

# Seasonal Variation and Health Risk Assessment of Polycyclic Aromatic Hydrocarbons in Miaoli City, Taiwan

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**Abstract** The ambient PAHs levels in the downtown area of a traditional small city were analyzed for winter and summer seasons. A total of 16 PAHs in gaseous and particulate phase were quantified. The average gaseous PAHs were  $2,189 \pm 1,194$  and  $623.8 \pm 545.1$  ng/m<sup>3</sup> in winter and summer seasons, respectively. For the PAHs in particulate phase, they were  $40.32 \pm 12.15$  and  $11.99 \pm 5.63$  ng/m<sup>3</sup> in winter and summer seasons, respectively. These values were comparable to those reported for large cities or even higher. The estimated BaPeq was  $12.32 \pm 6.34$  ng/m<sup>3</sup>. As low-molecular-weight PAHs primarily existed in gaseous phase, high-molecular-weight PAHs in particulate phase became a significant fraction of total particulate phase PAHs. Particulate phase PAHs was significantly inversely associated with the ambient temperature for each individual PAHs species. However, this relationship did not exist for high-molecular-weight PAHs in gaseous phase. The results indicated the photo-degradation of high-molecular-weight PAHs should warrant a further thoughtfully investigation.

**Keywords** Polycyclic aromatic hydrocarbons (PAHs) · Seasonal variation · Total suspended particles (TSP) · Miaoli city

Polycyclic aromatic hydrocarbons (PAHs) are released into the environment because of incomplete combustion of fossil fuels. Some PAHs are carcinogenic and mutagenic to human beings (ATSDR 1995). Thus their levels in the ambient air are of crucial concerns. These compounds are formed with more than 2 benzene rings and classified as persistent organic compounds (ATSDR 1995). High-molecular-weight PAHs, such as benzo[a]pyrene, benzo[b]fluoranthene, and benzon[ghi]perylene, primarily exist in particulate phase in the atmosphere because of their low vapor pressure. By contrast, low-molecular-weight PAHs with higher vapor pressure are predominant in gaseous phase.

The measurements of ambient PAHs have been conducting since the 1970s from the developed countries to Asia countries (Fang et al. 2004; Hien et al. 2007; Lu et al. 2007; Motelay-Massei et al. 2005; Valerio and Lazzarotto 1985; Ruchirawat et al. 2007). The ambient PAHs levels reported varied because of meteorological conditions and sampling locations. In general, their levels are higher in industrial areas than those in non-industrial areas, and higher in urban than those in suburban (Fang et al. 2004). In addition, their levels are usually higher in winter than those in summer (Ma et al. 2011) owing to their photo-degradation (Hien et al. 2007; Lu et al. 2007; Valerio and Lazzarotto 1985). However, most of studies have been focusing on industrial regions and large cities.

In Taiwan, there are many traditional small cities with a very high population density in downtown areas. In general, the public transportation is always inconvenient in

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these cities and thus the residents must commute with their own vehicles, in particular motorcycles. Traffic is demonstrated the most important source of ambient PAHs in urban (Olson and McDow 2009). In addition, these small cities usually have a canyon-like condition in their downtown areas. This study therefore aimed to investigate the ambient PAHs levels in a small city, both in gaseous and particulate phase, and their potential health risk.

## Materials and Methods

Miaoli city is a small traditional city in Taiwan, with a population of 91,000 and an area of 38 km<sup>2</sup>, where has been classified as a good air quality region because there are very few intensive stationary sources and its meteorological condition. Thus, very few data are available on its ambient pollutant levels. This study chose a total of 20 sites locating in the downtown area, because most of its residents live near streets. A total of 35 samples (January, 2010: 20; August, 2010: 15) were sampled during daytime (10 h) for gaseous and particulate phase PAHs to estimate its traffic-related PAHs levels that may represent the residents' exposure to ambient PAHs. These samples were collected at 1.5 m above ground. The particulate PAHs were collected by a 37 mm cassette containing PTFE filter (SKC Inc, PA) at a flow rate of 2,000 mL/min and gaseous PAHs were collected by a XAD-2 tube (SKC Inc, PA) at a flow rate of 200 mL/min with a SKC air sampling pumps (SKC PCXR8, SKC Inc., PA) simultaneously. The sampling flow rates were calibrated by a calibrator (Calibrator 520, SKC Inc., PA) both in the beginning and end of sampling.

The PTFE filters were conditioned at  $25 \pm 0.2^\circ\text{C}$  and R.H.  $30 \pm 1\%$  for at least 24 h before weighing. The gaseous PAHs were extracted from XAD-2 by an ultrasonic bath (DC400H; Delta, Taiwan) with 2 mL of dichloromethane (Merck, Germany) for 10 min. The procedure was carried out in dark and repeated for three times to ensure a complete extraction. The extraction procedure for particulate PAHs was briefly described as follows. Each sample was ultrasonically extracted by 40 mL dichloromethane for 1 h and then cleaned with solvent-rinsed glass wool (Sigma-Aldrich, USA) and concentrated to about 0.5 mL. Finally, the internal standards was added (Supelco, USA), and solution volume was adjusted to 1 mL with pure dichloromethane.

Sixteen organic PAH pollutants were identified and quantified by gas chromatography/mass spectrometry (GC/MS) (GCMS-QP2010; Shimadzu, Japan). The GC/MS conditions were as follows. Injection mode was splitless with an injection volume of 1  $\mu\text{L}$ . Injection port temperature was  $280^\circ\text{C}$ ; ion source temperature was  $230^\circ\text{C}$ ; and

interface temperature was  $290^\circ\text{C}$ . The initial temperature of the oven was maintained at  $70^\circ\text{C}$  for 3 min, and then increased at  $10^\circ\text{C}/\text{min}$  to  $300^\circ\text{C}$  and maintained at  $300^\circ\text{C}$  for 20 min. Analysis was conducted in selective ion monitoring (SIM) mode. The standards of the 16 organic PAH compounds (Naphthalene (Nap), Acenaphthylene (AcPy), Acenaphthene (Acp), Fluorene (Flu), Phenanthrene (Phen), Anthracene (Ant), Fluoranthene (FL), Pyrene (Pyr), Benzo[a]anthracene (BaA), Chrysene (Chr), Benzo[b]fluoranthene (BbF), Benzo[k]fluoranthene (BkF), Benzo[a]pyrene (BaP), Indeno[1,2,3-cd]pyrene (INP), Dibenzo[a,h]anthracene (DBA), and Benzo[g,h,i]perylene (BghiP) (AccuStandard, USA) with 5 internal standards (Naphthalene-D8, Acenaphthene-D10, Phenanthrene-D10, Chrysene-D12, Perylene-D12) were analyzed in the 5–800 ng/mL range to establish standard calibration curves to determine the concentrations of these 16 PAHs collected. All correlation coefficients for calibration curves were  $>0.995$ . Instrument detection limits were  $2.06 \times 10^{-4}$ – $2.76 \times 10^{-3}$  ng, determined by threefold standard deviation of seven measurements of the lowest concentration of a calibration curve (Table 1). An appropriate amount of PAHs was added to clean blank filters and XAD2 tubes, respectively, to estimate the recovery of extractions. Mean recoveries for all compounds in gaseous and particulate phase were 62.5%–84.2% and 69.0%–95.2%, respectively. In order to estimate potential contamination during sampling, transport, storage and extraction; field blanks were conducted and analyzed. In addition, we also analyzed reagent and lab blanks. All blanks were well below detection limits.

Data analysis was conducted by IBM SPSS 18. The ambient PAHs levels in winter and summer were compared with one-way ANOVA. The correlations between variables were estimated by general linear models and simple regression models.

## Results and Discussion

The ambient levels of gaseous and particulate phase PAHs were  $2,189 \pm 1,194$  and  $40.32 \pm 12.15$  ng/m<sup>3</sup> in winter, and  $623.8 \pm 545.1$  and  $11.99 \pm 5.63$  ng/m<sup>3</sup> in summer, respectively. The total PAHs concentration in winter (2,230 ng/m<sup>3</sup>) was much higher than that in summer (635.9 ng/m<sup>3</sup>), and the individual PAH levels were detailed in Table 2. The gaseous PAHs with low-molecular-weight (2, 3-rings) were the dominant species both in winter (86.0%) and summer (95.1%) seasons. On the other hand, the particle PAHs with 2,3-ring were 43.9 and 52.0% for winter and summer, respectively. In addition, the ratio of gaseous/particulate PAHs with smaller molecular weight (less than 5-ring, except for AcPy) in winter (ranging from

**Table 1** Recovery of Spiked Samples ( $n = 3$ ) and Instrument Detection Limits of 16 PAHs ( $n = 7$ )

Compound	XAD-2 (%)	PTFE filter (%)	Detection limit (ng)
NaP	$62.5 \pm 7.9$	$69.9 \pm 19.4$	$1.19 \times 10^{-3}$
AcPy	$62.2 \pm 2.8$	$69.0 \pm 7.2$	$2.53 \times 10^{-4}$
tAcp	$63.6 \pm 2.5$	$70.7 \pm 7.0$	$1.14 \times 10^{-3}$
Flu	$67 \pm 2.6$	$74.6 \pm 12$	$4.02 \times 10^{-4}$
Phen	$77.5 \pm 12.4$	$81.4 \pm 12.0$	$2.06 \times 10^{-4}$
Ant	$78.1 \pm 12.7$	$86.3 \pm 13.3$	$3.74 \times 10^{-4}$
FL	$79.1 \pm 12.8$	$78.4 \pm 15.2$	$3.36 \times 10^{-4}$
Pyr	$78.6 \pm 12.5$	$77.4 \pm 14.2$	$7.75 \times 10^{-4}$
BaA	$82.8 \pm 12.5$	$83.9 \pm 17.0$	$7.92 \times 10^{-4}$
Chr	$78 \pm 11.2$	$75.1 \pm 14.3$	$1.04 \times 10^{-3}$
BbF	$83.5 \pm 13.2$	$88.2 \pm 7.3$	$1.31 \times 10^{-3}$
BkF	$82.1 \pm 13.1$	$88.8 \pm 5.7$	$1.51 \times 10^{-3}$
BaP	$84.2 \pm 13.8$	$92.5 \pm 8.4$	$1.83 \times 10^{-3}$
IND	$82.6 \pm 11.1$	$95.2 \pm 5.4$	$1.58 \times 10^{-3}$
DBA	$81.7 \pm 11.6$	$90.3 \pm 5.3$	$1.44 \times 10^{-3}$
B(ghi)P	$78.3 \pm 9.1$	$84.2 \pm 5.0$	$2.76 \times 10^{-3}$

9.5 to 724.0 ng/m<sup>3</sup>) was significantly higher than that in summer (range: 3.8–463.3 ng/m<sup>3</sup>). By contrast, the ratio of gaseous/particulate PAHs with a high-molecular-weight ( $\geq 5$ -ring) was greater in summer. These phenomena may be a result of more incomplete combustion occurring during motor vehicular cold start during winter than that during summer. The ambient gaseous PAHs in Miaoli city dominated during winter because this city located in subtropics and the temperature was  $20.3 \pm 2.4^\circ\text{C}$  during the sampling period. In addition, a higher ambient temperature ( $29.5 \pm 0.8^\circ\text{C}$ ) during summer may result in more vaporization of PAHs with higher molecular weight from particle matter.

The photolysis of PAHs in aerosol is demonstrated (Hien et al. 2007; Lu et al. 2007; Valerio and Lazzarotto 1985) and may accelerate at a higher temperature (Kishida et al. 2009). The half-life of PAHs in aerosol is associated with their molecular weight; in general, a shorter half-life was observed for heavier PAHs (Lu et al. 2007). For example, the half-life of BkF, BaP, IND, DBA and BghiP was shorter than 1 h where the half-lives of Flu, Phen, Pyr were 3.61, 4.62 and 2.63 h, respectively (Lu et al. 2007). Our linear regression models indicated that the particulate phase PAHs concentrations were all significantly inversely associated with temperature with an exception of Nap. These results imply the photolysis of ambient PAHs in aerosol would occur. However, only Phen, Ant, FL, Pyr in gaseous phase were still significantly inversely associated with temperature in our regression models. Thus, the decrease of particulate phase PAHs with higher molecular weight might not be completely photo-degraded, some might just evaporate to gaseous phase. In fact, among BkF, BaP, IND, DBA, BghiP, only total IND

(gaseous + particulate phase IND) was significantly inversely associated with temperature. Hence, the photolysis of gas PAHs with greater molecular weight may warrant further studies to explain their environmental fate.

Compared with the reported data for ambient PAHs levels (Table 3), our results were higher with an exception for those collected near highways. The higher observations may be a result of our samples collected during daytime, so a greater traffic density would be expected than that for a 24 h sampling period. Secondly, the population density of Miaoli city was comparable to big cities. Third, motor vehicle, especially motorcycles, was the most important transportation for residents in Miaoli city. Forth, our samples were collected at 1.5 m height from ground on canyon-like streets; while some studies collected their samples at 10 m height from ground (Fang et al. 2004). It's noteworthy that these values were comparable to or even higher than those measured in some industrial areas (Fang et al. 2004). For example, a total of 21 PAHs was 1,605 ng/m<sup>3</sup> in an industrial area of Taichung city in Taiwan (Fang et al. 2004) which was significantly lower than our values of winter (2,230 ng/m<sup>3</sup>). This may be resulted from our canyon-like sampling sites.

The sources of ambient PAHs may be identified by some specific PAH ratios. For the combustion of gasoline and diesel, the ratios of FL/(FL + PYR) and IND/(IND + BghiP) are 0.4–0.5 and 0.2–0.5, respectively (Yunker et al. 2002). In addition, the ratio of PYR/BaP can be used to identify the source of gasoline or diesel driven vehicles, and they are 2–120 and 50–100, respectively (Masclet et al. 1986). The average ratio FL/(FL + PYR) for gaseous, particulate phase and total PAH were 0.46, 0.85, and 0.76, respectively. The average ratio IND/

**Table 2** Ambient Gaseous and TSP-PAHs concentrations in Maioli City (ng/m<sup>3</sup>)

Compounds	Winter (n = 20)			Summer(n = 15)		
	Gaseous PAHs	Particle-bounds PAHs	Σ PAHs	Gaseous PAHs	Particle-bounds PAHs	Σ PAHs
(Nap)	1,683 ± 1,143	2.80 ± 1.59	1,685 ± 1,142	569.3 ± 522.7*	1.85 ± 1.11*	571.1 ± 522.4*
(AcPy)	18.27 ± 29.30	0.99 ± 0.51	19.27 ± 29.30	4.40 ± 5.56	0.16 ± 0.20*	4.21 ± 5.65
(Acp)	24.90 ± 58.54	1.73 ± 1.80	26.63 ± 58.58	4.02 ± 5.16	0.75 ± 0.41*	4.78 ± 5.16
(Flu)	38.99 ± 85.71	2.85 ± 0.86	41.84 ± 85.63	8.77 ± 10.41	1.00 ± 0.39*	9.80 ± 10.52
(PA)	34.36 ± 17.40	4.11 ± 1.82	38.47 ± 17.80	4.33 ± 3.26*	1.55 ± 0.34*	5.88 ± 3.17*
(Ant)	87.88 ± 59.98	5.22 ± 2.26	93.09 ± 59.09	3.82 ± 0.99*	1.03 ± 0.71*	3.80 ± 3.79*
(FL)	85.91 ± 93.62	3.47 ± 1.84	89.38 ± 92.83	0.96 ± 0.64*	0.53 ± 0.29*	1.50 ± 0.69*
(Pyr)	80.05 ± 92.26	2.38 ± 2.04	82.44 ± 91.69	2.36 ± 2.08	1.32 ± 0.40	3.67 ± 2.02*
(BaA)	40.22 ± 151.8	1.52 ± 0.69	41.74 ± 151.65	3.02 ± 1.94*	0.61 ± 0.43*	3.66 ± 1.86
(CHR)	35.90 ± 110.6	2.01 ± 1.72	37.91 ± 110.4	1.40 ± 1.14	0.61 ± 0.44*	2.04 ± 1.03*
(BbF)	10.04 ± 19.53	1.84 ± 0.60	11.88 ± 19.52	1.40 ± 1.14*	0.52 ± 0.99*	4.26 ± 3.57
(BkF)	14.41 ± 23.74	1.90 ± 0.84	16.31 ± 23.77	2.59 ± 2.81*	0.36 ± 0.52*	2.97 ± 2.79*
(BaP)	10.56 ± 15.36	2.04 ± 0.82	12.60 ± 15.38	5.58 ± 5.33	0.37 ± 0.53*	5.96 ± 5.22
(IND)	9.49 ± 25.07	3.55 ± 1.62	13.04 ± 25.36	3.29 ± 2.75	0.35 ± 0.70*	3.65 ± 2.84*
(DBA)	7.54 ± 17.28	1.34 ± 0.41	8.88 ± 17.28	3.77 ± 3.55	0.39 ± 0.55*	4.18 ± 3.47
B(ghi)p	8.09 ± 12.53	2.54 ± 1.00	10.62 ± 13.05	3.86 ± 2.86	0.58 ± 1.29*	4.46 ± 2.88*
Σ 2,3-ring PAHs	1,887 ± 1,212	17.71 ± 6.30	1905 ± 1212	593.2 ± 539.1	6.29 ± 2.24	599.5 ± 538.6
Σ 4-ring PAHs	241.1 ± 393.4	9.39 ± 4.78	251.5 ± 391.5	7.75 ± 3.98	3.11 ± 1.30	10.86 ± 3.47
Σ 5,6-ring PAHs	60.11 ± 88.18	13.22 ± 3.69	73.33 ± 89.25	22.80 ± 9.64	2.69 ± 3.71	25.49 ± 9.02
Σ 16-PAHs	2,189 ± 1,194	40.32 ± 12.15	2230 ± 1196	623.8 ± 545.1	11.99 ± 5.63	635.89 ± 544.79
Σ BaPeq	28.31 ± 37.80	4.32 ± 0.96	32.68 ± 37.76	11.30 ± 6.53	1.02 ± 0.86	12.32 ± 6.34
Cancer Risk	3.1 × 10 <sup>-5</sup>	4.8 × 10 <sup>-6</sup>	3.6 × 10 <sup>-5</sup>	2.3 × 10 <sup>-5</sup>	3.2 × 10 <sup>-6</sup>	1.4 × 10 <sup>-5</sup>

\* ΣPAHs = gaseous PAH + particle-PAH

(IND + BghiP) for gaseous, particulate phase and total PAH were 0.41, 0.95, and 0.95, respectively. The average ratio PYR/BaP for gaseous, particulate phase and total PAH were 23, 44, and 79, respectively. According to Miaoli government's in-time traffic monitoring system, the traffic flow and average speed were obtained for our sampling period. Thus, a linear regression model analysis was conducted to study the relationship between ambient PAHs level and traffic flow. The logarithm transformed value of ambient PAHs concentration was used in these analyses, since the ambient PAHs concentration was log-normal distribution. The results showed the ambient PAHs were significantly influenced by traffic flow. The correlation coefficients for gaseous, particulate and total PAHs, and traffic flow were 0.47 ( $p = 0.014$ ), 0.666 ( $p < 0.001$ ), and 0.47 ( $p = 0.013$ ), respectively. Based on these phenomena, the gasoline driven vehicles, in particular motorcycle, were recognized as the predominant source of ambient PAHs for Miaoli city.

Because each PAH species does not pose similar toxicity and health risk, the total ambient PAHs cannot be used to estimate their potential health risk. Thus

higher ambient PAHs levels do not represent higher health risk. In order to estimate the potential health risk owing to PAHs exposure, BaPeq value was calculated with the potency equivalency factors suggested by Nisbet and LaGoy (1992). The estimated BaPeq values were presented in Table 2. Although the ambient PAHs levels in Miaoli city were higher than that reported in many other cities, the BaPeq values estimated here were not significantly higher because Nap was the predominant PAHs species in Miaoli city, which was much less toxic than BaP. A much higher level of Nap determined here may result from collecting sample at canyon-like street sites.

In conclusion, this study pointed out the ambient PAHs levels in a small traditional city, such as Miaoli city, is comparable to major cities and should not be neglected. The downtown area in such city is usually with narrower streets and intensive activities; more importantly, often with a higher residence density in Taiwan. In addition, a narrow street with high buildings may results in a canyon-like district that may accumulate ambient pollutants and obstruct their diffusion and circulation.

**Table 3** Ambient PAHs level in different cities worldwide (ng/m<sup>3</sup>)

	Sample site	Sampling time	Σg-PAHs	Σp-PAHs	ΣPAHs*	References
Bangkok, Thailand <sup>a</sup>	Urban roadside	8 h		41.14 ± 10.17		Ruchirawat et al. (2007)
	Rural			17.00 ± 0.40		
Beijing, China <sup>a,d,f</sup>	Suburb	24 h	5,108	581.7	5,690	Zhang et al. (2009)
Beijing, China <sup>a,d,g</sup>			12,505	22.51	12,528	
Northeastern China <sup>d,f</sup>	Urban		81–240			Ma et al. (2011)
Northeastern China <sup>d,g</sup>			13–50			
Raleigh, NC, USA <sup>b,c</sup>	Highway roadside	24 h		467		Olson and McDow (2009)
	10 m			319		
	Highway roadside					
	275 m					
Taichung, Taiwan <sup>e</sup>	Urban		1,070	115	1,220 ± 520	Fang et al. 2004
Taichung, Taiwan	Rural		735	79.1	831 ± 472	
Taichung, Taiwan	Industrial		1,530	122	1,605 ± 1,240	
Ho Chi Minh City, Vietnam <sup>a,i,n</sup>	Urban	24 h		4.28 ± 2.83		Kishida et al. (2009)
Ho Chi Minh City, Vietnam <sup>a,i,o</sup>				5.71 ± 8.21		
Mioali, Taiwan <sup>a,d,f</sup>	Urban	10 h	2,189 ± 1,194	40.32 ± 12.15	2,230 ± 1,196	This study
Mioali, Taiwan <sup>a,d,g</sup>			623.8 ± 545.1	11.99 ± 5.63	635.89 ± 544.8	

\* ΣPAHs = Σg-PAHs + Σp-PAHs; <sup>a</sup> TSP; <sup>b</sup> PM<sub>2.5</sub>; <sup>c</sup> 10PAHs; <sup>d</sup> 16PAHs; <sup>e</sup> 21PAHs; <sup>f</sup> winter; <sup>g</sup> summer

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