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Size Distribution of Particulate Polycyclic Aromatic Hydrocarbons in the Diluted Four-stroke Motorcycle Exhausts

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ABSTRACT

Twenty-one polycyclic aromatic hydrocarbons (PAHs) from a four-stroke motorcycle were collected with a micro-orifice uniform deposit impactor and measured in size-segregated particulate samples of 0.056–18 μm aerodynamic diameter. The motorcycle exhausts were passed through a dilution tunnel to simulate reactions between ambient air and exhaust. The PAHs were analyzed using Gas Chromatography/Mass Spectrometry (GC/MS). The size distribution of particulates was unimodal with a peak at 0.1–0.18 μm. Lower molecular weight PAHs predominate in motorcycle exhaust. The emission factors increase as the particle diameter decreases for PAHs with low molecular weights. Two peaks appear at 0.056–0.1 μm and 0.18–0.32 μm for most middle and higher molecular weight PAHs. For PAHs, the cumulative fractions attributable to diameters less than 1.0 and 2.5 μm were 77.0% and 84.3% for the test motorcycle. The mass median diameter of total PAHs was 0.45 ± 0.11 μm. The BaP_{eq} emission factor for 0.056–18 μm aerodynamic diameter is 70.8 ± 38.4 ng/km for the test motorcycle. High molecular weight PAHs have the highest BaP_{eq} emission factor, indicating that higher molecular weight PAHs are more carcinogenic.

Keywords: particulate matter, four-stroke motorcycle emission, PAHs, particle size distribution, carcinogenicity

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INTRODUCTION

The motorcycle is an essential transportation mode in tropical and subtropical countries. Taiwan has more than 12 million motorcycles, accounting for 67% of all motor vehicles. The number of motorcycles is expected to increase in the future due to their convenience and mobility. Extensive research has been conducted to measure the quantity of regulated air pollutants such as CO, HC and NOx from motorcycles (Chan et al., 1995; Tsai et al., 2001; Leong et al., 2002; Chen et al., 2003). In addition to these regulated air pollutants, the unregulated constituents can be significant because of smog formation in urban areas and their potential health effects on humans. Polycyclic aromatic hydrocarbons (PAHs) are organic compounds, the derivatives of which are widespread and harmful compounds formed by incomplete combustion. Some PAHs are potential mutagens and carcinogens, and are probably a significant cause of cancer (IARC, 1987). Motor vehicles are known to represent a significant source of PAH emissions (Lim et al., 1999; Larsen et al., 2003). Most carcinogenic PAHs have been found to associate with particulates, predominately with fine particulates (Westerholm et al., 1991). A study of the bioactivity of particulate matter both *in vivo* and *in vitro* shows particulate size may affect pulmonary inflammation and allergic asthma (Dick et al., 2003). Hence, particulate size distributions need to be analyzed when assessing the possible influence of PAHs on human health.

Yang et al. (2005a) investigated the size distributions of PAHs emitted from two-stroke motorcycle engine, which are generally run with a mixture of gasoline and lubricant, causing unburnt oil to be emitted. Two-stroke motorcycles have greater emission levels than four-stroke motorcycles for most regulated air pollutants and PAHs (Chan et al., 1995; Tsai et al., 2001; Leong et al., 2002; Yang et al., 2005b). Four-stroke carburetor motorcycles are currently popular worldwide; however the particulate and PAH size distribution in four-stroke motorcycle exhaust has not been sufficiently characterized for advanced health risk assessment.

In this study, PAHs in four-stroke motorcycle exhaust were size-segregated from 0.056 μm to 18 μm with a micro-orifice uniform deposit impactor (MOUDI). The cascade impactor has been used to investigate PAH size distributions emitted from diesel and gasoline vehicles (Kleeman et al., 2000; Zielinska et al., 2004). Twenty-one PAHs, including the 16 U.S. Environmental Protection Agency (USEPA) priority PAHs, plus cyclopenta[*cd*]pyrene, perylene, benzo[*e*]pyrene, benzo[*b*]chrysene and coronene were selected as the target compounds due to their carcinogenicity. The potential human health risk associated with inhalation of PAHs of the four-stroke motorcycle exhaust was assessed.

MATERIALS AND METHODS

MOTORCYCLE AND TESTING FUEL

A four-stroke carburetor motorcycle was used to study PAH size distribution in motorcycle exhaust. This one-cylinder motorcycle had a bore and stroke of 51 mm × 60 mm, total displacement 125 mL, compression ratio 9.5, maximum power 6.0 kW/7500 rpm, and maximum torque 0.85 kgm/6000 rpm. The motorcycle was a company-owned test vehicle generally used as quality control vehicle in routine emission tests. The accumulated mileage of the test motorcycle was about 12,000 km.

Commercial fuel from the Chinese Petroleum Company, which is the most widely used unleaded gasoline in Taiwan, was used as the test fuel. Table 1 lists the properties of the fuel.

DYNAMOMETER AND DRIVING CYCLE

The test motorcycle was driven on a Schenck GS-530 GS 30 chassis dynamometer located in a certified laboratory owned by the motorcycle manufacturer. The dynamometer system consisted of a fan, a dynamometer, a dilution tunnel, a constant-volume sampler unit (HORIBA, CVS-51S), a gas analyzer (HORIBA, MEXA-8320) and a personal computer. The exhaust from the test motorcycle was passed through a dilution tunnel and the sample stream was diluted by air that had passed through a HEPA filter, a desiccant and activated carbon beds. The dilution ratio is approximately 30. The exhaust gas temperature was approximately 25-30℃ after dilution. The dilution sampler flow supplied the particulate and PAH sampling. A MOUDI was connected to the size-segregated particulate and PAH samples.

Table 1. Properties of the test fuel.

Fuel parameter	Value	Analytical method
Density (g/mL, @15℃)	0.766	ASTM D5002
Octane number	95.2	ASTM D2699
RVP (kPa)	49.7	ASTM D5191
Distillation		ASTM D86
10 vol% (℃)	59.2	
50 vol% (℃)	102.2	
90 vol% (℃)	168.3	
FBP (℃)	204.7	
Residue (vol%)	1.5	
Sulfur content (ppm wt)	86.1	ASTM D5453
Aromatics (vol%)	38.7	ASTM D4420
Benzene (vol%)	0.78	
MTBE	7.75	ASTM D5599
Total O (wt%)	1.4	ASTM D5599

The European driving cycle (ECE) is the legislated standard for automotive emission certification in Taiwan (CNS 11386). One complete test cycle (780 s) includes idle (240 s), acceleration (168 s), cruising (228 s) and deceleration (144 s). The test applies four different cruising speeds (15, 32, 35 and 50 km/h). To enhance the PAH analytical sensitivity, ten continuous ECE cycles were conducted for MOUDI sampling.

SAMPLING OF PARTICULATE MATTER AND PAHS

A MOUDI (MSP Co., Model 110) was used to measure particulate matter and PAH mass size distributions. Available particle cut-size diameters of MOUDI are 0.056, 0.10, 0.18, 0.32, 0.56, 1.0, 1.8, 3.2, 5.6, 10 and 18 μm. Aluminum foil was used as collection media. The MOUDI was disassembled and cleaned before the test by rinsing with deionized water and n-hexane. A small amount of silicone-based lubricant was then applied to the O-ring to assemble and operate the instrument correctly.

PAH ANALYSIS

After sampling, the aluminum foil samples were brought to the laboratory and put in a dessicator for 8 h to eliminate moisture, and then weighed with a balance (Sartorius, Model CP225D) to determine the net mass of particulates collected. After final weighing, each PAH-containing sample was Soxhlet extracted with a mixed solvent (n-hexane 125 mL and dichloromethane 125 mL) for 24 h. The extract was then concentrated by purging with ultra-pure nitrogen to 2 mL for the cleanup procedure. The cleanup procedure was to remove pollutants which would coelute with PAHs from the gas chromatograph (GC) column. The cleanup column contained some glass wool in the bottom (I.D. = 1 cm). Seventeen grams of 6% deactivated silica gel was mixed with 60 mL n-hexane, loaded into the cleanup column, and topped with 1.5 cm of anhydrous sodium sulfate. Next, 60 mL of hexane was added to wash the sodium sulfate and the silica gel. Right before the sodium sulfate layer was exposed to the air, the elution of hexane was stopped and the eluant was discarded. The concentrated sample was then transferred onto the column, the vessel wall was rinsed twice with 2 mL hexane which was also added to the column. Next, 200 mL of 6% ethylether in hexane was added to the column and allowed to flow through the column at a rate of 3-5 mL/min. The eluant was then collected. The eluant collected from the cleanup procedure was reconcentrated to 0.5 mL with ultra-pure nitrogen. The concentrations of the following PAHs were determined: naphthalene (Nap), acenaphthylene (AcPy), acenaphthene (Acp), fluorene (Flu), phenanthrene (PA), anthracene (Ant), fluoranthene (FL), pyrene (Pyr), cyclopenta[*cd*]pyrene (CYC), benz[*a*]anthracene (BaA), chrysene (CHR), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene (BkF), benzo[*e*]pyrene (BeP), benzo[*a*]pyrene (BaP), perylene (PER),

indeno[*1,2,3,-cd*]pyrene (IND), dibenz[*a,h*]anthracene (DBA), benzo[*b*]chrysene (BbC), benzo[*ghi*]perylene (BghiP) and coronene (COR).

A gas chromatograph (Agilent 6890) with a mass selective detector (MS) (Agilent 5973N) and a computer workstation was used for the PAH analysis. This GC/MS was equipped with an Agilent capillary column (Agilent Ultra 2 - 50 m × 0.32 mm × 0.17 μm) and an Agilent 7673A automatic sampler. In this study, the injection volume was 1 μL and the ion source temperature was 310℃. The oven temperature increased from 50℃ to 100℃ at 20℃/min, 100℃ to 290℃ at 3℃/min and hold at 290℃ for 40 min. The masses of the primary and secondary PAH ions were determined by the scan mode for pure PAH standards. The PAHs were quantified using the selected ion monitoring mode. The recovery efficiency of each PAH was determined by processing a solution containing known PAH concentrations by the same experimental procedure used for the samples. The experimental results show that the recovery efficiency of PAHs varied between 75% and 94% and averaged 85%. Blank tests for PAHs were performed using the same procedure as the recovery-efficiency tests but without adding the standard solution before extraction. The detection limit of the 21 PAHs analyzed are as follows: Nap (0.74 μg/mL), AcPy (0.41 μg/mL), Acp (0.32 μg/mL), Flu (0.12 μg/mL), PA (0.06 μg/mL), Ant (0.09 μg/mL), FL (0.19 μg/mL), Pyr (0.57 μg/mL), CYC (0.05 μg/mL), BaA (0.02 μg/mL), CHR (0.09 μg/mL), BbF (0.14 μg/mL), BkF (0.04 μg/mL), BeP (0.04 μg/mL), BaP (0.02 μg/mL), PER (0.02 μg/mL), IND (0.07 μg/mL), DBA (0.15 μg/mL), BbC (0.17 μg/mL), BghiP (0.06 μg/mL) and COR (0.11 μg/mL). Analyses of field blanks including filters and PUF/XAD-16 cartridges found no significant contamination (GC/MS integrated area < detection limit).

RESULTS AND DISCUSSIONS

SIZE DISTRIBUTION OF PARTICULATE MATTER AND PAHS IN MOTORCYCLE EXHAUST

Figure 1 shows the particulate size distribution ($df/dlogd_p$ vs. d_p) in the test motorcycle exhaust. The size distribution of particulate matter was unimodal with a significant peak at 0.1–0.18 μm. The mode in this size range is recognized as accumulation mode (Friedlander et al., 2000). The accumulation mode particulates are solid carbonaceous agglomerates formed in the engine through incomplete combustion of fuel hydrocarbons (Zielinska et al., 2004; Vaaraslahti et al., 2004).

A previous study on emissions from a two-stroke motorcycle found two significant peaks in the exhaust (0.056–0.1 and 0.1–0.56 μm) (Yang et al., 2005a). Two-stroke motorcycles are run by a mixture of gasoline and lubricant. Four-stroke motorcycles use pure gasoline fuel, resulting in the dissimilitude of the particulate size distribution in emission exhaust.

Figure 2 shows the size distributions of the 21 analyzed PAHs in the tested motorcycle exhaust. The Y-axis is expressed as dE/dlogdp, where E denotes the emission factor of PAHs. The emission factors of the *i*th PAHs, EF*i* (ng/km), were calculated using the formula:

$$EFi = Wi/L \quad (1)$$

where W*i* (ng) denotes the weight of the *i*th PAHs, and L is the travel distance in kilometers (km). The PAH emission factors can be used to estimate the PAH emission inventories for various size ranges when the travel distance is available. Low molecular weight PAHs predominate in four-stroke motorcycle exhaust (Fig. 2), which is consistent with the results of analyses of gasoline and diesel vehicle exhaust (Cadle et al., 2001; Zielinska et al., 2004). The emission factors increase as the particle diameter decreases for lower molecular weight PAHs (Fig. 2). Most middle and higher molecular weight PAHs have two significant modes at 0.056–0.1 and 0.18–0.32 μm.

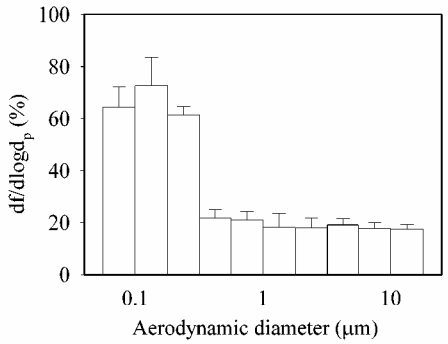

Figure 1. Size distribution of particulate matter in the test motorcycle exhaust.

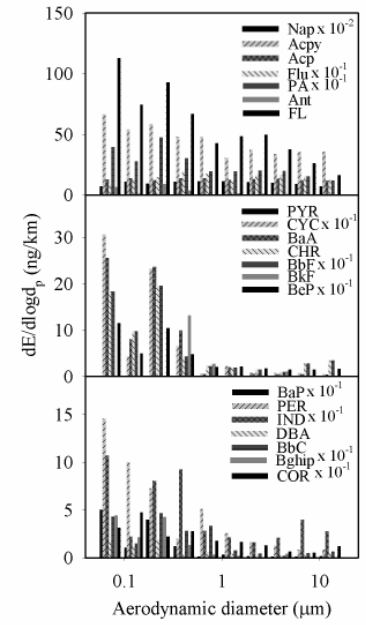

Figure 2. Size distributions of the 21 PAHs in the test motorcycle.

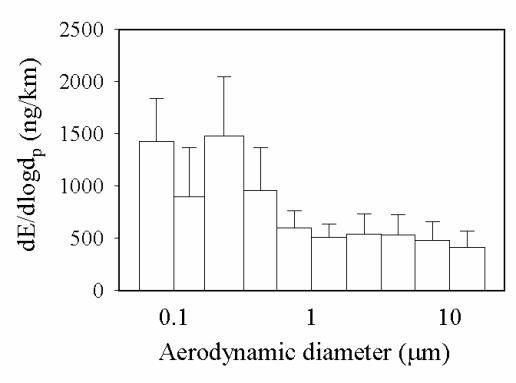

Figure 3. Size distributions of sum of 20 PAHs in motorcycle exhaust.

Figure 3 shows the sum of 20 individual PAH size distributions in the test motorcycle exhaust. Nap is excluded because it has a much higher emission rate than other species, and is recognized as volatile organic compound. Similar to most of the individual PAH size distributions in Figure 2, the size distribution of the summation of 20 PAHs shows two peaks in the 0.056–0.1 μm and 0.18–0.32 μm size ranges (Fig. 3). The two peaks seem to be caused by the noncombustion process of unburned fuel leading to the formation of PAHs primarily in the accumulation mode, and the combustion process associated with PAHs found mainly in the nuclei mode, respectively (Miguel et al., 1998).

CUMULATIVE SIZE DISTRIBUTIONS OF PARTICLES AND PAHS

The cumulative size distributions (%) of particles and PAHs are useful for comparing the contributions of fine and coarse particles and PAHs in the motorcycle exhaust, as well as in deriving the values of MMD (mass median diameter) and σ_g (geometric standard deviation). The extent to which fine particles influence human health is currently attracting significant attention. Figure 4a shows the cumulative size distributions of particles. For the four-stroke motorcycle, 78.4% and 86.5% of the mass are attributable to particles smaller than 1.0 and 2.5 μm, respectively (Fig. 4a). The results show that a great quantity of particles is in the respirable size ranges (≤ 2.5 μm), and a significant amount is in the ultrafine size ranges (≤ 1.0 μm).

To estimate the hazardous effect for different motorcycles, Figure 4a also shows the cumulative size distribution for a two-stroke motorcycle (Yang et al., 2005a). Higher percentages (89.7% and 92.8%) of the mass are attributable to particles smaller than 1.0 and 2.5 μm for the two-stroke motorcycle. For PAHs, the cumulative fractions attributable to diameters smaller than 1.0 and 2.5 μm are 77.0% and 84.3% for the four-stroke motorcycle. The large amount of PAHs in the submicron size ranges is highly

toxic and harmful to human health. High percentages (86.8% and 90.4%) of the PAHs are also attributable to particles smaller than 1.0 and 2.5 μm for the two-stroke motorcycle.

Mass median diameter (MMD) is the diameter dividing the total mass into two halves, and is obtained from the cumulative size distribution curve in Figure 4 and picked for the diameter at 50% of cumulative mass. The geometric standard deviation (σ_g) indicates the PAH size distribution. Table 2 shows the MMDs and σ_g of the 21 PAHs. The MMD values of all PAHs are 0.45 ± 0.11 μm. The MMD values of the lower molecular weight PAHs are higher than those of the higher molecular weight PAHs, indicating that higher molecular weight PAHs exist in finer particle size ranges. IARC (1987) notes that higher molecular weight PAHs are often more carcinogenic than lower molecular weight PAHs. The higher carcinogenic PAHs in the finer particles increase toxicity to humans.

Table 3. BaP_{eq} emission factors for various size ranges (ng/km).

Size ranges	LMW PAHs	MMW PAHs	HMW PAHs	Total PAHs
0.056–0.10	0.34 ± 0.12	0.87 ± 0.35	20.1 ± 8.40	21.3 ± 10.3
0.10–0.18	0.35 ± 0.08	0.14 ± 0.10	5.10 ± 2.22	5.59 ± 2.41
0.18–0.32	0.49 ± 0.13	0.76 ± 0.31	19.2 ± 11.1	20.5 ± 9.10
0.32–0.56	0.41 ± 0.10	0.20 ± 0.16	6.55 ± 3.80	7.16 ± 3.16
0.56–1.00	0.39 ± 0.09	0.03 ± 0.01	1.61 ± 0.78	2.03 ± 1.09
1.00–1.80	0.64 ± 0.08	0.07 ± 0.03	5.75 ± 1.49	6.47 ± 1.08
1.80–3.20	0.37 ± 0.04	0.04 ± 0.03	1.12 ± 0.90	1.53 ± 0.47
3.20–5.60	0.34 ± 0.03	0.03 ± 0.01	1.31 ± 1.04	1.68 ± 1.01
5.60–10.0	0.32 ± 0.03	0.03 ± 0.01	2.03 ± 0.95	2.37 ± 1.00
10.0–18.0	0.25 ± 0.08	0.02 ± 0.01	1.94 ± 1.08	2.21 ± 1.18
0.056–18.0	3.90 ± 0.95	2.19 ± 1.05	64.8 ± 25.4	70.8 ± 38.4

LMW PAHs: low molecular weight PAHs;
MMW PAHs: middle molecular weight PAHs;
HMW PAHs: high molecular weight PAHs.

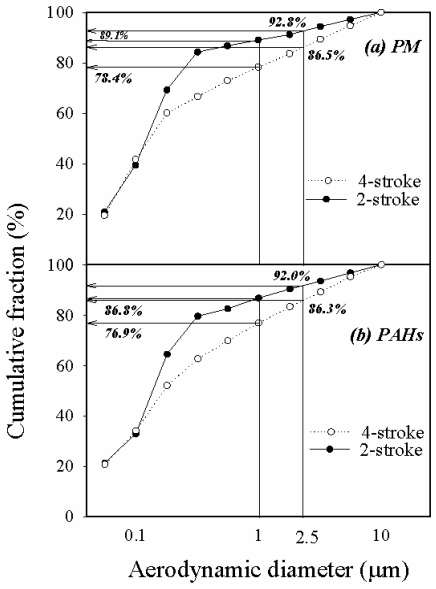

Figure 4. Cumulative size distributions of particles and PAHs.

EMISSION OF BAP EQUIVALENT CARCINOGENICITIES FOR VARIOUS SIZE RANGES

This study parameterizes the carcinogenicity of the analyzed PAHs for various size ranges by toxic equivalency factors (TEFs). TEFs were devised to compare the carcinogenic potency of the individual PAHs with the carcinogenicity of BaP (expressed as BaP_{eq}). This study used TEFs estimated by Nisbet and LaGoy (1992). BaP_{eq} emission rates for each individual PAH were computed by multiplying the emission factor with the corresponding TEF values. Table 3 shows BaP_{eq} emission factors for various size ranges. The BaP_{eq} emission factor for all size ranges was 70.8 ± 38.4 ng/km for the test motorcycle. The average emission factors of BaP_{eq} for all PAHs were much higher for particles 0.056–0.10 μm (84.7 ng/km) and 0.18–0.32 μm (72.5 ng/km) than other size ranges.

Table 3 also shows the BaP_{eq} emission factors of low molecular weight (LMW, molecular weight ≤ 178), middle molecular weight (MMW, 178 < molecular weight ≤ 228) and high molecular weight (HMW, molecular weight > 228) PAHs for various size ranges. The BaP_{eq} emission factor is highest for HMW-PAHs (64.8 ng/km). The results correlate with those found in IARC (1987), indicating that the higher molecular weight PAHs are more carcinogenic than the lower molecular weight PAHs.

CONCLUSIONS

This study used a four-stroke carburetor motorcycle to examine the PAH size distribution in the exhaust. The test motorcycle was driven on a chassis dynamometer. The exhaust gas was passed to the dilution tunnel. Particulate matter and PAH mass size distributions were measured by a MOUDI. The results indicate that lower molecular weight PAHs predominate in four-stroke motorcycle exhaust. Two significant peaks appear at 0.056–0.1 μm and 0.18–0.32 μm for most middle and higher molecular weight PAHs. The two peaks may be caused by the combustion process associated with PAHs found mainly in the nuclei mode, and the noncombustion process (unburned fuel) leading to the formation of PAHs primarily in the accumulation mode, respectively. The cumulative fractions attributable to diameters smaller than 2.5 μm are 86.5% and 84.3% for particulates and PAHs, respectively. The MMD value of total PAHs is 0.45 ± 0.11 μm. The lower molecular weight PAHs have higher MMD values than higher molecular weight PAHs, suggesting that higher molecular weight PAHs exist in finer particle size ranges. High molecular weight PAHs have a high BaP_{eq} emission factor, suggesting that the high molecular weight PAHs in emissions from motorcycles are more carcinogenic than low molecular weight PAHs.

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