DVs) constituted 10.2% of vehicle populations in China in 2016, but were responsible for over 99%, 68.7%, 12.6%, and 24.0% of all on-road PM, NO_x, CO, and hydrocarbon (HC) emissions, respectively. In Beijing, DVs contribute to over 80% to 90% and 60% of on-road PM and NO_x emissions, respectively (Shen et al., 2015; Wu et al., 2011). In addition, DVs were the key sources of organic carbon and organic aerosol based on the results of source apportionment in Beijing (Tang et al., 2018; Guo et al., 2012, 2013). Gertler (2005) reported that gasoline vehicles (GVs) are the primary sources of on-road PM emissions in the United States, based on EFs from tunnel tests. Dallmann et al. (2013) found that DVs accounted for $<$ 1% of all vehicles observed in the tunnel in the United States, but were responsible for (18 ± 3)%, (22 ± 6)%, and (45 ± 8)% of NO_x, organic aerosol, and black carbon emissions. The contribution of emissions from DVs, especially heavy-duty diesel vehicles (HDDVs), is generally underestimated as heavy-duty trucks (HDTs) are often not allowed access to the center city, where tunnel tests are often conducted.

There are discrepancies in the relative importance of contributions of GVs and DVs, HDDVs and non-HDDVs to ambient air pollutant concentrations (Dallmann et al., 2013). In 2014, the Ministry of Environmental Protection (MEP) of the People's Republic of China issued an 'on-road vehicle emission inventory guidebook' (EI guidebook), which recommended a series of vehicle EFs based on local studies. However, real-world emissions for on-road HDDVs and light-duty diesel vehicles are often underestimated and exceed certification limits (Anenberg et al., 2017; Lelieveld and Pöschl, 2017; Huang et al., 2017). The insufficient and dated data on real-world EFs from on-road vehicles lead to the uncertainty in current assessments of the relative contribution of on-road sources to the overall burden of air pollution.

China is experiencing a rapid improvement in vehicle emission control technologies, including the prioritization of public transportation policy, improvement of emission standards and fuel quality, in-use vehicle inspection and maintenance (I/M) programs, and odd-even license plate rules, all of which have improved air quality (Wu et al., 2011). However, few studies in the Beijing-Tianjin-Hebei (BTH) region have investigated real-world EFs for the on-road vehicle fleet, especially for HDDVs. The overall objective of this study was to determine local $PM_{2.5}$, NO-NO_2-NO_x, and CO EFs for the fleet, GVs and DVs, LDV and HDV, HDDVs and non-HDDVs. Moreover, we attempted to determine the relative contributions of DVs and HDDVs to overall on-road vehicle emissions under typical urban fleet structure. The results from this study enriched the database on the fleet-average emission factors of on-road vehicles for emission inventory, air quality modeling, and health effects studies, and provided new insights into the management and control of HDDV emissions in northern China.

2. Materials and methods

2.1. Tunnel description

The 2-week measurement campaign was conducted in the Wujinglu (WJL) tunnel in the central urban area of Tianjin (117°12′15″, 39°8′31″). The WJL tunnel has been in operation since 2010 and is an urban tunnel with north and south bores (three vehicle lanes and one walkway each), which are important parts of the Tianjin Railway Station Transport Hub as they effectively alleviate traffic pressure. Sampling stations were located at both the inlet and outlet of the north bore. The length of the tunnel is approximately 1.6 km, and there is a 4% downslope (upslope) approaching (exiting) the tunnel. The cross-sectional area of the tunnel is approximately 54 m^2. The vehicle speed limit in the tunnel is 40 km h^{-1}, and the average daily traffic volume is approximately 15,000 vehicles. There is no fresh air supply throughout the bores, therefore, the dilution of air pollutants was eliminated. The longitudinal jetting ventilation fans along the ceiling throughout the tunnel were inactive during the sampling periods. Ventilation was thus only induced by the flow of traffic through the tunnel and prevailing winds. As the traffic light control is located at least 250 m from the entrance of the tunnel, vehicle emissions from a cold start were negligible.

2.2. Vehicle classification and fleet composition

Traffic counts and vehicle speed were continuously monitored using roadside laser loop detectors (AxleLight RLU11) that were installed at both inlet and outlet of the WJL tunnel. The vehicles could be grouped into 14 categories based on wheelbases and axle-number.

A high-definition vehicle license plate recognition system was installed at a pedestrian overpass approximately 115 m from the exit of the tunnel. Additionally, video footage was also recorded for data validation and review. The license plates of vehicles passing through the WJL tunnel during the measurement campaign were matched with the registered vehicle database (up to August 2017) of Tianjin to obtain the in-use fuel type, emission standards, and detailed information about the vehicles.

Vehicle classification results were obtained from the car license plates in this study, instead of those from the loop detectors. The fleet was classified into GVs, DVs, and alternative-fuel vehicles based on the in-use fuel types. HDVs were divided into three types: heavy-duty buses (HDBUs), heavy-duty passenger vehicles (HDPVs), and heavy-duty trucks (HDTs). The other vehicles were classified into taxis (TAs), buses (BUs), mini passenger vehicles (MPVs), light-duty passenger vehicles (LDPVs), medium-duty passenger vehicles (MDPVs), mini-trucks (MT), light-duty trucks (LDTs), and medium-duty trucks (MDTs). The fleet comprised China 0 to 5 vehicle emission standards.

2.3. Field measurements and instruments

Measurements were continuously recorded at both the inlet (45 m from the tunnel entrance) and outlet (605 m from the tunnel entrance) sites on the same side of the north bore (effective length of 560 m) during August 4–18, 2017 (2 weeks).

Meteorological data, including temperature (T), relative humidity (RH), wind speed (WS), wind direction (WD), and atmospheric pressure (P), were measured by VAISALA WXT520 (Helsinki, Finland) automatic weather stations with a resolution of 1 min. Volumetric flow rates induced by the vehicle fleet and prevailing winds in the tunnel were continuously measured by an ultrasonic gas flowmeter (Flowsick-200 SICK MAIHAK, Germany) with a resolution of 1 min, which has been used in previous tunnel tests (Zhang et al., 2018b; Imhof et al., 2006).

The $PM_{2.5}$ mass concentrations at both the inlet and outlet of the tunnel were measured by Pegasor PPS-M (Pegasor Oy, Tampere, Finland) sensors (Amanatidis et al., 2016; Rostedt et al., 2014; Lanki et al., 2011), which use the escaping charge principle, with a resolution of 1 s. The sensor has been used in measuring diesel exhausts (Amanatidis et al., 2017), and urban aerosols both in clean (Järvinen et al., 2015) and highly polluted (Maso et al., 2016) areas.

The NO-NO_2-NO_x, O_3, and CO were measured by micro-monitoring station (Environnement S.A, France), which integrated AC32 M module for NO-NO_2-NO_x analyzer, O342 module for O_3 analyzer, and CO12 module for CO analyzer. The AC32 M measurement module was used to continuously measure NO-NO_2-NO_x, which operates on the principle that NO will emit light (chemiluminescence) in the presence of highly oxidizing O_3 molecules. The O342 measurement module (specifically recording low concentrations) uses the principle of O_3 detection by absorption in ultraviolet light. The continuous CO12 measurement module uses the principle of detection by absorption in infrared light. The minimum detectable limits for the NO-NO_2-NO_x, O_3, and CO analyzers are 0.4 ppb, 0.4 ppb, and 0.05 ppm, respectively. We regularly maintained, calibrated, and cleaned the instruments to ensure that the measurements were reliable. Datasets for NO-NO_2-NO_x, O_3 and CO were available as 1 min values, which were then aggregated to 1 h mean values. Additionally, ambient concentrations of air pollutants ($PM_{2.5}$, NO-NO_2-NO_x, O_3 and CO) during the measurement periods were collected from national air quality monitoring sites (NAQMS) (Song et al., 2017b).

2.4. Emission factor calculation

The fleet-average EFs for vehicles traveling through the tunnel were estimated using the following formula, which has been widely used in previous studies (Fang et al., 2018; Zhang et al., 2018b; Pierson et al., 1996; Pierson and Brachaczek, 1983).

$$EF_{fleet} = \frac{(C_{Outlet} - C_{Inlet}) \times A \times v \times T}{N \times L} \quad (1)$$

where EF (mg km^{-1} veh^{-1}) is the average emission factor. C_{Outlet} and C_{Inlet} (mg m^{-3}) are the mass concentrations of air pollutants at the outlet and inlet of the tunnel, respectively. A (m^2) is the cross-sectional tunnel area in m^2 (54 m^2). v (m s^{-1}) is the air velocity parallel to the tunnel, recorded by the ultrasonic gas flowmeters. N (veh) is the traffic count traveling through the tunnel during the time interval (1 h, $T = 3600$ s). L (km) is the distance (0.56 km) between sampling sites. To exclude O_3 titration reactions when estimating the primary EFs of NO and NO_2 (Yao et al., 2005), the $C_{Outlet} - C_{Inlet}$ were $([NO]_{Outlet} - [NO]_{Inlet}) + ([O_3]_{Inlet} - [O_3]_{Outlet})$ for NO, and $([NO_2]_{Outlet} - [NO_2]_{Inlet}) - ([O_3]_{Inlet} - [O_3]_{Outlet})$ for NO_2.

To differentiate the relative contributions from HDDVs (DVs, HDV) and non-HDDVs (GVs, LDV), the following equation can be used to derive HDDVs (DVs, HDV) and non-HDDVs (GVs, LDV) emission factors according to the regression model (He et al., 2008; Colberg et al., 2005; Gertler et al., 1996; Pierson et al., 1996):

$$EF_i = \alpha + \beta \times pCV_i + \epsilon_i \quad (2)$$

where α is the EF of non-HDDVs (GVs, LDV), $\alpha + \beta$ is the EF of HDDVs (DVs, HDV), pCV$_i$ is the proportion of HDDVs (DVs, HDV) in the fleet, and ϵ_i is the random error.

The total emissions from on-road vehicles in the current year was calculated using the following formula (Sun et al., 2016):

$$EM_i = \sum_{j=1}^{N} EF_{i,j} \times VKT_j \times P_j \quad (3)$$

where EM_i (mg) is the total annual emission of air pollutant i from on-road vehicles in Tianjin, $EF_{i,j}$ (mg km^{-1} veh^{-1}) is the EF of air pollutant i from vehicle category j, VKT_j is the annual vehicle kilometers traveled (VKT) by category j recommended in the 'EI guidebook' by the MEP, and P_j is the population of vehicle category j in the current year.

3. Results and discussion

3.1. Traffic characteristics

In the WJL tunnel, 14,866± 900 (mean ± stand deviation) vehicles traveled through per day during the measurement campaign.

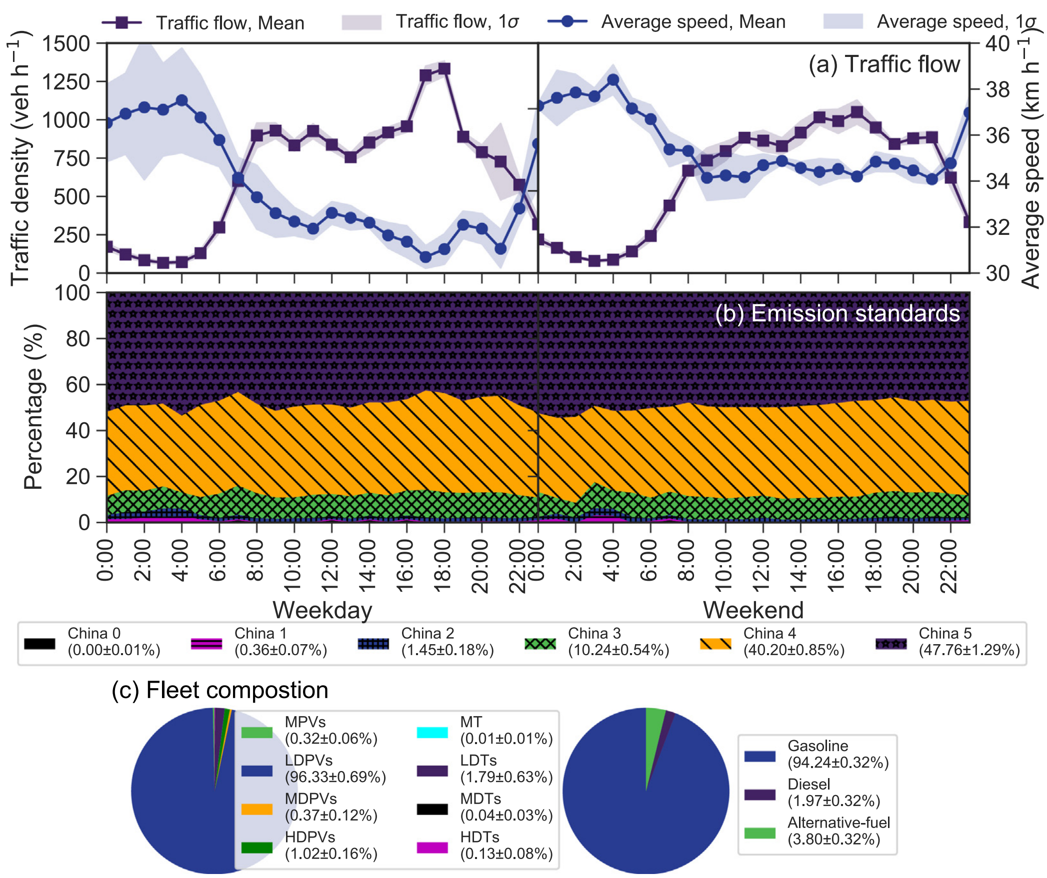

Fig. 1. Traffic characteristics (a: traffic flow including density and speed, b: emission standards, c: fleet composition) in the WJL tunnel during the measurement campaign.

As there is a 40-km h^{-1} speed limit in the tunnel, the average traffic speeds during the study period were 35.6 ± 1.7 km h^{-1} at the entrance and 38.7 ± 2.1 km h^{-1} at the exit of the tunnel. Fig. 1 shows the average traffic flow and emission standards for each hour during weekdays and weekends. The average vehicle flow and speed were 643 ± 385 veh h^{-1} and 38.8 ± 2.1 km h^{-1} on a weekday, and 573 ± 368 veh h^{-1} and 38.6 ± 2.1 km h^{-1} on a weekend. As shown in Fig. 1a, the traffic was heavier during the evening rush hour than it was during the morning rush hour, particularly on a weekday. The diurnal variations of traffic flow (including traffic density and speed) were related to local passenger travel behaviors. Vehicle flows during the peak hour reached 1333 ± 50 veh h^{-1} and 980 ± 73 veh h^{-1} on a weekday (18:00 LT) and weekend (17:00 LT), respectively. The diurnal variations in traffic speed were opposite to those of traffic flow, with higher average speeds at midnight (42.1 ± 1.7 km h^{-1}) and lower average speeds during rush hours (36.3 ± 0.5 km h^{-1}).

In the WJL tunnel, China 4 and 5 vehicles were most common, constituting $40.20 \pm 0.85\%$ and $47.76 \pm 1.29\%$ of the fleet, respectively (Fig. 1b), which was because the China 5 emission standard was implemented in Tianjin in 2016. As shown in Fig. 1b, diurnal variations of emission standards were steady, which indicates that it is difficult to differentiate between EFs from China 0 to 5 emission standards. The WJL tunnel was located in the center of Tianjin, where access for HDDVs ($0.35 \pm 0.35\%$) and MDDVs ($0.08 \pm 0.07\%$) was restricted. Therefore, the fleet in the tunnel primarily comprisedLDPVs (Fig. 1c), accounting for $96.33 \pm 0.69\%$ of the total vehicles. GVs were the most common in the fleet ($94.24 \pm 0.32\%$). With the implementation of strict vehicle emission control policies and the promotion of alternative-fuel (compressed natural gas, liquefied natural gas, liquefied petroleum gas, electronic, and hybrid) vehicles, alternative-fuel vehicles were emerging which accounted for nearly $3.80 \pm 0.32\%$ of the total vehicles, and were almost twice as common as DVs ($1.97 \pm 0.32\%$). This phenomenon further indicated the significance of non-exhaust in fleet emissions in the future.

The fleet structure changed in 2017 with the gradual elimination of older vehicles and the addition of new ones (Liu et al., 2017; Shen et al., 2015). In 2017, China 0, China 1, China 2, China 3, China 4 and China 5 accounted for 0.00%, 0.11%, 6.45%, 56.51%, 32.61%, and 4.32% of DVs; 0.01%, 0.66%, 4.10%, 19.01%, 48.47%, and 27.75% of GVs; 0.00%, 0.02%, 1.81%, 59.20%, 30.65%, and 8.32% of HDDVs; and 0.01%, 0.63%, 4.27%, 20.95%, 47.14%, and 27.00% of non-HDDVs in 2017, respectively. The emission standards for DVs and HDDVs (China 3 and China 4 dominance) are lagged by approximately one stage behind those for GVs and non-HDDVs (China 4 and China 5 dominance) due to differences in the implementation timetables of vehicle emission control in China (Wu et al., 2017).

Table 1 shows the contributions of vehicle categories to the total vehicle population (2.8 million) and mileage (59.6×10^9 km) in Tianjin in 2017. HDV, DVs (mostly LDTs and HDTs), and HDDVs accounted for 2.03%, 7.41%, and 1.75% of the vehicle population, and were responsible for 6.48%, 13.55%, and 5.69% of the total VKT in Tianjin in 2017, respectively. HDV, DVs (mostly LDTs and HDTs), and HDDVs only accounted for 1.20%, 2.10%, and 1.05% of the vehicle fleet traveled through the WJL tunnel, respectively, which were lower than those for the whole of Tianjin. The lower proportion of HDDVs

Table 1
Contributions of vehicle categories to the total vehicle population (2.8 million) and total mileage (59.6×10^9 km) of Tianjin in 2017.

Vehicle categories		Total vehicles (%)			Total mileage (%)		
		Gasoline	Diesel	Alternative-fuel	Gasoline	Diesel	Alternative-fuel
HDV	BUs	0.00	0.51	0.21	0.01	1.43	0.59
	HDPVs	0.04	0.14	0.01	0.11	0.38	0.04
	HDTs	0.00	1.10	0.01	0.01	3.88	0.03
Others	TAs	1.12	0.00	0.01	6.30	0.00	0.03
	BUs	0.00	0.03	0.00	0.01	0.08	0.00
	MPVs	0.59	0.00	0.06	0.50	0.00	0.05
	LDPVs	84.64	0.41	1.87	71.46	0.34	1.58
	MDPVs	0.25	0.18	0.00	0.36	0.27	0.00
	MT	0.01	0.00	0.07	0.02	0.00	0.09
	LDTs	3.54	4.71	0.19	4.98	6.62	0.27
	MDTs	0.00	0.33	0.00	0.00	0.54	0.00

(DVs, HDVs) in the tunnel fleet might be attributed to the restriction of HDTs in the central urban, which could further contribute to the underestimation of the fleet-average emissions in the whole city.

3.2. Meteorology and concentrations

The micro meteorological conditions in the WJL tunnel and ambient air parameters during the measurement campaign are shown in Fig. S1. Uncertainty in the average EFs estimated from the tunnel tests is also affected by the measurement of the effective ventilation flux in the tunnel. During the measurement campaign, wind speeds (WS) at the inlet and outlet of the tunnel, as well as those in ambient air, were measured continuously. Additionally, a Flowsic (ultrasonic gas flowmeters) device and a VAISALA WXT520 automatic weather station were installed to measure the WS at the outlet of the tunnel. The average wind speeds (m s^{-1}) were 1.49 ± 0.55 in ambient air, and 1.55 ± 0.47 and 0.83 ± 0.27 at the inlet and outlet of the tunnel, respectively. The WS measured at the entrance of the tunnel is mainly influenced by prevailing wind in the ambient air (Fig. S1a), with a Pearson correlation coefficient (r) of 0.23 ($p < 0.001$). Thus, the WS measured at the outlet of the tunnel is more representative of the actual volumetric flow rates. The WS measured by the VAISALA at the outlet of the tunnel was strongly correlated (Pearson's $r = 0.79$, $p < 0.001$) with those measured by the Flowsic (ultrasonic gas flowmeters) device. In our tunnel test, the WS measured by the Flowsic was utilized to represent the actual ventilation flux of the tunnel and to estimate the EFs. The average temperatures (°C) in the ambient air and at the inlet and outlet of the tunnel were 27.51 ± 2.71, 29.26 ± 2.98, and 29.66 ± 1.64, respectively. The average relative humidity (%) in the ambient air, at the inlet and outlet of the tunnel was 71.60 ± 9.62, 62.63 ± 12.36, and 61.79 ± 8.85, respectively. The average P (hpa) in the ambient air and at the inlet and outlet of the tunnel was 1002.50 ± 3.78, 1003.92 ± 3.63, and 1005.05 ± 3.80, respectively. The temperature and pressure at the outlet of the tunnel were slightly higher than those at the inlet of the tunnel and in the ambient air, which could due to the limited space in the tunnel and the ventilation flux pressure induced by the vehicle fleet. The micro meteorological conditions in the WJL tunnel were generally associated with those in the ambient air, and the WS measured at the outlet of the tunnel could be regarded as the actual ventilation flux of the tunnel.

Fig. 2 shows the $PM_{2.5}$, NO-NO_2-NO_x, O_3, and CO concentrations in the ambient air and at the inlet and outlet of the tunnel. The average $PM_{2.5}$ (NO, NO_2, NO_x, O_3 and CO) concentrations at the inlet and outlet of WJL tunnel were approximately 1.52 ± 0.67 (6.15 ± 7.60, 5.01 ± 2.56, 5.49 ± 2.87, 0.36 ± 0.20, and 2.31 ± 0.66), and 1.81 ± 0.89 (39.81 ± 30.81, 8.02 ± 5.12, 12.00 ± 7.63, 0.02 ± 0.04, and 2.62 ± 0.77) times of those in the ambient air, respectively. The concentrations of air pollutants (especially NO) at the entrance (inlet) and exit (outlet) of the tunnel were much higher than those measured at NAQMS, excluding O_3. The highest NO_2/NO_x was observed in the ambient air (0.86 ± 0.08), followed by that at the inlet (0.79 ± 0.12) and outlet (0.58 ± 0.15) of the tunnel. Our results suggested that the air pollution levels at roadside and tunnel micro-environments were far higher than those measured in the ambient air, indicating high traffic-related health risks because of the high population density living near major traffic roads. The traditional assessment of the health burden associated with exposure to air pollution might be underestimated due to the low spatial resolution of exposure assignment (Song et al., 2017a), and should be carefully adjusted to account for the high traffic-related exposure risks. The average oxidations ($NO_2 + O_3$) were the highest at the outlet (91.54 ± 27.93 ppb) of the tunnel, followed by that at the inlet (76.33 ± 16.91 ppb) of the tunnel, and that in the ambient air (64.49 ± 23.59 ppb). The time series of $PM_{2.5}$, NO-NO_2-NO_x, and CO at both the inlet and outlet of the tunnel show similar trends to those in ambient air, suggesting that the corresponding inlet concentrations should be subtracted to isolate the emission signals from vehicles traveling through the tunnel.

3.3. Emission factors

The diurnal variations of average EFs for $PM_{2.5}$, NO-NO_2-NO_x, O_3 and CO from the fleet in the WJL tunnel were calculated based on Eq. (1), as shown in Fig. 3. In summary, the average EFs (mg km^{-1} veh^{-1}) of the fleet were 9.21 (95% confidence interval: 1.60, 23.07) for $PM_{2.5}$, 62.08 (21.21, 138.25) for NO, 20.42 (0.79, 45.48) for NO_2, 83.72 (26.29, 162.87) for NO_x, and 284.54 (18.22, 564.67) for CO. The uncertainty of the calculated EFs was represented by 95% confidence interval (95% CI). The average EFs were compared with those presented in previous Chinese tunnel studies. Huang et al. (2017) reported that the EFs (mg km^{-1} veh^{-1}) of $PM_{2.5}$, NO_x, and CO tested in a tunnel (94.1% gasoline) in Shanghai in 2016 were 34.0 ± 23.5, 400 ± 250, and 1840 ± 900, respectively. EFs (mg km^{-1} veh^{-1}) of $PM_{2.5}$, NO_x, and CO were (2 ± 2)-(4 ± 3), (150 ± 70)-(330 ± 170), and (910 ± 470)-(1470 ± 630), respectively, were estimated through tunnel tests (light-duty dominance) conducted in Taiwan in 2006 (Chang et al., 2008). Zhang et al. (2015) conducted tunnel studies

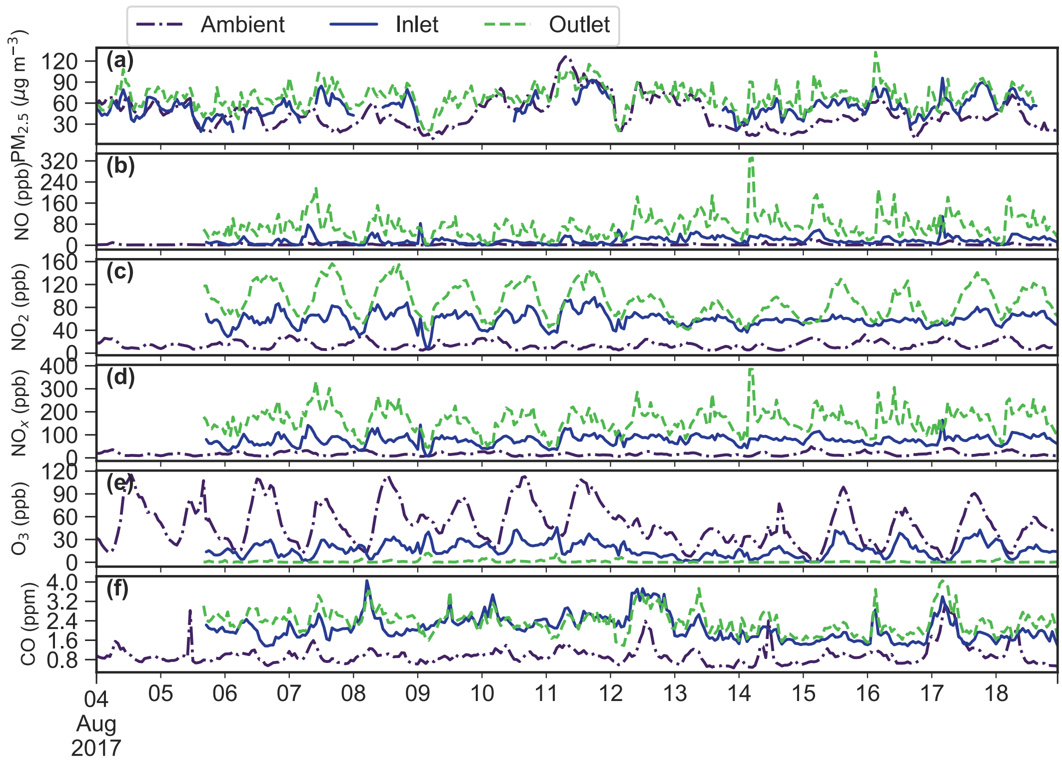

Fig. 2. $PM_{2.5}$, NO-NO_2-NO_x, O_3, and CO concentrations in the ambient air (represented by dashdot lines) and at the inlet (represented by solid lines) and outlet (represented by dashed lines) of the WJL tunnel.

(12% heavy-duty vehicles) in Guangzhou in 2014, and measured EFs (mg km^{-1} veh^{-1}) of $PM_{2.5}$, NO_x, and CO of 82.7 ± 28.3, 1290± 200, and 3100± 680, respectively. Our results were lower than those presented in previous studies, which was mainly due to the low proportion of HDDVs (0.35 ± 0.35%) in the fleet, the downslope gradient of -4%, and the strict enforcement of vehicle emission control strategies (China 4: 40.20 ± 0.85%, China 5: 47.76 ± 1.29%). According to the 'EI guidebook', the EFs (mg km^{-1} veh^{-1}) of $PM_{2.5}$, NO_x, and CO, should be 3, 17, and 460 for LDPVs with an emission standard of China 5; and 3, 32, and 680 for LDPVs with an emission standard of China 4, respectively. Our results were comparable to those published by the MEP.

As shown in Fig. 3, the average EFs of $PM_{2.5}$, NO, NO_2, NO_x, and CO showed similar diurnal variation, with the highest values before dawn (00: 00–05:00 LT). As tested by Zhang et al. (2015) in Guangzhou in 2014, the EF of $PM_{2.5}$ also varied diurnally, with the highest values at 22: 00–02:00. The average EFs of $PM_{2.5}$, NO, NO_2, NO_x, and CO before dawn (00: 00–05:00 LT) were 4.7, 2.8, 1.8, 2.1, and 2.5 times of those during the other hours (06: 00–23:00 LT) of the day, respectively, suggesting that the average EFs of the nighttime fleet were higher than those of the daytime fleet, and which could be due to the increased proportions of HDV and DVs in the fleet at night (Fig. 4a–b). This finding was also consistent with Shen et al. (2014)'s study in Beijing, which demonstrated that DVs contributed more to emissions at night than that during the daytime. In the WJL tunnel, the fractions of HDV and DVs in the fleet before dawn (00: 00–05:00 LT) were 1.5 and 1.9 times of those during the other hours (06: 00–23:00 LT) of the day, respectively. The proportion of HDV was strongly correlated (R^2=0.7) with that of DVs because DVs accounted for approximately 87.7% of the total HDV traversing the WJL tunnel.

The obviously diurnal variations of the proportions of HDV and DVs in the fleet allowed us to conduct linear regressions, as shown in Eq. (2). According to the regression results (Fig. 5), positive correlations between the average emission factors and the proportion of HDDVs were observed for both particular matter ($PM_{2.5}$) and gaseous pollutants (NO-NO_2-NO_x and CO), which further demonstrated the significant contribution of HDDVs to the before-dawn fleet in Tianjin. The average EFs of $PM_{2.5}$, NO, NO_2, NO_x, and CO from different vehicle categories (DVs and GVs; HDV and LDV; HDDVs and Non-HDDVs) are listed in Table 2. The average EFs (mg km^{-1} veh^{-1}) of HDDVs were 285.23 (190.94, 379.51) for $PM_{2.5}$, 1993.05 (1168.29, 2817.81) for NO, 308.19 (105.93, 510.44) for NO_2, 2312.52 (1304.71, 3320.33) for NO_x, and 4991.59 (1883.03, 8100.15) for CO. The average EFs (mg km^{-1} veh^{-1}) of non-HDDVs were 3.80 (2.69, 4.91) for $PM_{2.5}$, 24.61 (14.91, 34.31) for NO, 12.51 (10.13, 14.89) for NO_2, 35.35 (23.50, 47.20) for NO_x, and 149.97 (113.41, 186.53) for CO. According to our tunnel tests, the EFs of $PM_{2.5}$, NO, NO_2, NO_x, and CO from each HDDVs were approximately 75, 81, 24, 65, and 33 times of those from non-HDDVs, respectively. Additionally, the EFs of $PM_{2.5}$, NO, NO_2, NO_x, and CO for DVs were approximately 23, 28, 17, 26, and 21 times those for GVs. Our results are comparable with those from Dallmann et al. (2013)'s study, which reported that the EFs of organic aerosol and black carbon from heavy-duty diesel trucks were 10 and 50 times higher than those of LDV in the United States in 2010, respectively. However, due to the insignificant variation (Fig. 1b) of the compositions of the vehicle emission standards (from China 0 to 5) during the measurement campaign, the attempt to differentiate EFs under different emission standards by multiple linear regression (Zhang et al., 2018b; Hwa et al., 2002) failed. Although the EFs estimated by the tunnel tests were considered as

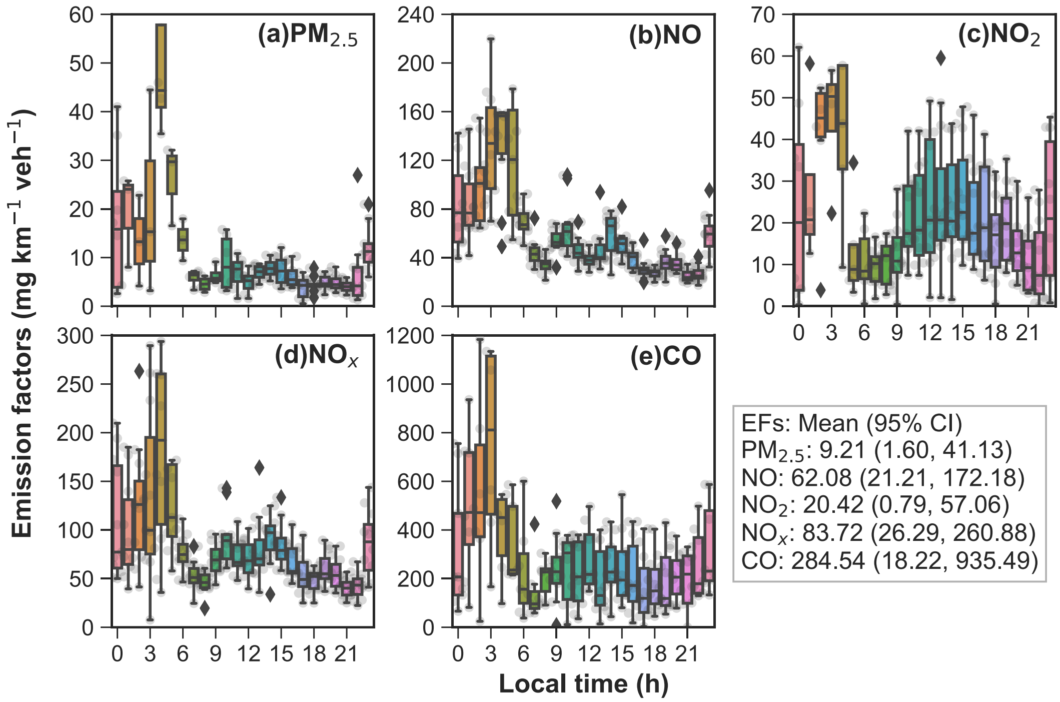

Fig. 3. Diurnal variations of average emission factors for $PM_{2.5}$, NO-NO_2-NO_x, and CO from the fleet in the WJL tunnel.

EFs in a real-world setting, there were still several limitations, such as the limited speeds and accelerations, and that most of the vehicles were operating under hot-stabilized conditions (Gertler, 2005).

HDV, DVs (mostly LDTs and HDTs), and HDDVs only accounted for 1.20%, 2.10%, and 1.05% of the fleet traveled through the WJL tunnel, respectively, which were lower than those for the whole of Tianjin (Table 1). Thus, the average EFs in the fleet in the WJL tunnel might underestimate those from on-road vehicles in Tianjin due to the strict tunnel access (located in the city center of Tianjin) for HDDVs and MDDVs. We used the fractions and EFs of diesel vehicles (HDV and HDDVs) to reconstruct the average EFs for on-road vehicles in Tianjin (as shown in Table 2), which gave values of (mg km^{-1} veh^{-1}) 20.19 ± 3.48 for $PM_{2.5}$, 108.01 ± 9.31 for NO, 30.25 ± 9.50 for NO_2, 164.79 ± 37.33 for NO_x, and 452.77 ± 143.22 for CO. The reconstructed average EFs of $PM_{2.5}$, NO, NO_2, NO_x, and CO for on-road vehicles in Tianjin were approximately 2.2, 1.7, 1.5, 2.0, and 1.6 times those in the WJL tunnel. Our results demonstrated that average EFs from tunnel tests should be carefully adjusted due to differences in fleet compositions between tested tunnels and the studied city.

According to the composition of vehicle populations in Tianjin and detailed EFs recommended in the 'EI guidebook', we calculated the weighted average EFs (as shown in Table 2) for DVs and GVs, HDV and LDV, and HDDVs and non-HDDVs, and the total fleet, under current compositions of emission standards. The EFs were adjusted by the fractions recommended in the 'EI guidebook' according to the speeds (30–40 km h^{-1}) in the tunnel. The reconstructed average EFs of $PM_{2.5}$ in the tunnel were approximately 1.7 times as much as

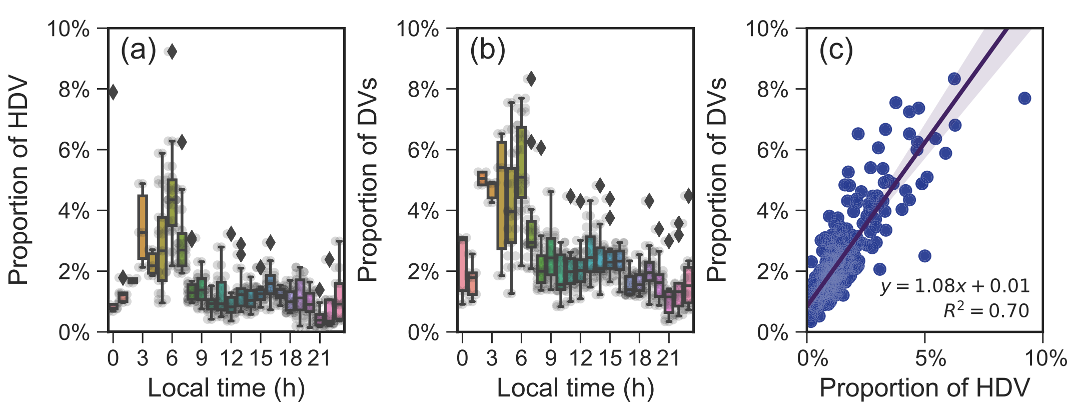

Fig. 4. Proportions of (a) HDV (heavy-duty vehicles) and (b) DVs (diesel vehicles) in the fleet, and (c) their relationships.

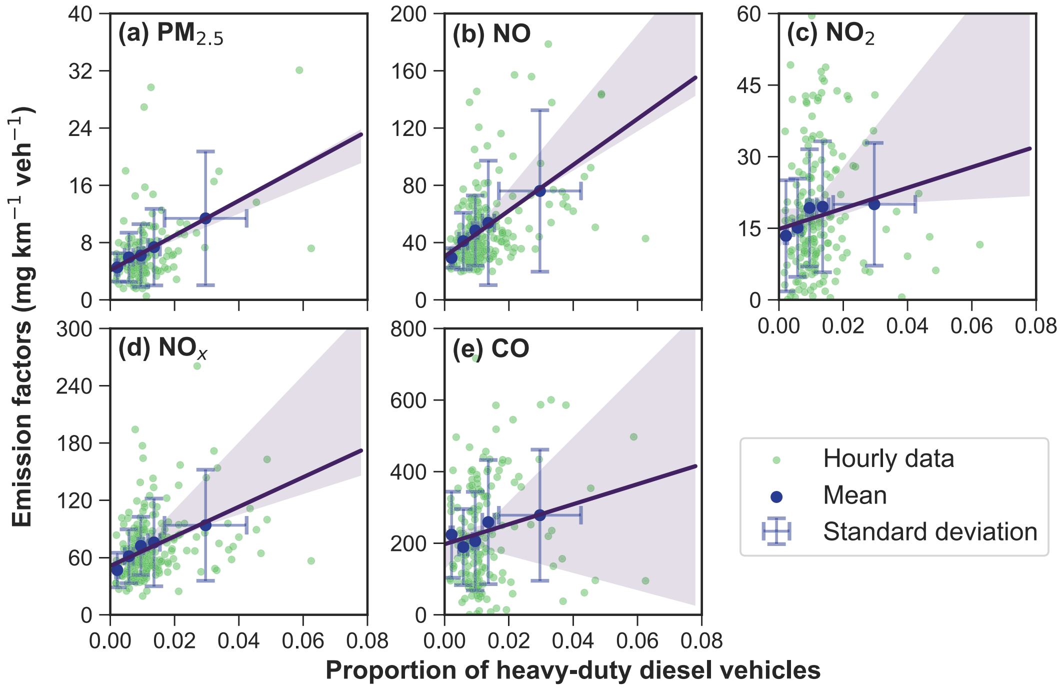

Fig. 5. Linear regressions between the average emission factors (a: $PM_{2.5}$, b: NO, c: NO_2, d: NO_x, and e: CO) and proportion of heavy-duty diesel vehicles (HDDVs). The fit lines were filled with 95% confidence intervals of regressions.

those estimated by the 'EI guidebook', which might be attributable to the high non-exhaust emissions. Generally, the vehicle EFs in the 'EI guidebook' were measured by portable emission measurement system (PEMS), which only measured the tailpipe emissions (Zhang et al., 2016), and ignored the non-exhaust emissions. The reconstructed average EFs of NO_x, and CO for on-road vehicles in Tianjin were approximately 0.4, 0.5 times as much as those estimated by the 'EI guidebook', which might be associated with the downslope gradient of −4% of the tested tunnel. The lower EFs for NO_x and CO were often observed in GVs tunnels, such as 145–331 mg

Table 2
Average emission factors (mg km^{-1} veh^{-1}) from tunnel tests in Tianjin, China.

	$PM_{2.5}$	NO	NO_2	NO_x	CO
Average EFs in the WJL tunnel					
Total fleet	9.21	62.08	20.42	83.72	284.54
DVs	137.72	979.18	216.05	1201.24	3419.96
GVs	5.99	35.19	12.59	45.37	166.74
HDV	194.00	1313.81	86.37	1153.31	2070.00
LDV	4.64	32.73	16.72	56.28	204.19
HDDVs	285.23	1993.05	308.19	2312.52	4991.59
Non-HDDVs	3.80	24.61	12.51	35.35	149.97
Average EFs in Tianjin					
Total fleet	20.19	108.01	30.25	164.79	452.77
DVs	137.72	979.18	216.05	1201.24	3419.96
GVs	5.99	35.19	12.59	45.37	166.74
HDV	194.00	1313.81	86.37	1153.31	2070.00
LDV	4.64	32.73	16.72	56.28	204.19
HDDVs	285.23	1993.05	308.19	2312.52	4991.59
Non-HDDVs	3.80	24.61	12.51	35.35	149.97
EFs estimated by 'EI guidebook'					
Total fleet	11.61	-	-	369.66	849.60
DVs	111.70	-	-	4073.42	1714.81
GVs	3.49	-	-	68.92	779.34
HDV	177.86	-	-	6891.31	3000.19
LDV	8.25	-	-	238.10	806.24
HDDVs	194.15	-	-	7240.30	2773.22
Non-HDDVs	8.31	-	-	245.49	814.83

km^{-1} veh^{-1} in the Hsuehshan Tunnel (Chang et al., 2008) and 110–130 mg km^{-1} veh^{-1} in the Loma Larga Tunnel (Araizaga et al., 2013; Mancilla and Mendoza, 2012).

3.4. On-road vehicle emissions

The reconstructed real-world EFs for on-road vehicle categories that were estimated through the tunnel tests (Table 1), annual VKT data recommended in the ‘ EI guidebook’ by the MEP, and the detailed vehicle populations from registered vehicle database (up to August 2017) were used to calculate on-road vehicle emissions in Tianjin in 2017 using Eq. (3). In 2017, Tianjin's on-road vehicles emitted approximately 1203± 207 tons of $PM_{2.5}$, 8249± 1414 tons of NO, 1801± 566 tons of NO_2, 9815± 2223 tons of NO_x, and 26,966 ± 8530 tons of CO. Using EFs recommended in the ‘EI guidebook’ by the MEP, the contributions of vehicle categories to the total vehicle emissions of $PM_{2.5}$ (1.25 × 10^3 tons), NO_x (4.20 × 10^4 tons), and CO (4.33 × 10^4 tons) estimated from the ‘EI guidebook’ in Tianjin in 2017 were shown in Table 3. Liu et al. (2008) reported that the on-road vehicle emissions of PM, NO_x, and CO in Tianjin in 2008 were 2.33 × 10^3 tons, 5.99 × 10^4 tons, 22.03 × 10^4 tons, respectively, by the IVE model. Zhang et al. (2017a) reported that the total vehicle emissions in Tianjin in 2013 were approximately 2.90 × 10^3 tons of $PM_{2.5}$, 3.20 × 10^3 tons of PM_{10}, 6.74 × 10^4 tons of NO_x, and 22.78 × 10^4 tons of CO, based on EFs from the ‘EI guidebook’. The total emissions estimated in our study were lower than those in previous studies due to the lower EFs estimated in the tunnel tests. We found that 78.28 (70.12, 82.74)%, 81.35 (67.71, 83.87)%, 72.90 (57.25, 77.89)%, 80.59 (71.75, 83.08)%, and 76.28 (65.76, 79.94)% of the total $PM_{2.5}$, NO, NO_2, NO_x, and CO emissions from on-road vehicles in Tianjin could be attributed to DVs, respectively (Fig. 6). Additionally, HDDVs were responsible for 81.92 (81.08, 82.35)%, 83.02 (82.55, 93.21)%, 59.79 (38.69, 67.42)%, 79.79 (77.02, 93.21)%, and 66.77 (50.05, 72.38)% of total $PM_{2.5}$, NO, NO_2, NO_x, and CO emissions from on-road vehicles in Tianjin, respectively (Fig. 6). DVs, especially HDDVs, are major sources of on-road $PM_{2.5}$, NO-NO_2-NO_x, and CO emissions in northern China, which is consistent with Shen et al. (2015)'s study in China and in contrast to Gertler (2005)'s study in the United States.

According to the contributions of vehicle categories to the total vehicle emissions estimated from the ‘EI guidebook’ (Table 3), DVs accounted for approximately 86.31%, 92.65%, and 36.26% of total $PM_{2.5}$, NO_x, and CO emissions in Tianjin, respectively (Fig. 6). Additionally, HDDVs accounted for approximately 52.52%, 57.33%, 20.89% of total $PM_{2.5}$, NO_x, and CO emissions in Tianjin, respectively (Fig. 6). The relative contributions of on-road HDDV emissions in the fleet calculated by the EFs from the ‘EI guidebook’ were underestimated, as compared to our results from the reconstructed real-world EFs inferred from tunnel tests, indicating that the EFs from on-road vehicles should be updated due to the rapid progression of vehicle technology combined with emission standards in China.

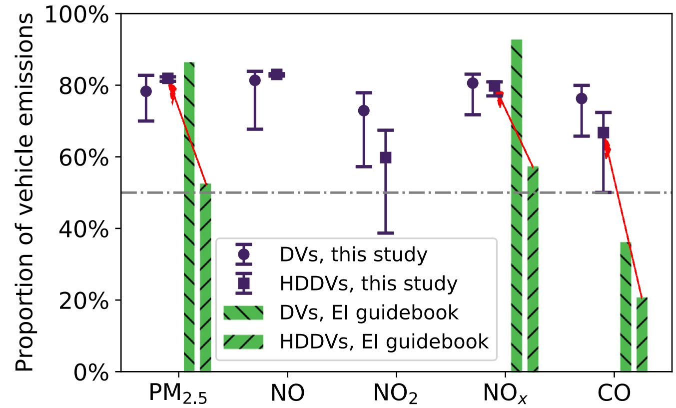

Fig. 6. Proportion of DV (diesel vehicle) and HDDV (heavy-duty diesel vehicle) emissions in the total on-road vehicle emissions from this study (represented by error bars with 95% confidence intervals) and ‘EI guidebook’ (represented by bars).

3.5. Implication for vehicle emission controls

For $PM_{2.5}$, NO-NO_2-NO_x, and CO emissions, HDDVs are the major contributors among all vehicle categories. The high emissions of real-world DVs, especially HDDVs, could be attributed to the following aspects. First, the rapid development of freight transportation during the last decade has led to a rise in VKT and in the HDDVs (DVs) fleet sizes. The VKT of HDTs was estimated to be approximately 3.4 to 4.2 times as much as that of LDPVs in the BTH region (Lang et al., 2012, 2014, 2016). In addition, the proportion of HDDVs (DVs, HDV) in vehicle populations of Tianjin increased from 0.7% (1.5%, 2.4%) in 2005 to 2.4% (8.8%, 3.9%) in 2017 (Fig. 7a), at a rate of 0.16% yr^{-1} (R^2=0.94, p<0.001) during the last decade. The running time and driving distance of HDDVs are much higher than LDPVs, which could be mainly attributable to the increasingly rigid demands of freight transportation. The highway transportation is the dominated freight transportation mode (Fig. 7b), accounted for 56.42±8.10% in Tianjin, 75.82±4.65% in the BTH region, and 74.74±1.67% in China. The share of highway transportation in freight

Table 3
The contributions of vehicle categories to the total vehicle emissions of $PM_{2.5}$ (1.25 × 10^3 tons), NO_x (4.20 × 10^4 tons), and CO (4.33 × 10^4 tons) estimated from ‘EI guidebook’ in Tianjin in 2017.

Vehicle categories		$PM_{2.5}$ (%)			NO_x (%)			CO (%)		
		Gasoline	Diesel	Alternative-fuel	Gasoline	Diesel	Alternative-fuel	Gasoline	Diesel	Alternative-fuel
HDV	HDBUs	0.68	17.15	0.01	0.37	17.80	0.07	1.77	7.56	0.04
	HDPVs	0.01	5.69	0.26	0.01	4.79	1.49	0.04	2.43	1.05
	HDTs	0.04	29.68	0.00	0.02	34.74	0.00	0.12	10.90	0.00
Others	LDPVs	0.09	0.43	11.62	0.02	0.34	3.98	0.39	0.06	46.76
	LDTs	0.00	28.50	0.00	0.00	29.44	0.00	0.00	13.24	0.00
	MDBUs	0.00	0.20	0.00	0.00	0.16	0.01	0.00	0.09	0.01
	MDPVs	0.00	1.35	0.14	0.00	1.14	1.09	0.00	0.61	1.31
	MDTs	0.00	3.31	0.00	0.00	4.23	0.00	0.00	1.37	0.00
	MPVs	0.00	0.00	0.11	0.00	0.00	0.04	0.00	0.00	0.39
	MT	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	TAs	0.00	0.00	0.73	0.00	0.00	0.26	0.08	0.00	11.78

transportation will maintain its continued growth trends unless the freight transportation structures are well adjusted and optimized. The promotion of railway transportation might be the solution to the increasingly rigid demands of freight transportation, and the reduction of HDDV emissions. Unfortunately, the proportions of freight transportation by railways are decreasing dramatically in Tianjin, the BTH region, and China (Fig. 7b). For Tianjin, one of the most port throughput in China and the world, the adjustment and optimization of freight transportation structures become particularly important.

Second, the fraudulent fuels and vehicle technologies of HDDVs are commonly observed according to results of sampling inspection. China has implemented the China 5 emission standards (from July 2017) for HDDVs, however, their real-world emissions were still significantly exceeding the regulatory limits. Chinese government should strength intensity of supervision and inspection, guarantee the supply of low sulfur diesel fuels, and strengthen after-treatment devices (e.g., selective catalytic reduction systems, diesel particulate filter, diesel oxidation catalyst) and On-Board Diagnostic (OBD), otherwise, the benefits of the implementation of more stricter emission standards and phasing out older vehicles could be offset. The real-world remote sensing measurement might be an effective way for screening the HDDV emissions that exceed the regulatory limits in China. With the emerging of new technology and methods, the I/M programs for in-use HDDVs should be further explored and reconsidered.

In summary, the adjustment and optimization of freight transportation structures, the strict inspections over production conformity and in-use compliance, the intelligent transport system, and the optimization of traffic network might be the solution of achieving low vehicle emissions.

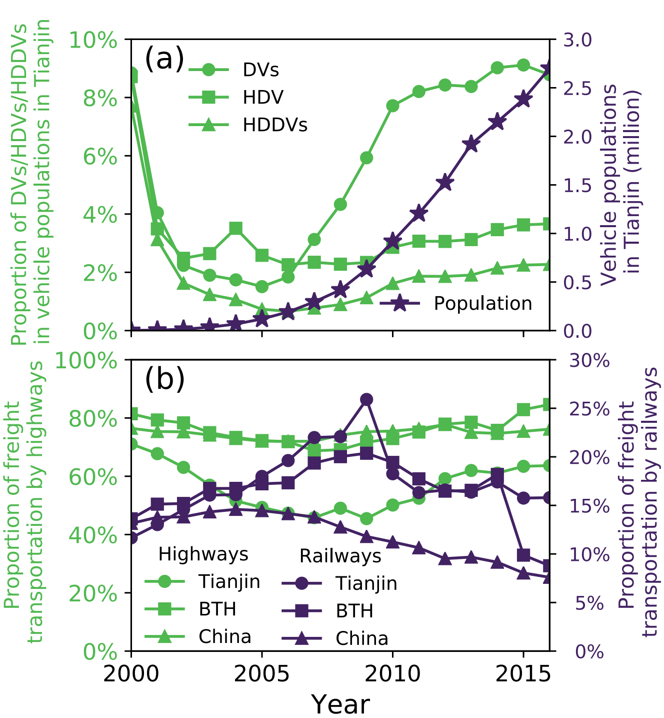

Fig. 7. (a) Proportion (left panel) of HDDVs (DVs, HDV) in the vehicle populations (right panel) in Tianjin from 2000 to 2016. (b) Proportions of freight transportation by highways (left panel) and railways (right panel) in Tianjin, Beijing-Tianjin-Hebei (BTH) region, and China from 2000 to 2016. The data in subplot (a) were obtained from the registered vehicle database (up to August 2017) of Tianjin. The data in subplot (b) were obtained from 'China statistical yearbook' compiled by the National Bureau of Statistics of China.

4. Conclusions

From the 2-week test of on-road vehicle emissions in the WJL tunnel in urban Tianjin, we updated EFs of $PM_{2.5}$, NO, NO_2, NO_x, and CO. The average EFs for the fleet varied diurnally, which could be due to diurnal variations in the proportions of DVs (1.97 ± 0.32%) and HDDVs (0.35 ± 0.35%) in the fleet, though the proportion of HDDVs (or DVs) never exceeded 10% in the tested tunnel during the study period. The obviously diurnal variations of the proportions of HDV and DVs in the fleet allowed us to conduct linear regressions to obtain EFs for GVs and DVs, LDV and HDV, and HDDVs and non-HDDVs. We used the fractions and EFs of DVs (HDV, and HDDVs) to reconstruct the average EFs for on-road vehicles in Tianjin, which were approximately 2.2, 1.7, 1.5, 2.0, and 1.6 times of those in the WJL tunnel. Our results demonstrated that the average EFs from previous tunnel tests should be carefully adjusted as there are differences in fleet compositions between the tested tunnels and cities, especially cities that HDTs are often not allowed to access to. DVs, especially HDDVs, are major sources of on-road $PM_{2.5}$, NO, NO_2, NO_x, and CO emissions in northern China. The relative contribution of on-road HDDVs to the total fleet emissions calculated by the EFs from the 'EI guidebook' was underestimated, as compared to our results from the reconstructed real-world EFs inferred from tunnel tests. The management and control of HDDV emissions have become urgent to reduction of on-road vehicle emissions. The adjustment and optimization of freight transportation structures, the strict inspections over production conformity and in-use compliance, the intelligent transport system, and the optimization of traffic network might be the solution to achieving low vehicle emissions.

The following are the supplementary data related to this article.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.04.387.

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