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Newer Generation of Scooters: Polychlorinated Dibenzo-*p*-dioxin and Dibenzofuran and Polychlorinated Biphenyl Reductions

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ABSTRACT

Scooter emissions have attracted attention in recent years because of human exposure to their direct effects in urban areas. Trace toxics, such as polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and polychlorinated biphenyls (PCBs) have thus become important in scooter emissions. In this work, ten Tier 5 and 6 scooters were tested using a 100-second model to analyze their PCDD/F and PCB emissions and compare the results with previous Tier 3 studies. Tier 5 and 6 scooters emitted 1.86–2.91 and 0.133–0.298 pg WHO-TEQ Nm^{-3} of PCDD/Fs and PCBs, respectively. It was interesting to find that the PCDD/Fs were reduced by 94.6–97.4% and 99.4–99.6% in Tier 5 and 6 motors, respectively. The congener profiles of PCDD/Fs were affected by improving the emission control. The domination of highly chlorinated congeners shown in Tier 3 was reduced in Tier 5 with increases in low chlorinated PCDFs. This showed that *de novo* synthesis occurred and could be inhibited by the OBD system in Tier 6. The tailpipe renews reduced 60.0–93.8% of PCDD/Fs and 85.3–97.7% of PCB emissions, but several cases would still exhibit a delay for stable operation of a catalytic converter. The annual emissions of PCDD/F TEQ was calculated based on the statistics in 2019 and tested as 1.63 g WHO-TEQ. It could be 99.7% reduced to 3.55 mg by replacing all scooters with Tier 6. Consequently, the improvement of electronic fuel injection and on-board diagnostics systems from a carburetor without feedback control not only reduced the regulated pollutants but effectively reduced PCDD/F emissions.

Keywords: Scooter; PCDD/Fs; PCBs; Electronic fuel injection; on-board diagnostics; Carburetor; Emission standards.

INTRODUCTION

The rapid economic development and increasing population leads to more environmental impact by human activities. The relative health risks are in terms of pollutant emission especially in the urban area (Goel and Guttikunda, 2015; Grivas *et al.*, 2018; Wu *et al.*, 2020). The highly intensive and complex industrial activities, the traffic emission is also an important contributor of urban atmospheric pollutants, including carbon monoxide (CO), hydrocarbons (HC), and nitrogen oxides (NO_x) (Cheng *et al.*, 2018, Tsai *et al.*, 2019a), and other toxic pollutants in the urban atmosphere (Grivas *et al.*, 2018, Dhital *et al.*, 2019). The volatile organic carbons (VOCs) emissions are also concerned in the densely populated urban area (Tsai *et al.*, 2018b). They could be contributed by both human life activity (Que *et al.*, 2019) and locally traffic emission, when the secondary pollutants subsequently occurs from the atmospheric chemical reactions

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and remained in the air around the residents (Liu *et al.*, 2017; Liu *et al.*, 2020; Wu *et al.*, 2020). Particulate matter (PM) pollution attracted more consideration from environmental and public health researchers. The physical and chemical characteristics of PM and their containing chemicals (e.g., heavy metals, air toxins, acids) could directly affect the human health (Tsai *et al.*, 2018a; Lin *et al.*, 2019b; Tsai *et al.*, 2019a; Zhang *et al.*, 2019). Ultrafine particle also becomes a recent issue for more deeply health effect on the respiratory system of the urban residents (Grana *et al.*, 2017, Xiang *et al.*, 2018, 2019). Fortunately, most of the gaseous pollutants and PM from on-road vehicles could be measured (or monitored) by either standard method certified by EPA, the consumer-grade air pollution measurement devices (Manibusan and Mainelis, 2020; Park *et al.*, 2020), or even portable monitors. The control strategy could be approached smoothly, while the emerging measures could be on time. The only challenge of pollution issues would be that they cannot quantify some pollutants in a quick time. Unfortunately, those pollutants are usually more persistent and even toxic, such as polyaromatics and their halogenated compounds (Durant *et al.*, 1996, 1999; Chen *et al.*, 2018).

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) and polychlorinated biphenyls (PCBs) are semi-volatile organic compounds (SVOCs) that could spread out in various media. They are considered as two important groups of persistence organic pollutants (POPs), which have a long half-life in the environment (Pirkle *et al.*, 1989, Sinkkonen and Paasivirta, 2000) and even in the human body (Flesch-Janys, 1996; Gao *et al.*, 2019). International Agency for Research on Cancer (IARC) classify 2378-TeCDD in Group 1 (carcinogenic to human), when the other PCDD/Fs and dioxin-like PCBs (dl-PCBs) were probable human carcinogens (IARC, 1997; Iwata *et al.*, 2004; IARC, 2016). They are reported to have enzyme induction and endocrine effect (Li and Hansen, 1996), bioaccumulation, and magnification in many organisms. This persistence property led to spread widely around the world even in the place off the beaten track (Booth *et al.*, 2013; Vecchiato *et al.*, 2015; Corsolini *et al.*, 2017; Ssebugere *et al.*, 2019). Therefore, the PCDD/F and PCB exposures attracted prominent concern.

PCDD/Fs and PCB were mainly emitted from waste incinerators, metallurgical processes, fossil fuel power plants, boilers, cement plants, and open burning when the on-road transport is a less contribution for PCDD/Fs and PCBs in most of the countries (Bawden *et al.*, 2004; Quass *et al.*, 2004; Lohmann *et al.*, 2007; Wang *et al.*, 2012). In Taiwan, the total annual PCDD/F emissions have been reduced by 83.8% from 329.5 g-TEQ yr^{-1} in 2002 to 53.3 g-TEQ yr^{-1} in 2016 (Tu, 2018). The dominant emission sources were changed from the electric arc furnace (54.6%), waste incinerators (19.4%), and sinter plant (11.4%) in 2002 to the boiler combustion (23.6%), electric arc furnace (22.1%), sinter plant (17.7%), waste incinerators (10.0%), non-ferrous metal secondary smelting processes (9.6%), and fugitive sources (6.9%) in 2012 and remained the similar flat-distribution of PCDD/F sources until today. This could be resulted from that the stricter emission standards for specific stationary sources were established within 2003–2010. The PCDD/F emission seems to be well controlled in Taiwan. However, the real near-ground exposure levels of PCDD/Fs and PCBs were still not clear, especially the peak value occurred by traffic sources, since they could not be online continuous monitored and could increase the potential health risk in the urban area (Oehme *et al.*, 1991; Broz *et al.*, 2000; Chuang *et al.*, 2010, Wheatley and Sadhra, 2010). More recently, the level of contribution of exhaust emissions from scooters and scooters to the contamination of ambient air was reported by 30% in Rome City (Grana *et al.*, 2017). A latest report showed there were 28% of PCDD/F came from traffic emissions in Busan, which is a heavily industrialized and densely populated city in South Korea (Jang *et al.*, 2020). There are still no regulations on PCDD/F and PCBs emissions from mobile sources, while several studies have reported the technologies to reduce their emissions.

The scooters could be considered a major contributor to traffic-related emissions in Asian cities (Chiang *et al.*, 2014; Macedo *et al.*, 2017; Hu *et al.*, 2018). There are over 350 million scooters around the world and keep increasing with the rise of the metropolitan population (Chiang *et al.*, 2014; Alves *et al.*, 2015; Chernyshev *et al.*, 2018). Among the global distribution of scooters, Asia is overriding followed by Europe, Latin America, North America, and Africa (DeMarini *et al.*, 2004; Costagliola *et al.*, 2014; Costagliola *et al.*, 2016). Scooters are more flexible and convenient, especially in urban areas where traffic is intensively crowded (Durant *et al.*, 1996, 1999). In recent years, the emission from the old model scooter was considered as the major source of air pollutants. It is expected to grow at a faster rate with no strategy to scrap off the older scooters (Alves *et al.*, 2015; TWEPA, 2020), which contributes more regulated and unregulated pollutants related to environmental and human health because of their poor combustion quality. Besides, the two-stroke scooter was not performed ideal air-fuel ratio and led to higher air pollutant emission (Prati *et al.*, 2011; Yao *et al.*, 2013; Platt *et al.*, 2014; Tsai *et al.*, 2017; Tsai *et al.*, 2020).

In Taiwan, the number of scooters slightly decreased from 15.9 million in 2010 to 14.0 million in 2019 (TWEPA, 2020). Unfortunately, the density of the resident population remained very high in the major city (e.g., 9,732 people km^{-2} in Taipei City), when the scooter density was also higher 3,232 than most of the cities around the world (as shown in Fig. 1). The raising health risk by the exposure of tailpipe emissions could be then expectable. Today, the treatment technologies for HC, CO, NO_x, VOC, and PM (Dhital *et al.*, 2019, Tsai *et al.*, 2019b) were improved along with the generations of scooter emission standards (Tier 1 to Tier 6). Fig. 1 also shows the amount of Tier 1+2+3 scooter decreased, when the Tier 4 and 5 scooters dominates the on-road vehicle emissions, leading to lower regulated pollutant levels. Fortunately, the motors fit Tier 6 and 7 (established in 2021) would increase. However, the unregulated toxic pollutants, such as PCDD/Fs and PCBs, were not showed to be simultaneously reduced by upgrading the older vehicles to the newer generations.

Therefore, this study focuses on the comparison of the PCDD/F and PCB emissions between Tier 3- and Tier 5-scooters, which have different fuel-feeding systems. Four-

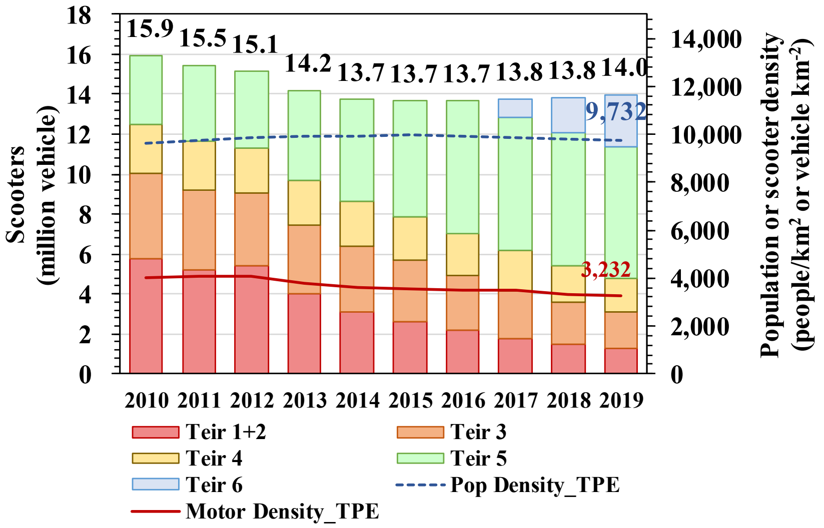

Fig. 1. The amount and density of population and various-generation scooters in Taiwan.

and two-stroke Tier 3 engines were also discussed for their different emissions. The effectivity of new fuel injection and treatment system on PCDD/F and PCB reductions were eventually confirmed.

MATERIALS AND METHODS

Tested Scooters

There were thirty scooters selected from three leading brands (over 90% market share) in Taiwan, selected as the tested vehicles in the current research. They all had 4-stroke (4-ST), 125 cm^3 displacement, and electronic fuel injection engines, which were manufactured in 2016, fitting the Tier 5 and Tier 6 emission standards (as shown in Table 1). The major improvement to approach the stricter emission standards from Tier 3 to 5 was replacing the carburetor by electronic fuel injection (EFI) system, when the on-board diagnostics with an O_2 sensor (OBD/O_2) was further added into Tier 6 scooter. These improvements would lower the pollutants in the exhaust. The testing fuels used in this study were provided by Chinese Petroleum Corporation. Their benzene, sulfur, and oxygen contents were 0.8 vol%, 10 mg kg^{-1}, and 2.7 wt%, respectively, as well as the fuel-containing aromatic and toxic substances were limited. For Tier 5 (abbr. T5) /Tier 6 (abbr. T6), the limits of CO, HC, and NOx were tightened to 2.0/1.14, 0.8/0.38 and, and 0.15/0.07 g km^{-1}, respectively, while the advanced electronic fuel injection system and on-board diagnostics (OBD) with O_2 sensor were equipped. The specifics of compared scooters selected from the previous studies were also listed in Table 1, includes six Tier 3, 4-stroke scooters (abbr. R3-4ST), and other six Tier 3, 2-stroke ones (abbr. R3-2ST). Besides, we investigated the emission effect of renewing the tailpipe from each brand of scooter. The replacement took place after the driving cycle, introduced in the following section tests for the older ones.

Dynamometer and Gaseous Pollutant Monitoring

The overall testing system for the current research could be classified into three parts, dynamometer unit, sampling unit, and vacuum system, which are illustrated in Fig. 2. The dynamometer unit is composed of an AXIS DYNO MOTO VX-12 (290 cm × 96.5 cm × 127 cm), a controlling computer, and the tested scooters. The maximum testing power, torque, and speed were 180 hp, 136 kg-m, and 262 km h^{-1}, respectively, when the system could be driven by 12 V/60 Hz electricity power.

The standard locomotive testing driving cycle in Taiwan is Urban Driving Cycle (UDC), which is a part of New European Driving Cycle (NEDC). This study further simplified the UDC to a 100-second model, which included 29.70% idling, 24.75% acceleration, 18.81% cruising, and 26.73% deceleration periods. This model operated all tested scooters, while the 100-second emission data of regulated pollutants, PCDD/F, and PCBs were simultaneously collected from an extended stack connected to the tailpipe of each scooter (as shown in Fig. 2). The gaseous pollutants were then monitored by an emission analyzer (HORIBA MEXA-584L), which used a non-dispersive infrared (NDIR) sensor for CO (0.00–10.00% vol.), HC (0–10,000 ppmv), and CO_2 (0.00–20.00% vol.), as well as a yttrium-stabilized ZrO_2 (YSZ) electrochemical sensor for NO_x (0–5,000 ppmv) and O_2 (0.00–25.00 vol%) on-line monitoring. The precisions of CO, HC, and CO_2 sensors were ±0.01% vol., ±3.3 ppmv, and 0.17% vol., respectively.

Sampling and Analyzes of PCDD/Fs and PCBs

An isokinetic flue gas sampling procedure referred to USEPA Method 23A and NIEA A808.75 was used to collect the PCDD/F and PCB samples from an extended stack connected with the scooters' tailpipes. The sampling train and vacuum system is illustrated in Fig. 2. The particulate phase of PCDD/Fs and PCBs were collected by a 70-mm quartz fiber filter paper (Pallflex®), which was pretreated by heating to 400°C for 6 hours with a high-temperature furnace and storage in electronic desiccator (47% relative humidity) for 24 hours before being weighed by an electronic balance (model CP225D, five-digit balance manufactured by Sartorius) to eliminate the interference of background medium. The

Table 1. Specifications of testing and referenced scooters.

Scooters type (amounts of samples)	Brand A (n = 4)	Brand B (n = 3)	Brand C (n = 3)	R3-4ST (n = 6)	R3-2ST (n = 6)
Engine type	4-stroke	4-stroke	4-stroke	4-stroke	2-stroke
Year of manufacture	2016	2016	2017	1998–2001	2003–2004
Displacement (cm^3)	125	125	125	50, 100, 125	50, 100
Fuel feeding system	EFI[1]	EFI[1]	EFI[1]	Carburetor	Carburetor
Mileage (km)	8,098–29,564	7,235–20,392	6,668–26,363	2,788–10,919	17,077–49,956
APCS[2]	PUFF[3] TWC[5]	PUFF[3] TWC[5]	PUFF[3] TWC[5] OBD/O_2	NFF[4]	NFF[4]
Emission standard	Tier 5	Tier 5	Tier 6	Tier 3	Tier 3
CO, gkm^{-1}	2.00	2.00	0.38	3.50	3.50
HC, gkm^{-1}	0.80	0.80	1.14	-	-
NO_x, gkm^{-1}	0.15	0.15	0.07	-	-
Data resources	This study			(Chuang *et al.*, 2010)	

[1] Electronic fuel injection; [2] Air pollution control system; [3] Non-woven fabric filter; [4] Polyurethane foam filter; [5] Three-way catalytic converters; [6] On-board diagnostics with O_2 sensor.

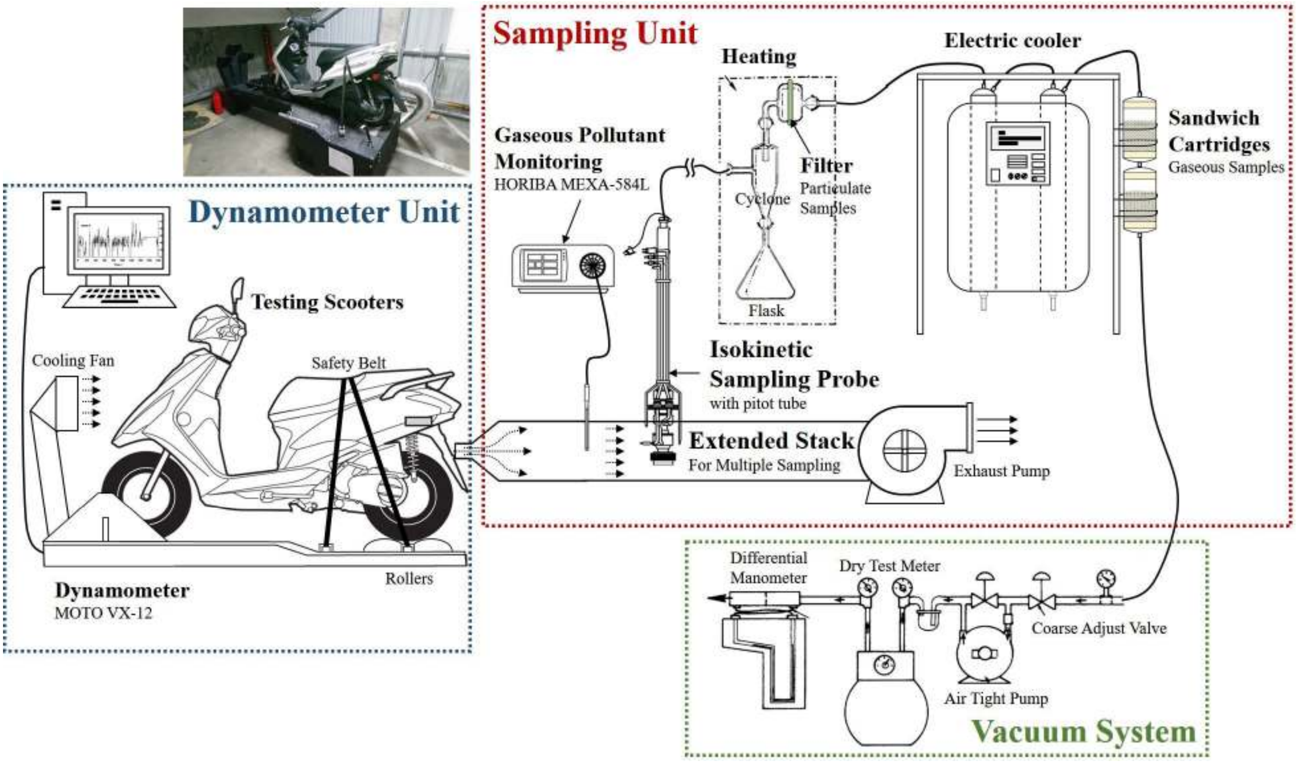

Fig. 2. Dynamometer and sampling system.

above particle collection unit was followed by an electric condenser (M&C Tech Group gas conditioning system, PSS-5/3) to remove moisture in exhaust and simultaneously reduce the temperature to 4°C to inhibit the evaporation loss of semi-volatile PCDD/Fs and PCBs. A preliminary test used a three-stage glass-cartridge adsorption unit packed with 60-g XAD-2® resin (Supelpak, Sigma-Aldrich Co., St. Louis, MO) for each cartridge to collect the gaseous PCDD/Fs and PCBs in the exhaust. Results showed that the masses of both PCDD/F and PCB congeners in the third-stage cartridge were only contributed 0.07–1.73% of the total three-cartridge adsorption unit. Therefore, a two-stage cartridge unit was used to collect the gaseous PCDD/Fs and PCBs in this study. All samples were spiked with PCDD/F and PCB surrogate standard solutions (SS) before the sample collection. Each sample was collected isokinetically from the tested scooter-dynamometer system for 60 minutes.

After the sample was transferred to the laboratory, each of them was added with the ^{13}C-labeled internal standard (IS) solution of PCDD/Fs and PCBs and later extracted by a 240-mL toluene-Soxhlet extraction for 24 hours (about 6 extraction cycles per sample). The extract was concentrated in a vacuum concentrator and transferred to a 6-dram sample tube. There were 30 µL of alternative standard (AS) solution subsequently added to the extract to estimate the recovery of analytes during the following clean-up processes. Furthermore, the sulfuric acid was added by 4 mL into the concentrate and well mixed by an ultrasonic homogenizer for 10 minutes. The solution was then transferred to an acid-silica-gel column and eluted with 20 mL of *n*-hexane. The eluent was collected, vacuum concentrated, transferred to an alumina column with *n*-hexane, and eluted by a mixture of 25-mL *n*-hexane and 15-mL dichloromethane/*n*-hexane (4/96, v/v). The non-planar PCBs passed through the column with the eluent as solution A. Furthermore, the alumina column was washed again with a 25-mL solvent composed of dichloromethane/*n*-hexane in 40/60, v/v. The eluent was collected and concentrated to the near-dry amount, transferred to the third column packed with activated carbon/diatomaceous earth, and eluted with 5 mL of toluene/methanol/ethyl acetate/n-hexane (5/5/10/80, v/v) when the planar PCBs could be eluted out as solution B. On the other hand, the PCDD/Fs were eventually washed out by eluting 40-mL toluene through the third packed column into solution C. the solution A and B were then mixed and added with 15-µL PCBs recovery standard (RS) solution, while the RS of PCDD/Fs was added into solution C before the instrumental analysis to evaluate the recovery process (EAL, 2019).

High-resolution gas chromatography coupled with high-resolution mass spectrometry (HRGC/HRMS) was employed to quantify the PCDD/F and PCB compounds. The HRGC (Hewlett-Packard 6970, CA) equipped a DB-5 column (L: 60 m, id: 0.25 mm, film thickness: 0.25 µm, J&W Scientific, CA) connected to the auto-injection port and liner. The oven temperature was set at 413 K for a minute and then raised to 473, 503, and 583 K at the rates of 30, 1.5, and 4 K min^{-1}, respectively, and hold for 4 minutes. The column again heated to 588 K at a rate of 20 K min^{-1} and held for the last 3.5 minutes for following ionization process. The HRMS (Micromass Autospec Ultima, Manchester, UK) with a positive electron impact source was operated at 523 K and 35 eV. The selected ion monitoring (SIM) mode was applied with > 10,000 resolution power to quantify the PCDD/F and PCB samples accurately (Lin *et al.*, 2019a).

RESULTS AND DISCUSSION

Transient Emissions of Pollutants from Scooters

The emissions of scooters are continuous during their operation. The emission concentration (or rate) would vary with the different engine speeds, that a transient cycle test is essential to be utilized for emission inspection. Fig. 3 shows the transient averaging emission concentrations of CO, HC, and NO_x from Tier 6 scooters, which has been expected to have the lowest emissions in the market so far. The idling period at the beginning of the 100-second cycle kept the CO, HC, and NO_x emission by 0.07%, 118, and 47 ppm, respectively, showing the relatively low emission from the steady engine speed. This observation did not conflict with the idling emission control strategy around the world, because the idling operation provides extra emission and exposure to the scooter driver nearby. Thus, the prohibition of idling operation or auto-start-stop system design is useful for emission reduction. The production of CO and HC are generally caused by incomplete combustion with unsteady air-fuel control (Yang *et al.*, 2007; Chiang *et al.*, 2014), while the oxidation rate of CO to CO_2 was sensitively affected by the local temperature in the combustion zone (Chang *et al.*, 2014). Therefore, the CO increased with the raising engine speed, which had a higher fuel injection rate and increased the equivalent ratio (Φ) of combustion and instantly lower flame temperature. The above phenomenon could further lead to both slower the thermal breakdown of residual fuel gases in the combustion area and increase the probability of the reaction among hydrocarbon radicals to form HC. That could be why CO emissions increased first and is then followed by the HC peak value. The OBD/O_2 sensor in the Tier 5 scooter could detect the abnormal Φ value and inform the next air-intake stroke (injection and air-intake valve) to stabilize the combustion. On the same time,

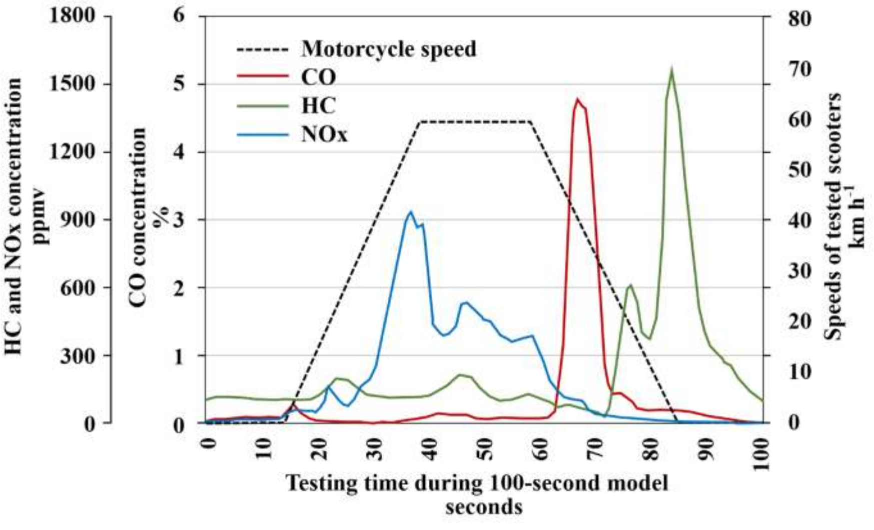

Fig. 3. Dynamic emissions of regulated gaseous pollutants along with testing cycle.

the three-way catalytic converter (TWC) was heated up by exhaust gas to its operating temperature (generally about 300–400°C), that started to convert the CO, HC, and NO_x to unharmful CO_2, H_2O, and N_2 simultaneously (Lin *et al.*, 2018). Therefore, the high emission concentration would not take long during the speed increasing period. Notably, the CO and HC occurred relatively high values during the slow-down period. The sudden release of the accelerator would quench the in-cylinder temperature and result for CO and HC productions. This period could also significantly produce PM, VOC, and SVOC, which include PCDD/Fs and PCBs.

PCDD/F and PCB concentrations

The mean PCDD/F concentrations of the brand-A and brand-B scooters (Tier 5, 4-stroke; abbr. T5-4ST) were 31.7 and 25.7 pg Nm^{-3}, respectively, as shown in Table 2. These PCDD/F mass emissions were 97.3–97.8% and 96.5–97.2% lower than those from 2-stroke (avg. 1170 pg Nm^{-3}) and 4-stroke (avg. 912 pg Nm^{-3}) scooters fit Tier 3 in the previous study (Chuang *et al.*, 2010). Meanwhile, the PCDD/F toxicity emissions from T5-4ST were 2.91 and 1.86 pg WHO-TEQ Nm^{-3}, showing 96.0–97.4% and 94.6–96.5% reductions from T3-2ST (72.7 WHO-TEQ Nm^{-3}) and T3-4ST (53.4 WHO-TEQ Nm^{-3}), respectively. More excitingly, the 4-stroke scooters approaching Tier 6 standard (T6-4ST) showed extremely lower mass and TEQ concentrations of PCDD/Fs by 6.68 pg Nm^{-3} and 0.314 pg WHO-TEQ Nm^{-3}, respectively, which showed 99.4 and 99.3% of mass and 99.6% and 99.4% of toxicity reductions from T3-2ST and T3-4ST, respectively.

The mean PCB concentration of the tested scooters were 433 and 351 pg Nm^{-3} for brand A and B (T5-4ST), respectively, when the level of brand C (T6-4ST) was 118 pg Nm^{-3} (as shown in Table 3). Additionally, the corresponding mean PCBs concentrations of toxicity were 0.133–0.298 pg WHO-TEQ Nm^{-3} for T5-4ST and 0.0496 pg WHO-TEQ Nm^{-3} for T6-4ST, respectively. There is almost no report on the PCB emissions from scooters. In comparison, the emission toxicity of total PCBs were 10 times lower than those of simultaneously emitted PCDD/Fs from the testing T5-4ST and T6-4ST scooters. Moreover, the sum of PCDD/F and PCB TEQ concentration from new generation motors were still much lower than those of older ones. However, the emissions of scooters were near to the ground surface, which could seriously increase the exposure of the residents in their daily life. The ongoing studies on air pollution in densely cities of Asian countries show that the two-wheel vehicles' emissions still dominated the contribution, even there were some prohibitions established in the metropolitan area (Yang *et al.*, 2005; Goel and Guttikunda, 2015; Liu *et al.*, 2017; Wu *et al.*, 2017). Therefore, the reduction of scooter emissions, especially toxic pollutants become an important and emerging issue.

As mentioned in the previous section, the fuel feeding system of Tier 3 was significantly improved by replacing the

Table 2. PCDD/F concentrations emitted from the testing and referenced scooters.

PCDD/Fs	Brand A (n = 4)		Brand B (n = 3)		Brand C (n = 3)		R3-4ST (n = 6)	R3-2ST (n = 6)	WHO_{2005}-TEFs
	Mean	SD	Mean	SD	Mean	SD	Mean	Mean	
2378-TeCDD	0.38	0.07	ND	NA	ND	1	6.19	8.30	1
12378-PeCDD	0.66	0.07	0.52	0.02	0.07	1	11.0	9.13	1
123478-HxCDD	0.25	0.01	0.22	0.01	ND	0.1	7.53	5.81	0.1
123678-HxCDD	0.68	0.05	0.37	0.16	ND	0.1	15.1	15.8	0.1
123789-HxCDD	0.47	0.03	0.46	0.17	0.09	0.1	12.9	10.8	0.1
1234678-HpCDD	2.41	0.22	2.82	0.89	0.80	0.01	81.7	115	0.01
OCDD	6.15	0.44	4.68	1.00	2.12	0.003	326	446	0.003
2378-TeCDF	2.75	0.35	1.40	0.27	0.29	0.1	37.2	50.6	0.1
12378-PeCDF	2.41	0.27	1.49	0.29	0.53	0.03	31.7	39.0	0.03
23478-PeCDF	2.57	0.04	1.51	0.04	0.32	0.3	49.0	58.9	0.3
123478-HxCDF	1.92	0.26	1.43	0.40	0.17	0.1	31.1	36.5	0.1
123678-HxCDF	1.19	0.26	1.48	0.43	0.20	0.1	31.5	39.8	0.1
123789-HxCDF	1.83	0.27	1.71	0.58	0.26	0.1	NA	0.83	0.1
234678-HxCDF	0.52	0.05	0.52	0.08	0.18	0.1	31.0	38.2	0.1
1234678-HpCDF	4.08	0.53	3.70	1.42	0.49	0.01	87.0	91.3	0.01
1234789-HpCDF	0.65	0.05	0.85	0.33	0.25	0.01	16.2	16.6	0.01
OCDF	2.79	0.09	2.59	0.90	0.92	0.003	137	187	0.003
PCDDs, pg Nm^{-3}	11.0	0.876	9.07	2.24	3.07	NA	460	612	NA
PCDFs, pg Nm^{-3}	20.7	2.17	16.7	4.74	3.61	NA	452	558	NA
PCDD/PCDF (mass)	0.531	0.403	0.54	0.47	0.85	NA	1.02	1.10	NA
Total PCDD/Fs, pg Nm^{-3}	31.7	3.51	25.8	7.59	6.68	NA	912	1170	NA
PCDDs, pg WHO-TEQ Nm^{-3}	1.20	0.15	0.66	0.06	0.08	NA	22.5	27.3	NA
PCDFs, pg WHO-TEQ Nm^{-3}	1.71	0.14	1.20	0.22	0.23	NA	30.9	45.4	NA
PCDD/PCDF (TEQ) (McKay, 2002)	0.70	1.01	0.55	0.28	0.37	NA	0.73	0.60	NA
Total TEQ, pg WHO-TEQ Nm^{-3}	2.91	0.29	1.86	0.27	0.31	NA	53.4	72.7	NA

ND: not detected; NA: not available.

Table 3. PCBs concentrations emitted from the testing and referenced scooters.

PCBs	Brand A (n = 4)		Brand B (n = 3)		Brand C (n = 3)		WHO$_{2005}$-TEFs
	Mean	SD	Mean	SD	Mean	SD	
Non-ortho-substituted PCBs							
33'44'-TeCB (PCB-77)	38.6	22.1	31.0	8.64	9.94	6.82	0.0001
344'5-TeCB (PCB-81)	3.30	20.3	6.11	7.73	0.984	1.14	0.0003
33'44'5-PeCB (PCB-126)	2.45	3.33	1.09	1.45	0.396	0.463	0.1
33'44'55'-HxCB PCB-169	1.23	2.26	0.309	0.357	0.184	0.246	0.03
Mono-ortho-substituted PCBs							
233'44'-PeCB (PCB-105)	98.9	77.2	79.0	19.4	27.3	18.9	0.00003
2344'5-PeCB (PCB-114)	9.44	5.72	4.30	5.42	1.91	2.34	0.00003
23'44'5-PeCB (PCB-118)	242	186	201	43.5	68.6	45.2	0.00003
2'344'5-PeCB (PCB-123)	16.4	13.5	13.0	11.1	1.88	2.66	0.00003
233'44'5-HxCB (PCB-156)	11.7	4.59	8.85	2.38	3.69	1.88	0.00003
233'44'5'-HxCB (PCB-157)	2.49	0.925	2.13	0.773	0.784	0.614	0.00003
23'44'55'-HxCB (PCB-167)	5.31	2.61	4.31	1.35	1.72	0.775	0.00003
(233'44'55'-HpCB (PCB-189)	0.893	0.875	0.633	0.739	0.285	0.356	0.00003
Total PCB, pgNm^{-3}	433	294	352	72.0	118	79.4	-
Total TEQ, pg WHO-TEQ Nm^{-3}	0.298	0.391	0.133	0.157	0.0496	0.0549	-

carburetor with the EFI system to accurately and precisely control the fuel injection timing and mass flow before the combustion in the power stroke. The more preferable equivalent ratios between fuel and air led to more complete combustion and lower the CO, HC, prompt NOx, as well as the PCDD/F and PCB emissions in the untreated exhaust gases. Moreover, the lower entreated pollutant level could further reduce the loading of a three-way catalytic converter (TWC) to have better performances. Nevertheless, Tier 6 (brand C) scooter showed an extremely low emission by equipping OBD/O_2 system. The frequently residual O_2 information feedback from the exhaust gas could sensitively tune the EFI system, improve its performance, and reduce the PCDD/F and PCB emission. Consequently, the PCDD/Fs and PCBs could be effectively reduced with the development of scooter generations without a specific treatment design for them. However, the mechanism of PCDD/F and PCB inhibition could not be identified, since they have two major formation pathways, including (1) high-temperature oxidation and chlorination of precursors and (2) heterogeneous reaction (*de novo* synthesis) in the post-combustion zone (Lin *et al.*, 2018). Therefore, an evaluation of their congener mass and TEQ distributions would be discussed in the following section.

Congener Profiles of PCDD/Fs and PCBs

The mass contributions of PCDD/F congeners were illustrated in Fig. 4(A). The dominant congeners in brand A and B (T5-4ST) were OCDD (18.2–19.4%), 1234678-HpCDF (12.9–14.4%), and OCDF (8.8–10.1%), when brand C (T6-4ST) had the same top three congeners, OCDD (31.8%), OCDF (13.8%), and 1234678-HpCDF (7.3%). A similar PCDD/F congener profile was observed for those of unled gasoline-fueled vehicles and diesel-fueled vehicles (U.S. EPA, 2001). Notably, the most significant change of congener profile from Tier 5 to Tier 6 scooters was the sharp increase of OCDD contributions (from 18.7 to 31.8%), while most of the lowly chlorinated PCDFs were inhibited.

The PCDD/F congener contributions of R3-4ST and R3-2ST were compared with the newer generation of scooters in Fig. 4(B). Their profiles were more similar to that of T6-ST. Highly chlorinated congeners, which could form by incompletely combustion of fuel, in the Tier 3 vehicles dominated the fingerprint of PCDD/Fs. This could be resulted by using carburetor as a fuel-feeding system. The carburetor is mechanically distributed the fuel by the intake-air velocity, which could not satisfy the various fuel demands from rapidly changing engine speeds. Therefore, the incompletely reacted aromatics were rapidly oxidized, chlorinated to form PCDD/Fs in high temperature (McKay, 2002; Stanmore, 2004) to form PCDDs. The mass ratios of PCDDs/PCDFs were then 1.10 and 10.2 for R3-4ST and R3-2ST, as well as their TEQ ratios, were 0.60 and 0.73, respectively (as shown in Table 2). These observations supported the PCDD formation mechanism from high-temperature precursor reactions (McKay, 2002).

Fortunately, the above problem could be overcome by using the EFI system in Tier 5 vehicles, providing a more complete combustion in the power stroke. However, there were traced amounts of residual soot, catalytic metals, C_2H_2 radicals, and aromatics remained in the post-combustion area. These residues could react with each other on both the inner surface of the exhaust pipe and even in TWC to form PCDFs by *de novo* synthesis (Huang and Buekens, 1995; Lin *et al.*, 2018). The latter mechanism would increase the contribution of lowly chlorinated PCDFs (4-Cl to 6-Cl substitutions) to around 39.3% in Tier 5 vehicles (as shown in Fig. 4(B)), and further affect the toxicity distribution of PCDD/F congeners. Interestingly, Tier 6 vehicles in the current study showed a congener profile very similar to those of Tier 3 vehicles, which had 175 and 234 times higher of total mass and TEQ concentrations of PCDD/Fs, respectively. The stroke number seems no significant effect. The aforementioned O_2 sensor feedback control could lead to this result in the OBD system. The TWC loading on CO, HC, and prompt NO_x treatments could be inhibited and

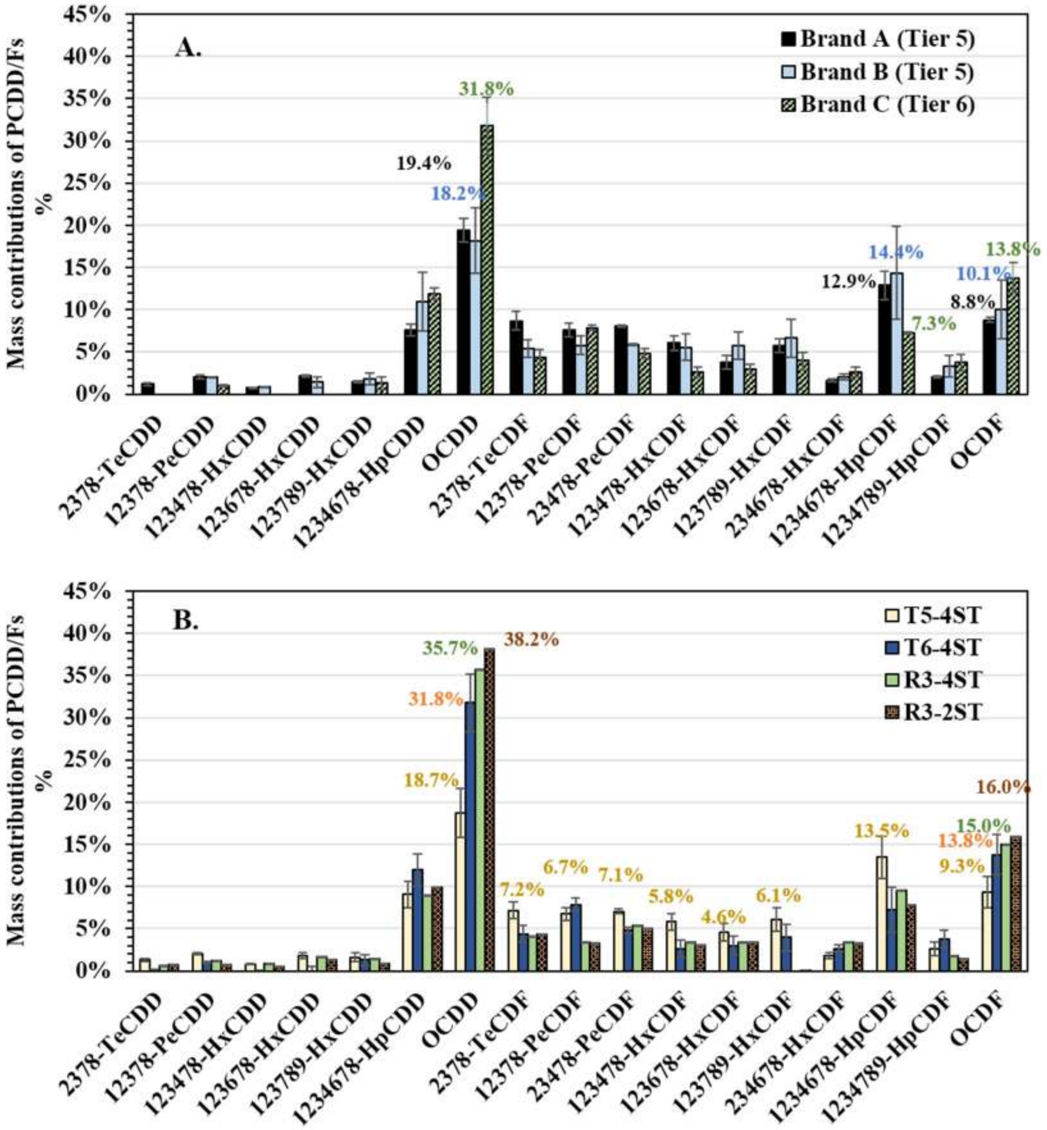

Fig. 4. Mass profiles of PCDD/F congeners from (A) three tested scooters (Tier 5 and Tier 6) and (B) their average compared with the 2-stroke and 4-stroke scooters (Tier 3 in reference).

released more capacity to oxidize the newly reformed PCDFs by *de novo* synthesis in the post-combustion zone. Therefore, the overall PCDD/F emission could be improved from Tier 5 vehicles, while the highly chlorinated PCDD/Fs predominated the congener profiles.

Fig. 5 illustrated the congener profiles of the PCB congeners. The dominant PCB congeners of Tier 5 and Tier 6 scooters were similar, when 23'44'5-PeCB (PCB-118) and 233'44'-PeCB (PCB-105) with *mono-ortho*-Cl substitutions and 33'44'-TeCB (PCB-77) with *non-ortho*-Cl substitutions contributed 55.9–58.3%, 22.5–23.2%, and 8.4–8.9% mass, respectively, of total PCBs (as shown in Fig. 5(A)). Although the most toxic *non-ortho*-congeners, 33'44'5-PeCB (PCB-126) and 33'44'55'-HxCB PCB-169, had relatively low concentrations in the Tier 5 and Tier 6 vehicle emissions, the contributions of emission toxicity among PCB congeners were still dominated by them (78.9–82.1% for PCB-126 and 6.9–12.4% for PCB-169) (as shown in Fig. 5(B)). Fortunately, all the toxicity equivalent factors of PCBs were lower than those of PCDD/Fs to reduce the toxicity contributions of them. However, the TEQ emissions of PCBs should be still involved in the calculation of real emission factors discussed in the following section.

Effects of Tailpipe Renew on PCDD/F and PCB Emissions

The particles from exhaust gases are very complicated, including metals, carbon, inorganic salts, acidic droplet, and various surfactants, which can inhibit the immune system of mammals (Zakharenko *et al.*, 2017). A recent study by Emissions Database for Global Atmospheric Research (EDGAR) showed that implementing the EURO standards led to the reduction of PM emissions in internal combustion engine exhaust by 60% worldwide (Crippa *et al.*, 2016). Therefore, the pollutants should be effectively removed before they escaped from the mobile source. The tailpipe is

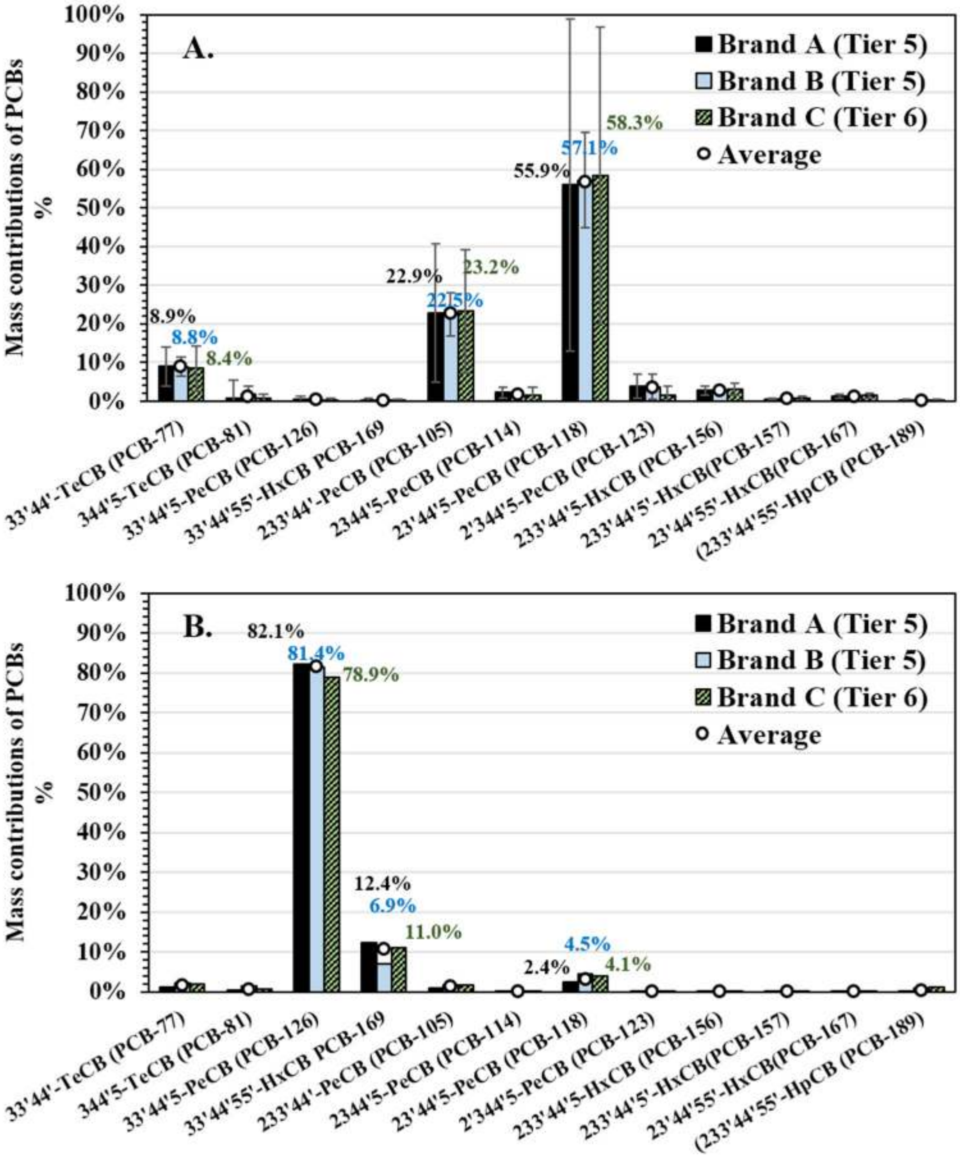

Fig. 5. (A) Mass and (B) TEQ congener profiles of PCBs in the exhausts of scooters.

the most conventional treatment technique to deal with the exhaust from scooters, when it is composed of silencer cotton, head cover, barrel body, back pressure structure, muffler cotton, tail cover, exhaust outlet, and catalyst. The particles in the exhaust might be partially trapped in the TWC converter and on the muffler cotton surface, when the exhaust gases went through the pipe. A double crisis would happen after a long-term operation of the scooters and their tailpipes. The catalytical efficiency of TWC would be inhibited by the physical and chemical poisoning, while the particles in the muffler cotton might become the carbon source for *de novo* synthesis to regenerate PCDD/Fs. Therefore, the exhaust pipe of the scooters was replaced and investigate the changes of the removal efficiencies on PCDD/Fs and PCBs emissions in this study.

Fig. 6 shows the changes of PCDD/F and PCB TEQ concentration by replacing three scooters with brand A (Tier 5), B (Tier 5), and C (Tier 6). For brand A, the PCDD/F emissions dropped 93.8% from 0.692 to 0.0431 pg WHO-TEQ km^{-1}, when the PCBs reduced 85.3% from 0.122 to 0.0179 pg WHO-TEQ km^{-1} after the recent tailpipe replacement. The significant reduction could also be found in brand B (Tier 5) scooter. Its PCDD/F emission was reduced 60.0% from 0.0865 to 0.0346 pg WHO-TEQ km^{-1} when PCB concentrations reduced 97.7% from 0.0021 to 0.0000479 pg WHO-TEQ km^{-1}. However, the reductions were not observed after the tailpipe renew of brand C. The PCDD/F emissions increased from 0.0343 to 0.0668 pg WHO-TEQ km^{-1}, while PCBs significantly increased from 0.000511 to 0.0237 pg WHO-TEQ km^{-1}. This abnormal phenomenon might not result from the quality of the TWC converter. It is believed that the manufacturer of brand C generally apply some waterproof glue and thermal insulation paint in the new tailpipe which could be released or even reacted to generate the SVOC by the hot exhaust gases passed through. Consequently, the tailpipe renew could effectively reduce

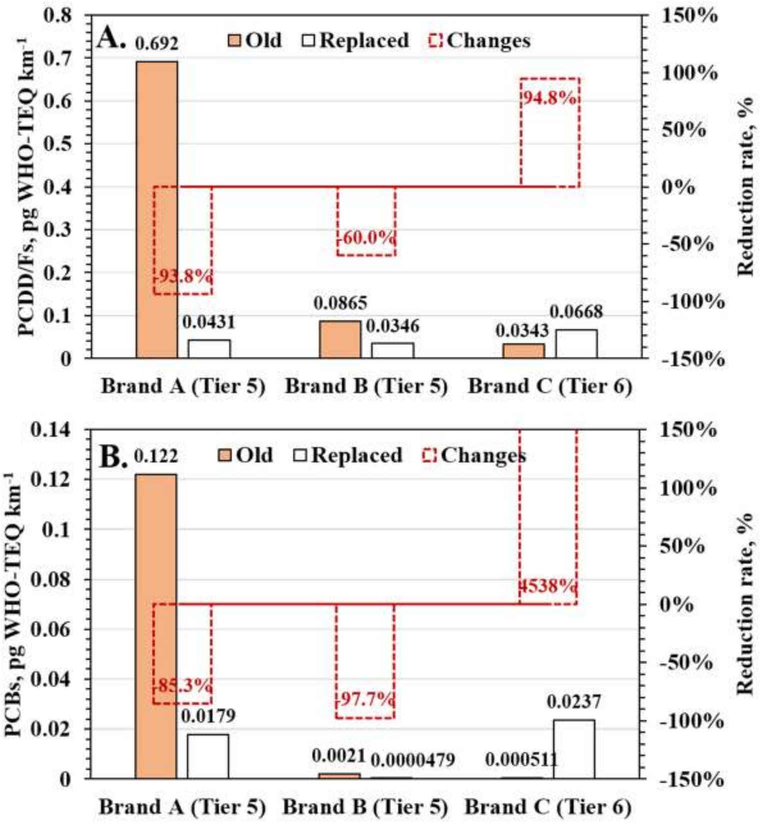

Fig. 6. Changes of (A) PCDD/F and (B) PCB emissions from scooters after the replacement of tailpipes.

the PCDD/F and PCB concentration in the exhaust, when several cases would still have a delay to perform enough removal efficiencies after a period of operation.

Emission Factors and Annual Emission Reduction

Emission factors (EFs) are normalized indexes for comparing the vehicles with various conditions, mileage, emission control equipment, feeding fuels etc. in the same base, while most of the emission standards refer to them. Table 4 showed the EFs of regulated pollutants, while the CO and NO_x emissions still excesses the limits of Tier 5 and Tier 6 standards in several cases. The trade-off between CO and NO_x emissions is always a dilemma, when the vehicle emission standards are getting stricter worldwide. The well-controlled combustion by OBD and high-quality TWC converter would be the essential solution for a new generation of scooters.

On the other hand, the EFs of total PCDD/Fs ($EF_{PCDD/Fs}$) of the R3-4ST and R3-2ST were 81.0 and 96.6 pg WHO-TEQ km^{-1}, respectively, as shown in Table 4 (Chuang *et al.*, 2010). The piston operation design in strokes led to different $EF_{PCDD/Fs}$. The 2-stroke engines had 19.3% higher $EF_{PCDD/Fs}$ than those of the 4-stroke ones, when PCDD/F concentration of R3-2ST showed 36.1% higher value than R3-4ST in tailpipe. Two-stroke scooter was one of the major sources of air pollution in many cities (Platt *et al.*, 2014), while there are scientific suggestions that scooters will emit more PAHs than all other vehicles combined in Europe by 2020 (Geivanidis *et al.*, 2008). Our finding indicated that the 4-stroke engine with separate intake and exhaust stroke could improve the combustion, decreased the incomplete oxidation of fuel, and eventually reduced the PCDD/F formation. The $EF_{PCDD/Fs}$ of T5-4ST were 1.18–1.21 pg WHO-TEQ km^{-1}, representing 98.7% reduction from R3-2ST scooters. The $EF_{PCDD/Fs}$ of T6-4ST was lower by 0.0715 pg WHO-TEQ km^{-1}, representing 99.9% reductions from R3-2ST scooters. By adding the EFs of PCBs (EF_{PCBs}) into total TEQ emission ($EF_{total} = EF_{PCDD/Fs} + EF_{PCBs}$), the EF_{total} of T5-4ST and T6-4ST became 1.30–1.34 and 0.0715 pg WHO-TEQ km^{-1}, respectively, which were still extremely lower than those from Tier 3 vehicles. That is to say, the improvement of fuel-feeding and OBE systems could not only reduce the regulated pollutants (CO, HC, and NO_x) but also could effectively reduce the PCDD/Fs emission (Yang *et al.*, 2005). The correlation between the emission factors of PCDD/Fs (or PCBs) and regulated pollutants (CO, HC, and NO_x) were not found, even the combustion condition would affect all of them simultaneously. This could result from the effective operation of the air pollution control system of the scooter.

This study estimated the annual reduction of the total PCDD/F TEQ emissions according to the following equation.

$$E_{PCDD/F-TEQ} = \sum_{1}^{n} N_i \times EF_i \times VKT_i \tag{1}$$

where $E_{PCDD/F-TEQ}$ is the annual TEQ emissions of PCDD/Fs; N_i, EF_i, and VKT_i represent the amount, TEQ emission

Table 4. PCDD/F emission factors of the testing and referenced scooters.

Tests	Brand A (n = 4)		Brand B (n = 3)		Brand C (n = 3)		R3-4ST (n = 6)		R3-2ST (n = 6)	
Engine type	4-stroke		4-stroke		4-stroke		4-stroke		2-stroke	
Year	2016		2016		2016		1999–2001		2003–2004	
Displacement (cm^3)	110–125		125		125		50, 100		50, 100, 125	
PCDDs, pg WHO-TEQ km^{-1}	0.486		0.446		0.0168		20.0		27.5	
PCDFs, pg WHO-TEQ km^{-1}	0.698		0.760		0.0449		61.0		69.1	
PCDD/Fs, pg WHO-TEQ km^{-1}	**1.18**		**1.21**		**0.0617**		**81.0**		**96.6**	
PCBs, pg WHO-TEQ km^{-1}	**0.155**		**0.0926**		**0.00977**		**-**		**-**	
Total TEQ, pg WHO-TEQ km^{-1}	**1.34**		**1.30**		**0.0715**		**81.0**		**96.6**	
Emission standard	Tier 5		Tier 5		Tier 6		Tier 3		Tier 3	
	Test	Standard	Test	Standard	Test	Standard	Test	Standard	Test	Standard
CO, gkm^{-1}	1.45–**5.22**	2.00	0.70–**3.22**	2.00	1.21–**2.12**	0.38	NA	3.50	NA	3.50
HC, gkm^{-1}	0.07–0.20	0.80	0.05–0.14	0.80	0.05–0.08	1.14	NA	-	NA	-
NO_x, gkm^{-1}	0.12–**0.26**	0.15	0.04–**0.30**	0.15	0.06–**0.09**	0.07	NA	-	NA	-
HC + NO_x, gkm^{-1}	0.19–0.42	-	0.10–**0.44**	-	0.12–0.17	-	NA	2.0	NA	2.0
Data resources	This study						(Chuang *et al.*, 2010)			

factors and the average vehicle kilometers traveled (VKT) of the scooter fit the specific Tier 1/2/3/4/5/6 standards in Taiwan. In 2019, the amounts of Tier 1 + 2, Tier 3, Tier 4, Tier 5, Tier 6-scooters were 1,279,389, 1,787,666, 1,741,999, 6,539,232, and 2,644,636 vehicles, respectively (as shown in Fig. 1). Since there is still very limit of international research on the PCDD/F and PCB emissions from scooters, the EFs of PCDD/F were then classified into only three groups in the current study. The first group used $EF_{R3\text{-}4ST}$ (81.0 pg WHO-TEQ km^{-1}), standing for the Tier 1–4 scooter emissions, when the other two used $EF_{T5\text{-}4ST}$ (1.195 pg WHO-TEQ km^{-1}) and $EF_{T6\text{-}4ST}$ (0.0617pg WHO-TEQ km^{-1}) to represent the Tier 5 and 6 emissions, respectively. The VKT of scooters reported by Taiwan Emission Database System 10.0 (TEDS 10.0) were about 3,471 to 5,070 km yr^{-1} in each administrative region. We selected an averaged VKT as 4,108 km yr^{-1} (as shown in Table 5) for further calculation (TWEPA, 2019). Finally, the original annual TEQ emissions in 2019 is then evaluated as 1.63 g WHO-TEQ, contributing only less than 2% of total annual PCDD/F TEQ emission in Taiwan. However, the scooter emission is much closer to the human life and exposure than those highly contributively stationary sources. Therefore, the annual emissions that replacing all scooters by Tier 6 levels was then estimated based on 2019 statistics. The improved annual emission became only 3.55 mg (> 99.7% reduction). This significant drop points out that the ongoing new emission standards for scooters could not only solve the problem of fine particle and regulated pollutant emissions but the potential risks from PCDD/F and PCB emissions around us, especially in the metropolitan area.

CONCLUSIONS

There were seven Tier 5 and three Tier 6 scooters tested with a 100-second transient model to analyze the PCDD/F and PCB emission and compare with those from the Tier 3 motors reported in the previous study. There are several important findings as follows.

1. PCDD/F toxicity emissions from Tier 5 and 6 scooters were 1.86–2.91 and 0.314 pg WHO-TEQ Nm^{-3}, representing 94.6–97.4% and 99.4–99.6% reductions from those of Tier 3 motors. Meanwhile, the toxicity emissions of PCBs were 0.133–0.298 and 0.0496 pg WHO-TEQ Nm^{-3} from Tier 5 and Tier 6 scooters, respectively, which were 10 times lower than those of PCDD/Fs.
2. PCDD/F were dominated by OCDD, 1234678-HpCDF, and OCDF from Tier 3 scooters, which were contained in the incompletely combusted soot and hydrocarbons. Lowly chlorinated PCDFs more contributed to Tier 5 emission, indicating the occurrence of *de novo* synthesis, when it could be inhibited by equipping OBD system in Tier 6. The PCB congeners were predominated by *mono-ortho*-PCB-118 and -105 and *non-ortho*-PCB-77 with from both Tier 5 and 6 scooters.
3. The improvement of EFI and OBD systems from a carburetor without feedback control not only reduced the regulated pollutants but also effectively reduces PCDD/F emissions.

Table 5. Estimation of annual PCDD/F TEQ emissions from scooters in both original and improved cases.

Scooter Gen.	VKT	EF	Original		Improved	
			Num. vehicle	Emission, g	Num. vehicle	Emission, mg
Tier 1-4	4108	81	4,809,054	1.60	0	0
Tier 5	4108	1.195	6,539,232	0.0321	0	0
Tier 6	4108	0.0617	2,644,636	0.000670	13,992,922	3.55
Annual Emission				1.63 g yr^{-1}		3.55 mg yr^{-1}

4. The tailpipe renew could effectively reduce 60.0–93.8% of PCDD/F and 85.3–97.7% of PCB concentrations in the exhaust, when several cases would still have a delay to perform enough removal efficiencies after a period of operation.
5. The annual TEQ emissions in 2019 is evaluated as 1.63 g WHO-TEQ and could be improved to 3.55 mg (> 99.7% reduction) by replacing all scooters by Tier 6 levels.

This is the first study that points out the emissions of PCDD/Fs and PCBs from scooters could be effectively reduced along with the regulated pollutants (CO, HC, and NO_x) by improving the fuel feeding and on-board diagnostic system. Furthermore, the exposure and health risk of human near the scooter emission could decrease in the urban area when the next generation of scooter comes.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

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