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Gaseous and Carbonaceous Composition of $PM_{2.5}$ Emitted from Rural Vehicles in China

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ABSTRACT

Rural vehicles (RVs) could contribute significantly to on-road vehicle emissions, especially $PM_{2.5}$ and NO_x; This study tested 10 three-wheeled (3-W) RVs and 8 four-wheeled (4-W) RVs on real roads in Hebei Province using a portable emissions measurement system to investigate gaseous concentrations (CO, NO_x, and HC) and the carbonaceous composition (EC and OC) of the $PM_{2.5}$ emitted. The results showed that the tightening emission standards resulted in the CO, HC, and $PM_{2.5}$ emissions for China II RVs decreasing, but may increasing NO_x emission for China II 3-W RVs. The emission level of $PM_{2.5}$ for China II RVs is between Euro II LDDTs and Euro III LDDTs. The emission factors (EFs) of OC and EC for 3-W RVs were 0.035 ± 0.019 and 0.058 ± 0.055 g km^{-1}, respectively, and for 4-W RVs, they were 0.046 ± 0.018 and 0.031 ± 0.024 g km^{-1}, respectively. The carbonaceous component represents the main fraction of $PM_{2.5}$ emitted from RVs (84.6% and 87.2% for 3-W and 4-W RVs, respectively), similar to other diesel vehicles. The average distance-based EFs of OC increased with increasing vehicle size (3-W RVs < 4-W RVs). The CO_2-based EFs of OC and EC decreased with increasing vehicle mass, consistent with the emission laws of light-, medium-, and heavy-duty diesel trucks. Driving cycles that included more cruise mode and less creep mode resulted in a higher average EC/OC ratio (1.57) for 3-W RVs than for 4-W RVs (0.63), and resulted in the average EC/OC ratios for both types of RV were lower than for highway LDDTs.

Keywords: Carbonaceous composition; Gaseous emission; Particulate matter; Rural vehicle; PEMS.

INTRODUCTION

Motor vehicles are a major source of air pollutant emissions in China (He *et al.*, 2002; Lang *et al.*, 2017), and diesel trucks are believed one of the most important sources of $PM_{2.5}$ (particulate matter with an aerodynamic diameter of < 2.5 µm) (Lv *et al.*, 2016). The characteristics of emissions by different types of regular motor vehicles have been estimated in previous studies (Guo *et al.*, 2007; Yao *et al.*, 2011a; Huo *et al.*, 2012a, b; Shen *et al.*, 2014; Cao *et al.*, 2016; He *et al.*, 2017). Rural vehicles (RVs) constitute a group of diesel-fueled vehicles used primarily in small towns and rural areas to transport people and goods. They are a very popular vehicle type in China and in other developing countries such as Thailand and Malaysia (Ellis and Hine, 1995; Ellis, 1997; Yao *et al.*, 2011a). In 2014, the total number of RVs in China was about 9.72 million (Ministry of Environment Protection of the People's Republic of China. 2016), but the population of RVs maybe underestimated by the Ministry of Environment Protection of the People's Republic of China because a lot of RVs were not registered in the government, the population of RVs estimated the sales data of RVs is 23.9 million in 2006 (Yao *et al.*, 2011a). They comprise an important part of the transportation network in China, and they have contributed greatly to the development and prosperity of China's rural economy. However, research on the emission factors (EFs) of RVs has rarely been reported, which constitutes an important obstacle to the improvement of the control of RV pollution. Research has shown that CO, HC, NO_x, and PM emissions by RVs in China in 2006 were 1409, 332, 933, and 54 Gg, respectively. These emissions accounted for > 40% of NO_x and PM emissions of on-road diesel vehicles, and > 14% of the total CO and HC emissions from all diesel vehicles (Yao *et al.*, 2011a). Phase 1 and phase 2 emission regulations for rural vehicles were introduced in China in 2006 and 2007, respectively (GB19756-2006, Limits and measurement methods of exhaust pollutants from diesel engines of 3-W and 4-W RVs). However, the lack of RV data from actual situations has made it difficult both to verify the validity of the relevant standards and to propose any future revisions (Xu *et al.*, 2014).

Particle composition of emissions is central to practical issues concerning health effects, climate change, source apportionment, and aerosol modeling. The chemical

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components of $PM_{2.5}$ include carbonaceous components (OC and EC), water soluble ions, elemental compounds, and unidentified compounds (Dellinger *et al.*, 2001; Xia and Nel, 2004; Shafer *et al.*, 2010; Stone *et al.*, 2010; Daher *et al.*, 2012; Zhou *et al.*, 2016). Emissions of carbonaceous components from diesel engines are often the focus of attention in relation to climate change, source resolution, and aerosol models (Maricq, 2007). It has been found that EC is a very suitable indicator for assessing the impact of traffic on PM (Minguillón *et al.*, 2014; Megido *et al.*, 2016). Furthermore, EC can reduce visibility through the absorption of light (Eldering and Cass, 1996; Eidels-Dubovoi, 2002; Yu *et al.*, 2016). Moreover, incomplete combustion and lubricating oil OC from fuels (Kittelson *et al.*, 2006; Li *et al.*, 2014; Wu *et al.*, 2016) include many carcinogenic and mutagenic substances (Chien *et al.*, 2009). The carbonaceous components of elemental carbon (EC) and organic carbon (OC) are the principal constituents of $PM_{2.5}$ emitted from diesel vehicles. Subramanian *et al.* (2009) found that the carbonaceous composition appeared relatively consistent among vehicle types, accounting for 57% of PM in 10 pickups, vans, heavy-duty trucks, and buses. The carbonaceous composition was found to account for 66% of PM in 39 light-duty and 54 heavy-duty engines based on chassis dynamometer tests (Kim Oanh *et al.*, 2010). Chiang *et al.* (2012) measured the emissions of six in-use light-duty diesel vehicles (LDDVs) using chassis dynamometer tests in Taiwan, and they found the carbonaceous constituents contributed 73% of the mass of $PM_{2.5}$. Shah *et al.* (2004) studied the EC, OC, and PM emission rates for a number of heavy heavy-duty diesel trucks (HDDTs) operating under real-world conditions using a unique mobile emissions laboratory. They found the carbonaceous fraction (CF = [OC × 1.2 + EC]/PM)) was approximately 0.90, 0.96, 0.96, and 0.94 for the idle, creep, transient, and cruise phases, respectively. Cheng *et al.* (2010) found that carbonaceous particles (the sum of organic material [OM = 1.2 × OC] and EC) were the dominant constituents of $PM_{2.5}$, on average, accounting for 82% and 70% of $PM_{2.5}$ emissions in a tunnel and at the three roadside sites, respectively. He *et al.* (2014) studied the characteristics of $PM_{2.5}$ and of its carbonaceous components emitted from heavy-duty diesel vehicles (HDDVs) using a portable emissions measurement system (PEMS). They found the carbonaceous components of $PM_{2.5}$ emitted from Euro I, Euro II, and Euro III HDDVs were 76.0%, 79.2%, and 77.8%, respectively. Zhang *et al.* (2015) found that carbonaceous components accounted for 89.6% of $PM_{2.5}$ emitted by pre-Euro I and Euro I HDDTs. Wu *et al.* (2015, 2016) also found that carbonaceous components were the main constituents (> 80%) of $PM_{2.5}$ emitted by Euro III and Euro IV diesel trucks. Jin *et al.* (2017) found that the total carbon (elemental carbon + organic carbon) was 90% of total DPM (Diesel Particulate Matter) mass for a heavy-duty diesel engine tested in ETC cycle with an engine dynamometer. Pervez *et al.* (2018) found that the OM was the major fraction (72–95%) of exhaust emissions. Carbonaceous components might also constitute the principal constituents of $PM_{2.5}$ emitted by RVs because RVs are diesel-fueled vehicles. Considering that RVs account for > 40% of PM emissions by on-road diesel vehicles, the total carbonaceous components of $PM_{2.5}$ emitted by RVs should be given greater attention; however, the carbonaceous composition of $PM_{2.5}$ emitted by RVs has not been measured before.

The carbonaceous composition of $PM_{2.5}$ emitted from diesel vehicles has been studied using chassis dynamometer tests, tunnel tests, roadside measurements, PEMS, and direct engine dynamometer measurements (Park *et al.*, 2003; Chellam *et al.*, 2005; Sharma *et al.*, 2005; Ntziachristos *et al.*, 2007; Biswas *et al.*, 2008; Kim Oanh *et al.*, 2010; May *et al.*, 2014; Alves *et al.*, 2015; Jin *et al.*, 2017). However, tests using chassis dynamometers cannot fully reveal all emission trends during a dynamic driving cycle (Holmen and Ayala, 2002; Saitoh *et al.*, 2003), tunnel and roadside tests cannot fully capture the range of driving cycles of diesel trucks (Geller *et al.*, 2005; He *et al.*, 2006; Zanini *et al.*, 2006), and direct engine dynamometer measurements do not fully represent emissions from diesel trucks (Park *et al.*, 2003; Sharma *et al.*, 2005). Recently, on-board PEMS tests have become important for vehicle emission research worldwide, because these methods can measure emission characteristics directly from the tailpipe in the real world (Durbin *et al.*, 2007; Johnson *et al.*, 2011; Giechaskiel *et al.*, 2014). Zhang *et al.* (2015) studied the chemical characterization of $PM_{2.5}$ emitted from on-road pre-EURO I and EURO I heavy-duty diesel trucks in China using PEMS, and Wu *et al.* (2015) studied the carbonaceous composition of $PM_{2.5}$ emitted from on-road China III diesel trucks in China using PEMS.

This study investigated the gaseous emissions and carbonaceous composition of $PM_{2.5}$ emissions from RVs using PEMS, provided the CO, NO_x, HC and $PM_{2.5}$ EFs of China II RVs and provided the carbonaceous composition EFs of RVs. The results of this study provide EFs that will be helpful for emission inventory establishment, model simulation, and source apportionment. In addition, the characterization of these emissions from RVs under real-world conditions is important for understanding where continuing efforts for improving air quality should be focused.

EXPERIMENTAL SECTION

Vehicle Test Fleet

In this study, 18 RVs (without exhaust emission control devices) comprising 10 three-wheeled (3-W) single-cylinder RVs and 8 four-wheeled (4-W) four-cylinder RVs, all equipped with manual transmissions, were tested in April 2015. Each vehicle was driven by the owner. Detailed information of the tested RVs is provided in Table 1. The average gross vehicle weight of the 3-W RVs was 817 kg, which was lower than the 4-W RVs (3,323 kg). The average rated power and engine displacement of the 3-W RVs were 12.9 kw and 988 mL, respectively, both lower than the 4-W RVs (75.3 kw and 3,468 mL, respectively). The average odometer reading of the 3-W RVs was 22,560 km, which was lower than the 4-W RVs (78,760 km).

Table 1. Information of the tested RVs.

TN	VT	CN	MY	GVW (kg)	EM	RP (kw)	ED (mL)	ODO (km)	ES
#1	3-W	1	2009	740	ZS1100-2	12.1	903	9000	China II
#2			2012	1130	KM130B	17.6	1290	9000	China II
#3			2011	1120	ZS1115EII	16.2	1194	12000	China II
#4			1999	700	ZS1100	11.0	903	115200	China 0
#5			1998	635	S1100	10.3	903	11900	China 0
#6			2009	820	S1100EII	12.1	903	8500	China II
#7			2005	665	ZS1100	12.1	942	3400	China 0
#8			2008	960	ZS1100E2	12.1	903	35100	China II
#9			2002	500	ZS1100	12.1	942	13000	China 0
#10			2005	900	ZS1105	13.2	996	8500	China 0
#11	4-W	4	2011	5700	YZ4DA7-30	83.0	3660	87186	China II
#12			2014	4800	YZ4DA7-30	83.0	3660	5265	China II
#13			2013	2720	YN38CR	88.0	3760	39264	China II
#14			2010	3300	4DX23-130E3	101.0	3857	95600	China II
#15			2007	2300	YZ4102QF	70.6	3857	150000	China I
#16			2009	2150	4DW91-63NG2	46.0	2545	11000	China II
#17			2006	2600	a	47.0	2545	99081	China 0
#18			2007	3015	4DX23	84.0	3857	142680	China I

TN: Test Number; VT: Vehicle Type; CN: Cylinder number; MY: Model Year; GVW: Gross Vehicle Weight; EM: Engine Model; RP: Rated Power; ED: Engine Displacement; ODO: Odometer; ES: Emission standard. a: Engine model was obscure and invalid.

Test Routes and Driving Cycles

Compared with light-duty diesel trucks (LDDTs), RVs have smaller engine power and lower design maximum speed (100 km h^{-1} for LDDTs and 50–70 km h^{-1} for RVs), which are major reasons why RVs are popular in developing countries (Yao *et al.*, 2011a). Driving patterns could affect vehicle emissions significantly and thus they constitute a very important element when estimating vehicle emissions (Hansen *et al.*, 1995; Kean *et al.*, 2003; Yao *et al.*, 2007; Shen *et al.*, 2014). Generally, the driving mode of RVs is different to LDDTs in China because RVs are not permitted on urban roads and highways. Additional details can be found in Yao *et al.* (2011a).

The test route used in this study is located in Gaobeidian in Hebei Province, close to Beijing. The selected test route included roads along which the drivers often drove. The 15-km route connects several villages and towns in a loop that includes 11.9 km of village cement roads and 3.1 km of secondary roads. On this route, the drivers followed the traffic and the driving cycle reflected the actual driving conditions. During the tests, the vehicles were unloaded, except that the total mass of the test equipment and testers was about 350 kg. At the time of the tests, the average atmospheric $PM_{2.5}$ concentration was about 0.39 mg m^{-3} and the air temperature was approximately 281–292 K.

Each vehicle was tested twice on the same route. Thus, 36 sets of valid data were obtained over the cumulative distance of 436 km (3-W RVs: 139 km and 4-W RVs: 297 km). The total test average speed for all RVs was 21 km h^{-1}. Typical examples of velocity profiles of the 3-W RVs and 4-W RVs are shown in Fig. 1. It is evident that the driving cycles of the 3-W RVs and 4-W RVs were different on the same test route. The average speed was 18.0 km h^{-1} for the 3-W RVs and 31.1 km h^{-1} for the 4-W RVs. The average speed for all 4-W RVs was always higher than for 3-W RVs during all tests.

Sampling System and Method

A PEMS based on our previous on-board emission system was used to collect data and samples (Huo *et al.*, 2012a, b; Yao *et al.*, 2014; Yao *et al.*, 2015; Wu *et al.*, 2015, 2016). The sampling system consisted of four main parts: the SEMTECH EFM-2 tube, SEMTECH DS mobile emission analyzer, SEMTECH MPS (Micro-proportional Sample System; Sensors Inc., Ann Arbor, MI, USA), and $PM_{2.5}$ sampling system. The SEMTECH EFM-2 tube measured the flow of the RV exhaust. The SEMTECH DS was used to analyze the instantaneous concentration of gaseous emissions (CO_2, CO, HC, and NO_x). The SEMTECH MPS was used to dilute the RV exhaust with clean air by a high efficiency particulate air (HEPA) filter. The $PM_{2.5}$ sampling system was used to collect the samples. Gravimetric filters are commonly used for PM mass measurements (Giechaskiel *et al.*, 2014). In this work, the sample time varied with different averaged speed, is generally about 30–50 mins; the sample flow was 10 L min^{-1}; the dilution rate varied with the volume of exhaust, because the SEMTECH MPS conducts constant proportion sampling of the vehicle exhaust flow and dilutes it with a certain amount of mixed gas (Shen *et al.*, 2014). The mass of $PM_{2.5}$ was determined by collecting particles on PTFE Teflon™ filters (47 mm) (R2PJ047, PALL, USA), which were weighed using a microbalance (MT5, Mettler Toledo, Switzerland). The $PM_{2.5}$ samples collected on 47-mm quartz filters (2500QAT-UP, PALL, USA), which were preconditioned at 650°C for 8 hr to eliminate organic pollutants, were used to analyze organic carbon (OC) and elemental carbon (EC) contents. A 1-cm^2 punched circle was cut from the quartz

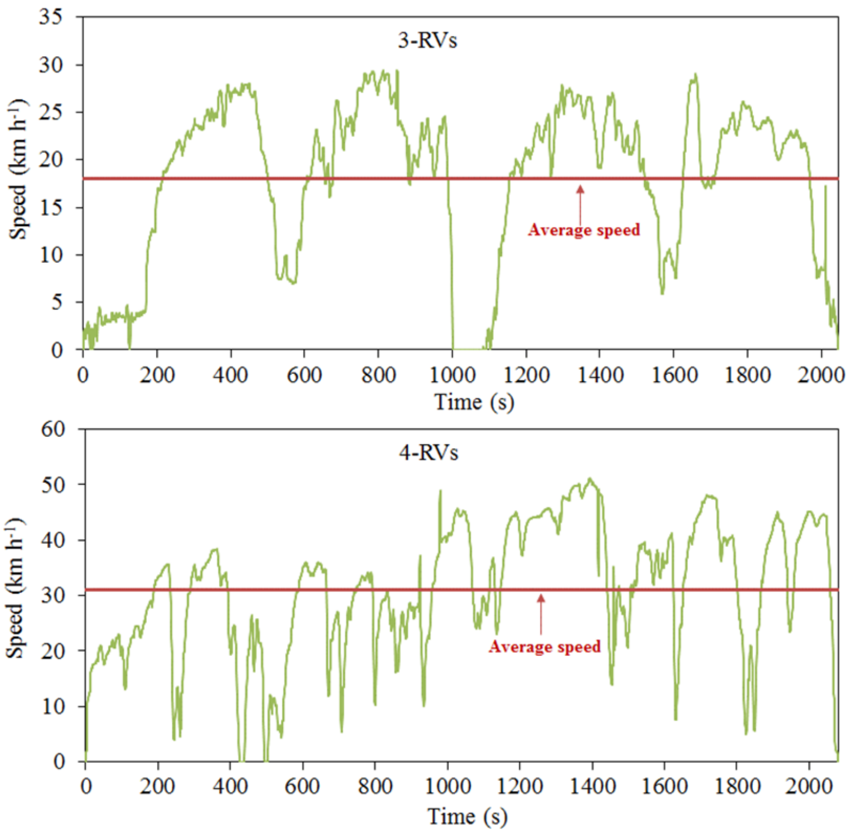

Fig. 1. Typical example of velocity profiles of 3-W RVs and 4-W RVs.

filter. This was analyzed using a Thermal/Optical Carbon Aerosol Analyzer (DRI Model 2001A, Atmoslytic Inc., USA) according to the NIOSH 5040 reference method, which is often used for assessing the OC and EC contents of diesel-laden aerosols. Additional detailed information on the system is provided in our previous study (Wu *et al.*, 2015).

RESULTS AND DISCUSSION

Emissions Factors of CO, HC, NO_x, and $PM_{2.5}$

The former study (Yao *et al.*, 2011a) showed that RVs are significant contributors to national emissions of CO, HC, NO_X, and $PM_{2.5}$ in China. And the driving patterns can affect vehicle emissions significantly (Yao *et al.*, 2007; Shen *et al.*, 2014). Therefore, when estimating vehicle emissions, it is important to compare the EFs of CO, HC, NO_x, and $PM_{2.5}$ for RVs with other studies and other vehicle types under real-world driving cycles. Fig. 2 illustrates the distance-based EFs (g km^{-1}) of CO, HC, NO_x, and $PM_{2.5}$ for 3-W RVs, 4-W RVs, LDDTs, and LDDVs. The RVs were divided into Pre-China II RVs (mode year before 2008) and China II RVs (mode year was and after 2008) in order to compare with previous studies. The CO, HC, and $PM_{2.5}$ distance-based EFs for China II 3-W RVs from this study were lower than those for Pre-China II 3-W RVs from this study and from Yao *et al.* (2011a), but for NO_x: Pre-China II from Yao *et al.* (2011a) < the China II from this study < Pre-China II from this study. For the HC and $PM_{2.5}$ distance-based EFs of 4-W RVs: Pre-China II from this study < the China II from this study < Pre-China II from Yao *et al.* (2011a); For NO_x distance-based EFs of 4-W RVs: Pre-China II from Yao *et al.* (2011a) < the China II from this study < Pre-China II from this study; For CO distance-based EFs of 4-W RVs: Pre-China II from Yao *et al.* (2011a) < Pre-China II from this study < the China II from this study.

The averaged distance-based EFs of CO, HC, and NO_x for China II 3-W RVs were all lower than Euro III LDDTs from Shen *et al.* (2015), the averaged distance-based EFs of $PM_{2.5}$ for China II 3-W RVs was between Euro II and Euro III LDDTs from Shen *et al.* (2015); The EFs of CO and $PM_{2.5}$ for China II 3-W RVs were lower than the LDDVs (without pollution control equipment) from Chiang *et al.* (2012), but for HC and NO_x, they were higher than the LDDVs. The averaged distance-based EFs of NO_x and $PM_{2.5}$ for China II 4-W RVs was between Euro II and Euro III LDDTs from Shen *et al.* (2015); That of CO for China II 4-W RVs was between Euro II and Euro III LDDTs, those of HC for China II 4-W RVs was lower than Euro III LDDTs. The EFs of CO, HC, and NO_x for China II 4-W RVs were lower than the LDDVs from Chiang *et al.* (2012), but the $PM_{2.5}$ had opposite trend.

The average EFs of CO, HC, NO_x, and $PM_{2.5}$ for all 3-W RVs under real-world driving cycles were 1.666 (0.489–3.321), 0.382 (0.077–1.350), 1.518 (1.387–6.418), and 0.081 g km^{-1} (0.029–0.339 g km^{-1}), respectively. The average EFs of CO, HC, NO_x, and $PM_{2.5}$ for all 4-W RVs under real-world driving cycles were 2.952 (1.937–4.989), 0.696

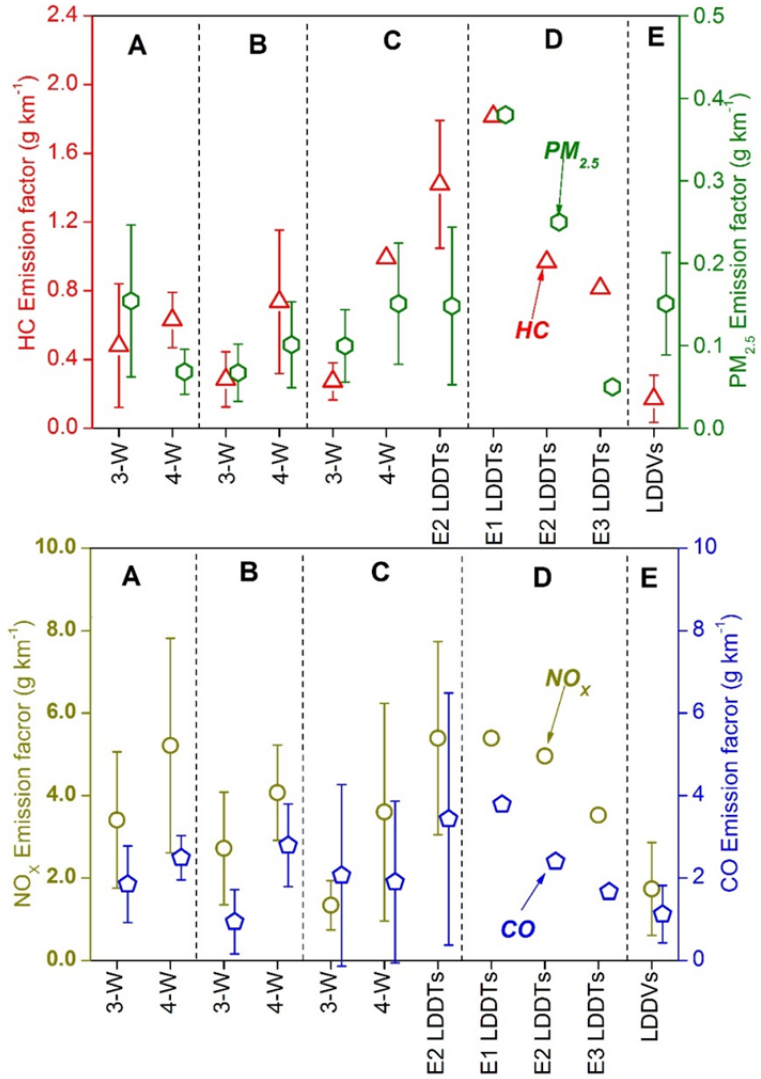

Fig. 2. Distance-based emission factors (g km^{-1}) of RVs, LDDTs, LDDVs (error bars show standard deviations). A: The Pre-China II RVs in this study; B: The China II RVs in this study; C: Yao *et al.* (2011a) (Pre-China II RVs); D: Shen *et al.* (2015); E: Chiang *et al.* (2012); E1: Euro I; E2: Euro II; E3: Euro III.

(0.283–1.614), 4.499 (2.160–8.668), and 0.046 g km^{-1} (0.032–0.218 g km^{-1}), respectively. It is evident that the EFs of CO for RVs were between those of Euro I LDDTs and Euro III LDDTs from Shen *et al.* (2015), lower than Euro II LDDTs from Yao *et al.* (2011a), and higher than LDDVs from Chiang *et al.* (2012).The EFs of HC for 3-W and 4-W RVs from this study and 3-W RVs from Yao *et al.* (2011a) were lower than Euro III LDDTs from Shen *et al.* (2014) and higher than LDDVs from Chiang *et al.* (2011). The EFs of NO_x for 3-W RVs from this study were similar to Euro III LDDTs from Shen *et al.* (2015). The EFs of NO_x for 4-W RVs from this study and Yao *et al.* (2011a) were between Euro II LDDTs and Euro III LDDTs from Shen *et al.* (2015). The EFs of $PM_{2.5}$ for all RVs were between Euro II LDDTs and Euro III LDDTs from Shen *et al.* (2015). Driving patterns, fuel quality, and types of diesel engine could all affect vehicle emissions significantly (Kean *et al.*, 2003; Yao *et al.*, 2007, 2011a; Wang *et al.*, 2018), which could account for the differences in the EFs of CO, HC, NO_x, and $PM_{2.5}$ between these various studies.

Fuel-based EF data are influenced less by average speed, vehicle weight, and engine power (Pierson *et al.*, 1996; Tong *et al.*, 2000; Kean *et al.*, 2003); therefore, the fuel-based EFs in this study were calculated to eliminate the impact of the weight and engine power of the vehicle as much as possible. Fig. 3 illustrates the fuel-based EFs (g kg^{-1} diesel) of CO, HC, NO_x, and $PM_{2.5}$ for 3-W RVs, 4-W RVs, LDDTs (Yao *et al.*, 2011a), and LDDVs (Chiang *et al.*, 2012). The RVs were divided into Pre-China II RVs (mode year before 2008) and China II RVs (mode year was and after 2008) in order to compare with previous studies. The CO, HC, and $PM_{2.5}$ fuel-based EFs for China II 3-W RVs from this study were lower than those for Pre-China II 3-W RVs from this study and from Yao *et al.* (2011a), but for NO_x, the fuel-based EFs for China II 3-W RVs from this study were higher than those for Pre-China II 3-W RVs from this study and from Yao *et al.* (2011a). The reason for phenomenon may be the improvement of engine technologies

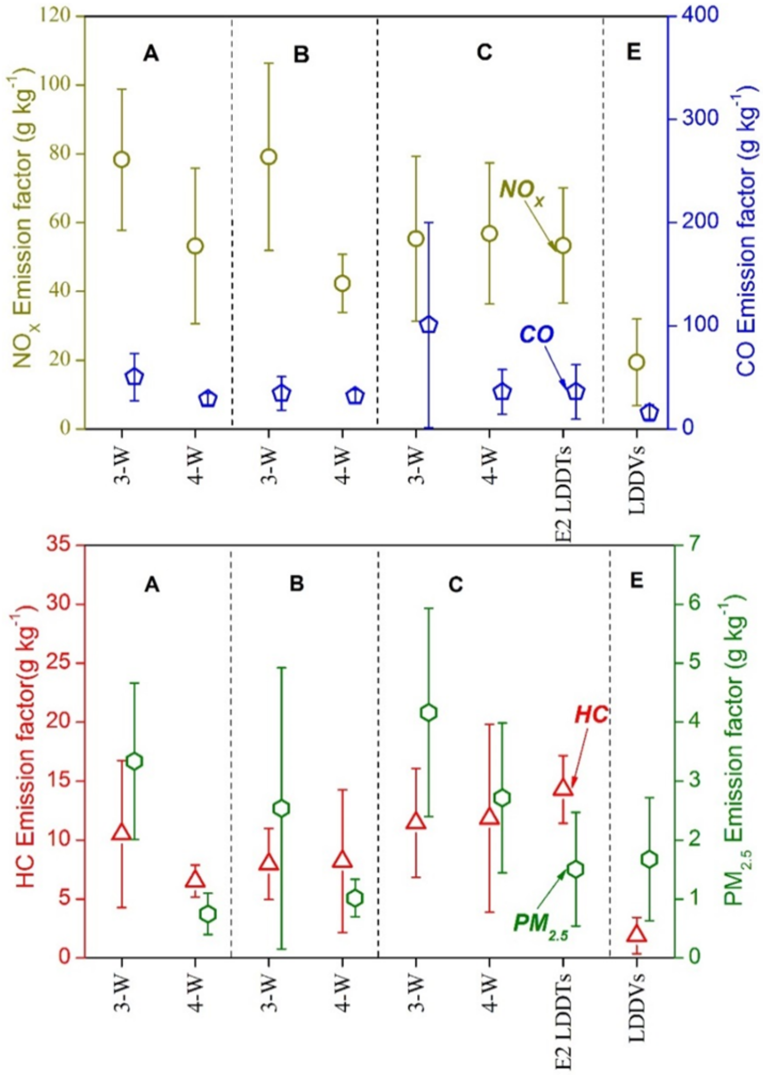

Fig. 3. Fuel-based emission factors (g kg^{-1}) of RVs, LDDTs, and LDDVs (error bars show standard deviations). A: The Pre-China II RVs in this study; B: The China II RVs in this study C: Yao *et al.* (2011a) (Pre-China II RVs); E: Chiang *et al.* (2012); E2: Euro II;

used in 3-W RVs caused the increasing of combustion efficiency, and then increasing NO_x emission, reducing CO, HC, and $PM_{2.5}$ emissions. The CO, HC, NO_x, and $PM_{2.5}$ fuel-based EFs for China II 4-W RVs from this study were lower than those for Pre-China II 4-W RVs from Yao *et al.* (2011a), but were higher than those for Pre-China II 4-W RVs from this study except NO_x, this phenomenon may be caused by small sample size for 4-W RVs in this study.

The average fuel-based EFs of CO, HC, NO_x, and $PM_{2.5}$ for all 3-W RVs were 42.4 (16.3–99.0), 9.2 (3.8–25.8), 78.7 (48.2–123.8), and 2.9 g kg^{-1} diesel (0.7–7.1 g kg^{-1} diesel), respectively. The average fuel-based EFs of CO, HC, NO_x, and $PM_{2.5}$ for all 4-W RVs were 30.7 (20.2–42.4), 7.6 (3.0–22.1), 46.4 (29.6–82.9), and 0.9 g kg^{-1} diesel (0.3–1.6 g kg^{-1} diesel), respectively. The fuel-based EFs of CO, HC, NO_x, and $PM_{2.5}$ for 3-W RVs were higher than 4-W RVs and close to the results for Euro II LDDTs; the trend was the same as the results from Yao *et al.* (2011a). All the EFs of CO, HC, and NO_x of the RVs in this study were higher than the results of Chiang *et al.* (2012).

Analysis of the Particulate Carbonaceous Component

Fig. 4 illustrates the dispersion of EFs for $PM_{2.5}$, OC, and EC based on distance for all test vehicles in the form of box and whisker plots. There is no previous research on the EFs of OC and EC for RVs; therefore, we compared them with the EFs of OC and EC for LDDTs using the PEMS (Wu *et al.*, 2015) and chassis dynamometer tests (Kim Oanh *et al.*, 2010). The EFs of $PM_{2.5}$, OC, and EC for 3-W RVs were 0.111 ± 0.079, 0.035 ± 0.019, and 0.058 ± 0.055 g km^{-1}, respectively. The EFs of $PM_{2.5}$, OC, and EC for 4-W RVs were 0.089 ± 0.046, 0.046 ± 0.018, and 0.031 ± 0.024 g km^{-1}, respectively. The EFs of $PM_{2.5}$, OC, and EC for 3-W RVs varied more widely than for 4-W RVs. Compared with results in the literatures (Kim Oanh *et al.*, 2010; Wu *et al.*, 2015), the average $PM_{2.5}$ and OC EFs for 3-W RVs and 4-W RVs were higher than Euro III LDDTs. The OC in vehicle exhausts is derived from unburned fuel

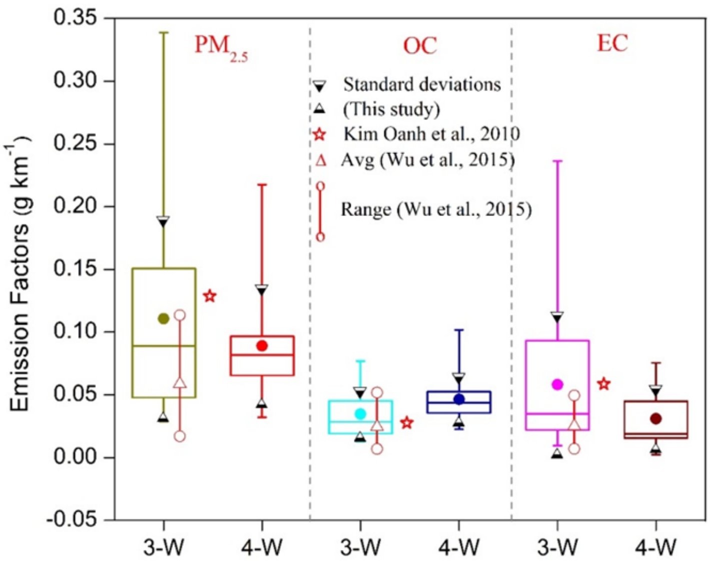

Fig. 4. Distance-based EFs of $PM_{2.5}$, OC, and EC in various studies. The box shows the interquartile range, line in the box shows the median, and circle shows the mean. The box range represent the upper 25% and lower 25%, and the whiskers represent the maximum and minimum EFs of $PM_{2.5}$, OC, and EC.

oil, combustion byproducts, and lubricating oil. In this study, the average EFs of OC increased with increasing vehicle size (Gross vehicle weight and engine displacement; 3-W RVs < 4-W RVs), similar to results in the literature (Wu *et al.*, 2015). The reason for this trend is larger vehicles produce greater quantities of unburned fuel oil, combustion byproducts, and lubricating oil per kilometer. The EC in vehicle exhausts is formed in the fuel-rich zone under high pressure and temperature, especially when the engine load increases and more fuel is injected into the cylinder. The 4-W RVs had higher engine displacement and had higher maximum engine output torque, so had lower engine load compare with 3-W RVs at normally driving condition. Therefore, the average EF of EC for 3-W RVs was higher than for 4-W RVs, and the EFs of EC for both 3-W RVs and 4-W RVs were higher than Euro III LDDTs, indicating that the engine operating condition of the 3-W RVs is worse than 4-W RVs, and worse than Euro III LDDTs.

Many studies have shown that the EFs of EC and OC from diesel engines vary with engine load, mode of vehicle operation, vehicle type, vehicle age, fuel quality, and ambient conditions (Park *et al.*, 2003; Chellam *et al.*, 2005; Sharma *et al.*, 2005; Ntziachristos *et al.*, 2007; Biswas *et al.*, 2008; Kim Oanh *et al.*, 2010; May *et al.*, 2014; Wu *et al.*, 2015). To exclude the influence of engine size, the EFs in this study were normalized to the amount of CO_2 produced (CO_2 can be used as a proxy for fuel consumption). The CO_2-based EFs (g (kg of CO_2)$^{-1}$) of the RVs are presented as box and whisker plots in Fig. 5.

The average EFs of CO_2 for 3-W RVs and 4-W RVs were 121 ± 50 and 300 ± 58 g km^{-1}, respectively. The distance-based average EFs of CO_2 for 4-W RVs was 1.5 times higher than that of 3-W RVs because 4-W RVs have greater engine displacement. After normalization, the EFs decreased with increasing vehicle mass, which is consistent with the emission laws of LDDTs, medium-duty diesel trucks (MDDTs), and HDDTs (Wu *et al.*, 2015). The CO_2-based EFs of $PM_{2.5}$, OC, and EC for 3-W RVs were 2.2, 1.2, and 3.5 times higher than those of 4-W RVs, respectively, indicating that 4-W RVs had greater combustion efficiency than 3-W RVs.

The CO_2-based emissions per vehicle showed greater dispersion for 3-W RVs than 4-W RVs. The EFs of #2, #4, and #5 were much higher than the other 3-W RVs. Possible reasons for this might be that the engine models of #2 and #5 were very different to the other 3-W RVs, and that the odometer reading of #4 was higher than the other RVs.

The fuel-based EFs of $PM_{2.5}$, OC, and EC for RVs were also calculated in this study, shown in Table 2. The fuel-based EFs of $PM_{2.5}$, OC, and EC for China II 3-W RVs were 2.537±2.262 g kg^{-1}, 2.537 ± 2.262 g kg^{-1}, and 2.537 ± 2.262 g kg^{-1}, respectively; The fuel-based EFs of $PM_{2.5}$, and EC for China 0 3-W RVs were 32% and 82% higher than China II 3-W RVs, respectively. But the fuel-based EFs of OC for China 0 3-W RVs were 32% lower than China II 3-W RVs. The fuel-based EFs of $PM_{2.5}$, and EC for China II and China I 4-W RVs were lower than those for China 0 4-W RVs, but the fuel-based EFs of $PM_{2.5}$, OC, and EC for China II were higher than China I 4-W RVs. Overall, the fuel-based EFs of $PM_{2.5}$, OC, and EC for China II 4-W RVs did not show a significant decrease trend compared with pre-China II 4-W RVs.

Mass Ratios of EC/OC, OC/$PM_{2.5}$, and EC/$PM_{2.5}$

Table 3 provides information on the mass ratios of EC/OC, OC/$PM_{2.5}$, and EC/$PM_{2.5}$ and other related information concerning this and other studies. None of the vehicles in any of the studies was equipped with pollution control equipment.

The average ratios of total carbon (TC: TC = OC + EC), OC, and EC to $PM_{2.5}$ for 3-W RVs were 84.6% (70.7%–91.6%), 37.2% (17.8%–57.6%), and 47.4% (25.7%–69.8%),

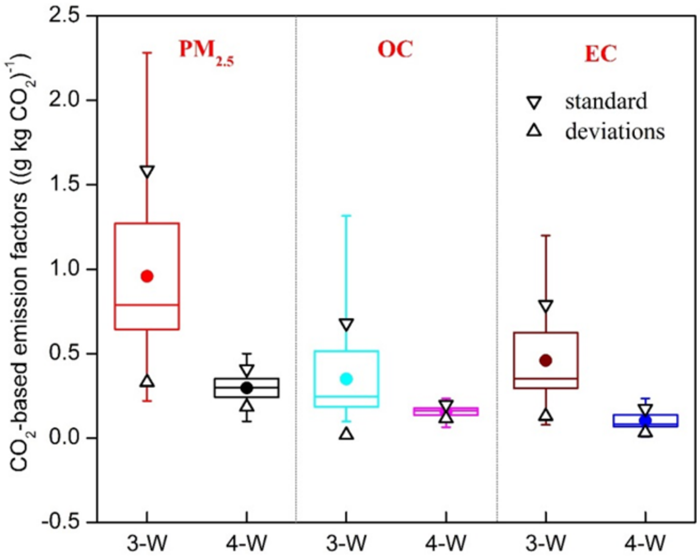

Fig. 5. CO_2-based EFs of $PM_{2.5}$, OC, and EC for RVs. The box shows the interquartile range, line in the box shows the median, and circle shows the mean. The box range represent the upper 25% and lower 25%, and the whiskers represent the maximum and minimum EFs of $PM_{2.5}$, OC, and EC.

Table 2. The fuel-based EFs of $PM_{2.5}$, OC, and EC for RVs.

VT	3-W RVs (g kg^{-1})		4-W RVs (g kg^{-1})		
ES	China II	China 0	China II	China I	China 0
$PM_{2.5}$	2.537 ± 2.262	3.339 ± 1.258	1.018 ± 0.301	0.593 ± 0.286	1.056 ± 0.038
OC	1.364 ± 1.354	0.932 ± 0.194	0.539 ± 0.071	0.358 ± 0.151	0.453 ± 0.039
EC	1.056 ± 0.621	1.920 ± 1.021	0.349 ± 0.208	0.141 ± 0.109	0.511 ± 0.098

VT: Vehicle Type; ES: Emission standard.

respectively. The average ratios of TC, OC, and EC to $PM_{2.5}$ for 4-W RVs were 87.2% (81.1%–93.5%), 56.4% (37.8%–80.0%), and 30.8% (6.9%–55.7%), respectively. In this study, EC was the main component of $PM_{2.5}$ emitted by 3-W RVs and OC was the main component of $PM_{2.5}$ emitted by 4-W RVs. The proportion of carbonaceous components in the $PM_{2.5}$ emitted by the RVs was > 80%, i.e., the same as found in other diesel vehicles using PEMS (Shah *et al.*, 2004; He *et al.*, 2014; Wu *et al.*, 2015; Zhang *et al.*, 2015; Wu *et al.*, 2016). However, the proportion of carbonaceous components in $PM_{2.5}$ emitted by diesel vehicles was found to be < 80% using chassis dynamometer (Subramanian *et al.*, 2009; Kim Oanh *et al.*, 2010; Chiang *et al.*, 2012) and tunnel tests (Cheng *et al.*, 2010). The EC/OC ratio clearly reflects the difference in carbon composition, a finding that might aid apportionment through models (Watson *et al.*, 1994; Wu *et al.*, 2015). The average mass ratios of EC/OC for 3-W RVs and 4-W RVs were 1.57 (0.45–3.70) and 0.63 (0.24–1.47), respectively. The higher value found for 3-W RVs compared with 4-W RVs might be attributable to their driving cycles. Although the driving cycles of both 3-W RVs and 4-W RVs are non-highway, the driving cycles of 3-W RVs include longer periods of cruise mode, as shown in Fig. 1. Cruise mode produces more EC but less OC (Shah *et al.*, 2004, 2006; Wu *et al.*, 2015, 2016); thus, the mass ratio of EC/OC of 3-W RVs is higher than 4-W RVs. The average mass ratios of EC/OC for 3-W RVs and 4-W RVs were both lower than highway LDDTs. The reason for this might be that the driving cycle of highway LDDTs includes longer periods of cruise mode and less creep mode than RVs.

CONCLUSIONS

In this study, 18 RVs (10 3-W RVs and 8 4-W RVs) were tested on real roads in Hebei Province (China) using the PEMS. Gaseous emissions (CO, NO_x, and HC) and carbonaceous composition (EC and OC) of the $PM_{2.5}$ emitted from the RVs were investigated.

With the tightening emission standards and the application of advanced emission control technologies, the CO, HC, and $PM_{2.5}$ distance-based and fuel-based EFs for China II 3-W RVs all had a significant downward trend compare with Pre-China II 3-W RVs, but NO_x fuel-based EFs for China II 3-W RVs had a upward trend compare with Pre-China II 3-W RVs. The CO, HC, NO_x, and $PM_{2.5}$ distance-based and fuel-based EFs for China II 4-W RVs all had a significant downward trend compare with Pre-China II 4-W RVs from previous study. The emission level of $PM_{2.5}$ for China II RVs is between Euro II LDDTs and Euro III LDDTs, The emission level of CO, HC, and NO_x for China II 3-W RVs were better than Euro III LDDTs.

The EC fuel-based EFs for China II 3-W RVs had a downward trend compare with Pre-China II 3-W RVs, but

Table 3. Mass ratios of EC/OC, OC/$PM_{2.5}$, and EC/$PM_{2.5}$ for different vehicles.

Source	Location	Method	Size	Driving Cycle	OC/$PM_{2.5}$ (%)	EC/$PM_{2.5}$ (%)	EC/OC
This study	HeBei	PEMS	3-W RV		37.3 ± 12.8	47.4 ± 12.5	1.57 ± 0.99
			4-W RV	NHW[a]	56.4 ± 12.7	30.8 ± 13.6	0.63 ± 0.41
Wu *et al.*, 2015	Beijing	PEMS	LDDT	NHW-I	55 ± 10	28 ± 5	0.54 ± 0.18
				NHW-II	40 ± 8	44 ± 8	1.16 ± 0.38
				HW[b]	27 ± 7	60 ± 6	2.42 ± 0.18
Subramanian *et al.*, 2009	Bangkok	chassis dynamometer	Pickups, Vans, HDT, Bus	POINT/ETC	17 ± 1	40 ± 2	2.35
Kim Oanh *et al.*, 2010	Bangkok	chassis dynamometer	LDDT, HDDT	EUDC/ETC	19	47	2.47
Chiang *et al.*, 2012	Taiwan	chassis dynamometer	LDDV	FTP-75	25	48	1.95
Cheng *et al.*, 2010	Hong Kong/Mainland	On-road tunnel	diesel-fueled vehicle		26	51	1.93

[a] NHW: non-highway.
[b] HW: highway.

OC fuel-based EFs for China II 3-W RVs had an upward trend compare with Pre-China II 3-W RVs. The OC fuel-based EFs for China II 4-W RVs had an upward trend compare with Pre-China II 4-W RVs, but the fuel-based EFs of EC for China II 4-W RVs did not show a significant decrease trend compared with pre-China II 4-W RVs.

The carbonaceous component is the main fraction of $PM_{2.5}$ emitted by RVs (84.6% for 3-W RVs and 87.2% for 4-W RVs), it is similar to the other diesel vehicles. The average distance-based EFs of OC increased with increasing vehicle size (3-W RVs < 4-W RVs), however, the average CO_2-based EFs of OC increased with decreasing vehicle size (3-W RVs < 4-W RVs), this is because more fuel is consumed to power the larger engines. The EFs of EC for both 3-W RVs and 4-W RVs were higher than Euro III LDDTs, The CO_2-based EFs of $PM_{2.5}$, OC, and EC for 3-W RVs were 2.2, 1.2, and 3.5 times higher than those of 4-W RVs, respectively. The CO_2-based EFs decreased with increasing vehicle mass, consistent with the emission laws of LDDTs, MDDTs, and HDDTs. The average mass ratios of EC/OC of 3-W RVs and 4-W RVs were 1.57 (0.45–3.70) and 0.63 (0.24–1.47), respectively. Driving cycles that included more cruise mode and less creep mode resulted in a higher average EC/OC ratio for 3-W RVs compared with 4-W RVs, and the average EC/OC ratios for both types of RV were lower than highway LDDTs.

The results of this study can supplement the database of gaseous EFs of RVs in China and result in a better understanding of the carbonaceous composition of $PM_{2.5}$ for RVs. This study will help other researchers to develop a $PM_{2.5}$ inventory, and provide source contributions of $PM_{2.5}$ from RVs in China. Our results can also be used for global and regional $PM_{2.5}$ emission inventories, and as a database for developing countries similar to China.

There are many other aspects needed to be explored to improve the estimates of RVs emissions because several uncertainties remained in our test. For example, measurement uncertainties related to random analysis errors and sampling might have contributed to our results (Giechaskiel *et al.*, 2014). Many factors (engine type, operating conditions, fuel type, malfunction, etc.) can affect emissions (Maricq, 2007). The other chemical components of $PM_{2.5}$ (water soluble ions, elemental compounds) were not measured in this study. Therefore, more RVs should be tested in future studies to understand the effects of driving conditions.

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