Emissions of Polychlorinated Dibenzo-\(p\)-dioxin and Polychlorinated Dibenzofuran from Motorcycles

Shiunn-Cheng Chuang\(^1\), Shui-Jen Chen\(^1\)*, Kuo-Lin Huang\(^1\), Guo-Ping Chang-Chien\(^2\), Lin-Chi Wang\(^2\), Yi-Chu Huang\(^1\)

\(^1\) Department of Environmental Science and Engineering, National Pingtung University of Science and Technology, 1 Shieh-Fu Rd., Nei Pu 91207, Pingtung, Taiwan

\(^2\) Department of Chemical and Materials Engineering, and Super Micro Mass Research & Technology Center, Cheng Shiu University, Kaohsiung County 833, Taiwan

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/Nm\(^3\), with a corresponding mean total PCDD/F I-TEQ of 0.0671 ng I-TEQ/Nm\(^3\). The mean PCDD/F emission concentration of 2-stroke engine motorcycles (1.17 ng/Nm\(^3\), 0.0727 ng I-TEQ/Nm\(^3\)) was more than that (0.912 ng/Nm\(^3\), 0.0534 ng I-TEQ/Nm\(^3\)) 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: \(2.1 \times 10^7\) vehicles (68% motorcycles, 30% gasoline cars and light trucks, and 0.8% heavy duty diesel vehicles) over an area of \(3.6 \times 10^4\) km\(^2\) (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/km\(^2\), far transcends those of England (5.2 vehicle/km\(^2\)) and Canada (0.1 vehicle/km\(^2\)), and exceeds those of Singapore (211.8 vehicle/km\(^2\)) and Hong Kong (46.6 vehicle/km\(^2\)) (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
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 $2.8 \times 10^3$ to $1.1 \times 10^4$ km and from $1.7 \times 10^4$ to $5.0 \times 10^4$ 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
PCDD/F Emission Concentrations from Motorcycles

The mean PCDD/F concentration of the tested motorcycles was 1.06 ng/Nm³ (in the range 0.823–1.25 ng/Nm³) (Table 1). The corresponding mean PCDD/F I-TEQ concentrations were 0.0671 ng I-TEQ/Nm³ (ranging within 0.0443–0.0784 ng I-TEQ/Nm³). For 2-stroke engines, the mean and range of PCDD/F emission concentrations were 1.17 and 1.07–1.25 ng/Nm³, respectively, with the corresponding mean and range of PCDD/F I-TEQ concentrations of 0.0727 and 0.0699–0.0784 ng I-TEQ/Nm³, respectively. For 4-stroke engines, the mean PCDD/F emission concentration and its I-TEQ were 0.912 ng/Nm³ (range: 0.823 to 1.01 ng/Nm³) and 0.0534 ng I-TEQ/Nm³ (range: 0.0443 to 0.0586 ng I-TEQ/Nm³), 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 pg I-TEQ/Nm³) 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 ng I-TEQ/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) Ave. | RSD | 4-stroke (n = 6) Ave. | RSD | Mean (n = 12) 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|                      |     | 0.0111            | 115 |
| 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-HpCDF| 0.140               | 16.0| 0.139                | 28.1| 0.140             | 21.7|
| OCDD           | 0.558                | 24.6|                      |     | 0.538             | 50.2|
| 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|                       |     | 1.51              | 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 that of power plants. The mean PCDD/F emission factor of PCDDs 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.

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/Nm³ | 0.550 μg I-TEQ/tonne-waste | Wang et al. (2007) |
| Sinter plants                 | 0.995–2.06 ng I-TEQ/Nm³ with SCR | 0.970 μg I-TEQ/ton with SCR | Wang et al. (2003b) |
| Electric arc furnaces         | 0.172 ng I-TEQ/Nm³     | 3.10 μg I-TEQ/ton without SCR | Wang et al. (2010) |
| Aluminum smelter plants       | 9.02 ng I-TEQ/Nm³      | 50.1 μg I-TEQ/ton-feedstock | Chen et al. (2004) |
| Crematories                   | 0.322–2.36 ng I-TEQ/Nm³ | 6.11–13.6 μg I-TEQ/body | Wang et al. (2003a) |
| Power plant                   | 0.017 ng I-TEQ/Nm³     | 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/Nm³   | 94.2 pg I-TEQ/km | This study               |

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

| Motorcycle | Lubricant | F1 Used | F1 New | F3 Used | F3 New | Reduction (%) | Reduction (%) |
|------------|----------|---------|--------|---------|--------|---------------|---------------|
|            |          | ng/Nm³  |        | ng/Nm³  |        | ng/km         | ng/km         |
| Motorcycles|          | 1.058   | 0.587  | 0.916   | 0.68   | 45            | 26            |
|            |          | 1.357   | 0.754  | 1.211   | 0.899  |               |               |
|            |          | 0.086   | 0.032  | 0.050   | 0.030  | 63            |               |
|            |          | 0.11    | 0.041  | 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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