



## Risk Assessment of PAH Exposure Involving Metal Working Fluids in Fastener Manufacturing Industries

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### ABSTRACT

Thirty-four air samples from different working areas of three fastener factories were collected to assess the gaseous/particulate-phase polycyclic aromatic hydrocarbon (PAH) levels as well as to assess the total-PAH level associated to health risks in fastener manufacturing workers.

The higher total-PAH level were found in heat treatment area (1221 ng m<sup>-3</sup>), threading area (920 ng m<sup>-3</sup>), forming area (897 ng m<sup>-3</sup>), packing area (878 ng m<sup>-3</sup>), and acid-cleaning area (850 ng m<sup>-3</sup>); in addition, higher BaP<sub>eq</sub> levels were found in packing area (216 ng m<sup>-3</sup>), threading area (103 ng m<sup>-3</sup>), heat treatment area (99.3 ng m<sup>-3</sup>), forming area (91.1 ng m<sup>-3</sup>), and surface treatment area (87.1 ng m<sup>-3</sup>). Because metal working fluids are widely used in fastener manufacturing factories, PAH emissions in the gas phase (over 80%) were predominant in these working areas except for in packing area.

Although the average total BaP<sub>eq</sub> exposure levels were lower than the permissible exposure limit set by the Occupational Safety and Health Administration (OSHA, 8 h time-weighted average: 0.2 mg m<sup>-3</sup>), the lung cancer risks associated with the above mentioned PAH exposures were  $4.36 \times 10^{-4}$ – $1.14 \times 10^{-3}$ , and the gaseous-phase PAH exposure risk was much higher than that from particulate-phase exposure in workers exposed to oil mists in a fastener manufacturing factory. The results of this study suggest that the occupational hazards of PAHs cannot be neglected for workers in the fastener manufacturing industry; occupational hazard prevention must focus on gaseous- as well as particulate-phase PAH exposure.

**Keywords:** PAH; Fastener; Benzo[a]pyrene; BaP; Health risk assessment.

### INTRODUCTION

Previous studies have reported that workers in the fastener manufacturing industry may have high risks of lung and skin cancer due to exposure to polycyclic aromatic hydrocarbons (PAHs) in oil mist (Chen *et al.*, 2007, 2008). According to statistical data from the Taiwan government for 2012, there were 1310 fastener manufacturing factories and approximately 28,000 employees in Taiwan. The fastener manufacturing process includes cleaning, wire drawing, forming, threading, surface treatment/heat treatment, and packing. Mineral-oil-based metal working fluids (MWFs) are applied throughout the manufacturing process, including in the cutting process, machining, forming, heat treatment, as lubrication during threading, and for corrosion inhibition

to improve the surface roughness (Debnath *et al.*, 2014). During the heat treatment process, the temperature is raised to 650–1500°C and then cooling, which can increase the tensile strength of the fasteners to make the products more resistant to breakage. The high cutting temperatures can also result in the production of oil mists (Sujová, 2012; Li *et al.*, 2014). Moreover, MWFs contain petroleum oils, which are expected to contain semi-volatile organic compounds, such as PAHs, may be emitted during cleaning, forming, threading, and surface/heat treatment (Chen *et al.*, 2008).

Exposure of PAH-related occupations are associated with increasing risks of lung, bladder, and urinary cancers (Gaertner and Thériault, 2002; Bosetti *et al.*, 2007), as well as higher risks of DNA damage or lesions related to oxidative damage in workers exposed to fireproof materials and in bitumen coke-oven workers (Cheng *et al.*, 2005; Cavallo *et al.*, 2006; Chao *et al.*, 2008).

For workers with occupational PAH exposure, naphthalene (Nap), consists of two aromatic rings, is considered to be a typical representative PAH in the gaseous phase (Rappaport *et al.*, 2004). In the particulate phase of PAHs, five and six

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aromatic rings are predominant, and three- and four-ring are both in the gaseous and particulate phases; these vary by the environmental conditions (Pleil *et al.*, 2004). Although in regard to toxicity concerns, the high potency of carcinogenic PAHs, especial five and six rings are predominant in the particulate phase, the gaseous PAHs comprise the majority of the total-PAH in occupational environments cannot be negligence (Chen *et al.*, 2008; Liu *et al.*, 2010). PAH exposure through inhalation, ingestion and dermal contact (Shimmo, 2004; Chen *et al.*, 2008; Cheruiyot *et al.*, 2015), but inhalation pathway is the major one in working environments. The gaseous/particulate distribution of PAHs may shift base on ambient temperature (Tsapakis and Stephanou, 2005). Thus, the heat treatment involving high temperatures during the manufacturing processes in the fastener industry is dominant to affect PAHs emission in this kind of work. To date, not all the fastener manufacturing industries include all of the manufacturing processes; therefore, in this study, the gaseous/particulate-phase PAHs in three fastener manufacturing factories with alternating manufacturing processes are investigated. Meanwhile, workplace air samples are obtained and subjected to PAH analysis to assess the health risks to fastener manufacturing workers that are related to PAH exposure.

## MATERIALS AND METHODS

### *Sampling Selection in Fastener Manufacturing Factories*

The study was conducted in three typical fastener manufacturing factories in Taiwan. Factory A is responsible for the later manufacturing process, including forming and threading (80 workers); Factory B is small, with a floor area of 300 m<sup>2</sup> (30 workers), but the manufacturing process includes acid cleaning, forming, threading, surface treatment/

heat treatment, and packing, all situated in a small factory; the same manufacturing process takes place in Factory C (419 workers). There are no partitions in any of the three factories and the ventilation is poor in Factory B.

A pre-sampling walk-through was conducted to determine the layout of each work site and its borders. In principle, the zones in the fastener manufacturing factories were based on various operational functions. Several areas inside the factories, including acid cleaning, forming, threading, heat treatment, packing, and office areas, as well as areas outside the factories, were selected for this study. A total of 34 air samples were taken inside the fastener manufacturing factories, and eight air samples were taken in office spaces and outside of the three factories.

### *Personal Air Samples*

Personal air samples were collected and analyzed using a modification of NIOSH method 5515. The sampling train included IOM inhalable dust samplers (SKC Inc. Eighty Four, PA, USA), equipped with glass fiber filters 25 mm in diameter (Whatman Inc., Grade G653, Cat. No.18209932) to collect PAHs in the particulate phase. A series connection using a sorbent tube (polyurethane foam, denoted PUF plug 3.5g Amberlite (tm) XAD-2 (denoted XAD-2) resin/PUF separation layer/0.5g XAD-2 resin/PUF plug (SKC Inc., Lot 9597, Cat. No.226-30-04, Made in the USA)) was linked to the IOM sampler. With regard to the personal air pump samplers (Gillian), the flow rate was set at 2 L min<sup>-1</sup> for the IOM sampler, and the flow rate for the sorbent tube was 0.2 L min<sup>-1</sup>, which was adjusted by a triangular flow adjustment kit designed by Tsai *et al.* (the sampling train is shown in Fig. 1) (Chen *et al.*, 2008). The glass fiber filters were conditioned at the same temperature and humidity. In addition, the filters were weighed 48 h post-conditioning.



**Fig. 1.** The sampling train of gaseous and particulate phase PAHs.

The post-sampling weights were subtracted from the pre-sampling weights to provide the particle mass in the ambient sample. During sample transportation, sampling, and storage, the sorbent tubes were covered with aluminum foil to avoid photolysis degradation.

#### PAH Analysis

After sampling, the sorbent tubes were cleaned by microwaving and extracted with a 1:1 solution (Merck) of n-hexane and acetone (v/v) for 40–60 min, before warming to 125°C. The samples were digested using a 1600 W microwave oven (Mars, microwave digestion system, CEM) set at 50% power for 10 min, and then held for 2 min and cooled down for 30 min.

The subsequent procedure involved the removal of any pollutants to avoid contaminating the gas chromatograph (GC) column. Following the re-concentration procedure, 1.0 mL of extract was obtained after ultra-pure nitrogen treatment. All of the sampling solvents were analyzed using a GC (Agilent 7890A)/mass-selective (MS) detector (Agilent 5975C) equipped with a GC capillary column (HP 5 ms–30 m, ID 0.25  $\mu\text{m}$ , Agilent Technology Inc.) and an automatic injection device (Agilent 7683B series, Agilent Technology Inc.). The injection volume was 1  $\mu\text{L}$ . A computer workstation was used for the PAH analysis. The temperatures for the injector, ion source, and quadrupole were 280°C, 230°C, and 230°C, respectively. The oven temperature began at 100°C and was increased to 200°C at a rate of 6°C per min and then was further increased from 200°C to 300°C at a rate of 4°C per min; the final temperature of 300°C was held for 8 min.

In this study, a total of 16 PAH species were analyzed, including Nap, acenaphthylene (AcPy), acenaphthene (Acp), fluorene (Flu), phenanthrene (PA), 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-c,d]pyrene (IND), dibenz[a,h]anthracene (DBA), and benzo[g,h,i]perylene (BghiP).

#### Quality Control

After consecutive sampling for 8 h, the filters and sorbent tube were stored at  $-20^{\circ}\text{C}$ . The masses of the molecular and fragment PAH ions were determined by using the scan mode for 16 pure PAH standards (2000  $\mu\text{g mL}^{-1}$ ; SUPELCO 48905-U). Quantification of the PAHs was performed using the selected ion monitoring mode. The instruments were calibrated using at least five standard concentrations. The correlation coefficient of the calibration curve was  $>0.995$  for a linear least-squares fit of the data. The detection limit was determined from a selected concentration that was slightly lower than the lowest concentration of the calibration curve. Measurements at this concentration were repeated seven times to estimate the standard deviation. The detection limit was determined to be three times the standard deviation. The detection limit of the GC/MS for the 16 PAHs was between 0.39  $\text{ng mL}^{-1}$  and 1.36  $\text{ng mL}^{-1}$ . In this study, PAH recovery efficiency tests were accomplished by performing the same analysis procedure of samples

with addition of the standard solution before extraction. The recovery efficiency of the PAHs ranged from 85% to 116% (average 89.1%).

#### Health Risk Assessment

Many researchers have calculated the cancer risk of PAH exposure based on the toxic equivalency factors, determined using BaP<sub>eq</sub> (Nisbet and LaGoy, 1992; Fang *et al.*, 2004; Tsai *et al.*, 2004) because BaP<sub>eq</sub> was a better indicator than the total PAH level for characterizing the carcinogenic potency of PAHs. The cancer risks herein were calculated as described by Nisbet *et al.* (1992) to assess workers' excessive lung cancer risks associated with 25 year-long occupational exposure. The assumptions for cancer risk assessment were that adult workers worked for 8 h a day, 5 days a week, for 25 years during their lifetime (78.74 years). The unit risk obtained in the current study was  $7 \times 10^{-5}$  (BaP<sub>eq</sub>  $\text{ng m}^{-3}$ )<sup>-1</sup> (Lin *et al.*, 2008).

#### Data Analysis

The JMP 5.0 (SAS Institute, Cary, NC, USA) and STATISTICA software (version 6.0, StatSoft Inc.) packages were used for data management and principal component analysis (PCA).

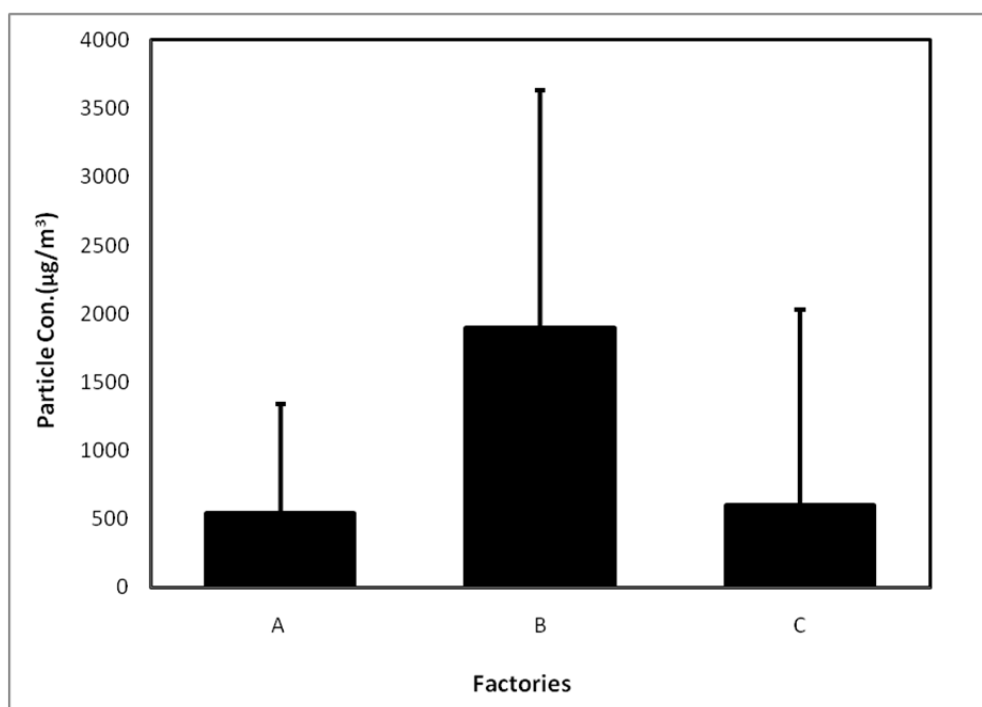
## RESULTS AND DISCUSSION

#### Particle Levels in Three Fastener Manufacturing Industries

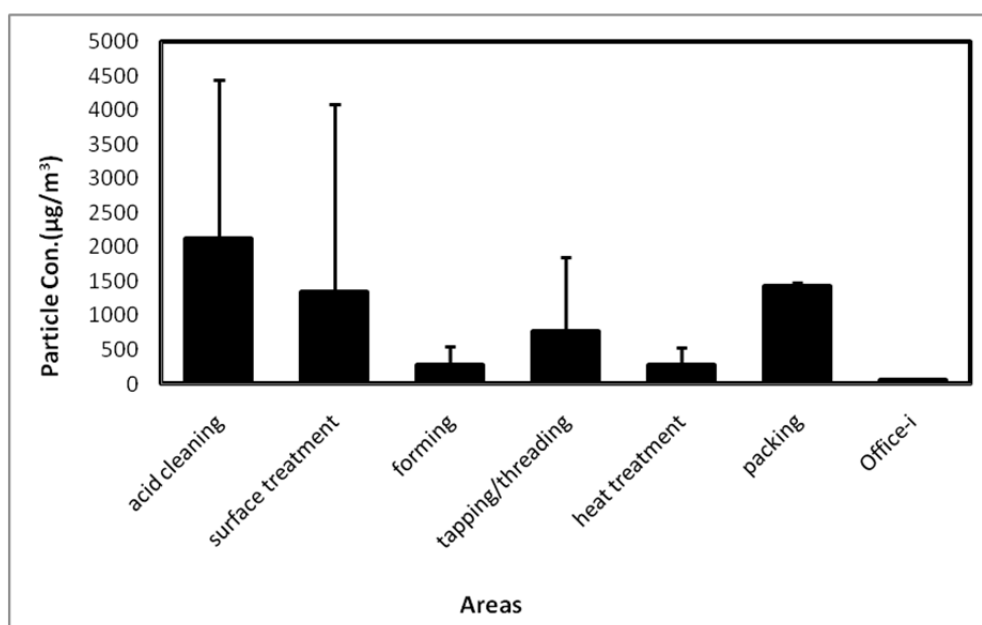
The average particle levels in Factory A, B and C were 542  $\mu\text{g m}^{-3}$  (standard deviation, 838), 1893  $\mu\text{g m}^{-3}$  (standard deviation, 1836) and 599  $\mu\text{g m}^{-3}$  (standard deviation, 1508), respectively (Fig. 2). The levels were different in the three factories, and those in Factory A and C were obviously lower than average particle levels ranged from 1.64  $\text{mg m}^{-3}$  to 1.87  $\text{mg m}^{-3}$  in foundries where handling metal manufacturing process (Liu *et al.*, 2010). The particle measurements in different areas showed that the highest level was found in acid cleaning area (2126  $\mu\text{g m}^{-3}$ ), the second highest level was in packing area (1429  $\mu\text{g m}^{-3}$ ), the surface treatment area (1346  $\mu\text{g m}^{-3}$ ), and others ranged 270–758  $\mu\text{g m}^{-3}$ , whereas the lowest one was in office (53  $\mu\text{g m}^{-3}$ ), respectively (Fig. 3). From the present study, concentration of inhalable particles were comparable to the personal inhalable oil mist sampling in threading workers (2.11  $\text{mg m}^{-3}$ ); forming workers (1.58  $\text{mg m}^{-3}$ ); heat treatment workers (0.08  $\text{mg m}^{-3}$ ) from a previous study in fastener manufacturing industry (Chen *et al.*, 2007). Chen *et al.* (2007) have mentioned PAH emitted from MWFs and may condensed as oil mist to effect the workers' health, meanwhile, among 34 samplings inside the manufacturing process in the current study, a sampling in acid cleaning area (1/4, 25%) and a sampling in heat treatment area (1/16, 6.25%) have over PEL-TWA of 5  $\text{mg m}^{-3}$  for oil mist. Except for the particulate phase hazard, the gaseous hazards should be considered simultaneously in the industry using MWFs.

#### PAH Levels in Three Fastener Manufacturing Industries

The average PAH level inside Factory C was 1126  $\text{ng m}^{-3}$ ,



**Fig. 2.** The particle concentration ( $\mu\text{g m}^{-3}$ ) and 95% upper limit levels in three industries.



**Fig. 3.** The particle concentration ( $\mu\text{g m}^{-3}$ ) and 95% upper limit levels in different working departments (Office-i means inside the office).

which was higher than those in Factories B ( $860 \text{ ng m}^{-3}$ ) and A ( $760 \text{ ng m}^{-3}$ ). The gaseous total-PAH levels in Factories A, B, and C were  $663 \text{ ng m}^{-3}$ ,  $662 \text{ ng m}^{-3}$ , and  $1014 \text{ ng m}^{-3}$ , respectively, and the contributions of the gaseous phase to the total PAHs were 87.2%, 77.0%, and 90.1%, respectively (Table 1). The BaPeq level inside Factory B was  $127 \text{ ng m}^{-3}$ , which was higher than those in Factories C ( $101 \text{ ng m}^{-3}$ ) and A ( $81 \text{ ng m}^{-3}$ ) (Table 2). Meanwhile, the gaseous/particulate-phase BaPeq levels were 91.9%, 56.3%, and 87.2% in

Factories A, B, and C, respectively. The five- and six-ring and two-ring PAHs contributed the most to the BaPeq levels in these factories, including BaP, DBA, and IND. When compared to environmental samplings, total concentration of PAHs in gas phase ranged from  $662 \text{ ng m}^{-3}$  to  $1014 \text{ ng m}^{-3}$  in this occupational study, which are obviously higher than those ranging from  $44.3 \text{ ng m}^{-3}$  to  $129.2 \text{ ng m}^{-3}$  and  $6.89 \text{ ng m}^{-3}$  to  $124 \text{ ng m}^{-3}$  in urban regions of Greece (Tsapakis and Stephanou, 2005, Vasilakos *et al.*, 2007). As compared

**Table 1.** Average concentrations of 16 gaseous and particulate PAHs in three fastener manufacturing industries.

PAHs (ng m <sup>-3</sup> )	Factory A (n = 7)				Factory B (n = 6)				Factory C (n = 21)			
	Particle	Gas	Total	Gas%	Particle	Gas	Total	Gas%	Particle	Gas	Total	Gas%
Nap	4.0	63.0	67.0	94.0	6.0	93.0	100	93.0	8.0	92.0	100	92.0
AcPy	2.0	77.0	79.0	97.5	11.0	43.0	55.0	78.2	3.0	73.0	76.0	96.1
Acp	2.0	76.0	78.0	97.4	7.0	61.0	68.0	89.7	3.0	127	130	97.7
Flu	2.0	69.0	72.0	95.8	6.0	51.0	57.0	89.5	3.0	97.0	101	96.0
PA	3.0	96.0	100	96.0	15.0	90.0	105	85.7	4.0	199	203	98.0
Ant	49.0	48.0	97.0	49.5	36.0	54.0	90.0	60.0	54.0	75.0	129	58.1
FL	4.0	47.0	51.0	92.2	9.0	41.0	50.0	82.0	3.0	60.0	64.0	93.8
Pyr	5.0	40.0	44.0	90.9	11.0	37.0	48.0	77.1	3.0	62.0	65.0	95.4
BaA	1.0	19.3	20.3	95.1	5.0	20.3	25.3	80.2	1.0	20.3	21.3	95.3
CHR	1.3	12.0	13.3	90.2	12.3	12.0	24.7	48.6	1.3	14.3	15.3	93.5
BbF	2.0	11.3	13.3	85.0	1.7	14.3	15.7	91.1	1.3	22.3	23.7	94.1
BkF	2.7	7.7	10.3	74.8	11.0	12.0	23.3	51.5	2.0	26.3	28.3	92.9
BaP	2.7	46.0	48.7	94.5	48.7	38.0	87.0	43.7	8.0	47.3	55.3	85.5
IND	11.0	15.7	26.7	58.8	6.0	43.0	49.3	87.2	10.3	51.7	62.0	83.4
DBA	1.7	21.7	23.3	93.1	3.7	23.0	26.7	86.1	2.7	27.0	29.7	90.9
Bghip	2.3	13.7	16.0	85.6	5.7	29.7	35.3	84.1	3.0	19.7	22.7	86.8
Total	95.7	663	760	87.2	195	662	860	77.0	111	1014	1126	90.1

**Table 2.** Average BaPeq concentrations of 16 gaseous and particulate PAHs in three fastener manufacturing industries.

PAHs (ng m <sup>-3</sup> )	Factory A (n = 7)				Factory B (n = 6)				Factory C (n = 21)			
	Particle	Gas	Total	Gas%	Particle	Gas	Total	Gas%	Particle	Gas	Total	Gas%
Nap	0.004	0.06	0.07	85.7	0.006	0.09	0.1	93.0	0.008	0.09	0.1	90.0
AcPy	0.002	0.08	0.08	100	0.01	0.04	0.06	71.7	0.003	0.07	0.08	87.5
Acp	0.002	0.08	0.08	100	0.007	0.06	0.07	87.1	0.003	0.13	0.13	100
Flu	0.002	0.07	0.07	100	0.006	0.05	0.06	85.0	0.003	0.10	0.10	100
PA	0.003	0.10	0.10	100	0.02	0.09	0.11	81.8	0.004	0.20	0.20	100
Ant	0.49	0.48	0.97	49.5	0.36	0.54	0.90	60.0	0.54	0.76	1.30	58.5
FL	0.004	0.05	0.05	100	0.009	0.04	0.05	82.0	0.003	0.06	0.06	100
Pyr	0.005	0.04	0.04	100	0.01	0.04	0.05	74.0	0.003	0.06	0.07	85.7
BaA	0.11	1.93	2.05	94.1	0.50	2.04	2.54	80.3	0.11	2.04	2.15	94.9
CHR	0.01	0.12	0.13	92.3	0.13	0.12	0.25	48.0	0.01	0.14	0.15	93.3
BbF	0.20	1.15	1.35	85.2	0.15	1.42	1.57	90.4	0.14	2.24	2.37	94.5
BkF	0.27	0.76	1.03	73.8	1.12	1.20	2.32	51.7	0.21	2.63	2.84	92.6
BaP	2.63	46.1	48.7	94.7	48.8	38.1	86.9	43.8	8.00	47.3	55.3	85.5
IND	1.09	1.58	2.67	59.2	0.60	4.30	4.93	87.2	1.03	5.17	6.20	83.4
DBA	1.81	21.6	23.4	92.3	3.73	23.1	26.8	86.2	2.83	27.0	29.8	90.6
Bghip	0.02	0.14	0.16	87.5	0.06	0.30	0.35	85.7	0.03	0.20	0.23	87.0
Total	6.66	74.4	81.0	91.9	55.5	71.5	127	56.3	12.9	88.1	101	87.2

to total concentration of PAHs in particle phase, the average ambient air concentration of 16 PAHs of PM<sub>2.5</sub> downwind of petrochemical industries was 0.389 ng m<sup>-3</sup> (Yuan *et al.*, 2015) and 11.4 ng m<sup>-3</sup> to 44.9 ng m<sup>-3</sup> and 0.44 ng m<sup>-3</sup> to 13.2 ng m<sup>-3</sup> in the particulate phase in urban regions of Greece (Tsapakis and Stephanou, 2005, Vasilakos *et al.*, 2007), which was lower than the amount of inhalable particles measured in the current study (95.7–195 ng m<sup>-3</sup>). While compared the current result to other occupational studies, the total-PAH exposure levels (760 to 104 ng m<sup>-3</sup>) was lower than that of dayshift workers in highway toll station (12,300 ng m<sup>-3</sup>) (Tsai *et al.*, 2004), as well as lower than total PAH exposure level (88300 ng m<sup>-3</sup>) in a fastener manufacturing plant (Chen *et al.*, 2008).

in heat treatment area (1221 ng m<sup>-3</sup>), threading area (920

ng m<sup>-3</sup>), forming area (897 ng m<sup>-3</sup>), packing area (878 ng m<sup>-3</sup>), and acid-cleaning area (850 ng m<sup>-3</sup>); in addition, higher BaPeq levels were found in the packing area (216 ng m<sup>-3</sup>), threading area (103 ng m<sup>-3</sup>), the heat treatment area (99.3 ng m<sup>-3</sup>), the forming area (91.1 ng m<sup>-3</sup>), and the surface treatment area (87.1 ng m<sup>-3</sup>).

The 16 total-PAH levels in different working departments were shown in Table 3. The higher concentrations of 16 total-PAHs were found in heat treatment area (1221 ng m<sup>-3</sup>), threading area (920 ng m<sup>-3</sup>), forming area (897 ng m<sup>-3</sup>), packing area (878 ng m<sup>-3</sup>), and acid-cleaning area (850 ng m<sup>-3</sup>). Meanwhile, of the 16 total-PAHs measured in these factories, except for packing area, over 80% were found to be in the gaseous phase. Table 4 presented the BaPeq levels of the 16 PAHs in different fastener manufacturing areas of

**Table 3.** Average concentrations (ng m<sup>-3</sup>) of 16 gaseous and particulate PAHs by the working process.

Department	acid cleaning	surface treatment	forming	threading	heat treatment	packing	Office-i
N=	4	6	4	4	14	2	4
Nap	110.0	80.0	90.0	70.0	100.0	70.0	70.0
AcPy	60.0	50.0	50.0	120.0	90.0	40.0	40.0
Acp	70.0	70.0	70.0	100.0	160.0	70.0	50.0
Flu	60.0	60.0	70.0	90.0	120.0	60.0	50.0
PA	110.0	80.0	110.0	120.0	260.0	90.0	70.0
Ant	80.0	110.0	100.0	100.0	140.0	110.0	100.0
FL	50.0	40.0	50.0	60.0	70.0	50.0	40.0
Pyr	50.0	40.0	50.0	60.0	70.0	50.0	30.0
BaA	26.7	20.0	20.0	23.3	23.3	26.7	13.3
CHR	30.0	13.3	13.3	16.7	16.7	16.7	10.0
BbF	16.7	20.0	33.3	16.7	20.0	13.3	10.0
BkF	26.7	23.3	50.0	13.3	20.0	16.7	13.3
BaP	40.0	46.7	30.0	66.7	60.0	183.3	40.0
IND	50.0	70.0	113.3	23.3	36.7	46.7	23.3
DBA	30.0	23.3	36.7	26.7	26.7	23.3	16.7
Bghip	46.7	23.3	16.7	16.7	23.3	16.7	10.0
Particulate -PAHs	166 ± 116	110 ± 16	105 ± 10	93 ± 21	112 ± 21	256	105 ± 19
Gas-PAHs	684 ± 165	659 ± 123	792 ± 694	827 ± 422	1109 ± 1028	622	480 ± 70
Total-PAHs	850 ± 99	769 ± 131	897 ± 704	920 ± 439	1221 ± 1030	878	585 ± 71
Gas%	80.5	85.7	88.3	89.9	90.8	70.8	82.1

Note: Office-i means inside the office.

**Table 4.** Mean BaP<sub>eq</sub> levels (ng m<sup>-3</sup>) of 16 gaseous and particulate PAHs by the working process.

Department	acid cleaning	surface treatment	forming	threading	heat treatment	packing	Office-i
N=	4	6	4	4	14	2	4
Nap	0.11	0.08	0.09	0.07	0.1	0.07	0.07
AcPy	0.06	0.05	0.05	0.12	0.09	0.04	0.04
Acp	0.07	0.07	0.07	0.1	0.16	0.07	0.05
Flu	0.06	0.06	0.07	0.09	0.12	0.06	0.05
PA	0.11	0.08	0.11	0.12	0.26	0.09	0.07
Ant	0.81	1.08	0.97	0.99	1.4	1.08	1.08
FL	0.05	0.04	0.05	0.06	0.07	0.05	0.04
Pyr	0.05	0.04	0.05	0.06	0.07	0.05	0.03
BaA	2.50	1.98	1.95	2.41	2.15	2.60	1.39
CHR	0.29	0.13	0.14	0.17	0.16	0.15	0.10
BbF	1.62	2.09	3.48	1.64	1.88	1.48	1.03
BkF	2.57	2.48	4.89	1.16	1.99	1.81	1.31
BaP	39.4	47.3	29.8	67.4	59.2	182	40.0
IND	5.10	6.97	11.23	2.40	3.73	4.60	2.30
DBA	29.2	24.4	38.0	25.9	27.7	22.1	15.9
Bghip	0.45	0.23	0.16	0.17	0.23	0.16	0.10
Particulate -PAHs	16.3 ± 8.0	9.8 ± 7.1	7.0 ± 2.8	7.5 ± 6.4	14.4 ± 20.5	134	10.7 ± 4.4
Gas-PAHs	66.1 ± 42.0	77.3 ± 22.0	84.1 ± 75.7	95.4 ± 40.4	84.9 ± 33.9	82.4	52.9 ± 22.7
Total-PAHs	82.4 ± 40.8	87.1 ± 28.2	91.1 ± 78.5	103 ± 44.9	99.3 ± 40.2	216	63.6 ± 27.0
Gas%	80.2	88.7	92.3	92.8	85.5	38.1	83.2

Note: Office-i means inside the office and Office-o means outside the office.

the three factories. Higher levels of BaP<sub>eq</sub> were found in packing area (216 ng m<sup>-3</sup>), threading area (103 ng m<sup>-3</sup>), heat treatment area (99.3 ng m<sup>-3</sup>), forming area (91.1 ng m<sup>-3</sup>), and surface treatment area (87.1 ng m<sup>-3</sup>). The current data was slightly higher than the fugitive emissions of PM<sub>10</sub> in the vicinity of an integrated iron and steel plant situated in India (2.4 to 231.7 ng m<sup>-3</sup>) (Khaparde *et al.*, 2016).

The gaseous/particulate ratio varied more for BaP<sub>eq</sub> levels than for 16 PAH levels. The gaseous/particulate ratios in packing area were found to differ from those in the other working areas.

When compared to other occupational groups, the particle-PAH concentrations in the different working areas (93–256 ng m<sup>-3</sup>) were lower than those in the continuous casting

factory (465–936 ng m<sup>-3</sup>) (Bergamaschi *et al.*, 2005), and the concentrations of gaseous plus particle PAHs (769–1221 ng m<sup>-3</sup>) was also lower than those obtained in the fastener manufacturing factory (88.3 μg m<sup>-3</sup>) (Chen *et al.*, 2008) and a sinter plant (8.37–30.4 μg m<sup>-3</sup>) (Lin *et al.*, 2008). What were the differences between the metal-related and fastener manufacturing factories? The reasons for the differences between their PAH emissions may be interpreted as follows: (1) MWFs are widely used in fastener manufacturing factories, such as in forming, threading, and de-oiling via heat treatment, and PAH emission is predominant in these working areas. (2) The literature have shown that high temperature will cause the PAHs emission (Chen *et al.*, 2014) and the gaseous PAHs will elevated in summer (Cheruiyot *et al.*, 2015). Therefore, the heat treatment processes may be the primary sources of PAHs due to the high temperatures involved in these processes. However, the cover on top of the furnace is usually closed and is only opened when a treatment process is completed; thus, emission from this source takes place only when the cover is opened. (3) The particulate concentrations and PAH emissions in the packing and acid cleaning areas are unexpectedly high, compared to those in the forming and threading areas, which are expected to be a source of PAH emission. This scenario may be due to packing and acid cleaning areas existing in only Factory B, which is a small factory (the acid cleaning area is 150 m<sup>2</sup> in a separated space, and the packing area is situated in a plant of 150 m<sup>2</sup> without any partition) with poor ventilation and no air-conditioning equipment. (4) Comparing the proportions of gaseous-phase and particulate-phase PAHs, the proportions of gaseous-phase PAHs are 80.5% and 70.8% in the acid cleaning area

and the packing area, which are lower than in other areas (85.7%–90.8%). This data indicates that the sources of PAHs may be different in the two working areas.

In order to compare the patterns in the air samples containing PAHs in the three fastener factories, we used PCA on the congener profiles, with 16 PAH concentrations as variables. Factors 1 and 2 explained 43.8% and 26.9%, respectively, of the total variance; together, they account for 70.7% of the total variance (Fig. 4). The score plot showed that the samplings were clustered into two groups: Group 1 (three- and four-ring PAHs, including AcPy, Acp, Ant, PA, FL, and Pyr), and Group 2 (five- and six-ring PAHs, including BbF, DBA, BkF, and IND), which represented Factors 1 and 2, respectively, that influenced the fastener factory air samples. Five- and six-ring PAHs were often predominant in the particulate phase, but the gaseous and particulate phases varied according to the environmental conditions (Pleil *et al.*, 2004). Meanwhile, five- and six-ring PAHs were predominantly toxic (Chen *et al.*, 2008; Liu *et al.*, 2010), and BaP was the predominant source of the toxicity in this type of environment.

In the current study, the gaseous phase contributed 77.0–90.1% of the 16 PAH levels in Factories A, B, and C. These values were lower than 97.4%–98% of contributions in the workplace atmospheres of iron foundries, sinter plants (Omland *et al.*, 1994; Lin *et al.*, 2008, 2010), fastener manufacturing factory (Chen *et al.*, 2008) and the value of almost 90% that was found in suburban areas (Vasilakos *et al.*, 2007). Shimmo *et al.* (2004) and Cheruiyot *et al.* (2015) indicated that the total particulate PAH concentrations were higher for particles between 440 nm and 2.5 μm. Meanwhile, it has been suggested that shifts in the gaseous/particulate

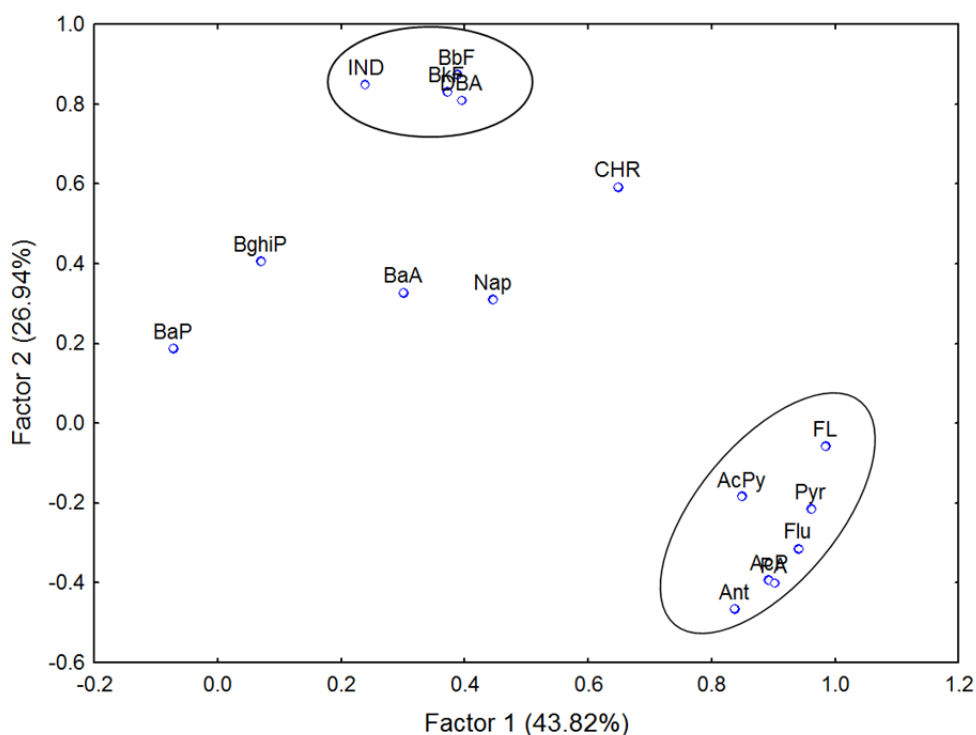


Fig. 4. The score plot from PCA by using gaseous PAHs concentrations in the air samples as the variables.

distribution were due to differences in the ambient temperature, which accounted for seasonal variations in the concentrations of particulate PAHs. Because high temperatures were used only in heat treatment processes in fastener factories, the possible influencing factors might include the type of MWFs used (Park *et al.*, 2009), the particle size, and the temperatures in the working areas, such as those in the forming, threading, and heat-treatment areas.

### Health Risk Assessment of PAHs Exposure in Different Working Departments

Regarding to the inhalation pathway, lung cancer risk calculations based on time-weighted average exposure levels of BaP<sub>eq</sub> for fastening industry workers were presented in Table 5. The risk unit of BaP<sub>eq</sub> was adopted from Nisbet and LaGoy (1992) and Lin *et al.* (2008). The cancer risks due to PAH exposure in the packing, threading, heat treatment, forming, surface treatment and acid cleaning areas were  $1.14 \times 10^{-3}$ ,  $5.45 \times 10^{-4}$ ,  $5.25 \times 10^{-4}$ ,  $4.82 \times 10^{-4}$ ,  $4.61 \times 10^{-4}$ , and  $4.36 \times 10^{-4}$ , respectively. For the particulate-phase risk, except for that in the packing area, which was  $7.09 \times 10^{-4}$ , those in other working areas were all below  $10^{-4}$ . For the gaseous phase, the highest cancer risk was in the threading area ( $5.05 \times 10^{-4}$ ). Chen *et al.* (2008) mentioned that the estimated lifetime lung cancer risk ( $1.64 \times 10^{-2}$ ) was higher than the skin cancer risk ( $9.72 \times 10^{-3}$ ) in threading workers in a fastener manufacturing factory. Although the average total BaP<sub>eq</sub> exposure levels were lower PEL-TWA ( $0.2 \text{ mg/m}^3$ ), the lung cancer risks associated with the above PAH exposures ranged from  $4.36 \times 10^{-4}$  to  $1.14 \times 10^{-3}$ , which were equal to or higher than those for foundry workers ( $9.06 \times 10^{-4}$ – $1.09 \times 10^{-3}$ ) (Liu *et al.*, 2010), coking wastewater treatment workers ( $0.1 \times 10^{-3} \pm 0.1 \times 10^{-4}$  to  $5.2 \times 10^{-3} \pm 0.5 \times 10^{-3}$ ) (Zhang *et al.*, 2012), sinter plant workers ( $3.18 \times 10^{-5}$ – $4.98 \times 10^{-5}$ ), and temple workers ( $10^{-6}$ – $10^{-4}$ ) who had occupational PAH exposure (Lin *et al.*, 2008; Chiang *et al.*, 2009), whereas the lung cancer risk in the current study was lower than that for carbon black workers ( $4.35 \times 10^{-2}$ ) (Tsai *et al.*, 2001) and fastener workers ( $1.64 \times 10^{-2}$ ) (Chen *et al.*, 2008). Compared to the general environmental risk, the cancer risk in the current study was higher than the PAH exposure risk from settled house dust

and street dust ( $> 10^{-4}$ ) in China (Wang *et al.*, 2013), as well as the risks for kitchen workers (cancer risk based on dust sampling:  $6.8 \times 10^{-5}$ ) (Singh *et al.*, 2016), traffic assistants in China (the cancer risk from particle PAHs is approximately  $1.05 \times 10^{-4}$ ) (Xue *et al.*, 2014) and general from the ambient air in cities (Wu *et al.*, 2014). Whereas it was equal to the cancer risk obtained from environmental sampling near three large power plants ( $4.7 \times 10^{-4}$ – $2.4 \times 10^{-3}$ ) (Wang *et al.*, 2015). The previous study have mentioned that there are general risk of exposure to PAHs emissions, such as during household use of fuels ( $3.84 \times 10^{-6}$  to  $9.11 \times 10^{-5}$ ) (Tiwari *et al.*, 2015) and ingestion of house dust ( $8.525 \times 10^{-4}$  and  $2.558 \times 10^{-3}$ ) (Pongpiachan *et al.*, 2016) in Asian people of their lifetime, but the occupational risk of PAHs exposure was relative important if a worker's job was related to the fastener manufacturing process (maximum:  $1.14 \times 10^{-3}$ ). The occupational hazard prevention should not only focus on particulate-phase, but also on gaseous-phase, PAH exposure prevention.

### CONCLUSIONS

The particle measurements in the fastener manufacturing factories were significantly higher than the offices inside the factories. Only a sampling in acid cleaning area (1/4, 25%) and a sampling in heat treatment area (1/16, 6.25%) have over PEL-TWA of  $5 \text{ mg m}^{-3}$  for oil mist. The contribution of gaseous phase to total PAHs was over 80% in air samples taken inside the industries. As Wu *et al.* (2014) mentioned that seasonal high temperature in summer may release much higher levels of gas phase PAHs, the high temperature in heat treatment processes may cause the release of PAHs with the widely usage of MWFs. Although the average total BaP<sub>eq</sub> exposure levels were lower than PEL-TWA ( $0.2 \text{ mg m}^{-3}$ ), the lung cancer risks of PAH exposure in different working areas ranged from  $4.36 \times 10^{-4}$  to  $1.14 \times 10^{-3}$ . The lung cancer risk due to gaseous phase PAH exposure was much higher than that of particulate phase PAH exposure, therefore, the occupational hazard prevention was not only focused on particle exposure prevention, but also in gaseous PAHs exposure concern if the worker's job integrates the manufacturing fastener processes.

**Table 5.** Time-weighted average exposure levels of total BaP<sub>eq</sub> and the cancer risk of workers exposed to gaseous/particle phase PAHs between different working areas.

	acid cleaning	Surface treatment	forming	tapping/ threading	Heat treatment	packing	Office-i
Unit risk <sup>#</sup>	$7.00 \times 10^{-5}$						
BaP <sub>eq</sub> (ng m <sup>-3</sup> )							
Particle -phase	48.1	28.2	19.8	21.5	42.1	400.4	31.1
Gas -phase	196.5	230.0	250.5	283.8	251.4	245.4	156.9
Cancer risk <sup>1</sup>							
Particle -phase	$8.63 \times 10^{-5}$	$5.19 \times 10^{-5}$	$3.70 \times 10^{-5}$	$3.97 \times 10^{-5}$	$7.62 \times 10^{-5}$	$7.09 \times 10^{-4}$	$5.66 \times 10^{-5}$
Gas -phase	$3.50 \times 10^{-4}$	$4.09 \times 10^{-4}$	$4.45 \times 10^{-4}$	$5.05 \times 10^{-4}$	$4.49 \times 10^{-4}$	$4.36 \times 10^{-4}$	$2.80 \times 10^{-4}$
Total cancer	$4.36 \times 10^{-4}$	$4.61 \times 10^{-4}$	$4.82 \times 10^{-4}$	$5.45 \times 10^{-4}$	$5.25 \times 10^{-4}$	$1.14 \times 10^{-3}$	$3.37 \times 10^{-4}$

<sup>#</sup> The unit risk was adopted from Lin *et al.* (2008), unit (ng m<sup>-3</sup>)<sup>-1</sup>

<sup>1</sup> Risk of cancer = Air Levels (μg m<sup>-3</sup>) × Unit Risk (μg m<sup>-3</sup>)<sup>-1</sup> × 8/24 × 25/78.74 × (5 × 10<sup>-4</sup> – 4)/7.



## NOMENCLATURE AND ABBREVIATIONS

PAHs: polycyclic aromatic hydrocarbons,  
 total-PAH: 16 polycyclic aromatic hydrocarbons  
 BaP<sub>eq</sub>: benzo[a]pyrene toxic equivalent,  
 Nap: naphthalene,  
 AcPy: acenaphthylene,  
 AcP: acenaphthene,  
 Flu: fluorene,  
 PA: phenanthrene,  
 Ant: anthracene,  
 FL: fluoranthene,  
 Pyr: pyrene,  
 BaA: benzo[a]anthracene,  
 CHR: chrysene,  
 BbF: benzo[b]fluoranthene,  
 BkF: benzo[k]fluoranthene,  
 BaP: benzo[a]pyrene,  
 IND: indeno[1,2,3-c,d]pyrene,  
 DBA: dibenz[a,h]anthracene  
 BghiP: benzo[g,h,i]perylene

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