

Research Article

Assessing Consumer Exposure to Particle-Bound Polycyclic Aromatic Hydrocarbons in Parking Areas

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Background: The rapid growth of industrial activities, population and traffic density, people in Taiwan are facing serious air pollution problems. Studies show that vehicle exhaust from traffic has been proven to be one of the largest contributors of PAHs in urban areas; while large-scale enclosed and semi-enclosed vehicle parking areas were also should be concerned.

Methods: We used a 15-channel dust monitor (Grimm 1.108) and combine with particle-bound PAHs (p-PAHs) detection device (Model #130) for measuring the real-time p-PAHs concentration in parking area. Consumer exposure parameters were estimated *via* questionnaire survey. Moreover, a probabilistic risk assessment for different transportation populations (car- and scooter-using) was conducted based on the Incremental Lifetime Cancer Risk (ILCR) model with a Monte Carlo simulation.

Results: We discovered that the fraction of fine p-PAHs (0.23–1 μm) to total PAHs to be nearly 55%, 45–46%, and 32–37% in outdoor, first floor, and basement floor in parking areas, respectively. Furthermore, the fraction of PAHs measuring <10 μm to total PAHs averaged 88%. These findings implicate fine p-PAHs caused by vehicle exhaust as a matter of health concern. Our results indicate that the 95% confidence intervals for benzo[a]pyrene equivalent concentrations (BaP_{eq}) and the inhalation risk were 1.52–6.05 and 9.11×10^{-8} for car-using and 1.70–5.42 $\mu\text{g}/\text{m}^3$ and 1.04×10^{-7} for scooter-using populations, respectively.

Conclusion: We conclude that consumers exposed to p-PAHs in parking areas are not likely to encounter a potential cancer risk. However, the long-term exposure for combined with other exposure routes and sources should be concerned.

Keywords: Benzo[a]pyrene (BaP); Exposure; Parking area; Inhalation risk

Introduction

Polycyclic Aromatic Hydrocarbons (PAHs) are a large group of organic contaminants that composed of particulate or gaseous matter. According to the U.S. Environmental Protection Agency, there are 16 priority pollutant PAHs that are classified as probable human carcinogens [1,2]. Moreover, the carcinogenic effects of PAHs have been observed experimentally and epidemiologically [3,4].

The major sources of PAHs include fossil fuel combustion, vehicle exhaust, and industrial emissions, because PAHs are mainly formed *via* the incomplete combustion or pyrolysis of organic materials such as coal, oil, and petrol [1,2]. Studies show that vehicle exhaust from traffic has been proven to be one of the largest contributors of PAHs in urban areas; this along with population density usually causes these regions to contain the highest concentrations of PAHs [5,6].

In the past decades, studies have characterized PAH emissions by vehicle exhaust [7], indicating an association with fuel type (leaded/unleaded gasoline, diesel) [8]. Spatial variation was estimated from traffic-related sampling locations such as highway tunnels [9], roadsides and intersections [8,10], and large-scale enclosed and semi-enclosed vehicle parking areas [11]. Moreover, it is important to

evaluate traffic-related workers' exposure to PAH concentrations as a result of vehicle exhaust. Hu et al. [12] and Tsai et al. [13] assessed the inhalation risk for traffic policemen and highway toll station workers from vehicle emissions, respectively, implicating the potential risk and importance of human exposure to particle-bound PAHs (p-PAHs).

Recently, Li et al. [11] estimated the carcinogenic potential of airborne PAHs in enclosed and semi-enclosed vehicle parking areas. They indicated that the PAH concentrations of these parking areas ranged from 1,178–4,793 ng/m^3 and the Benzo[a]pyrene equivalent (BaP_{eq}) values varied from 11.0–98.0 ng/m^3 , exceeding the air quality standard of WHO (1.0 ng/m^3). Lovinsky-Desir et al. [14] also evaluated the differences in PAHs between streets (open spaces) and alleys (semi-closed space) in New York City. The small-scale spatial variations of PAH concentration were also highlighted [14].

In central Taiwan, for the year 2015, the population density of Taichung city and county was estimated at 587.23 people per km^2 on average. As of 2015, the numbers of cars and scooters were estimated to be 379.89 and 601.53 per thousand people, respectively [15]. Parking is one of the most widespread and frustrating problems afflicting consumers when shopping at places like department stores or supermarkets. The worst air quality can be found in enclosed

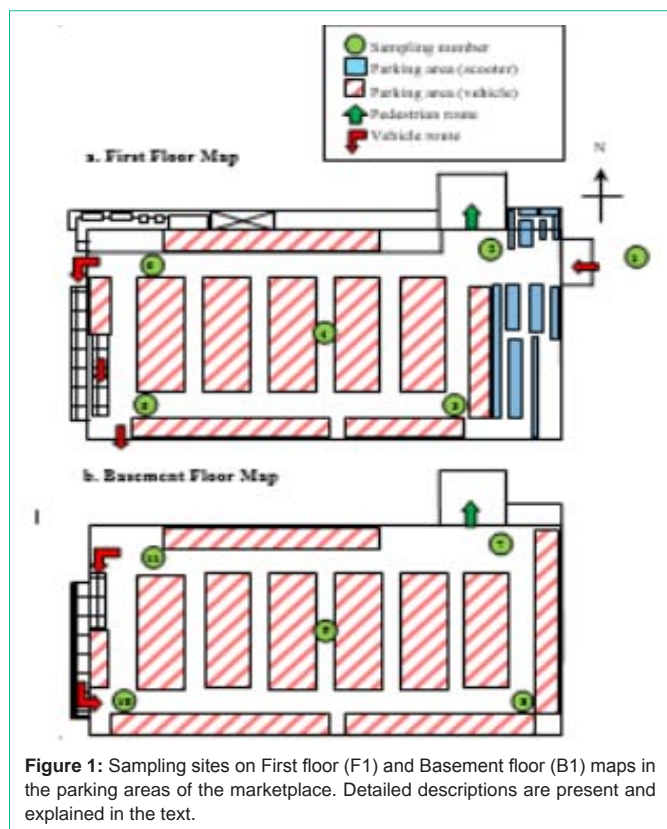


Figure 1: Sampling sites on First floor (F1) and Basement floor (B1) maps in the parking areas of the marketplace. Detailed descriptions are present and explained in the text.

parking towers or underground parking areas. Hence, in our study, we conducted sampling in parking areas to investigate the exposure factors for consumers given a questionnaire.

We previously assessed the health risks for humans exposed to environmental PAH pollution sources and estimated the inhalation risk for consumer exposure to p-PAHs in Taiwanese night market [16,17]. Thus, the aim of this study is to estimate consumer exposure to p-PAHs in parking areas in Taiwan, especially for different transportation modes and fuels used. Specifically, our aims were (i) to conduct a survey assessing the exposure factors, (ii) to estimate a time-dependent p-PAH concentrations across enclosed and semi-enclosed parking areas based on experimental sampling, and (iii) to assess the probabilistic risk assessment for different groups inhaling p-PAHs based on an Incremental Lifetime Cancer Risk (ILCR) model.

Methods and Materials

Study design and sampling

The present study measured the p-PAHs concentrations in one marketplace in central Taiwan from 30 July through 4 August 2015, during which four weekdays (THU, FRI, MON and TUE) and two weekends (SAT and SUN) were included. In considering the difference of human activity during specific times, we set the sampling time to be 17:00–20:00 and 13:00–16:00 on weekdays and weekends, respectively, based on the busiest times.

Figure 1 presents the sampling sites and positions of the marketplace on the First floor (F1) and Basement floor (B1). The second floor is marketplace only. A total of 11 sampling sites were chosen; Site number 1 (S1), numbers 2–5 (S2–S5), and numbers 6–11

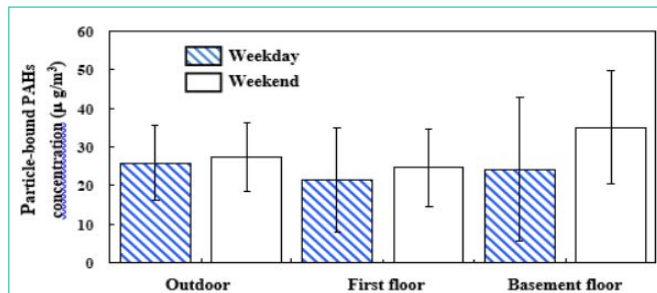


Figure 2: Total p-PAHs concentration ($\mu\text{g}/\text{m}^3$) at the First floor (F1), Basement floor (B1) and outdoor areas. The histogram bars indicate the Standard Deviation (SD) estimations.

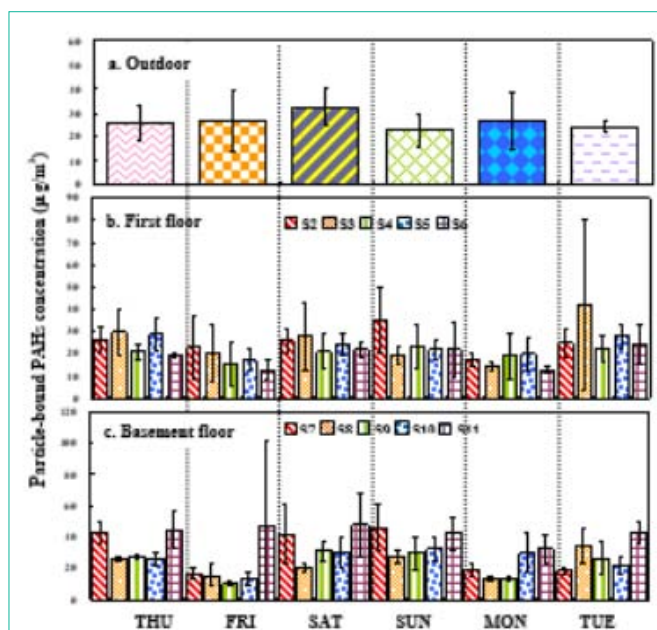


Figure 3: Total p-PAHs concentration ($\mu\text{g}/\text{m}^3$) (a) outdoors, (b) on the First floor (F1), and (c) on the Basement floor (B1) of each Sampling site (S1 – S11) and sampling day. The histogram bars indicate the estimation of Standard Deviation (SD).

(S6–S11) represent the outdoor areas of F1, indoor areas of F1 and indoor areas of B1, respectively, where the average concentration of p-PAHs was collected. S2 and S7 were the nearest entrances for consumers between parking areas and the marketplace. S5 was the only way to exit the parking area for vehicles. We followed the sampling numbers from S1 to S11 for 10 minutes at each site. The average concentration of p-PAHs at each site was analyzed. In (Figure 1), blue and red areas represented the scooter and car parking lots with 226 and 466 parking spaces, respectively; and there were 833 m² and 7,936 m² contained by the F1 and B1 indoor areas, respectively. The green and red arrows indicate pedestrian and vehicle routes, respectively.

Real-time p-PAHs were detected by a particle-bound PAH detection device (Model #130) which was incorporated into a 16-channel dust monitor (Grimm 1.108, Germany). Detailed descriptions with Grimm 1.108 and instrument principle were adopted from Chen et al. [17]. We also recorded temperature, RH, and traffic volume during the sampling duration. An LED car counter

for parking places was utilized.

Exposure assessment

A survey was conducted from 13:00 to 16:00 during 13–14 August, 2015. The study populations were chosen randomly from consumers who arrived at the marketplace by car or other transportation. In order to avoid a selection bias, we limited questionnaire responders to a maximum of two participants from one family or one close group.

Participants were asked to provide information regarding gender, age (<10, 10–19, 20–29, 30–39, 40–49, 50–59 or >60 years old), education level (graduate school, university/college, senior high school, junior high school, or elementary school), body weight (kg), types of transportation used (car, scooter, or electric scooter), and types of fuel used (98/95/92 lead-free gasoline or diesel) at the beginning of the questionnaire. We also investigated the exposure items including visit frequency (1, 2, 3, 4, 5 times/month), time taken for parking (<1, 1–3, 3–5, 5–10 or >10 minutes), time to marketplace (<5, 5–10, 10–15, 15–20 or >20 minutes), and first time to the marketplace (<1, 1, 2, 3 or >3 years ago) (Appendix A). The survey questions and answer types were reviewed by three experts to confirm both the effectiveness of the questionnaire and that accurate information was provided. This questionnaire was also approved by the Institutional Review Board of the Ethical Committee of Chung Shan Medical University (CSMUH No: CS16046).

Based on the survey, we analyzed the frequency and probabilistic distribution for each exposure factor (variables) including “visit frequency”, “time for parking”, “time for marketplace”, and “first time to this marketplace” (Appendix A). A Monte Carlo (MC) analysis was used to incorporate variability among different parameters and to estimate the uncertainty of inhalation risk for consumers. The MC simulation was implemented using Crystal Ball software (Version 2000.2, Decisioneering Inc., Denver, CO, USA).

Potential toxicity based on carcinogenic PAHs

Toxic Equivalency Factors (TEFs) were estimated carcinogenicity relative to benzo[a]pyrene and used to assess the risks associated with consumer exposure to PAHs. Each BaP equivalent (BaP_{eq}) were calculated for car-using (i=1) and scooter-using populations (i=2) by multiplying each PAH concentration with its corresponding TEF as follows:

$$BaP_{eq} = \sum_{j=1}^{21} C_i \times F_j \times TEF_j$$

Where $C_{i=1}$ and $C_{i=2}$ represent the consumer exposure PAH concentration for car-using and scooter-using populations, respectively. For estimating exposure concentration, we averaged PAH concentrations at sample sites 2 to 11 and assumed it to be the concentration for the car-using population, and averaged PAH concentrations at sample sites 2 and 3 for the scooter-using population. F_j Expresses the fraction of each individual p-PAH to total PAHs [18]. TEF_j Expresses the TEF for each individual PAH j ($j = 1 - 21$). Sheu et al. [18] reported 21 p-PAH concentrations in three sampling sites including a traffic intersection, urban site, and petrochemical industrial plant in Tainan, Taiwan. We estimated the average fractions from particles sized 0.056 to 10 μm in traffic exhaust only (Appendix B).

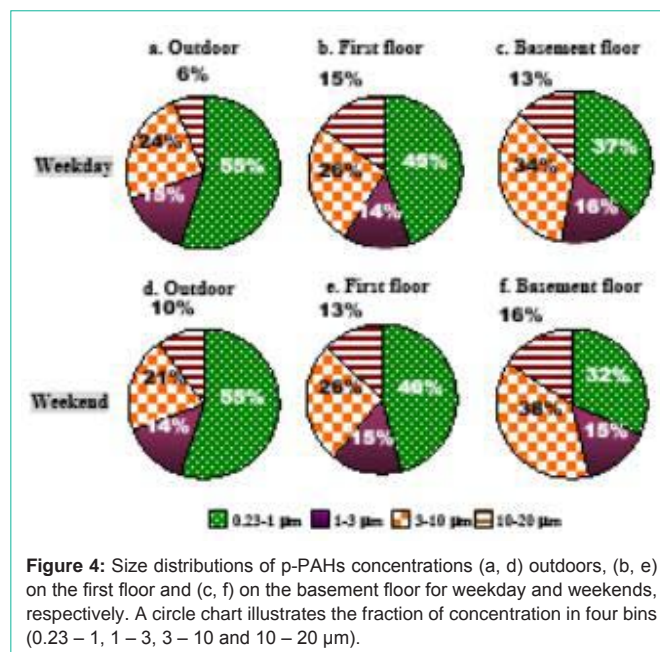


Figure 4: Size distributions of p-PAHs concentrations (a, d) outdoors, (b, e) on the first floor and (c, f) on the basement floor for weekday and weekends, respectively. A circle chart illustrates the fraction of concentration in four bins (0.23 – 1, 1 – 3, 3 – 10 and 10 – 20 μm).

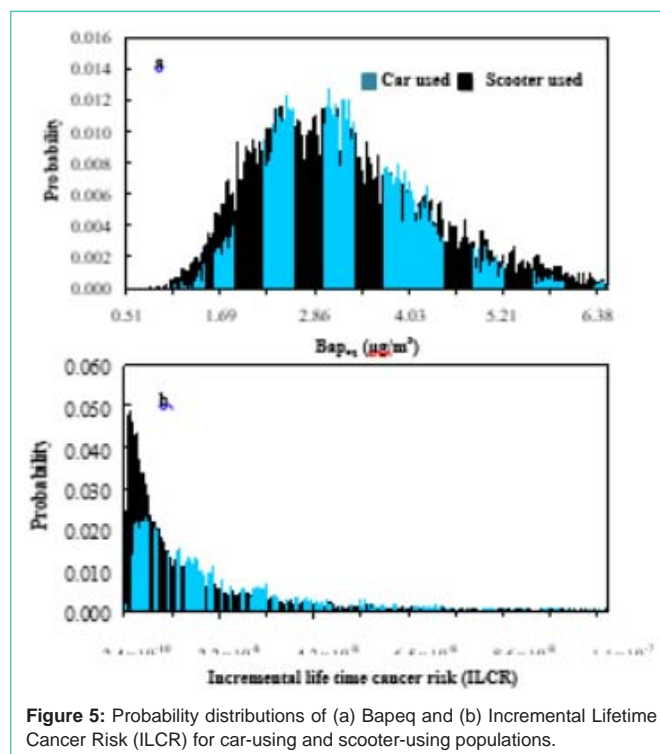


Figure 5: Probability distributions of (a) BaPeq and (b) Incremental Lifetime Cancer Risk (ILCR) for car-using and scooter-using populations.

Inhalation exposure risk

Our probabilistic risk model integrates BaPeq values and the Incremental Lifetime Cancer Risk (ILCR) approach to quantitatively estimate PAH exposure risk for car-using and scooter-using populations. The ILCR model for human inhalation was defined as:

$$R = BaP_{eq} \times CSF \times IR \times EF \times ED / AT \times BW$$

Whereis the incremental individual lifetime cancer risk by the inhalation route, BaPeq is the BaP equivalent concentration for the car-using and scooter-using populations, CSF is the inhalation cancer

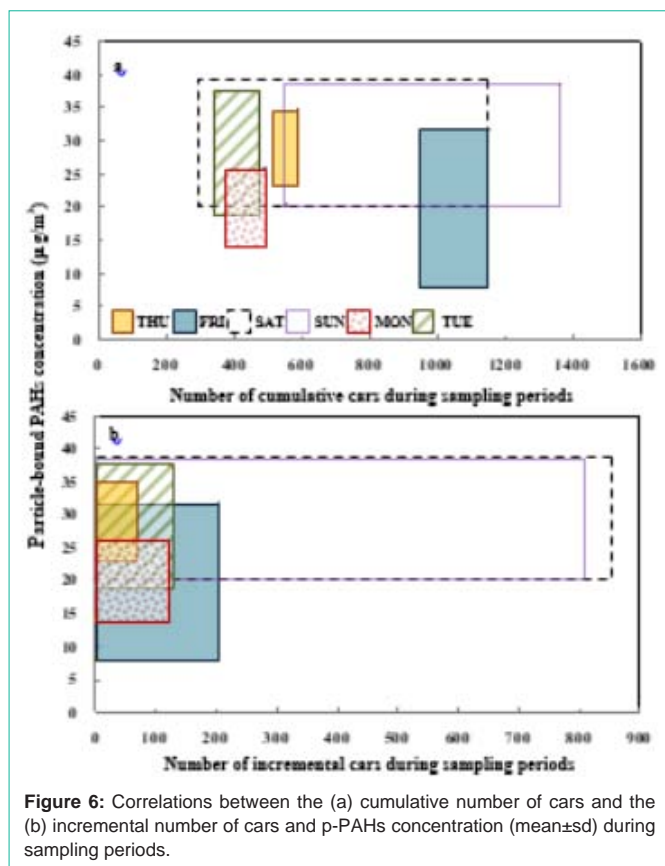


Figure 6: Correlations between the (a) cumulative number of cars and the (b) incremental number of cars and p-PAHs concentration (mean±SD) during sampling periods.

slope factor (mg/kg-day)⁻¹. We estimated a log-normal distribution with a geometric mean of 3.14 (mg/kg-day)⁻¹ and a geometric standard deviation of 1.80 based on hamsters exposure to BaP experiment from Collins et al. [19]. IR is the inhalation rate (m³/day), EF is the exposure frequency (day/year), ED is the exposure duration (year), AT is the averaging time for carcinogenic effects (days), and BW is the body weight (kg). Exposure Frequency (EF) and Exposure Duration (ED) were adopted from the questionnaire. For the car-using population, EF was estimated by “visit frequency” multiplied by “time to marketplace”; for the scooter-using population, EF was estimated by “visit frequency” multiplied by “time for parking” plus “time to marketplace”. ED was cited from the variable of “first time to the marketplace”. An averaging time of 365 d/year for 70 years was used to characterize lifetime exposure for cancer risk.

We treated CSF, IR, EF, ED, and BW in Eq. (2) probabilistically. For explicating quantify the uncertainty of the data, a MC simulation was performed with 10,000 iterations via the random sampling method to obtain a 95% confidence interval.

Results

PAH concentrations in parking area

Figure 2 shows total p-PAH concentrations (µg/m³) at First floor (F1), Basement floor (B1) and outdoors. In parking areas, PAH concentrations in B1 exhibited higher levels than those in F1: 24.33 ± 18.71 (mean ± SD) and 21.51 ± 3.54 µg/m³ for weekdays, respectively, versus 35.18 ± 14.66 and 24.67 ± 10.15 µg/m³ for weekends. The main reason could arise from the ventilation conditions in the indoor

environment. There are three walls that have outside facing windows in F1, however B1 is a window-free parking space. Temperature and relative humidity ranged from 28–34°C and 45–65%, respectively, during the sampling period.

Figure 3 demonstrates the detailed PAH concentrations during each day and at each sampling site. For the F1 parking area, the average PAH concentrations at sites 2 and 3 showed higher levels, estimated to be 25.32 and 25.72 µg/m³, respectively. For the B1 parking area, the average PAH concentrations at sites 7 and 11 showed higher levels and were estimated to be 30.98 and 42.82 µg/m³, respectively. Based on the map in (Figure 1), sites 2 and 7 were the nearest locations to the marketplace. There are barrier machines at the parking exit (site 5) and parking entrance (site 2). On the other hand, there is no outside-facing window at site 3.

Size distribution of particle-bound PAHs

Particle sizes were divided into four bins (0.23–1, 1–3, 3–10, and 10–20 µm) for describing the fraction of each size distribution with fine and coarse particles (Figure 4). The fraction of particle sizes between weekdays and weekends was not significantly different (Figure 4a *verse* 4d, Figure 4b *verse* 4e, and Figure 4c *verse* 4f) at the same sampling site. However, we discovered that the fraction of fine p-PAHs (0.23–1 µm) to total PAHs to be nearly 55%, 45–46%, and 32–37% in outdoor, first floor, and basement floor, respectively. Furthermore, the fraction of PAHs measuring <10 µm to total PAHs averaged 88%. These findings implicate fine p-PAHs caused by vehicle exhaust as a matter of health concern.

Participant demographics

Table 1 lists the statistical summary for the questionnaire. The effective population size was 182 with a response rate of 91%. Characteristics of participants include a majority being female (55%) and in the age groups between 20–60 years (97%). Transportation by car or scooter was estimated to be 80% and 20%, respectively. Fuels used for lead-free gasoline were 98 (8%), 95 (78%), and 92 (10%). The results for exposure factors indicate that 43% of consumers visited the marketplace once per month and the time taken for parking at the marketplace was <1 minutes for 33% and <5 minutes for 75% of respondents. In total, 51% of participants visited the marketplace less than once per year.

Bap_{eq} and incremental lifetime cancer risk

In terms of (Equation 1), C₁ and C₂ the respective particle-bound PAH concentrations for the car- and scooter-using populations were estimated to be 25.97 ± 9.61 (mean ± SD) and 25.52 ± 7.69 µg/m³, respectively. Therefore, Bapeq was 3.24 ± 1.19 (mean ± SD) and 3.19 ± 0.95 µg/m³ for the car- and scooter-using populations, respectively. The related estimated process was described in (Appendix B).

Table 2 shows the exposure factors, units, and estimations for different exposure groups of inhalation lifetime cancer risk caused by exposure to p-PAHs in these parking areas. The probabilistic distributions and statistical descriptions of the parameters “exposure frequency”, “exposure duration”, and “body weight” were estimated by goodness of fit with chi-square test, and found *via* the best fitted distribution for survey data (Appendix C and D).

Our results indicated that the 95% confidence intervals for BaP_{eq} were 1.52–6.05 and 1.70–5.42 µg/m³ for the car- and scooter-using

Table 1: Participant demographics ($n = 182$).

Variables	Items	Number	%
Gender	Male	82	45.1
	Female	100	54.9
Age (year)	<10	1	0.5
	10-19	3	1.6
	20-29	40	22.0
	30-39	66	36.3
	40-49	59	32.4
	50-59	11	6.0
	>60	2	1.1
Educational level	Graduate school	98	53.8
	University/College	51	28.0
	Senior High school	27	14.8
	Junior high school	5	2.7
	Elementary school	1	0.5
Body weight (kg)	<40	1	0.5
	40-49	19	10.4
	50-59	61	33.5
	60-69	40	22.0
	70-79	49	26.9
	80-89	6	3.3
	>90	6	3.3
Types of transportation	Car	146	80.2
	Scooter	36	19.8
	Electric scooter	0	0
Types of fuel	98 lead-free gasoline	15	8.2
	95 lead-free gasoline	142	78.0
	92 lead-free gasoline	19	10.4
	Diesel	6	3.3
Visit frequency (times/month)	1	78	42.9
	2	47	28.8
	3	19	10.4
	4	6	3.3
	5	32	17.6
Time for parking (minutes)	<1	60	33.0
	1-3	50	27.5
	3-5	46	25.3
	5-10	12	6.6
	>10	14	7.7
Time to marketplace (minutes)	<5	137	75.3
	5-10	32	17.6
	10-15	3	1.6
	15-20	1	0.5
	>20	9	4.9
First time to this marketplace (years ago)	<1	93	51.1
	1	22	12.1
	2	30	16.5
	3	20	11.0
	>3	17	9.3

populations (Figure 5a). The 97.5 percentile for inhalation risk was 9.11×10^{-8} and 1.04×10^{-7} for car- and scooter-used populations, respectively (Figure 5b). Liao and Chiang [20-23] indicate an ILCR between 10^{-6} and 10^{-4} expresses a potential risk, whereas an ILCR $> 10^{-4}$ indicates a high potential health risk. Therefore, our results imply that consumers exposed to p-PAHs in parking areas are not likely to be exposed to a potential cancer risk.

Discussion

This study conducted a questionnaire and measured the particle-bound PAH concentrations in parking areas. We used a probabilistic ILCR model to estimate the inhalation risk for consumers and treated several input parameters probabilistically. Generally, exposure assessment was a critical step in the risk assessment process. Previous studies always made assumptions to estimate the exposure factors and parameters. However, in our study, a questionnaire was used to investigate and characterize the visiting frequency, duration, and even the body weight of consumers. It is a more precise method for quantitatively estimating inhalation risk exposure to p-PAHs concentration.

Based on our results, several points were provided to discuss including (i) change in particle size distribution within different sampling locations, and (ii) correlation analysis between traffic volume and PAHs levels.

Variability in the particle size distribution within different sampling locations was also observed. Measurement results indicate that the fraction of fine p-PAHs (0.23–1 μm) to total PAHs to be nearly 55% and 32–37% in outdoor roadside and basement floor parking areas, respectively. The fraction in the outdoor roadside was reasonable because the fresh aerosols generated from traffic sources were primarily composed of fine particles. The result agrees with Sheu et al.'s [18] study indicating that a smaller particle has a higher total PAH content. This is due in part to the fact that soot from combustion sources is primarily made up of fine particles. In the basement floor, however, poor ventilation might cause the coagulation process to occur and gradually decrease the fraction of fine-mode particles. Meanwhile, particles with a size of 10–20 μm might be deposited on the floor surface and increase the fraction of particles with 3–10 μm diameters. This phenomenon of size change is reflected in other studies [20,21].

Figure 6 shows the correlation between traffic volumes and PAH levels. We illustrated the cumulative or incremental car numbers with PAH levels during each sampling date range. Results show that the maximum cumulative car counts were present on weekends (SAT and SUN) with 1,152 and 1,361 cars, respectively. There were nearly 1.86 times the maximum cumulative car counts on weekends compared to weekdays (Figure 6a). Particle-bound PAH concentrations highly varied with the number of incremental cars, especially on weekends. On weekends, there were nearly 6.42 times the incremental car numbers than on weekdays. The linear equation was estimated to be ($R_2 = 0.25$). It might imply that exhaust from car emissions highly contributed as sources of PAHs (Figure 6b).

In this study, several limitations were included. First, the individual PAH concentration was estimated by fractional values from Sheu et al. [18]. Thus, the estimation process for each individual

Table 2: Exposure scenarios and exposure factors.

Exposure factors	Symbols (unit)	Exposure population		Data sources
		Car-used (n=146)	Scooter-used (n=36)	
Exposure concentration	BaP _{eq} (µg/m ³)	3.24 ^a	3.19 ^b	Estimated by Eq. (1)
Cancer slope factor	CSF (mg/kg-day) ⁻¹	LN (3.14, 1.8) ^c	LN (3.14, 1.8)	Collins et al. [19]
Inhalation rate	IR (m ³ /day)	LN (7.01, 1.27)	LN (7.01, 1.27)	ICRP [23]
Exposure frequency	EF (day/year)	0.217 ^d	0.234 ^e	Questionnaire (Appendix C and D)
Exposure duration	ED (year)	LN (0.99, 2.17) ^f	U (0.41, 2, 3.59) ^{f, g}	Questionnaire (Appendix C and D)
Body weight	BW (kg)	U (34.1, 64.5, 94.9)	U (43.1, 69.5, 95.9)	Questionnaire (Appendix C and D)
Averaging time	AT (days)	25550	25550	-

a) Average PAH concentrations at sample sites 2 to 11.

b) Average PAH concentrations at sample sites 2 and 3.

c) LN = lognormal distribution with geometric mean (gm) and geometric standard deviation (gsd).

d) Exposure frequency = Visit frequency (times/month)*Time to marketplace (minutes spent each time)*12 (months/year).

e) Exposure frequency = Visit frequency (times/month)*[Time for parking + time to marketplace](minutes spent each time)*12(months/year).

f) Exposure duration was gathered from the variable "First time to this marketplace"

g) U = Uniform distribution with min, mean, and max values.

PAH compound had biases. The gas-phase PAH concentration was not included in calculating BaP_{eq}. Additionally, this study only considered the inhalation risk, which does not include dermal contact and ingestion exposure pathways.

Conclusion

Overall, in the parking area, the basement floor shows higher PAH levels than those on the first floor. The highest PAH levels were discovered at the location nearest to the marketplace and in unventilated areas. Correlation analysis also shows that the exhaust from vehicle emissions in parking areas highly contributed as a source of PAHs. In addition, we determined that the inhalation risk for cancer among both car- and scooter-using populations was acceptable. However, the 95% confidence intervals for BaP_{eq} were 1.52–6.05 and 1.70–5.42 µg/m³ for car- and scooter-using populations, respectively, exceeding the air standards of Europe and WHO.

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Authors Contributions

SC initiated the study and YY conducted the experimental survey and questionnaire survey. SC drafted the manuscript and writing of the manuscript, contributed the interpretation of the study results, and approved the final version of the manuscript submitted for publication.

Ethics Approval and Consent to Participate

This questionnaire was also approved by the Institutional Review Board of the Ethical Committee of Chung Shan Medical University (CSMUH No: CS16046).

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