Particle size distribution of polycyclic aromatic hydrocarbons in motorcycle exhaust emissions

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Abstract

The size distribution of polycyclic aromatic hydrocarbons (PAHs) in emission of a two-stroke carburetor motorcycle was studied. The exhaust gas from the test motorcycle was passed to a dilution tunnel and collected using a 10 cascade micro-orifice uniform deposit impactor (MOUDI) of 0.056–10 μm aerodynamic diameter fitted with aluminum substrates. All MOUDI substrates were analyzed for particulate mass and for PAHs by GC/MS. Most of the 21 analyzed PAHs have two significant modes that peak at <0.1 and 0.18–0.32 μm. For some PAHs, a third peak appears around 1.8 μm. MOUDI impactor samples show that 88.9% particulate and 89.6% PAH mass distributed smaller than 2.5 μm. Mass median diameters of PAHs are about 0.2 μm. Total benzo[*a*]pyrene toxic equivalency emission factor was 440 ± 13.8 ng/km for the test motorcycle. An average of 90.3% of carcinogenicity is observed in particulate smaller than 1.0 μm. The results suggest that submicron particulates predominate in the exhaust from motorcycle and exhibit high carcinogenic potency for these particulate.
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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are products of incomplete combustion, and in urban and industrial atmosphere, are almost entirely anthropogenic in origin. Most carcinogenic PAHs have been found to associate with particulate, predominately with fine particulate [1]. Some PAHs are strong carcinogens and can cause pulmonary inflammation and allergic asthma in human airway cells [2,3]. A study of the bioactivity of particulate matter both in vivo and in vitro indicates that the size of particulate may play a role in the effect on pulmonary inflammation and allergic asthma [4]. Particulates in the low-micrometer range traverse deep into lungs and inflict more death damage than that of larger particles that are arrested in the upper respiratory tract removed

by mucociliary action. Thus, it is important to analyze particulate size distributions when assessing the possible influence of PAH on human health.

Motor vehicles are proved to be a significant source of PAH emission [5,6]. Particulate emissions from motor vehicles are among the major contributors to fine particle concentrations in urban atmosphere [7–9]. Furthermore, particulate PAHs measured in roadway tunnels and in dynamometer measurements have been found in the respirable size range. In order to assess health risk, it is necessary to specify both the size distribution and the chemical composition of particulate emissions as they occur at their source. Size distributions of PAHs emitted from diesel and gasoline vehicles have been studied based on samples collected in roadway tunnels [10,11] and using a dynamometer facility [9,12]. All these studies utilized a cascade impactor as sampling device. To our knowledge, there are no data on the particulate and PAH size distribution pertaining to emissions from motorcycles in current literature.

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In many countries, the motorcycle is one of the important transportation modes. In Taiwan, for example, there are over 12 million motorcycles, which account for 67% of all motor vehicles. The health risk from motorcycle in these countries would be high. PAHs, including the 16 USEPA priority PAHs plus cyclopenta[*cd*]pyrene, perylene, benzo[*e*]pyrene, benzo[*b*]chrysene and coronene were selected as the target compounds in this study for their carcinogenicity. In this study, the 21 PAHs in particulate were size-segregated from 0.056 to 10 μm with a micro-orifice uniform deposit impactor (MOUDI) for the test motorcycle. The potential human health risk associated with inhalation of carcinogenic PAHs was assessed by considering the appropriate toxic equivalency factors (TEFs) for the analyzed PAHs.

2. Materials and methods

2.1. Motorcycle and testing

The motorcycle tested in this study for the investigation of PAH emission is a two-stroke carburetor motorcycle, which is one of the most popular motorcycle type in Taiwan. It has one cylinder and bore and stroke 40 mm × 39 mm, with a total displacement of 50 mL, maximum power 3.5 kW/7000 rpm and maximum torque 5.0 N m/6500 rpm. This motorcycle is a company-owned vehicle, which is normally used as QA/QC vehicle in routine emission test. The accumulated mileage of the test motorcycle is 15,000 km. The commercial fuel from the Chinese Petroleum Company, which was the most widely used unleaded gasoline in Taiwan, was used as the test fuel. The properties of the fuel were listed in Table 1.

The test motorcycle was driven on a Schenck GS-530 GS 30 chassis dynamometer, which was located in a certified laboratory owned by the motorcycle manufacturer. The dynamometer system comprised a fan, a dynamometer, a dilution tunnel (length is 3 m), a constant-volume sampler unit (HORIBA, CVS-51S), a gas analyzer (HORIBA, MEXA-8320) and a personal computer. The exhaust from

Table 1
Properties of the test fuel

Fuel parameter	Value	Analytical method
Density (g/mL) at 15 °C	0.766	ASTM D5002
Octane number	95.2	ASTM D2699
RVP (kPa)	49.7	ASTM D5191
Distillation		ASTM D86
10 vol% (°C)	59.2	
50 vol% (°C)	102.2	
90 vol% (°C)	168.3	
FBP (°C)	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 test motorcycle was passed to the dilution tunnel. The sample stream was diluted by air that had passed through a HEPA filter, desiccant and activated carbon beds. The dilution ratio is approximately 30. After dilution, the temperature of the exhaust gas was approximately 25–30 °C. The dilution sampler flow supplied two sampling trains. One of which (train 1) was connected to sampling bags for the analysis of regulated air pollutants (CO, NO_x) and CO_2. Sampling train 2 supplied the particulate and PAH sampling. A MOUDI was connected to sampling train 2 to size-segregate particulate and PAH samples.

The European driving cycle (ECE) is the legistive cycle used 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), and four different cruising speeds (15, 32, 35 and 50 km/h) are applied in the test. To enhance the PAH analytical sensitivity, 10 continuous ECE cycles (130 min) were conducted for MOUDI sampling. Three successful samples were collected for this study.

2.2. MOUDI

Particulate matter and PAH mass size distributions were measured with a MOUDI (MSP Co., Model 110). The MOUDI is a 10-stage cascade impactor with a flow rate of 30 L/min. 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 and 10 μm. Aluminum foils were used as collection media. Before test, the MOUDI was disassembled and cleaned by rinsing with deionized water and *n*-hexane. A small amount of silicone-based lubricant then is applied to the O-ring in order to properly assemble and operate the instrument. The cascade impactors have been adopted for PAH sampling for vehicle exhaust emission from dynamometers [9,10,12,13].

2.3. PAH analysis

After the sampling the aluminum foils were brought to the laboratory and put in a desiccator for 8 h to remove moisture. They were weighed by 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 GC column. The cleanup column was filled with 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 which was then 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 eluent

was discarded. The concentrated sample was then transferred onto the column, the wall of vessel 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, and the eluent was collected. The collected eluent 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 (GC) (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), an Agilent 7673A automatic sampler, injection volume 1 μL, splitless injection at 310 °C, ion source temperature at 310 °C, oven from 50 to 100 °C at 20 °C/min; from 100 to 290 °C at 3 °C/min and hold at 290 °C for 40 min. The masses of primary and secondary ions of PAHs were determined by using the scan mode for pure PAH standards (Merck). Quantitation of PAHs was performed by using the selected ion monitoring mode. 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). PAHs analytical procedure is described in detail elsewhere [14].

3. Results and discussions

3.1. Size distribution of particulate matter

The weights of particulate collected on the 10 stage filters of MOUDI were recorded for calculating the mass percentage of each stage to total weight. The size distribution of particulate (df/dlog d_p versus d_p) from the test motorcycle is shown in Fig. 1. MOUDI impactor samples show that 83.5 and 88.9% of the particulate mass was distributed in the sizes smaller than 1 and 2.5 μm, respectively. The particulates smaller than 2.5 μm traverse easily through the upper respiratory tract into the bronchioles and alveoli of the lungs, and pose a direct health effect. This gives important information related to health hazards of motorcycle exhaust.
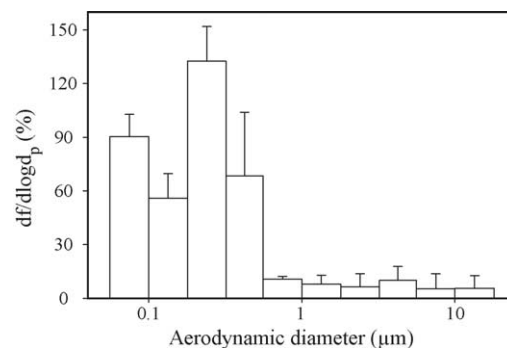

Fig. 1. Size distribution of particulate matter measured with a MOUDI.

In this study, it was found that particulates amass in the 0.1–0.56 μm diameter range. These particles are carbonaceous agglomerates loaded with adsorbed matters. The second peak of the mass distribution occurs at 0.056–0.1 μm, which is recognized as nuclei or ultra-fine mode [15]. The particles in this mode are formed during exhaust dilution and cooling, and may also include solid carbon [16]. While particles in micrometer size may be innocuous, in ultra-fine and nanosize particles are considered health hazardous [17]. In view of the adverse health effects, the reduction of ultra-fine particles emitted from motorcycles is urgent. A less-definite mode (coarse mode) between 1.8 and 10 μm diameter range was found in this study. It is possible that this mode consists of particles that have deposited on cylinder and exhaust system surfaces and later reentrained [16]. Previous studies by other researchers on emissions from diesel and gasoline vehicles have shown that the particle size distributions are bimodal [11,12] or trimodal [16]. This study shows that the particle size distribution of motorcycle exhaust is similar to those of diesel and gasoline vehicles.

3.2. Size distribution of PAHs

Twenty-one PAH size distributions (dE/dlog d_p versus d_p, E represents emission factor of PAHs) between the MOUDI stages for the test motorcycle are shown in Fig. 2. Although five continuous ECE driving was conducted in this study, some PAHs collected on the MOUDI impactor stages are below the detection limit of GC/MS, especially for larger particulates. The results in Fig. 2 are average of the three tests of the experiment. Low-molecular-weight PAHs (especially Nap) are dominating in the exhaust gas for all size ranges (Fig. 2). This observation agrees with the result obtained by the previous studies on light-duty gasoline vehicle [13] and diesel vehicle [12]. It is generally assumed that the aluminum foil has very low affinity for gaseous organic compounds, and thus is not prone to positive sampling artifacts. The result indicates that the low-molecular-weight PAHs are present in particulate phase in measurable amounts for all stages.

Most of the 21 PAHs have a major peak (in particulate) in the 0.18–0.32 μm size range (Fig. 2). A second peak is also apparent in particle size smaller than 0.1 μm
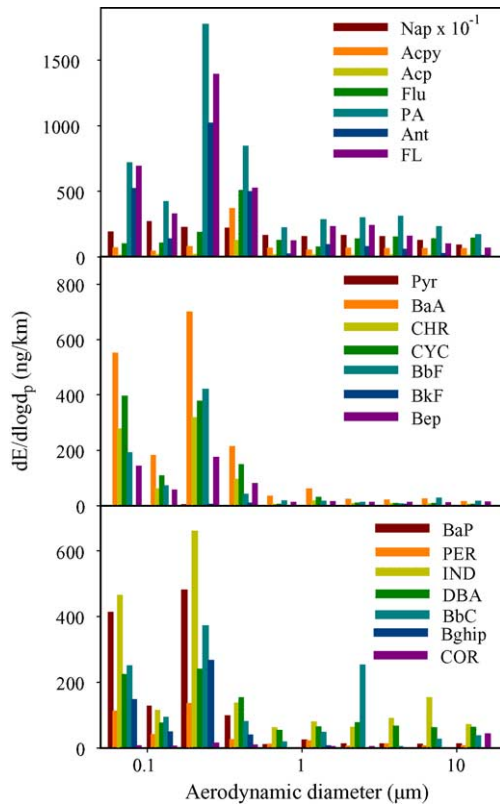
Fig. 2. Size distribution of 21 individual PAHs measured with a MOUDI.

aerodynamic diameter. It is concluded that the major peak is caused by the noncombustion process (unburnt fuel or lubricant) that leads to the formation of PAHs primarily in the accumulation mode. The second peak is resulted from the combustion process associated with PAHs that is found mainly in the nuclei mode [11]. A third mode for some PAHs occurs around 1.8 μm. The bi- and trimodal nature of the distribution is characteristic of the PAH species. The size distribution of aggregated 21 PAHs is bimodal with modes in size smaller than 0.1 μm and 0.18–0.32 μm (Fig. 3). Of the mass, 84.2 and 89.6% are attributable to particulates smaller than 1.0 and 2.5 μm, respectively, for the test motorcycle. PAHs will pose stronger toxicity and then do greater
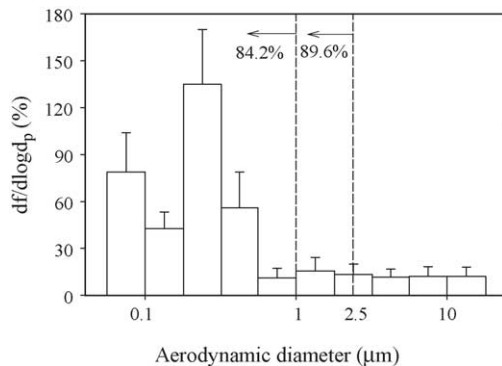

Fig. 3. Size distribution of sum of 21 PAHs measured with a MOUDI.

harm to human health when associated with submicron particles.

Mass median diameter (MMD) is the diameter dividing the total mass into two halves. The geometric standard deviation (σ_g) indicates the variability of PAHs. The MMDs and σ_g of the 21 PAHs analyzed are shown in Table 2. The MMDs of the 21 PAHs are between 0.15 and 0.42 μm and the MMD of total particulate is 0.19 μm. It is anticipated that MMD of organic compounds is smaller than that of the total particulate if a condensation mechanism prevails in forming organic particles. The MMD values of some low-molecular-weight PAHs calculated in this study were larger than that of particulate. This suggests that mechanisms other than condensation may be dominating these PAH formation.

3.3. Size distribution of BaP equivalent carcinogenicity

The particulates from motorcycles responsible for the carcinogenicity are little known. To better parameterize the carcinogenicity of the analyzed PAHs of various size ranges, TEFs are introduced in this study. These TEFs have been devised as a way of comparing the carcinogenic potency of the individual PAHs with the carcinogenicity of BaP, and expressed in terms of BaP equivalent emission (BaP$_{eq}$). There are proposals of TEFs for PAHs available currently [18–20]. In this study, TEFs estimated by Nisbet and LaGoy [18] were adopted (Table 3), as these have been demonstrated to be a better reflection of the actual state of knowledge on the toxic potency of each PAH species relative to BaP [14,21].

BaP$_{eq}$ emission for each individual PAH was calculated by multiplying emission factor with corresponding TEF values. Table 3 presents the BaP$_{eq}$ emission factors and their importance as the percentages of the sum of total emission.

Table 2
MMD (μm) and σ_g of the 21 analyzed PAHs

PAHs	MMD	σ_g
Nap	0.41	5.67
AcPy	0.39	6.47
Acp	0.39	6.09
Flu	0.42	6.23
PA	0.23	6.03
Ant	0.20	6.23
FL	0.21	2.09
Pyr	0.17	1.48
CYC	0.16	1.83
BaA	0.15	1.58
CHR	0.15	1.58
BbF	0.17	2.41
BkF	0.25	1.97
BeP	0.17	2.40
BaP	0.15	1.66
PER	0.18	2.68
IND	0.18	3.44
DBA	0.15	2.25
BbC	0.21	1.59
BghiP	0.16	1.47
COR	0.20	1.30

Table 3
BaP$_{eq}$ emission factor of the 21 PAHs for the sum of all size ranges

PAH	TEFs[a]	BaP$_{eq}$ emission factor (ng/km)	BaP$_{eq}$ (%)
Nap	0.001	4.40 ± 0.11	1.00
AcPy	0.001	0.23 ± 0.01	0.05
Acp	0.001	0.07 ± 0.03	0.02
Flu	0.001	0.42 ± 0.02	0.01
PA	0.001	1.34 ± 0.07	0.30
Ant	0.010	6.25 ± 0.38	1.42
FL	0.001	0.98 ± 0.04	0.22
Pyr	0.001	2.21 ± 0.04	0.50
CYC	–[b]	–	–
BaA	0.100	20.1 ± 0.68	4.57
CHR	0.010	2.72 ± 2.16	0.62
BbF	0.100	21.0 ± 1.04	4.78
BkF	0.100	0.59 ± 0.52	0.14
BeP	–	–	–
BaP	1.000	303 ± 12.5	68.9
PER	–	–	–
IND	0.100	48.2 ± 1.00	10.9
DBA	0.100	26.9 ± 2.63	6.10
BbC	–	–	–
BghiP	0.010	1.31 ± 0.06	0.30
COR	–	–	–
Total	–	440 ± 13.8	100

[a] Nisbet and LaGoy [18].
[b] No TEF has been suggested.

The total BaP$_{eq}$ emission factor was 440 ± 13.8 ng/km for the test motorcycle. By using TEFs, the carcinogenic properties of the individual PAHs can be assessed and compared. From the carcinogenicity point of view, BaP, IND and DBA are important compounds in emissions from motorcycles. BaP$_{eq}$ emission for various size ranges (dBaP$_{eq}$/dlog d_p versus d_p) is shown in Fig. 4. Two distinct peaks are at 0.18–0.32 and 0.056–0.1 μm, respectively. Based on BaP$_{eq}$, 90.3 and 92.7% of carcinogenicity are attributable to particulate smaller than 1.0 and 2.5 μm, respectively, for the test motorcycle. These distributions are similar to those of PAH compounds measured in this study, as shown in Fig. 3. These findings suggest that fine particulates from motorcycle are influential vectors for carrying various carcinogenic compounds into the human body.
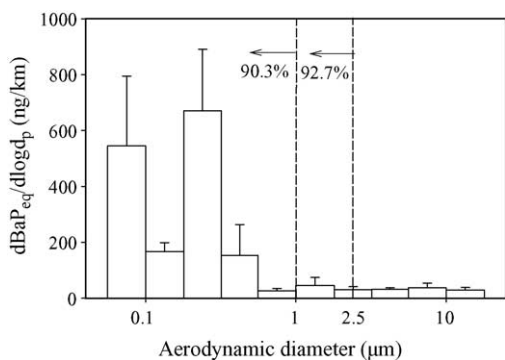

Fig. 4. Size distribution of BaP equivalent carcinogenicity.

4. Conclusions

In this study, a two-stroke carburetor motorcycle was driven on the chassis dynamometer to investigate the size distributions of particulate, PAHs and their carcinogenic potencies in the exhaust. The analyzed 21 PAHs show bi- and trimodal size distributions. The major peak occurs in the 0.18–0.32 μm size range and the second peak occurs at the aerodynamic diameter less than 0.1 μm. A third mode appears for some PAHs that locate around 1.8 μm. The MMDs of the PAHs are between 0.15 and 0.42 μm and the MMD of total particulate is 0.19 μm. The MMD values of some low-molecular-weight PAHs were larger than that of total particulates, which suggest that condensation may not be the only mechanism for PAH formation.

Of the total mass, 84.2 and 89.6% are present in particulates smaller than 1.0 and 2.5 μm, respectively, for the test motorcycle. To assess the carcinogenicity of the PAHs, BaP$_{eq}$ emission was calculated by using TEFs in this study. The BaP$_{eq}$ emission factor was 440 ± 13.8 ng/km. BaP, IND and DBA are the most important carcinogenic compounds in the exhausts from motorcycles. Based on BaP$_{eq}$, 90.3 and 92.7% of carcinogenicity are attributable to particulates smaller than 1.0 and 2.5 μm, respectively. With this apparent dominance by fine particles and for the fact that fine particles traverse deep into respiratory system, it is imperative to control motorcycle emission to alleviate the health hazards.

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