



Emissions of Polychlorinated Dibenzo-*p*-dioxin and Polychlorinated Dibenzofuran from Motorcycles

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ABSTRACT

This study presents the first investigation of polychlorinated dibenzo-*p*-dioxin and polychlorinated dibenzofuran (PCDD/F) emission from (six 2-stroke and six 4-stroke engine) motorcycles using chassis dynamometer tests. Effects of engine type and lubricant renewal on PCDD/F emission were also evaluated. The mean total PCDD/F emission concentration of tested motorcycles was 1.06 ng/Nm³, with a corresponding mean total PCDD/F I-TEQ of 0.0671 ng I-TEQ/Nm³. The mean PCDD/F emission concentration of 2-stroke engine motorcycles (1.17 ng/Nm³, 0.0727 ng I-TEQ/Nm³) was more than that (0.912 ng/Nm³, 0.0534 ng I-TEQ/Nm³) of 4-stroke engine motorcycles. The PCDD/F emission factors of motorcycles were comparable to those of some types of vehicles, although the tested motorcycles equipped with cylinders much smaller than those of vehicles. The dominant PCDD/F congeners of 2-stroke and 4-stroke motorcycles in emission priority were OCDD, OCDF, 1,2,3,4,6,7,8-HpCDD, and 1,2,3,4,6,7,8-HpCDF, accounting for 37%, 15%, 10% and 9% of the total PCDD/F emission, respectively. The reductions of PCDD/F and I-TEQ emissions after lubricant renewal of the 4-stroke motorcycles were 26%–45% and 41%–63%, respectively.

Keywords: PCDD/F; Motorcycle; Lubricant.

INTRODUCTION

Traffic transportations and industrial activities have long been associated with air pollution in the world. In Taiwan, the control of air pollution has suffered from heavy vehicular transportation: 2.1×10^7 vehicles (68% motorcycles, 30% gasoline cars and light trucks, and 0.8% heavy duty diesel vehicles) over an area of 3.6×10^4 km² (May, 2009). Motorcycles are popular in both metropolitan and suburban areas for all Taiwan residents due to shuttle mobility and reasonable price when compared to passenger cars. The density of motorcycles on the island is 385.3 vehicle/km², far transcends those of England (5.2 vehicle/km²) and Canada (0.1 vehicle/km²), and exceeds those of Singapore (211.8 vehicle/km²) and Hong Kong (46.6 vehicle/km²) (Taiwan MOTC, 2008). These motorcycles contribute more to air pollution in Taiwan than in the mentioned countries.

Recently, public concerns on air pollutant emissions from on-road vehicles have emerged and therefore pressed the authorities to impose more stringent vehicle emission standards, especially for the motorbike fleets due to their high contribution of air pollutant emission (Lin *et al.*, 2006; Lin *et al.*, 2008; Alvarez *et al.*, 2009). In general, governments hope to minimize the motorcycle pollution problem with more stringent emission standards that request initial reductions on traditional air pollutants such as HC, NO_x and CO.

Although interests in PCDD/F emissions have considerably increased in recent years, the PCDD/F emissions from on-road vehicles are still largely unexplored. Intensive studies have focused on the PCDD/F emissions from various sources, such as municipal waste, metallurgical activities and temples, due to their vast majority of the total PCDD/F contribution (Lee *et al.*, 2004; Li *et al.*, 2007; Wang and Chang-Chien, 2007; Hu *et al.*, 2009a-c; Wang *et al.*, 2010a-b). Only limited studies paid attention to traffic PCDD/F emissions (Marklund *et al.*, 1987; Bingham *et al.*, 1989; Hagenmaier *et al.*, 1990; Marklund *et al.*, 1990; Oehme *et al.*, 1991; Geueke *et al.*, 1999; U.S. EPA, 2001; Kim *et al.*, 2003), and no information on PCDD/F emission from motorcycles is

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available in literature. Because of exclusive proportion in transportation, motorcycle fleets (dominated by four-stroke engines) are considered more important in traffic and air pollution control than on-road vehicles in many countries. The vehicular PCDD/F emission factors are important for authorities to estimate the total PCDD/F emissions along with their health effects. Therefore, this study investigated the tailpipe exhausts of motorcycles to elucidate the PCDD/F emissions and the congener characteristics.

Depending on different experimental conditions (e.g., tunnel study or bench testing) and because of poor reproducibility, remarkable variation in PCDD/F level may occur for traffic emissions in different studies. In addition, some well-known factors such as engine type, the contents of fuel, catalyst installation, lubricant oil contents and renewal, and the difference of driving pattern, may affect the motorcycle emissions in tailpipe exhaust gases (Joumard *et al.*, 2000; Weilenmann *et al.*, 2005; Dyke *et al.*, 2007). In this study, emission tests for 12 motorcycles (including six 2-stroke engines and six 4-stroke engines) were performed using the regulated driving cycles to obtain the emission concentrations, emission factors, and I-TEQ data of PCDD/Fs according to their toxic equivalency factors (TEFs). This paper provides essential information for conducting inventory of PCDD/F emissions and referable discussion for setting up PCDD/F emission standards. Also, the effects of lubricant replacement on PCDD/F emission reduction were evaluated.

MATERIALS AND METHODS

Selection of Motorcycles

Test motorcycles were adopted with the priority of engine type and sale volume. This study tested 12 motorcycles including six 2-stroke engine and six 4-stroke engine in-use motorcycles. The characteristics of the tested motorcycles are provided in Table 1. The mileages of the 2- and 4-stroke engine motorcycles ranged from 2.8×10^3 to 1.1×10^4 km and from 1.7×10^4 to 5.0×10^4 km, respectively. The 2- and 4-stroke motorcycles were compliant to the Taiwan Environmental Protection Administration (TWEPA) motorcycle emission regulation Phase III (1998–2003) and Phase IV (2004–2007), respectively. In addition, each tested motorcycle was fueled with standard test gasoline and equipped with a two-way catalytic converter.

Test Procedure

The class A urban driving pattern, used for the motorcycles with the maximum velocity over 50 km/h to simulate the on-road emission status, adopts the standard test procedures similar to those of Economic Commission for Europe (ECE) cycle (Taiwan EPA, 1996). A complete test cycle (195 s) composed of the stages of idle (60 s), acceleration (42 s), cruising (57 s), and deceleration (36 s) operations. The maximum, minimum, and mean velocities were 50, 0, and 16.1 km/h, respectively.

Sampling Work

The chassis dynamometer (SCHENCK 500/GS112) is

Table 1. Mean PCDD/F concentrations (ng/Nm³) emitted from the tested motorcycles.

Engine Type	2-stroke		Ave. 2-stroke		4-stroke			Ave. 4-stroke		Mean	RSD (%)
	T1 (n = 2)	T2 (n = 2)	T3 (n = 2)	(n = 6)	F1 (n = 2)	F2 (n = 2)	F3 (n = 2)	(n = 4)	(n = 12)		
Motorcycle	2004	2003	2003		1999	2001	1998				
Year	5171–9188	2788–10919	6574–8544		36742–36753	26645–26753	17077–49956				
Mileage (km)	50	100	100		50	100	125				
Volume (cm ³)											
Concentration											
Total PCDD/Fs (ng/Nm ³)	1.25	1.07	1.18	1.17	0.823	0.908	1.01	0.912	1.06	19	
PCDDs (ng/Nm ³)	0.626	0.516	0.613	0.585	0.405	0.485	0.531	0.474	0.545	19	
PCDFs (ng/Nm ³)	0.624	0.558	0.571	0.584	0.418	0.423	0.474	0.438	0.517	22	
Total I-TEQ (ng I-TEQ/Nm ³)	0.0784	0.0699	0.0697	0.0727	0.0586	0.0443	0.0573	0.0534	0.0671	28	

located in Environmental Protection & Energy Test Lab of Automotive Research & Test Center, which is fully accredited by Taiwan EPA in executing emission tests of gasoline/diesel and alternative fuel vehicles, and the tests of middle/heavy duty diesel engines. All the tested motorcycles were fueled with a certified reference gasoline in order to minimize the interference of fuel composition variation.

Samplings and chemical analyses of PCDD/Fs for motorcycle exhaust flue gases were performed by an accredited laboratory (Super Micro Mass Research and Technology Center, Cheng Shiu University) in Taiwan. The sampling procedures followed the U.S. EPA modified Method 23. The sampling train adopted in this study was comparable with that specified by the U.S. EPA modified Method 5. All the exhaust gas samples were collected isokinetically under the aforesaid test cycles and implemented for a complete run period test.

Analysis

Prior to sampling, XAD-2 resin was spiked with PCDD/F surrogate standards prelabeled with isotopes, including $^{37}\text{C}_{14}$ -2,3,7,8-TCDD, $^{13}\text{C}_{12}$ -1,2,3,4,7,8-HxCDD, $^{13}\text{C}_{12}$ -2,3,4,7,8-PeCDF, $^{13}\text{C}_{12}$ -1,2,3,4,7,8-HxCDF, and $^{13}\text{C}_{12}$ -1,2,3,4,7,8,9-HpCDF. The recoveries of the spiked PCDD/F surrogate standards were 93.7–103% and 82.0–116% for the 2- and 4-stroke motorcycles, respectively. All the samples met the recovery criteria within 70–130%, with a relative standard deviation (RSD) less than 15%, revealing no PCDD/F breakthrough appeared. Details are similar to that given in our previous work (Wang *et al.*, 2003a).

High-resolution gas chromatograph/high-resolution mass spectrometer (HRGC/HRMS) was used for PCDD/Fs analyses. The HRGC (Hewlett-Packard 6970 Series gas chromatograph, CA) was equipped with a DB-5 fused silica capillary column (L = 60 m, i.d. = 0.25 mm, film thickness = 0.25 μm) (J&W Scientific, CA) with a splitless injection. The oven temperature program was set according to the following: beginning at 150°C (held for 1 min), followed at 30 °C/min to 220°C (held for 5 min), then at 1.5 °C/min to 240°C (held for 5 min), and finally at 1.5 °C/min to 310°C (held for 20 min). The HRMS (Micromass Autospec Ultima, Manchester, UK) mass spectrometer was equipped with a positive electron impact (EI+) source. The analysis mode of the selected ion monitoring (SIM) was used with resolving power at 10000. The electron energy and source temperature were specified at 35 eV and 250°C, respectively.

Background samples acquired from the dynamometer laboratory indoor air were simultaneously analyzed. Results indicated the PCDD/F concentration was 0.4–0.5 pg/Nm^3 and the PCDD/F I-TEQ concentration was 0.02–0.03 $\text{pg I-TEQ}/\text{Nm}^3$, revealing a low mean concentration as compared to the investigations from Lee *et al.* (2004), who found the PCDD/F concentration from the residential, traffic and industrial area, 1.4, 1.2, and 2.1 pg/Nm^3 , respectively, and the corresponding I-TEQ concentrations were 0.088, 0.073 and 0.15 $\text{pg I-TEQ}/\text{Nm}^3$,

respectively. The low PCDD/F concentrations of indoor air background suggest that the indoor air could be regarded as unpolluted by PCDD/Fs.

RESULTS AND DISCUSSION

PCDD/F Emission Concentrations from Motorcycles

The mean PCDD/F concentration of the tested motorcycles was 1.06 ng/Nm^3 (in the range 0.823–1.25 ng/Nm^3) (Table 1). The corresponding mean PCDD/F I-TEQ concentrations were 0.0671 $\text{ng I-TEQ}/\text{Nm}^3$ (ranging within 0.0443–0.0784 $\text{ng I-TEQ}/\text{Nm}^3$). For 2-stroke engines, the mean and range of PCDD/F emission concentrations were 1.17 and 1.07–1.25 ng/Nm^3 , respectively, with the corresponding mean and range of PCDD/F I-TEQ concentrations of 0.0727 and 0.0699–0.0784 $\text{ng I-TEQ}/\text{Nm}^3$, respectively. For 4-stroke engines, the mean PCDD/F emission concentration and its I-TEQ were 0.912 ng/Nm^3 (range: 0.823 to 1.01 ng/Nm^3) and 0.0534 $\text{ng I-TEQ}/\text{Nm}^3$ (range: 0.0443 to 0.0586 $\text{ng I-TEQ}/\text{Nm}^3$), respectively. Thus, the 2-stroke engine motorcycles had more PCDD/F emission than the 4-stroke engines. Some previous studies also found that the hazard potential of a 2-stroke engine was higher than that of a 4-stroke engine for the comparison of air pollutants (e.g., hydrocarbons and volatile organic compounds) emission from different engine type motorcycles (Tsai *et al.*, 2003a-b). The PCDD/F I-TEQ emission concentrations in this study are also higher than those (0.93–45 $\text{pg I-TEQ}/\text{Nm}^3$) for unleaded gasoline vehicles in literature (Marklund *et al.*, 1987; Bingham *et al.*, 1989; Hagenmaier *et al.*, 1990; Marklund *et al.*, 1990; Oehme *et al.*, 1991).

Congener Profiles and Gas/Particulate Phase Distributions

Fig. 1 shows the congener profiles of the seventeen 2,3,7,8-substituted PCDD/Fs illustrated as the signatures of motorcycle tailpipe emissions. The fraction (%) of a congener is its concentration percentage normalized by a corresponding total PCDD/F emission concentration. The 2-stroke and 4-stroke motorcycles exhibited similar 2,3,7,8-substituted PCDD/F congener profiles. The dominant PCDD/F congeners of motorcycles in emission priority were OCDD, OCDF, 1,2,3,4,6,7,8-HpCDD, and 1,2,3,4,6,7,8-HpCDF, accounting for 37, 15, 10, and 9%, respectively. A similar PCDD/F congener profile was also observed for those of unleaded gas-fueled vehicles and diesel-fueled vehicles (US EPA, 2001). The emissions of four of the tested motorcycles were surveyed to analyze the proportions (%) of gaseous and particulate PCDD/Fs (Table 2). The particle-phase PCDD/Fs dominated the emission, accounting for 71 and 68.7% of total PCDD/F and I-TEQ emissions, respectively.

PCDD/F Emission Factors

The mean PCDD/F emission factor of all tested motorcycles was 1.51 ng/km (range = 0.746–2.25 ng/km , RSD = 26.6%) corresponding to the mean emission factor of 0.0942 $\text{ng I-TEQ}/\text{km}$ (RSD = 29.2%) for the total PCDD/F I-TEQ (Table 3). For 2-stroke motorcycles, the

mean PCDD/F emission factor was 1.56 ng/km (range = 1.30–1.93 ng/km, RSD = 16.9%) and the corresponding mean PCDD/F I-TEQ emission factor was 0.0966 ng I-TEQ/km (range = 0.086–0.114 ng I-TEQ/km, RSD = 11.9%). For 4-stroke motorcycles, the mean PCDD/F and

I-TEQ emission factors were 1.41 ng/km (range = 0.746–2.25, RSD = 37.2%) and 0.081 ng I-TEQ/km (range = 0.04–0.138 ng I-TEQ/km, RSD = 46.1%), respectively. This tendency is consistent with that of motorcycle PCDD/F emission concentrations.

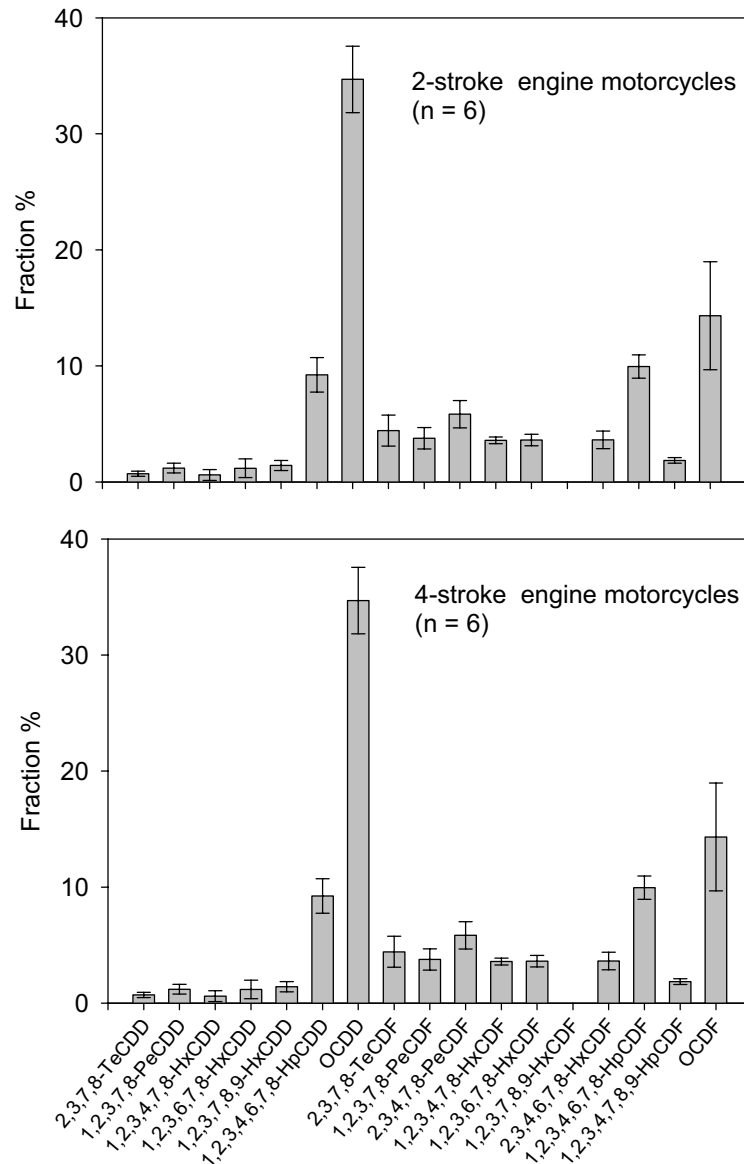


Fig. 1. Congener profiles of seventeen 2,3,7,8-PCDD/Fs in the exhausts of motorcycles.

Table 2. Proportions of gaseous and particulate PCDD/Fs emissions from the tested motorcycles.

	Fraction % (n = 4)		RSD (%)	
	Particulate	Gaseous	Particulate	Gaseous
PCDDs	31.6	19.8	21.1	94.7
PCDFs	39.4	9.13	17.5	45.5
Total PCDD/Fs	71.1	28.9	14.5	37.1
PCDDs (I-TEQ)	16.4	7.41	24.3	113
PCDFs (I-TEQ)	68.7	7.47	13.2	78.4
Total I-TEQ	85.1	14.9	64.6	64.9

Table 3. Mean PCDD/F emission factors (ng/km) with relative standard deviations (RSD, %) of the tested motorcycles.

PCDD/Fs (ng/km)	2-stroke (n = 6)		4-stroke (n = 6)		Mean (n = 12)	RSD
	Ave.	RSD	Ave.	RSD		
2,3,7,8-TeCDD	0.0106	23.9	0.010	66.7	0.0113	33.3
1,2,3,7,8-PeCDD	0.0189	34.3	0.011	115	0.0202	30.9
1,2,3,4,7,8-HxCDD	0.0129	26.4	0.007	131	0.0132	36.9
1,2,3,6,7,8-HxCDD	0.0259	21.7	0.019	92.0	0.0274	32.0
1,2,3,7,8,9-HxCDD	0.0221	30.1	0.013	125	0.0232	35.0
1,2,3,4,6,7,8-HpCDD	0.140	16.0	0.139	28.1	0.140	21.7
OCDD	0.558	24.6	0.538	50.2	0.548	37.3
2,3,7,8-TeCDF	0.0637	17.2	0.061	49.7	0.0622	34.8
1,2,3,7,8-PeCDF	0.0543	12.6	0.047	37.8	0.0508	26.4
2,3,4,7,8-PeCDF	0.0840	10.2	0.071	40.0	0.0775	27.2
1,2,3,4,7,8-HxCDF	0.0533	12.0	0.044	36.5	0.0488	25.8
1,2,3,6,7,8-HxCDF	0.0539	13.7	0.048	32.7	0.0510	23.7
1,2,3,7,8,9-HxCDF	NA	NA	0.001	245	0.00300	141
2,3,4,6,7,8-HxCDF	0.0531	16.4	0.046	29.6	0.0494	23.3
1,2,3,4,6,7,8-HpCDF	0.149	21.7	0.110	28.4	0.130	28.1
1,2,3,4,7,8,9-HpCDF	0.0277	14.5	0.020	54.1	0.0261	17.9
OCDF	0.235	32.0	0.225	63.6	0.230	47.4
Total PCDD/Fs	1.56	16.9	1.41	37.2	1.51	26.6
PCDDs/PCDFs ratio	1.01	11.9	1.10	13.7	1.08	12.9
Total I-TEQ (ng I-TEQ/km)	0.0966	11.9	0.0810	46.1	0.0942	29.2
PCDDs/PCDFs (TEQ) ratio	0.398	21.6	0.328	47.1	0.461	27.0

ND: not detected; NA: not available.

The mean ratio of the PCDD to PCDF emission factors (PCDDs/PCDFs) in the exhaust gases of motorcycles was 1.08 while the corresponding mean PCDD/F I-TEQ ratio (PCDDs/PCDFs) was smaller (0.461). Accordingly, although the mean emission factor of PCDDs was similar to that of PCDFs, the total PCDD/Fs toxicity was dominated by the equivalent toxicity of PCDFs.

Table 4 listed the PCDD/F concentrations and emission factors of several stationary and mobile sources. The PCDD/F concentrations in the exhaust of motorcycles in this study were lower than those of mostly stationary emission sources, except for power plants. However, the mean PCDD/F emission factor (94.2 pg I-TEQ/km) of motorcycles was higher than those (1.5–2.6 pg I-TEQ/km for unleaded gasoline cars and 2.4 pg I-TEQ/km for diesel cars, respectively) reported by Hagenmaier *et al.* (1990) and Hutzinger *et al.* (1992). A wide divergence in vehicular PCDD/F emission factors (0.7–2010 and 241–9500 pg/km for unleaded gasoline cars and diesel trucks, respectively) can be found in literature (Geueke *et al.*, 1999). Our data demonstrate that the PCDD/F emissions from motorcycles were comparable to those from vehicles, although motorcycles equipped with cylinders (50–125 cm³) much smaller than those of vehicles.

Effects of Lubricant Renewal on PCDD/F Emissions

It is common to use lubricant oils to keep the interior of

engine lubricated, cool and clean. However, the long-term use of lubricant oils may pollute surface of the interiors of engine cylinders and even worse wears internal combustion chambers, leading to lower engine efficiencies and more pollutant emission. Therefore, the two 4-stroke motorcycles (F1 and F3) were also tested to explore effects of lubricant renewal on PCDD/F emissions of motorcycles. After lubricant renewal, the PCDD/F emissions dropped. As expected, the reductions of total PCDD/F and I-TEQ emissions from the lubricant renewal were 26–45 and 41–63%, respectively (Table 5). Pedersen *et al.* (1980) indicated that PAH content of lubricants increased linearly with time of use, which caused a significant increase in particle-bonded PAH emission (Pedersen *et al.*, 1980). Such PAHs play the role of aromatic precursors in PCDD/F formation and that of degenerated graphitic soot structures in de novo synthesis (Gullett *et al.*, 2002). Furthermore, the accumulated heavy metals resulted from mechanical rubs in engines may also influence PCDD/F formations. Therefore, regularly replacing lubricant oil is helpful for reducing PCDD/F emission from motorcycles.

CONCLUSIONS

Twelve motorcycles (including 2- and 4-stroke engines) were tested in this study to characterize their PCDD/F emissions. The mean PCDD/F emission factor of all tested

Table 4. Comparisons between the PCDD/F emissions among different stationary and mobile sources.

Sources	PCDD/F concentration	PCDD/F emission factor	References
Municipal solid waste incinerators	0.0725 ng I-TEQ/Nm ³	0.550 µg I-TEQ/tonne-waste	Wang et al. (2007)
Sinter plants	0.995–2.06 ng I-TEQ/Nm ³ with SCR 3.10 ng I-TEQ/Nm ³ without SCR	0.970 µg I-TEQ/ton with SCR 3.13 µg I-TEQ/ton without SCR	Wang et al. (2003b)
Electric arc furnaces	0.172 ng I-TEQ/Nm ³	carbon steel EAFs: 1.6–2 µg I-TEQ/tonne-feedstock; stainless steel EAFs: 0.52 µg I-TEQ/tonne-feedstock	Wang et al. (2010)
Aluminum smelter plants	9.02 ng I-TEQ/Nm ³	50.1 µg I-TEQ/ton-feedstock	Chen et al. (2004)
Crematories	0.322–2.36 ng I-TEQ/Nm ³	6.11–13.6 µg I-TEQ/body	Wang et al. (2003a)
Power plant	0.017 ng I-TEQ/Nm ³	0.62 µg I-TEQ/tonne-coal	Lin et al. (2007)
Unleaded gasoline vehicles	–	1.5–2.6 pg I-TEQ/km	Hutzinger et al. (1992)
Diesel-fueled vehicles	–	2.4 pg I-TEQ/km	Hagenmaier et al. (1990)
Motorcycles	0.0664 ng I-TEQ/Nm ³	94.2 pg I-TEQ/km	This study

Table 5. Reductions of PCDD/F emissions from motorcycles after lubricant renewal.

Motorcycle	F1			F3		
	Used	New	Reduction (%)	Used	New	Reduction (%)
Total PCDD/Fs						
ng/Nm ³	1.058	0.587		0.916	0.68	
ng/km	1.357	0.754	45	1.211	0.899	26
Total I-TEQ						
ng I-TEQ/Nm ³	0.086	0.032		0.050	0.030	
ng I-TEQ/km	0.11	0.041	63	0.066	0.039	41

motorcycles was 1.51 ng/km and the mean I-TEQ emission factor of total PCDD/F was 0.0942 ng I-TEQ/km. The 2-stroke engines had more PCDD/F emission than the 4-stroke engines, and this trend was also true for their PCDD/F emission factors. The PCDD/F emission factors of motorcycles are comparable to those of vehicles, although the former equipped cylinders much smaller than the latter. In addition, particulate PCDD/F emission was higher than gaseous PCDD/F emission. The fraction of particle-phase PCDD/Fs were 71% and 85% of total PCDD/F and I-TEQ emissions, respectively. The dominant congeners of the 2-stroke and 4-stroke motorcycles in emission priority were OCDD, OCDF, 1,2,3,4,6,7,8-HpCDD, and 1,2,3,4,6,7,8-HpCDF. The renewal of lubricant oil is helpful for reducing PCDD/F emission from the 4-stroke engine motorcycles.

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