



Characteristics, Sources, and Health Risks of Atmospheric PM_{2.5}-Bound Polycyclic Aromatic Hydrocarbons in Hsinchu, Taiwan

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ABSTRACT

This study investigated PM_{2.5}-bound polycyclic aromatic hydrocarbons (PAHs) in order to determine the seasonal changes in total benzo[*a*]pyrene equivalent (BaPeq) concentrations and to identify contamination sources by using a positive matrix factorization model, a conditional probability function, and characteristic ratios of PAHs in Hsinchu. The sampling period was from September 2014 to August 2015. PM_{2.5} samplers equipped with 47-mm quartz membrane filters were operated at a flow rate of 16.7 L min⁻¹ for 48 h. The concentrations of 20 PAHs were determined through gas chromatography–mass spectrometry. The results revealed the PM_{2.5}, total PAHs, and BaPeq mass concentrations in the four seasons ranged from 4.91 to 58.5 μg m⁻³, 0.21 to 8.08 ng m⁻³, and 0.03 to 0.78 ng m⁻³, respectively. The PM_{2.5}, total PAHs, and BaPeq mass concentrations were in the order winter > autumn > spring > summer and exhibited significant seasonal variations. The carcinogenic potency of PAHs in winter was approximately 6.21 times higher than that in summer. The major BaPeq contributors were BaP, BbF, INP, and DBA. BaP accounted for 49.0% of BaPeq concentrations in PM_{2.5} in all four seasons. The annual average lifetime excess cancer risk of PM_{2.5}-bound PAHs (1.60 × 10⁻⁵) was higher than that specified in the United States Environmental Protection Agency guidelines (10⁻⁶). The two major sources were stationary emission sources and unburned petroleum and traffic emissions, which together accounted for 90.3% of PM_{2.5}-bound PAHs.

Keywords: PM_{2.5}; Polycyclic aromatic hydrocarbon; Total benzo[*a*]pyrene equivalent; Hsinchu.

INTRODUCTION

The World Health Organization (WHO) International Agency for Research on Cancer (IARC) indicated that outdoor air pollutants and particulate matter have been identified as human carcinogens (IARC, 2013). Long-term exposure to mass concentrations of particulate matter with an aerodynamic diameter ≤ 2.5 μm (PM_{2.5}) is an important risk factor for cardiopulmonary and lung cancer mortality (Pope *et al.*, 2002). Additionally, PM_{2.5} can easily penetrate or be deposited on the alveolus zone of the lung, and these particles can adsorb numerous toxic compounds such as polycyclic aromatic hydrocarbons (PAHs), heavy metals, aldehyde, organic carbon, elemental carbon, sulfate, and nitrate. Hence, PM_{2.5} has received more academic attention in recent years because it can bind with numerous toxic

compounds and cause adverse health effects.

PAHs comprise a group of semivolatile organic pollutants containing at least two fused aromatic rings. They are primarily derived from the incomplete combustion and pyrolysis of organic substances and are produced through both natural and anthropogenic sources. Forest fires and volcanic eruptions (Kim *et al.*, 2003) produce natural PAHs, whereas the burning of coal, wood, incense, candles, cigarettes, mosquito coils, household fuel, gasoline, diesel, and the use of cooking and industrial processes produce anthropogenic PAHs (Mastral and Callen, 2000; Bzdusek *et al.*, 2004; Orecchio, 2011; Yang *et al.*, 2012; Cheruiyot *et al.*, 2015; Tiwari *et al.*, 2015; Tsai *et al.*, 2015; Wang *et al.*, 2015; Yang *et al.*, 2015). The following chemicals have been recognized as confirmed (1), probable (2A) and possible (2B) carcinogens for humans (IARC, 2016): benzo[*a*]pyrene (BaP) (1), dibenz[*a,h*]anthracene (DBA) (2A), benzo[*a*]anthracene (BaA) (2B), chrysene (Chr) (2B), benzo[*b*]fluoranthene (BbF) (2B), benzo[*k*]fluoranthene (BkF) (2B), indeno[1,2,3-*cd*]pyrene (INP) (2B), and naphthalene (Nap) (2B).

Approximately 434,060 people live in Hsinchu City, which houses Hsinchu Science Park, a major semiconductor

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manufacturing region in Taiwan. Two highways (Highways 1 and 3) pass through Hsinchu City. In addition to Hsinchu Science Park, the Hsinchu neighborhood has numerous stationary emission sources (e.g., Jiu Zhan Company factory, Asia Cement, chemical fiber plants, Hsin Tao Power, Chang Chun plant, Taiwan Glass Company factory, and Hsinchu incineration plant). In recent years, ambient PM_{2.5} mass concentrations have produced highly polluted air during winter in Taiwan. However, information on the concentrations, characteristics, and sources of ambient PM_{2.5}-bound PAHs and their associated health risks relation to seasonal change is very limited in Hsinchu area.

Hence, this study examined seasonal changes in PM_{2.5}-bound PAHs and equivalent benzo[*a*]pyrene (BaP_{eq}) concentrations in order to identify potential contamination sources by using a positive matrix factorization (PMF) model, a conditional probability function (CPF), and the characteristic ratios of PAHs in Hsinchu. The experimental results can help assess health risks, to formulate control strategies for PM_{2.5}-bound PAHs, and to develop a BaP_{eq} regulation reference.

METHODS

Sampling Sites and Period

The sampling site was located on the roof of the Jixian building (21 m above ground level) of Yuanpei University of Medical Technology (24°46'N, 120°56'E) in Hsinchu City, approximately 5 km from the Hsinchu Environmental Protection Agency (EPA) air quality station (Fig. 1). The overall sampling period was from September 2014 to August 2015. Specifically, the autumn sampling period was from September to November 2014 (12 days), the winter sampling period was from December 2014 to February 2015 (24 days), the spring sampling period was from March to May 2015 (20 days), and the summer sampling period was from June to August 2015 (18 days).

The PM_{2.5} samplers (PQ200, BGI Incorporated, USA) equipped with PM_{2.5} cut cyclones (VSCC) and 47-mm quartz filters (7201, Pall, USA) were operated at a flow rate of 16.7 L min⁻¹ for 48 h. The change in the weight of the quartz filters before and after sampling was measured using a microbalance (CP2P-F, Sartorius AG, Goettingen, Germany), which has a sensitivity of ± 1 µg, to determine the mass of the particles. Field quartz blanks were used to monitor the contamination level in the sampling process. Sampled quartz filters were then stored in a refrigerator at 4°C for subsequent analysis.

Extraction of PAHs

The PAHs were extracted as follows: the sampled quartz filters were inserted into 50-mL brown sampling vials and extracted using 25 mL of dichloromethane (UN1539, Merck, Germany). The mixtures were ultrasonicated (DC400H, Delta, Taiwan) for 15 min, with a rest period of 5 min (Yang *et al.*, 2013). This process was repeated three times. The resultant solutions were filtered using a 20-mL glass syringe and a 13-mm disposable syringe filter unit (0.5 µm, Advantec, Inc., Japan). The filtered solutions were concentrated using a

rotary evaporator (N-1000S, Eyela, Japan) with a vacuum controller (NVC-2100, Eyela, Japan) at 650 hPa and a thermostatic bath (OSB-2100, Eyela, Japan) at 33°C. The final volume, approximately 1 mL, was transferred into a 1-mL volumetric flask and was further reduced to 0.8 mL by using nitrogen gas. After the sample had spiked at 0.1 mL internal standards (Supelco, USA), the solution volumes were adjusted to 1 mL by adding pure dichloromethane.

Chemical Analysis of PAHs

A total of 20 PAH of organic pollutants were identified from the atmosphere and quantified through gas chromatography–mass spectrometry (GC–MS; GCMS-QP2010, Shimadzu, Japan). The analytical conditions of the GC–MS instrument were established in a previous study (Yang *et al.*, 2013). The compounds were as follows: Nap, 2-Methylnaphthalene (2-M-Nap), acenaphthylene (AcPy), acenaphthene (Acp), fluorene (Flu), phenanthrene (Phen), anthracene (Ant), fluoranthene (FL), pyrene (Pyr), BaA, Chr, BbF, BkF, benzo[*e*]pyrene (BeP), BaP, perylene (PER), INP, DBA, benzo[*g,h,i*]perylene (BghiP) (Wellington Laboratories, Canada), and coronene (Cor) (AccuStandard, USA) (1 µg mL⁻¹) were used as a PAH standard stock solution. Additionally, five internal standards (naphthalene-D₈, acenaphthene-D₁₀, phenanthrene-D₁₀, chrysene-D₁₂ and perylene-D₁₂) (Supelco, USA) (0.5 µg mL⁻¹) were used as an internal standard stock solution. Seven concentrations of the 20 PAH standards (5–100 ng mL⁻¹), with the same concentrations as the internal standards in the calibration curve, were measured through GC–MS. The concentrations of the 20 PAH standards corresponded to the ratio of the PAH target crest area to the special internal standard target crest area, and the standard quantitative calibration curves were obtained through linear regression. All correlation coefficients for the calibration curves of the 20 PAHs were higher than 0.995. Finally, the concentration of each of the 20 PAHs was determined using the calibration curve. In this study, three times the mean of standard deviation of the seven measurements of the lowest concentration of the calibration curve was defined as the detection limit (DL). The DL range for the 20 PAHs was 0.005–0.022 ng m⁻³. Blank filters and suitable amounts of spiked PAHs were used in each experiment. The background values of the blank filter were subtracted from all samples. If the result was less than the DL, the sample was replaced with one with a value of DL × ((2) ^ 0.5)⁻¹ for the appropriate compound (Hoffman *et al.*, 2015). The cleaned quartz blank filter was added to the known amount of PAH to determine the recovery yield. The mean recovery yield of the 20 PAHs was 76.4%–96.8%.

RESULTS AND DISCUSSION

PM_{2.5} Mass Concentrations in Relation to Seasonal Change

During the sampling period, the average PM_{2.5} mass concentrations in spring, summer, autumn, and winter were 16.5 ± 9.3, 14.7 ± 8.4, 26.5 ± 7.5, and 30.6 ± 15.1 µg m⁻³, respectively (Table 1). The PM_{2.5} mass concentrations

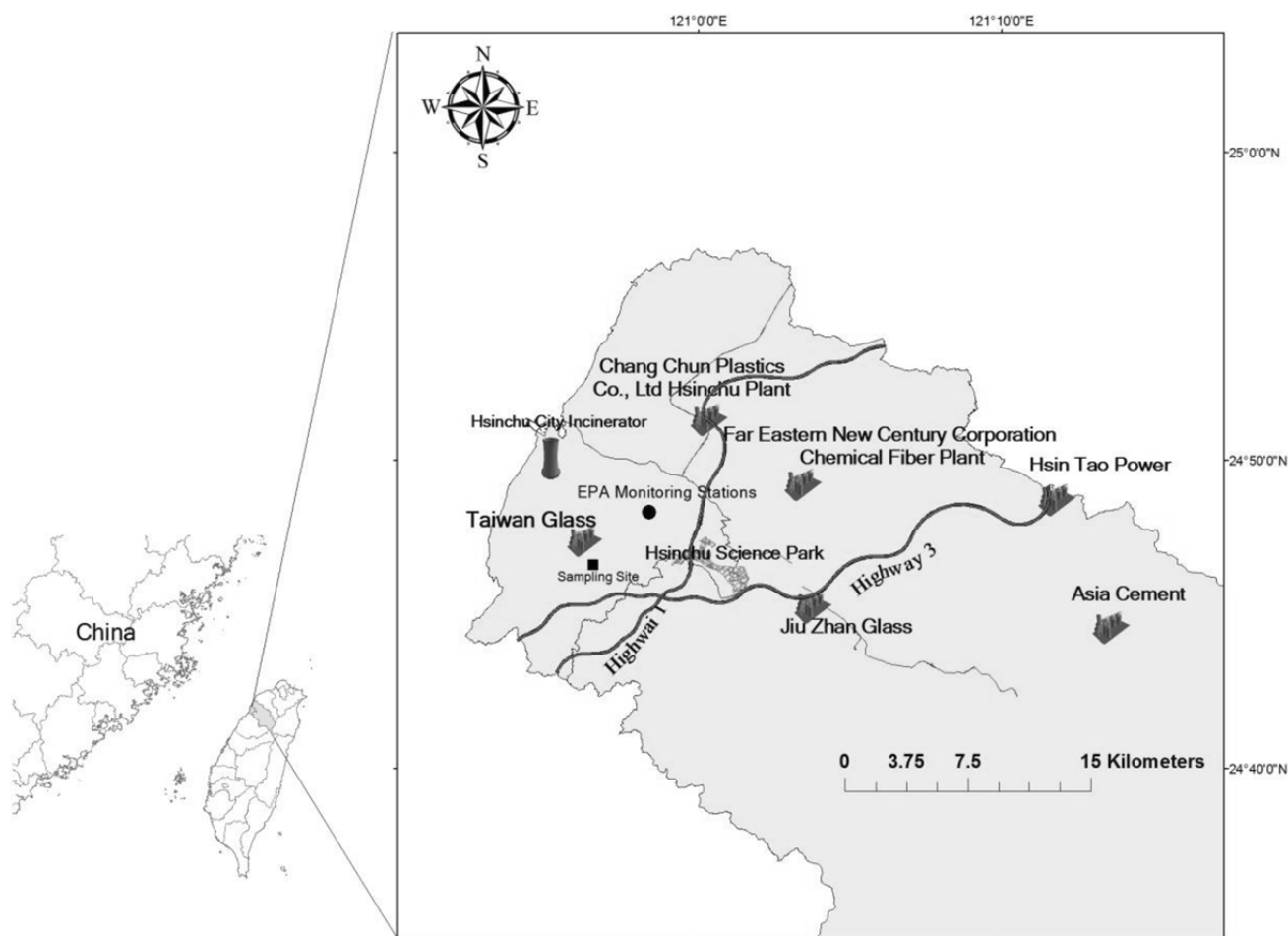


Fig. 1. Overview of the sampling site (square) in Hsinchu City.

during the four seasons were in the order winter > autumn > spring > summer, with significant seasonal variations (analysis of variance (ANOVA) $p < 0.05$). The annual $PM_{2.5}$ mass concentration was $22.3 \pm 12.8 \mu\text{g m}^{-3}$, which exceeded the $PM_{2.5}$ concentration standard for annual air quality set by the Taiwan EPA ($15 \mu\text{g m}^{-3}$). Furthermore, a linear and highly positive correlation was observed between the $PM_{2.5}$ mass concentrations obtained in this study and that reported by the EPA Hsinchu air quality station (regression formula $Y = 1.05X - 4.32$, $R^2 = 0.86$, and $p < 0.0001$, where y represents the $PM_{2.5}$ mass concentrations (Fig. 2). The $PM_{2.5}$ mass concentrations detected in this study were 1.05 times those detected by the EPA Hsinchu air quality station; the slight difference is likely attributable to the differences in the sampling instrumentation.

PAH Mass Concentrations of $PM_{2.5}$ in Relation to Seasonal Change

The average total PAH mass concentrations in $PM_{2.5}$ in spring, summer, autumn, and winter were 0.58 ± 0.38 , 0.42 ± 0.14 , 1.14 ± 0.45 , and $4.04 \pm 2.02 \text{ ng m}^{-3}$, respectively (Fig. 3). The total average PAH concentrations ($0.21\text{--}8.08 \text{ ng m}^{-3}$) in $PM_{2.5}$ were close to those measured by Kume *et al.* (2007) in Shizuoka, Japan ($1.0\text{--}8.4 \text{ ng m}^{-3}$), by Li *et al.* (2009) in Atlanta, USA (3.16 ng m^{-3}), by Martellini *et al.*

(2012) in Tuscany, Italy (5.52 ng m^{-3}), by Wang *et al.* (2015) in Sanya, China ($3.8\text{--}9.9 \text{ ng m}^{-3}$), by Callén *et al.* (2014) in Zaragoza, Spain ($1.31\text{--}2.84 \text{ ng m}^{-3}$), by Odabasi, *et al.* (2015) in Izmir, Turkey (8.3 ng m^{-3}), and by Oliveira *et al.* (2016) in Portugal ($1.45\text{--}4.23 \text{ ng m}^{-3}$). Moreover, the total average PAH concentrations in this study were substantially lower than those reported earlier by numerous researchers such as Li *et al.* (2005) in Guangzhou, China ($15.8\text{--}84.8 \text{ ng m}^{-3}$); Wu *et al.* (2014) in Ordos, China (68.8 ng m^{-3}); Krugly *et al.* (2014) in Kaunas, Lithuania ($20.3\text{--}131 \text{ ng m}^{-3}$); Zhu *et al.* (2015) in Jinan, China ($30.8\text{--}140 \text{ ng m}^{-3}$); and Liu *et al.* (2015) in Guangzhou, China (33.9 ng m^{-3}). The aforementioned studies had focused on $PM_{2.5}$, not total suspended particles (TSPs). Deng *et al.* (2006) and Tan *et al.* (2011) have indicated that the ratio of $PM_{2.5}$ -bound PAHs to TSP-bound PAHs were 0.71 and 0.79, respectively. Therefore, the presented $PM_{2.5}$ -bound PAH data for Hsinchu City indicate a substantial amount of particle-bound PAHs in ambient air.

The mass percentages of two-ring (Nap, 2-M-Nap), three-ring (AcPy, Acp, Flu, Phen, Ant), four-ring (FL, Pyr, BaA, Chr), five-ring (BbF, BkF, BeP, BaP, PER, DBA), six-ring (INP, BghiP), and seven-ring (Cor) PAH levels in $PM_{2.5}$ for the four seasons were 0.29%–4.56%, 3.49%–14.6%, 9.01%–40.1%, 29.6%–44.0%, 15.5%–36.0% and

Table 1. PM_{2.5} and PAHs mass concentrations in spring, summer, autumn, and winter.

Chemicals	Spring (N = 10)			Summer (N = 9)			Autumn (N = 6)			Winter (N = 12)			Annual (N = 37)							
	Mean	S.D. ^c	Max.	Mean	S.D.	Max.	Mean	S.D.	Max.	Mean	S.D.	Max.	Mean	S.D.	Max.					
NaP	0.004	0.002	0.003	0.003	<0.001	0.003	0.003	0.003	0.003	0.011	0.012	0.006	0.003	0.021	0.007	0.005	0.003	0.021		
2-M-Nap	0.007	<0.001	0.007	0.007	<0.001	0.007	0.007	<0.001	0.007	0.007	0.008	0.002	0.007	0.014	0.007	0.001	0.007	0.014		
AcPy	0.009	<0.001	0.009	0.009	<0.001	0.009	0.009	<0.001	0.009	0.009	0.017	0.009	0.009	0.037	0.012	0.006	0.009	0.037		
AcP	0.007	<0.001	0.007	0.007	<0.001	0.007	0.007	<0.001	0.007	0.007	0.007	<0.001	0.007	0.007	<0.001	0.007	0.007	0.007		
Flu	0.007	<0.001	0.007	0.007	<0.001	0.007	0.007	<0.001	0.007	0.007	0.020	0.010	0.007	0.041	0.012	0.008	0.007	0.041		
Phan	0.018	0.020	0.004	0.063	0.008	0.003	0.004	0.012	0.032	0.027	0.014	0.085	0.305	0.193	0.013	0.682	0.111	0.174		
Ant	0.005	<0.001	0.005	0.005	<0.001	0.005	0.005	<0.001	0.005	0.005	0.014	0.008	0.005	0.034	0.008	0.006	0.005	0.034		
FL	0.045	0.051	0.006	0.160	0.017	0.007	0.009	0.028	0.087	0.076	0.038	0.238	0.586	0.356	0.025	1.28	0.220	0.327		
Pyr	0.039	0.036	0.015	0.118	0.019	0.005	0.015	0.029	0.077	0.050	0.038	0.174	0.395	0.233	0.015	0.854	0.156	0.214		
BaA	0.019	0.008	0.013	0.034	0.013	<0.001	0.013	0.013	0.034	0.015	0.020	0.052	0.116	0.063	0.013	0.265	0.051	0.058		
CHR	0.037	0.031	0.015	0.100	0.018	0.006	0.015	0.030	0.072	0.042	0.038	0.152	0.335	0.187	0.023	0.751	0.135	0.177		
BbF	0.087	0.063	0.021	0.202	0.054	0.019	0.023	0.087	0.180	0.072	0.094	0.273	0.594	0.288	0.056	1.17	0.259	0.290		
BkF	0.025	0.018	0.009	0.056	0.013	0.008	0.009	0.028	0.059	0.023	0.037	0.088	0.171	0.088	0.009	0.346	0.075	0.086		
BeP	0.045	0.030	0.015	0.099	0.030	0.013	0.012	0.056	0.092	0.034	0.051	0.132	0.279	0.132	0.028	0.528	0.125	0.133		
BaP	0.035	0.024	0.009	0.074	0.024	0.010	0.009	0.041	0.070	0.028	0.036	0.103	0.218	0.108	0.009	0.433	0.097	0.106		
Pery	0.014	<0.001	0.014	0.014	0.014	<0.001	0.014	0.014	0.014	<0.001	0.014	0.033	0.015	0.014	0.063	0.020	0.012	0.014		
INP	0.070	0.048	0.010	0.148	0.056	0.022	0.024	0.104	0.153	0.055	0.082	0.231	0.390	0.177	0.051	0.679	0.184	0.181		
DBA	0.011	<0.001	0.011	0.011	0.025	0.042	0.011	0.136	0.020	0.010	0.011	0.036	0.040	0.018	0.011	0.080	0.025	0.025		
BghiP	0.071	0.043	0.025	0.142	0.071	0.042	0.019	0.164	0.150	0.058	0.079	0.236	0.356	0.164	0.044	0.629	0.176	0.162		
COR	0.029	0.016	0.010	0.054	0.026	0.009	0.015	0.045	0.064	0.022	0.038	0.101	0.140	0.059	0.024	0.247	0.070	0.062		
ΣPAHs (Cons.) ^a	0.58	0.38	0.21	1.31	0.42	0.14	0.23	0.64	1.14	0.45	0.66	1.79	4.04	2.02	0.37	8.08	1.76	1.98	0.21	8.08
PM _{2.5} mass (Cons.) ^b	16.5	9.3	4.91	36.5	14.7	8.4	5.45	30.0	26.5	7.5	16.2	38.7	30.6	15.1	9.71	58.5	22.3	12.8	4.91	58.5

^aThe unit of PAHs mass concentration was ng m⁻³. ^bThe unit of PM_{2.5} mass concentration was μg m⁻³. S.D.: Standard deviation.

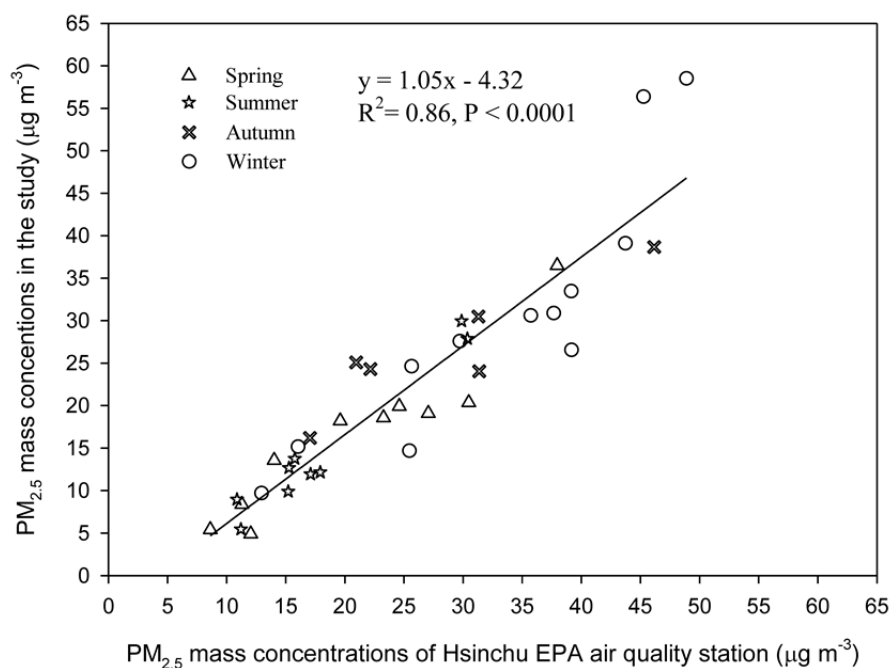


Fig. 2. Linear correlation between the PM_{2.5} mass concentrations in this study and those from the EPA Hsinchu air quality station.

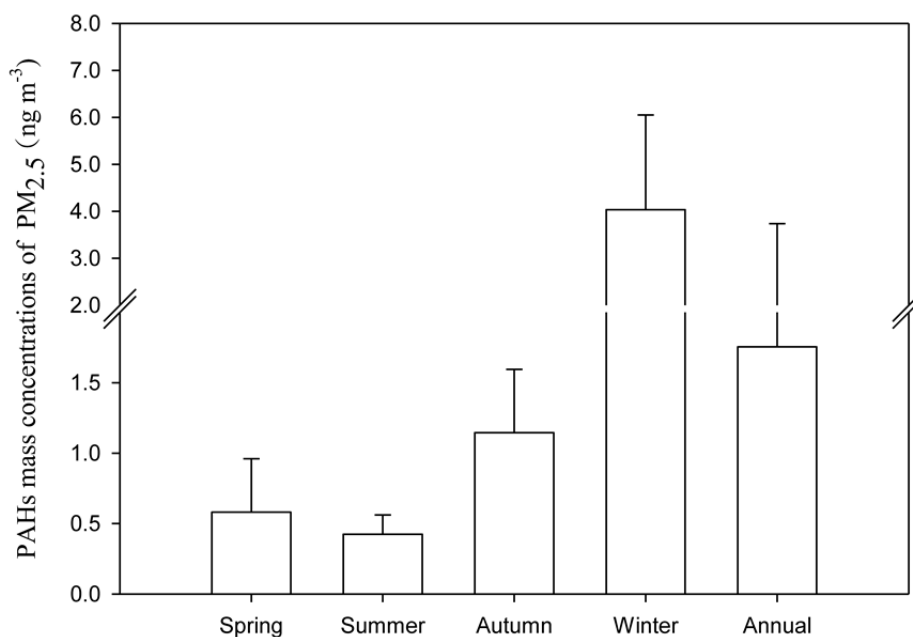


Fig. 3. Total PAH concentrations of PM_{2.5} in spring, summer, autumn, and winter (Bar: mean ± standard deviation).

2.51%–7.57%, respectively (Table 1). The four-, five-, and six-ring PAHs predominated in PM_{2.5}. The average annual contributions of PAHs with low (2- and 3-ring), median (4-ring), and high (greater than 4-ring) molecular weight to total PAH levels were 10.1%, 24.4%, and 65.5%, respectively, which is consistent with those reported by Delgado-Saborit *et al.* (2013). The ability of the particles to carry PAH depends on their molecular weight and vapor pressure. PAHs exhibiting high molecular weight and low vapor pressure exhibit an increased absorption rate in the particle phase

(Delgado-Saborit *et al.*, 2013). Furthermore, the high percentage of PAHs with a high molecular weight indicated that the predominant PAH sources were high-temperature processes, such as the combustion of fuels in engines (Tobiszewski and Namieśnik, 2012). The dominant species in spring, summer, and autumn were BbF, INP, and BghiP. The dominant species in winter were BbF, FL, and INP (Table 1). INP, BbF, and BghiP have also been reported to be predominant compounds in São Paulo, Brazil, (Bourotte *et al.*, 2005) and Porto Alegre, Brazil (Dallarosa *et al.*, 2005).

The total average PAH mass concentrations in PM_{2.5} among the four seasons were in the order of winter > autumn > spring > summer. The total PAH mass concentrations of PM_{2.5} exhibited significant seasonal variations (ANOVA $p < 0.05$) (Fig. 3). Agudelo-Castañeda and Teixeira (2014) indicated that the total PAH mass concentrations of PM₁ for summer and winter were 1.32 and 2.02 ng m⁻³ by a roadside in Canoas, Brazil, and 1.57 and 3.05 ng m⁻³ by a roadside in Sapucaia do Sul, Brazil. These results are consistent with the results of this study, wherein greater PAH concentration levels were observed in winter than in summer. Furthermore, the fractions of total PAHs in PM_{2.5} for spring, summer, autumn, and winter were $0.39 \times 10^{-2}\%$, $0.78 \times 10^{-2}\%$, $0.44 \times 10^{-2}\%$, and $1.37 \times 10^{-2}\%$, respectively. The PAH fraction in winter was the largest. The values for the four seasons were less than those ($2.2 \times 10^{-2}\%$ to $1.2 \times 10^{-1}\%$) obtained by Mannino and Orecchio (2008).

Meteorological Data, Other Air Pollutants, and Total PAH Correlation

The Hsinchu metropolitan area is located in subtropical Taiwan. Southwesterly winds prevail in summer, and northeasterly monsoons occur in winter. Based on data from the EPA air quality station in Hsinchu, meteorological data and other air pollutants observed during the sampling period were as follows: the atmospheric temperature and relative humidity ranged from 12.8–30.6°C and 53.2%–88.8%, respectively; the O₃ (ozone), CO (carbon monoxide), NO_x (nitrogen oxide), and SO₂ (sulfur oxide) concentrations, and the PM_{2.5} mass concentrations ranged from 14.5–48.1 ppb, 0.27–0.87 ppm, 10.2–48.4 ppb, 0.78–3.99 ppb, and 8.63–48.9 µg m⁻³, respectively.

There was a significant negative correlation between the total PAH concentration in PM_{2.5} and atmospheric temperature, with a negative correlation coefficient of $r = 0.71$, $p < 0.05$. However, the total PAH concentration did not exhibit a statistical correlation with relative humidity. This indicates that total PAH concentration of PM_{2.5} decreases with increase in atmospheric temperature. A possible mechanism for this phenomenon is that the low-molecular-weight PAHs in the particle phase can easily move in the vapor phase and higher mixing layer at higher temperatures (summer); conversely, these low-molecular-weight PAHs are easily adsorbed in the particulate phase and lower mixing layer at lower temperatures (winter) (Křůmal *et al.*, 2013). The mean temperature in winter (17.5°C) was lower than that in summer (29.9°C). The mean concentration of PAHs in winter was approximately 9.50 times higher than that in summer. PAHs are more easily photochemically degraded in summer than in winter (Křůmal *et al.*, 2013; Agudelo-Castañeda and Teixeira, 2014). Moreover, the PM_{2.5} mass concentrations in this study exhibited a strong positive correlation ($r = 0.68$, $p < 0.05$) with total PAHs. A weak linear correlation between total PAHs and CO/NO_x was observed, with positive correlation coefficients $r = 0.43$, $p < 0.05$ and $r = 0.36$, $p < 0.05$, respectively, indicating that PAH, PM_{2.5}, CO, and NO_x concentrations were derived from similar emission sources in Hsinchu. Furthermore, the total PAH concentration did not significantly correlate

with the concentrations of O₃ and SO₂.

Degree of Carcinogenicity of the PAHs of PM_{2.5} in Relation to Seasonal Change

The total BaP_{eq} concentration is the sum of the concentrations of the individual PAH multiplied by their respective toxic equivalent factors (TEQs) (Nisbet and LaGoy, 1992). Thus, the carcinogenic potency of the total PAH mass concentrations was represented as total BaP_{eq}. The average total BaP_{eq} concentrations of the 20 PAHs in the PM_{2.5} for spring, summer, autumn, and winter were 0.07 ± 0.04 , 0.06 ± 0.04 , 0.14 ± 0.05 , and 0.39 ± 0.19 ng m⁻³, respectively. The total BaP_{eq} concentrations of PM_{2.5} exhibited significant seasonal variations (ANOVA $p < 0.05$). The total BaP_{eq} concentrations during the four seasons were in the order winter > autumn > spring > summer (Fig. 4). In this study, the BaP, DBA, BbF, and INP were the major carcinogenic toxins in the 20 PAHs in PM_{2.5} for all seasons, comprising 49.0%, 19.7%, 12.7%, and 10.3% of the total carcinogenic toxicity, respectively. Delgado-Saborit *et al.* (2011) identified the major compounds contributing to total carcinogenic potency as BaP, DBA, and BbF. Results from the literature and those in this study both indicate that BaP is the primary contributor to overall carcinogenic potency.

Lifetime excess cancer risk (ECR) was calculated using the total BaP_{eq} concentrations of the 20 PAHs, multiplied by the unit risk (UR = 8.7×10^{-5} per ng m⁻³) of BaP inhalational exposure (WHO, 2000). Accordingly, the average ECR of PM_{2.5}-bound PAHs for spring, summer, autumn, and winter were 5.83×10^{-6} , 5.52×10^{-6} , 1.18×10^{-5} , and 3.43×10^{-5} , respectively, which is higher than the guideline (10^{-6}) provided by the United States EPA (USEPA). The ECR of PM_{2.5}-bound PAHs during the four seasons was in the order winter > autumn > spring > summer, indicating that people with exposure to PM_{2.5}-bound PAHs are at an increased risk of carcinogenic exposure in cold winters than in hot summers.

Characteristic Ratios of PAHs

The characteristic ratios of Ant/(Ant + Phen), FL/(FL + Pyr), BaA/(BaA + Chr), and INP/(INP + BghiP) were used to identify various contamination sources (Yunker *et al.*, 2002). The mean ratios of Ant/(Ant + Phen) for spring, summer, autumn, and winter were 0.33, 0.40, 0.17, and 0.07, respectively (Table 2). The Ant/(Ant + Phen) ratio < 0.1 in winter indicated a low-temperature source (petroleum sources) and a ratio > 0.1 indicated that combustion sources dominated. The mean ratio of FL/(FL + Pyr) was 0.47 in spring and summer and ranged from 0.51 to 0.59 in autumn and winter (Table 2). The FL/(FL + Pyr) ratio ranged from 0.4 to 0.5, which indicated liquid fossil fuel (vehicle and crude oil) combustion sources, and > 0.50 came from grass, wood, and coal combustion sources. The mean ratios of BaA/(BaA + Chr) in spring, summer, autumn, and winter were 0.39, 0.42, 0.33, and 0.27, respectively (Table 2). The BaA/(BaA + Chr) ratio ranged from 0.20 to 0.35, indicating either petroleum or combustion sources dominated in autumn and winter. The BaA/(BaA + Chr) ratios > 0.35 indicated

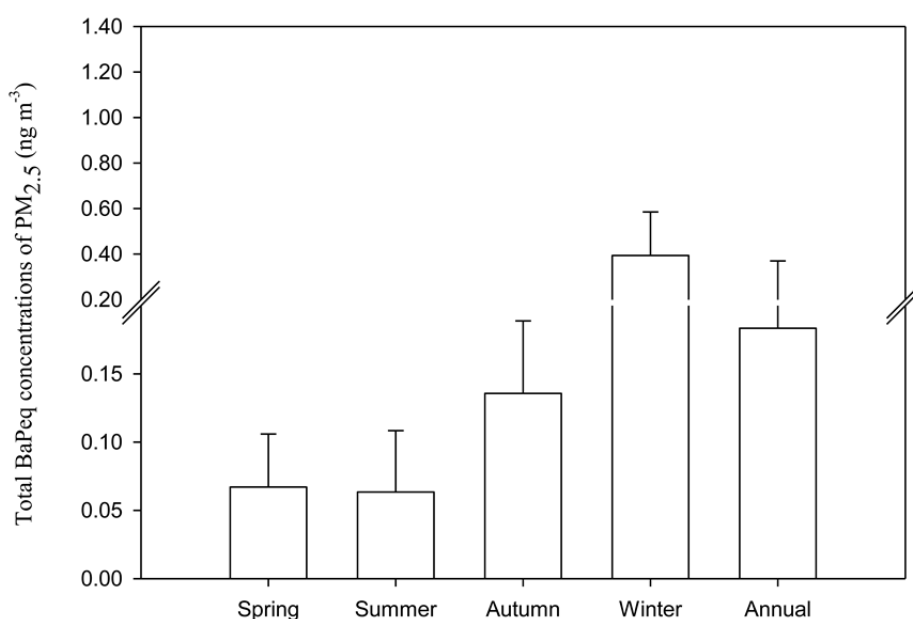


Fig. 4. Total BaPeq concentrations of PAHs in PM_{2.5} in spring, summer, autumn, and winter (Bar: mean ± standard deviation).

Table 2. Characteristic ratios of Ant/(Ant + Phen), FL/(FL + Pyr), BaA/(BaA + Chr), INP/(INP + BghiP), BghiP/BeP, Cor/BeP and BaP/(BaP + BeP) for PM_{2.5} in spring, summer, autumn, and winter.

Characteristic ratios	Spring (N = 10)		Summer (N = 9)		Autumn (N = 6)		Winter (N = 12)		Annual (N = 37)	
	Mean	S.D. ^a	Mean	S.D.	Mean	S.D.	Mean	S.D.	Mean	S.D.
Ant/(Ant + Phen)	0.33	0.18	0.40	0.10	0.17	0.07	0.07	0.07	0.24	0.18
FL/(FL + Pyr)	0.47	0.11	0.47	0.05	0.51	0.05	0.59	0.02	0.52	0.08
BaA/(BaA + Chr)	0.39	0.09	0.42	0.06	0.33	0.05	0.27	0.04	0.35	0.09
INP/(INP + BghiP)	0.47	0.07	0.46	0.07	0.51	0.02	0.52	0.01	0.49	0.06
BghiP/BeP	1.66	0.25	2.38	1.26	1.63	0.26	1.31	0.16	1.72	0.75
Cor/BeP	0.71	0.22	0.90	0.18	0.73	0.16	0.54	0.12	0.70	0.22
BaP/(BaP + BeP)	0.42	0.04	0.44	0.02	0.43	0.03	0.42	0.06	0.43	0.04

^a S.D.: Standard deviation.

that combustion sources dominated in spring and summer. The mean ratios of INP/(INP + BghiP) ranged from 0.47 to 0.46 in spring and summer and were 0.51 and 0.52 in autumn and winter, respectively (Table 2). The INP/(INP + BghiP) ratio ranged from 0.2 to 0.5, indicating that liquid fossil fuel combustion sources dominated, whereas > 0.50 were from grass, wood, and coal combustion sources.

BghiP/BeP and Cor/BeP ratios were utilized to identify traffic and nontraffic contributions (Nielsen *et al.*, 1996). The BghiP/BeP ratios from traffic and nontraffic contributions were 2.02 and 0.8, respectively (Nielsen, 1996). The mean ratios of BghiP/BeP for spring, summer, autumn, and winter in our study were 1.66, 2.38, 1.63, and 1.31, respectively (Table 2). The results clarify the traffic contribution during summer and the traffic and nontraffic contributions during all other seasons. The Cor/BeP ratios from traffic and nontraffic contributions were 1.54 and 0.3, respectively (Rogge *et al.*, 1993). The mean ratios of Cor/BeP among the four seasons ranged from 0.54 to 0.90 (Table 2), inclusive of both the traffic and nontraffic contribution. The BaP/(BaP + BeP) ratio is approximately 0.50 for fresh particulate emissions

(Oliveira *et al.*, 2011). The mean ratios of BaP/(BaP + BeP) among the four seasons in our study ranged from 0.42 to 0.44 (Table 2), indicating that the sample was from fresh particulate emissions.

Positive Matrix Factorization

We used 20 species of measured components to develop a PMF model. Three main sources based on annual data were identified: stationary emissions (56.2%), unburned petroleum and traffic emissions (34.1%), and steel industry emissions (9.7%). Fig. 5 displays the modeled source profiles for each identified source, which was obtained by collecting annual PAH data. We also determined the geographical origins of each PAH source by conducting a CPF (Fig. 6) to provide insights on source localization.

The first source was stationary emissions. A high percentage of FL (91.1%) and Pyr (82.2%), Phen (95.4%), Chr (72.6%), BaA (54.0%) and BbF (52.1%). FL, Pyr, Phen, and Chr are typically markers of petroleum and oil burning. Chr, BbF, and BeP have been demonstrated to be stationary emission sources (Yang *et al.*, 1998; Kulkarni

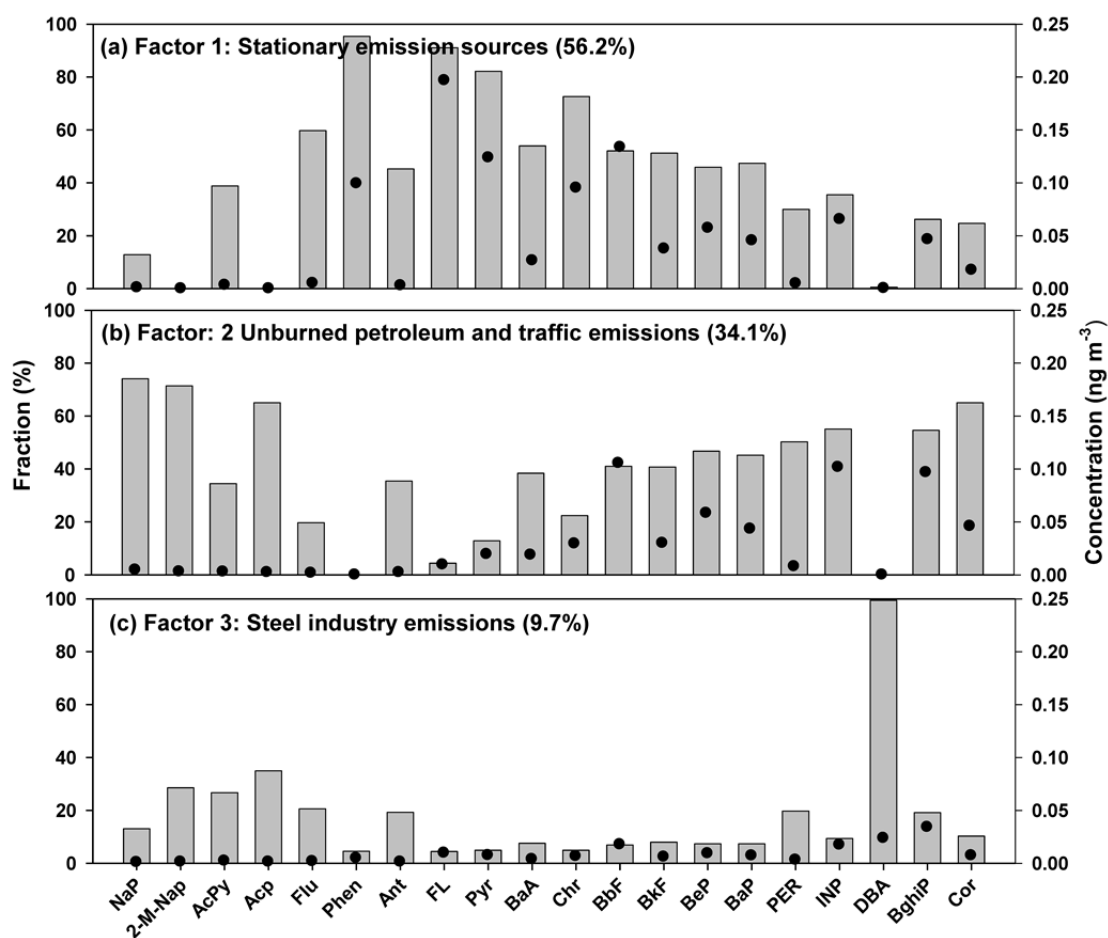


Fig. 5. Profiles of three sources identified from the PMF model for PAHs in $PM_{2.5}$.

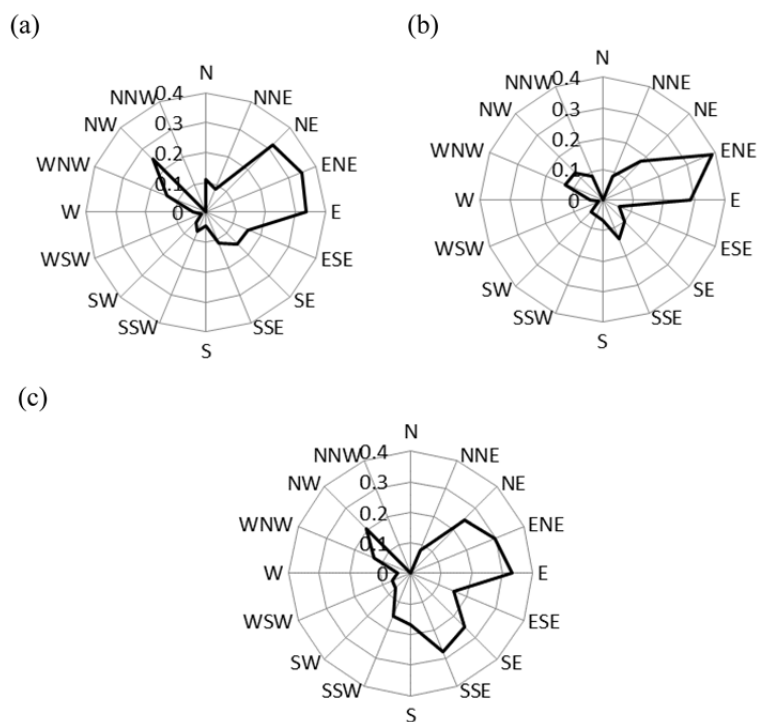


Fig. 6. Likely source area of (a) stationary emission sources (b) unburned petroleum and traffic emissions, and (c) steel industry emissions sources in the study area resulting from CPF.

and Venkataraman, 2000). Moreover, Pyr and BaA were identified as distinctive markers of coal combustion (Khalili et al., 1995; Teixeira et al., 2013). The diagnostic ratios for FL/(FL + Pyr) and BaA/(BaA + Chr) in this study indicated stationary emission sources. The CPF plot in Fig. 6(a) displays this factor originating predominantly from northeast, east-northeast, east, and northwest directions, which is in strong agreement with the actual location of the local sources (Fig. 1).

The second source was unburned petroleum and traffic emissions. A high percentage of contributions by NaP (74.1%), 2-M-Nap (71.4%), AcP (65.0%), PER (50.2%), INP (55.1%), BghiP (54.6%), and Cor (65%). Yunker et al. (2002) indicated that the predominance of 2- and 3-ring PAHs is associated with petrogenic sources of unburned petroleum, gasoline, and diesel. Additionally, BghiP and Cor with high loading have been suggested for gasoline-powered vehicles (Khalili et al., 1995; Schauer et al., 2002). Guo et al. (2003) also reported BghiP as a source marker for gasoline emissions. Yao et al. (2016) reported that 2-M-Nap indicates a petroleum-related source. The diagnostic ratio of BaP/(BaP + Chr) (= 0.49) has been used to identify diesel emissions (Khalili et al., 1995). The 0.48 value for BaP/(BaP + Chr) was calculated on the basis of the PAH concentration (Table 1). Vehicular transport likely releases associated aerosol compounds along highways and city roads, particularly during rush hour. Hence, unburned petroleum and traffic emissions were identified. The CPF plot in Fig. 6(b) indicates the prevailing east-northeast and easterly directions as possible source regions, where Highway 1, Highway 3, and the city roads are located.

The third source was steel industry emissions, which contributed little (< 10%) to PM_{2.5}-bound PAHs. The dominant contribution (99.4%) of DBA to the source profile suggests that this source is related to steel industry emissions. Ravindra et al. (2006) and Yang et al. (2002) reported that DBA was a typical marker of steel industry emissions, produced through processes such as sintering. Additionally, a study in central Taiwan attributed a high contribution of DBA to steel industry emissions (Chen et al., 2016). The CPF plot in Fig. 6(c) indicates that the high concentration of PAHs in this factor were from east-northeast, east, and south-southeasterly directions.

CONCLUSIONS

The concentrations of PM_{2.5} and PM_{2.5}-bound PAHs collected in Hsinchu were measured in spring, summer, autumn, and winter periods from September 2014 to August 2015 to assess the characteristics and sources of the PM_{2.5}-bound PAHs and their potential risks. The PM_{2.5} mass, total PAHs, and BaPeq concentrations of the four seasons were 4.91–58.5 μg m⁻³, 0.21–8.08 ng m⁻³, and 0.03–0.78 ng m⁻³, respectively. The predominating species among the 20 PAHs were BbF and INP for all four seasons. BaP, BbF, INP, and DBA were major contributors to the total PM_{2.5} carcinogenic toxins for all four seasons. BaP accounted for 49.0% of total BaPeq concentrations. The PM_{2.5}, total PAHs, and BaPeq mass concentrations were in the order

winter > autumn > spring > summer. Seasonal variation was observed in the PM_{2.5}, total PAHs, and BaPeq mass concentrations (ANOVA $p < 0.05$). The carcinogenic potency of PAHs in winter was approximately 6.21 times higher than that in summer. The annual average ECR of PM_{2.5}-bound PAHs was 1.60×10^{-5} , which is higher than that specified in the USEPA guideline (10^{-6}). Such ECRs from PM_{2.5}-bound PAHs likely underestimate the risk of cancer because gaseous PAHs were not determined in this study. Finally, the two major PAH sources were stationary emission sources and unburned petroleum and traffic emissions, which combined, accounted for 90.3% of PM_{2.5}-bound PAHs in Hsinchu City. Our experimental results on the characteristics and sources of PM_{2.5}-bound PAHs in Hsinchu can be used to implement PM_{2.5} control strategies, to develop BaP regulations, and to serve as a reference to determine methods for improving environmental health.

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