

# Indoor Exposure and Health Risk of Polycyclic Aromatic Hydrocarbons (PAHs) in Public Facilities, Korea

Ho-Hyun Kim, Young-Wook Lim, Jun-Min Jeon<sup>1)</sup>, Tae-Hun Kim, Geon-Woo Lee, Woo-Seok Lee<sup>2)</sup>  
Jung-Yun Lim<sup>2)</sup>, Dong-Chun Shin<sup>3)</sup> and Ji-Yeon Yang\*

The Institute for Environmental Research, Yonsei University College of Medicine, 250 Seongsan-Ro, Seoul 120-749, Korea

<sup>1)</sup>Department of Civil & Environmental Eng, Suncheon first College, Suncheon, Korea

<sup>2)</sup>Indoor Air and Noise Research Division, National Institute of Environmental Research (NIER), Korea

<sup>3)</sup>Department of Preventive Medicine, Yonsei University College of Medicine, 250 Seongsan-Ro, Seoul 120-749, Korea

\*Corresponding author. Tel: +82-2-2228-1896, E-mail: jyyang67@yuhs.ac

## ABSTRACT

In the study, pollution levels of indoor polycyclic aromatic hydrocarbons (PAHs) in public facilities (vapor phase or particulate phase) were evaluated, and a health risk assessment (HRA) was carried out based on exposure scenarios. Public facilities in Korea covered by the law, including underground subway stations, funeral halls, child care facilities, internet cafes (PC-rooms), and exhibition facilities (6 locations for each type of facility, for a total of 48 locations), were investigated for indoor assessment. For the HRA, individual excess cancer risk (ECR) was estimated by applying main toxic equivalency factor (TEF) values suggested in previous studies. Among the eight public facilities, internet cafes showed the highest average PM<sub>2.5</sub> concentration at 110.0 µg/m<sup>3</sup> (range: 83.5-138.5 µg/m<sup>3</sup>). When assuming a risk of facility exposure time based upon the results of the surveys for each public facility, the excess cancer risk using the benzo(a)pyrene indicator assessment method was estimated to be 10<sup>-7</sup>-10<sup>-6</sup> levels for each facility. Based on the risk associated with various TEF values, the excess cancer risk based upon the seven types cancer EPA (1993) and Malcolm & Dobson's (1994) assessment method was estimated to be 10<sup>-7</sup>-10<sup>-5</sup> for each facility. The excess cancer risk estimated from the TEF EPA (2010) assessment was the highest: 10<sup>-7</sup>-10<sup>-4</sup> for each facility. This is due to the 10-fold difference between the TEF of dibenzo(a,e)fluoranthene in 2010 and in 1994. The internet cafes where smoking was the clear pollutant showed the highest risk level of 10<sup>-4</sup>, which exceeded the World Health Organization's recommended risk of 1 × 10<sup>-6</sup>. All facilities, with the exception of internet cafes, showed a 10<sup>-6</sup> risk level. However, when the TEFs values of the US EPA (2010) were applied, the risk of most facilities in this study exceeded 1 × 10<sup>-6</sup>.

**Key words:** Polycyclic aromatic hydrocarbons, Public facilities, Indoor and outdoor, Toxic equivalency factor, Health risk assessment

## 1. INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are the most common types of polycyclic organic matter (POM) in the environment (US EPA, 2007, 1993; NIEHS, 1998). Since PAHs are classified as carcinogenic, they are of scientific interest and have been the subject of several recent reports (Kameda *et al.*, 2005; Brown *et al.*, 1999). Of suspended particulate phases, 44-56% consists of fine particles (PM<sub>2.5</sub>), and more than 80% of PAHs are closely related to the PM<sub>2.5</sub> (Ohura *et al.*, 2004). Benzo[a]pyrene (BaP), benz[a]anthracene (BaA), and dibenz[a,h]anthracene (DBahA) are classified as carcinogen grade (IARC, 1987). In general, humans are exposed to PAHs via foods and indoor and outdoor air (Menzie *et al.*, 1992). These substances accumulate in the body mainly through respiratory pathways, but exposure can also occur through skin contact in workspaces (Tsai *et al.*, 2002).

The most common PAH pollutants are generated by the incomplete combustion of organic matter such as coal, oil, gas, and wood, and can be generated by natural sources such as forest fires and volcanic activities (NRC, 1983). Other than smoking, the main sources of PAHs in indoor air include repellents, heating fuels, burnt food, and coal tar in shampoos and other household goods (including construction materials). The proximity of roads to residences and indoor spaces and the volume of traffic are also known to affect indoor PAH concentrations (Peng *et al.*, 2011). Candice Lung *et al.* (2002) assessed the geometric mean of the total gaseous and particulate PAH concentrations in the two shops were 1.01 and 0.46 µg/m<sup>3</sup>, respectively; but with

higher concentrations in the mall and food court and lower values in the hospital and library (Levy *et al.*, 2002).

In addition, studies have shown that PAH concentration is generally higher indoors than outdoors and that indoor pollutants are the primary carcinogenic factors (Froehner *et al.*, 2010; Abas *et al.*, 2003). Among PAHs, many individual substances are regulated as proven carcinogens or possible carcinogens in various international and national organizations. Their known effects on the human body include lung cancer, anemia, leukemia, lymphoma, and disorders of development, reproductive function, and the nervous system (Baird *et al.*, 2007; Colombo *et al.*, 2006). Most PAHs are carcinogens or mutagens. PAHs are by products of incomplete combustion during fuel burning, smoking, and cooking, and can be found in both indoor and outdoor air.

The US EPA (2002) has provided various methods to estimate human risk from PAHs corresponding to complex substance characteristics. These methods are mainly used as estimates. First, the surrogate method is based upon unconfirmed PAH mixtures (i.e., a mixture of PAHs, so-called “interested mixtures” that requires that surrogate PAHs be diluted). Second, the relative potency factor (RPF) method is in regards to the components. Using this method, degrees of cancer development are evaluated by selecting individual substances (such as BaP) from among PAHs, and the degrees of initial cancer development are then estimated by summing the degrees of carcinogenesis. Recently, this method was used in EPA provisional guidelines for assessing PAHs risks, and it was recommended that BaP be used as a standard substance. In general, previous studies have provided the results of applying TEFs in terms of BaP of individual substances when estimating PAH risks (Orecchio, 2011; Larsen and Baker, 2003; Nisbet and LaGoy, 1992).

In other countries, assessment research of household PAHs in relation to smoking and carcinogenic potential has been conducted since the late 1980s. Recently, the appropriateness of indoor air quality management was evaluated utilizing individual concentrations, body exposure paths, and the contributions of PAHs to carcinogenic potential in various indoor spaces (houses, child care centers, coffee shops) and in terms of the use of indoor heating fuels and other factors (Orecchio, 2011; Mannino and Orecchio, 2008). Previous studies have also concluded that effective environmental management is necessary through health risk assessment (HRA) of hazardous pollutants such as PAHs (Bai *et al.*, 2009; Asante-Duah, 2002). The risk associated with the inhalation of particulate PAHs indoors showed that the contribution of BaP to the total carcinoge-

nic potential was dominate in the range of 51% to 64% (Ohura *et al.*, 2004). According to the USEPA,  $1 \times 10^{-6}$  is the acceptable risk (1 in 1 million) (Froehner *et al.*, 2011). According to the previous studies (Froehner *et al.*, 2011; Bai *et al.*, 2009), the range of  $10^{-5}$ - $10^{-3}$ , higher than the acceptable risk level ( $1 \times 10^{-6}$ ), and lower than the priority risk level ( $1 \times 10^{-3}$ ).

These HRAs should be based upon research on actual PAH conditions in the indoor spaces of public facilities used by all age groups. In Korea, however, research data regarding indoor PAHs pollution level are very limited, and this study is expected to facilitate the gathering of more data in the field.

The main objectives of this study were to understand the distributions of indoor PAH concentrations (vapor and particulate phase) and to perform HRAs based on exposure scenarios targeting representative law-enforced public facilities in Korea.

## 2. EXPERIMENTS AND METHODS

### 2.1 Selection of Public Facilities

For this study, eight facilities were selected as investigation targets which included underground subway stations, funeral halls, child-care facilities, super markets, indoor parking lots, and terminal waiting rooms, and two facilities were selected that were included in new regulations in 2012, including internet cafes (PC-rooms) and exhibition facilities. The distribution characteristics of fine dusts ( $PM_{2.5}$ ) and particulate/vapor-phase PAHs substances were investigated at six locations in each facility.

Onsite investigation was performed in 48 facilities (6 location in each of the 8 facilities in each facility category) over a period of about five months from 25 May 2011 to 26 October 2011. Outdoor air was also investigated at the same time at 11 locations: 3 large cities (Seoul, Daejeon, and Gwangju), 3 medium-small cities (Suncheon, Gwangyang, and Yeosu) in Korea.

### 2.2 Sampling

Methods suggested by the Indoor Air Quality Process Test Standards of the Ministry of Environment, the National Human Exposure Assessment Study (NHEXAS) of the US EPA, or the Pilot Study of Children’s Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants (CTEPP) were used for sampling indoor and outdoor air samples. PAH assessments through sampling of particulate matter of diameter  $< 2.5 \mu m$  ( $PM_{2.5}$ ) were carried out as described in previous studies (Sugiyama *et al.*, 2010; Ohura *et al.*, 2004).

A preliminary investigation was conducted to con-

firm the indoor PAH detection rates. The PAH detection rates (more than 80%) were favorable in a sampling flow range of 4-5 L/min. A sampling time of a minimum of 24 hr was selected to minimize the number of errors in regards to the morality calculation of PM<sub>2.5</sub> and in order to consider PAH characteristics that had a high loss rate during preprocessing. In addition, PAHs, which were the subject of this study, were analyzed using a PAH standard (Quebec Ministry of Env. PAHs 24 Mix) provided by AccuStandard (AccuStandard Inc., US). The PAH standard substances used in the preliminary investigation were purchased from Supelco (EPA 610 mix, US). Preprocessing of PAH samples was performed based upon the EPA TO-13A method and domestic indoor air quality process test standard (ES 01552.1).

A Mini-Vol portable sampler (PAS-201, Air Metrics, US) was used to sample the PM<sub>2.5</sub> and PAHs in indoor air. A PTFE filter (Teflon 47 mm, 1.0 µg/m, PALL Life Sciences, US) and a polyurethane foam (PUF) glass cartridge were connected to a small sampler, and air was sampled with a flow rate of 5 L/min continuously for 24 hr. To eliminate organic impurities, the PTFE filters to used sample PM<sub>2.5</sub> and PAHs were sonicated in a solution of acetone: methanol (7 : 3, v/v) for 2 hr and dried with high purity nitrogen (N<sub>2</sub>) before being used. To measure the PM<sub>2.5</sub> mass concentration, the preprocessed filter was incubated in a desiccator for 24 hr to determine constant weight, and the weight of the filters was then measured. Filters were stored and

transported by sealing in polystyrene petri dishes (50Φ) until sampling. The PTFE sampling filters were stored in a constant temperature and humidity chamber for 24 hr both before and after the sampling. The weight of the filters was measured using an analytical balance with a sensitivity higher than 0.001 mg (AT261, Mettler-Toledo, Switzerland) in triplicate, and the means of the triplicates were calculated. In order to minimize weight measurement errors as a result of temperature and humidity when calculating the PM<sub>2.5</sub> morality, it was done using the same method with the sampled filter before and after the sampling, and the weight difference was measured before and after sampling for each sampling filter.

Vapor phase PAHs sampling absorbents (PUF) were washed in a Soxhlet extraction apparatus (6 cycles/hr) with an order of methylene chloride and acetone before being used, dried, and stored in glass bottles by sealing with aluminum foil until sampling was carried out.

PAH analysis utilized analysis procedures of the EPA TO-13A Method and domestic indoor air quality process test standard (ES 01552.1) with an Agilent GC/MS (HP-6890/HP-5973N). Sample volumes of 1-2 µL were injected on to the GC column (Length 30 m, outside diameter 0.32 mm, inside diameter 0.25 µg/m in HP-5).

### 2.3 Assessment of PAHs

This study investigated 20 PAH substances selected

**Table 1.** Molecular weight and toxic equivalency factor about compound of PAHs.

Compound	Molecular weight (g mol <sup>-1</sup> )	TEF 1 <sup>a</sup>	TEF 2 <sup>b</sup>	TEF 3 <sup>c</sup>
Naphthalene (Nap)	128			
Acenaphthylene (AcPy)	152		0.001	
Acenaphthene (AcP)	154		0.001	
Flourene (Flu)	165		0.001	
Phenanthrene (PA)	178		0.001	
Anthracene (Ant)	178		0.01	
Fluoranthene (FL)	202		0.001	0.08
Pyrene (Pyr)	202		0.001	
Benzo(a)anthracene (BaA)	228	0.1	0.1	0.2
Chrysene (CHR)	228	0.001	0.01	0.1
Benzo(b)fluoranthene (BbF)	252	0.1	0.1	0.8
Benzo(j)fluoranthene (BjF)			0.1	0.3
Benzo(k)fluoranthene (BkF)	252	0.1	0.1	0.03
Benzo(e)pyrene (BeP)	252		0.01	
Benzo(a)pyrene (BaP)	252	1	1	1
Indeno(1,2,3-c,d)pyrene (IND)	276	0.1	0.1	0.07
Dibenzo(a,h)anthracene (DBA)	278	1	1	10
Benzo(g,h,i)perylene (BghiP)	276		0.01	0.009
Benzo(a,i)perylene (BaiP)				0.6
Benzo(a,l)perylene (BalP)				30

<sup>a</sup>value adopted from US EPA (1993)

<sup>b</sup>value adopted from Malcom and Dobson (1994)

<sup>c</sup>value adopted from US EPA (2010)

out of 32 substances known to be carcinogenic or potentially carcinogenic by the International Agency for Research on Cancer (IARC) and the US EPA (Table 1).

## 2.4 Quality Control

QA/QC was performed using PAH standard substances, surrogate standard substances, and internal standard substances in order to increase the reliability of the analyzed data. Quality control was done by evaluating the calibration curve linearity, the reproducibility of exposure time, detection limits (instrument detection and method detection limits), sample preprocessing factors, the recovery factors of the preprocessing equipment using standard substances, and blank tests.

The linearity evaluation of the calibration curves indicated that most of the substances showed favorable linearity with correlation coefficients ( $R^2$ ) of higher than 0.98, and the reproducibility of the exposure time (RSD%) was also lower than 0.1%. The reproducibility of the instrument detection limit revealed that the reproducibility based on BaP was relatively favorable, with a RSD of 5.18%. The reproducibility of the method detection (MDL) limit indicated that the mass concentration of MDL based on BaP was 0.13 ng/ $\mu$ L, and the detection limit, when converted to the concentration in atmosphere by applying the onsite sampling quantity (7,200 L), was 0.02 ng/m<sup>3</sup>.

Assessment of the PAH extraction recovery rate regarding the preprocessing equipment (the Soxhlet extraction apparatus) revealed that the average recovery rate was 88.8% and that the reproducibility was 11.1% of the relative standard deviation (RSD). In addition, the assessment of the recovery rates (%) of surrogate and internal standard substances regarding whole measured samples (particulate phase and vapor phase) showed that the average recovery rates of vapor-phase sampling media (PUF) were naphthalene-d8 65.4%, acenaphthene-d10 127.2%, phenanthrene-d10 112.5%, chrysene-d12 78.1%, and perylene-d12 60.7%, and that the average recovery rates of particulate-phase sampling media (filter) were naphthalene-d8 77.6%, acenaphthene-d10 132.5%, phenanthrene-d10 127.6%, chrysene-d12 73.1%, and perylene-d12 57.7%; these findings are in agreement with the recovery rate range (60-120%) recommended by US EPA Method 8100.

## 2.5 Risk Assessment

For risk assessment, excess cancer risk (ECR) was estimated by applying various TEF values suggested in previous studies. We conducted our assessment by applying a Relative Potency Factors (RPF) approach (Chen and Liao, 2006; Collins *et al.*, 1998; Nisbet and LaGoy, 1992), which was initially calculated using

the concentration and carcinogenic potentials of the surrogate approach, i.e. the BaP indicator. Secondly, we carried out calculations by applying EPA (1993) TEF for seven types of carcinogenic PAHs among the 24 total analyzed types. Thirdly, we used Malcolm & Dobson's study (1994), which provided TEF values from among 24 types in total. Lastly, we used the TEF values from EPA (2010) to calculate the final risk level of PAHs in order to classify them into four categories.

The average users for each facility were determined based upon the characteristics of the public facilities, the ages and genders of the average users, and the representative values of the exposure factors (body weight, inhalation rate, exposure time, and number of uses) consisting of WIES and MIES were determined. Surveys were carried out in order to obtain information regarding the average number of users of the subject facilities, the age and gender of users, and when the facilities are typically used. A total of 144 subjects, including facility users and employees, participated in the survey, and exposure factors were determined based upon the survey results.

Body exposure quantity can be calculated based upon pollution concentrations, inhalation rates, body weight, exposure frequencies, exposure duration, and lifetime. For daily inhalation rates, the assumed average exposure duration and 24-hour exposure of facility users were applied using survey data.

The daily inhalation rate of an adult, 13.3 m<sup>3</sup>/day, was applied as recommended by the US EPA recommendations. When calculating body weight, the average body weight of Korean adults (60 kg) according to the Ministry of Health and Welfare was applied as a representative value. The expected life expectancy (70 years) for Koreans according to the National Statistical Office was also applied.

The average exposure duration for each facility was investigated using surveys, and the resulting rates by location were as follows: underground subway stations (0.9 hr/day), funeral halls (2.89 hr/day), child-care facilities (6.7 hr/day), super markets (1.89 hr/day), indoor parking lots (2.17 hr/day), terminal waiting rooms (0.86 hr/day), internet cafes (4.17 hr/day), and exhibition facilities (2.31 hr/day). The exposure frequencies were also investigated by survey and were as follows: subway stations (15.4 times/month), funeral halls (0.97 times/month), child-care facilities (20.11 times/month), super markets (4.85 times/month), indoor parking lots (3.34 times/month), terminal waiting rooms (6.78 times/month), internet cafes (8.61 times/month), and exhibition facilities (1.56 times/month). Smoking was allowed in all internet cafes, and all indoor parking lots were non-smoking areas, with the exception of one location.

Inhalation exposure reference values (RfC) were

calculated by determining various toxicity indicators for PAH carcinogens (carcinogenic potentials, unit risks, exposure references, and POD) and applying safety coefficients based upon the collected body toxicity data for PAH non-carcinogenic assessment. Carcinogenic potential assessment or inhalation unit risks were calculated based upon body carcinogenic data collected for PAH non-carcinogenic assessment. Since PAHs are mixtures consisting of individual PAHs, complex toxicity assessment is required (Reeves *et al.*, 2001; Collins *et al.*, 1998; Nisbet and LaGoy, 1992). Therefore, the risks were calculated in the study using two methods: a method using TEF so that individual PAH shows relative carcinogenic power regarding BaP (Yang *et al.*, 2007), and a method using the toxic equivalent quotient (TEQ) of PAHs using the TEF of individual PAHs (2) (Chen and Liao, 2006).

### 3. RESULTS AND DISCUSSION

#### 3.1 PM<sub>2.5</sub> Concentration Distribution

For internet cafes (PC-rooms) in particular, the average concentration of PM<sub>2.5</sub> was 110.0 µg/m<sup>3</sup> (range: 83.5-138.5 µg/m<sup>3</sup>). This finding is higher than the level of PM<sub>10</sub> (i.e., 100 µg/m<sup>3</sup>) allowed by the Public Facilities Indoor Air Quality Management Act (the standard PM<sub>2.5</sub> in the atmosphere is projected to be 50 µg/m<sup>3</sup> in 2015 in Korea). The high level of PM<sub>2.5</sub> seen in internet cafes was likely due to indoor smoking in most cases. Smoking is the major source of indoor pollution (Ohura *et al.*, 2002; Liu *et al.*, 2001). Although Ohura *et al.* (2004) could not clearly determine the effects of smoking, they described the impact of smoking on PAHs. A previous study which compared two different coffee shops reported that a higher PM<sub>2.5</sub> was seen in smoking facilities (108.1 µg/m<sup>3</sup>, 83.5 µg/m<sup>3</sup>) compared to nonsmoking facilities (87.5 µg/m<sup>3</sup>, 61.8 µg/m<sup>3</sup>; Lung *et al.* (2004)).

In another study, Levy *et al.* (2002) reported that PM<sub>2.5</sub> (DustTrak-measured) values were 7 µg/m<sup>3</sup>, 36 µg/m<sup>3</sup>, 25 µg/m<sup>3</sup>, 19 µg/m<sup>3</sup>, and 200 µg/m<sup>3</sup> in hospitals, malls, coffee shops, libraries, and food courts, respectively. Compared to other facilities, levels in food courts were significantly higher relative to outdoor measurements, and the indoor-outdoor ratio was higher than 1. This finding clearly indicates the presence of indoor pollutants, and it is likely explained by contributions from cooking.

In other facilities, including hospitals (outdoor 15 µg/m<sup>3</sup>), malls (outdoor 44 µg/m<sup>3</sup>) and libraries (19 µg/m<sup>3</sup>), the indoor-outdoor ratio did not exceed 1. Although there were no indoor pollutants, the indoor air at these locations was affected by outdoor pollutants

(e.g. locations in hospitals near ambulances and locations in libraries adjacent to bus stops).

#### 3.2 PAHs Composition and Concentration Distribution

There are 24 types of PAHs that are frequently detected in vapors at high levels; of these PAHs, naphthalene, a compound with 2-4 rings, was detected 100% and in the highest amounts in internet cafes (PC-rooms) (15.72 µg/m<sup>3</sup>), followed by underground subway stations (5.17 µg/m<sup>3</sup>), child care facilities (4.87 µg/m<sup>3</sup>), super markets (3.12 µg/m<sup>3</sup>), funeral halls (2.87 µg/m<sup>3</sup>), exhibition facilities (2.54 µg/m<sup>3</sup>), and indoor parking lots (1.72 µg/m<sup>3</sup>), respectively (Table 2).

Pyrene, fluorine, and phenanthrene were also detected more than 95% (Fig. 1). PAHs are organic substances that generally consist of more than two aromatic (benzene) rings. Low molecular weight PAHs (two and three rings) are mainly distributed in the atmosphere as a vapor phase (WHO, 2000). It has been reported that indoor occurrences are generally composed of PHAs with 2-3 rings, while PAHs with larger molecular weights are found outdoors (Masih *et al.*, 2010; Zhu *et al.*, 2009; Mannino and Orecchio, 2008; Ohura *et al.*, 2004). In this study, PAHs with two to four rings were emitted from indoor sources, whereas PAHs with five to six rings were expected to be mainly found outdoors. This trend was also reported in previous studies (Li *et al.*, 2005; Naumova *et al.*, 2002).

In this study, the levels of PAHs with two to three rings, including acenaphthene (22.42 ng/m<sup>3</sup>), phenanthrene (4.93 ng/m<sup>3</sup>), naphthalene (4.62 ng/m<sup>3</sup>) and fluorene (3.23 ng/m<sup>3</sup>) were also high. Both internet cafes and underground subway stations showed high levels of acenaphthene and naphthalene. Zhu and Wang (2003) reported that naphthalene is one of the most noticeable PAHs in commercial kitchens (3.0 µg/m<sup>3</sup>), non-smoking household kitchens (2.7 µg/m<sup>3</sup>), and smoking household kitchens (9.9 µg/m<sup>3</sup>). Naphthalene in the vapor phase accounts for more than 69% of PAHs (Zhu *et al.*, 2009).

Individual naphthalene, acenaphthene, phenanthrene, and anthracene with two to three rings, and individual benzo(b+j)fluoranthene, benzo(k)fluoranthene, benzo[e]pyrene, and benzo[a]pyrene with five rings were detected 100%. Fluorene, fluoranthene, benz[a]anthracene, chrysene, and pyrene had detection rates of 83-92%. Multi-ringed PAHs (five rings or more) exist mainly as particles (WHO, 2000), and are known to be increased in the winter season due to increased fossil fuel use (Ticombe *et al.*, 2011; Masih *et al.*, 2010; Ohura *et al.*, 2004).

In the case of particle PAHs, acenaphthylene, which

**Table 2.** Results of PAHs concentration measured in various public facilities.

PAHs	Underground subway station		Funeral hall		Child care facility		Super market		Indoor parking lot		Terminal waiting room		Internet cafe		Exhibition facility	
	Vapor (n=6)	Particle (n=6)	Vapor (n=6)	Particle (n=6)	Vapor (n=6)	Particle (n=6)	Vapor (n=6)	Particle (n=6)	Vapor (n=6)	Particle (n=6)	Vapor (n=6)	Particle (n=6)	Vapor (n=6)	Particle (n=6)	Vapor (n=6)	Particle (n=6)
Naphthalene	5.17	0.60	2.87	1.05	4.87	1.00	3.12	0.46	1.72	1.01	0.99	1.17	15.72	4.33	2.54	0.35
Acenaphthylene	0.26	0.05	0.35	0.15	1.37	0.23	0.89	ND	0.11	ND	0.39	0.18	0.92	0.41	0.54	0.11
Acenaphthene	28.17	33.36	10.61	42.14	13.54	35.92	14.35	31.51	24.52	54.48	19.49	18.15	38.37	35.23	26.61	25.75
Fluorene	4.82	0.36	2.63	0.36	1.32	0.37	4.69	0.38	0.78	0.58	2.21	1.75	5.82	2.47	3.12	0.35
Phenanthrene	5.18	0.45	2.97	0.66	3.83	0.71	4.63	0.66	1.48	0.92	3.80	3.34	13.57	5.12	3.62	0.45
Anthracene	2.73	0.46	1.31	0.68	4.97	0.75	3.08	0.66	1.18	0.93	0.74	0.78	3.50	1.29	0.96	0.44
Fluoranthene	0.72	0.27	0.70	0.43	1.09	0.43	0.36	0.52	0.39	0.89	0.61	0.78	1.13	1.45	0.50	0.49
Pyrene	0.94	0.19	1.07	0.27	1.04	0.28	0.69	0.41	0.95	0.54	0.52	0.43	1.21	1.22	0.40	0.35
Benzo[c]phenanthrene	1.62	0.61	ND	0.52	0.44	0.31	0.10	0.44	0.86	1.33	0.56	3.70	0.21	1.22	0.27	0.42
Benzo[a]anthracene	0.72	0.40	0.60	0.66	1.20	0.89	0.29	0.29	0.51	1.02	0.85	0.99	0.91	1.45	0.35	0.43
Chrysene	0.47	0.38	0.54	0.60	1.05	0.74	1.15	0.59	0.53	1.02	0.79	1.04	0.86	2.66	0.28	0.41
Benzo(b+j)fluoranthene	1.27	0.71	0.67	0.99	1.74	1.05	0.83	0.97	0.88	1.94	1.35	0.94	1.62	3.63	0.89	0.79
Benzo(k)fluoranthene	0.71	0.36	0.33	0.47	1.24	0.48	1.04	0.41	0.63	0.81	0.61	0.42	0.74	1.39	0.46	0.35
DMBA	2.11	0.56	ND	0.43	ND	0.30	3.42	0.43	ND	3.96	0.98	2.51	2.91	ND	1.63	1.10
Benzo(e)pyrene	1.68	0.78	0.83	1.16	0.86	1.20	1.38	1.06	0.98	1.50	1.53	1.23	1.81	1.85	0.78	0.77
Benzo(a)pyrene	2.75	0.93	1.15	1.63	2.75	1.56	1.82	1.40	1.34	2.07	2.12	1.47	2.52	2.34	0.88	1.11
3-Methylcholanthrene	ND	1.07	1.53	2.33	2.88	1.45	ND	0.82	1.32	1.75	1.14	5.14	2.92	4.22	0.60	2.78
I[23P]	0.03	1.26	0.09	1.09	3.57	1.23	1.03	0.65	1.38	2.70	0.25	2.30	2.98	5.57	0.20	0.93
Dibenz(a,h)anthracene	ND	0.79	ND	0.51	ND	0.40	ND	2.14	0.84	1.89	ND	1.52	0.37	17.52	0.27	0.59
Benzo(g,h,i)perylene	ND	0.90	ND	0.73	0.14	0.48	1.18	1.64	1.20	2.10	0.28	2.46	ND	11.53	0.32	0.75
Dibenzo(a,h)pyrene	ND	0.62	ND	ND	ND	0.70	ND	1.92	1.46	1.11	ND	2.54	3.42	2.07	0.68	1.51
Dibenzo(a,i)pyrene	ND	1.34	ND	0.92	ND	0.77	ND	ND	ND	0.88	ND	0.72	ND	ND	ND	0.41
Dibenzo(a,l)pyrene	ND	0.42	ND	ND	ND	ND	ND	ND	1.24	0.75	ND	0.63	ND	0.99	0.18	ND

(ND: Not detected, less than 0.02 ng/m<sup>3</sup>)

\*DMBA: 7,12-Dimethylbenz(a)anthracene

I[23P: Indeno[1,2,3-cd]pyrene

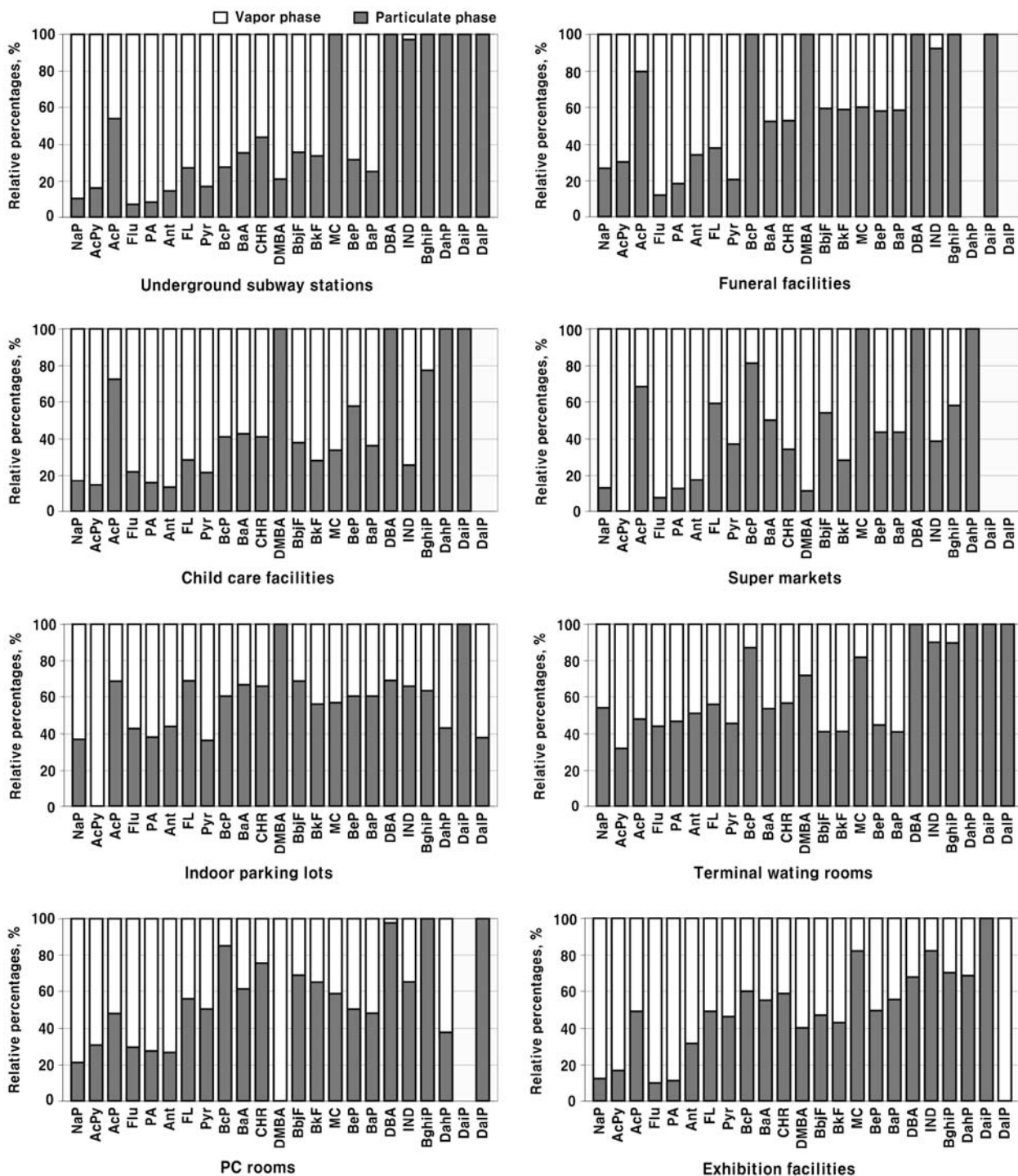


Fig. 1. Distribution of PAHs between vapor and particulate phase in various public facilities.

consists of three rings, was highest ( $34.57 \text{ ng/m}^3$ ) as compared to substances with five rings (Table 2). In a previous study conducted in China, 12 PAHs were investigated in kitchens, and it was found that PAHs

with three to four rings had the highest detection rates and concentrations, a finding that was similar to the results of the present study. For the rest, dibenz(a,h) anthracene was highest ( $4.04 \text{ ng/m}^3$ ), followed by benzo

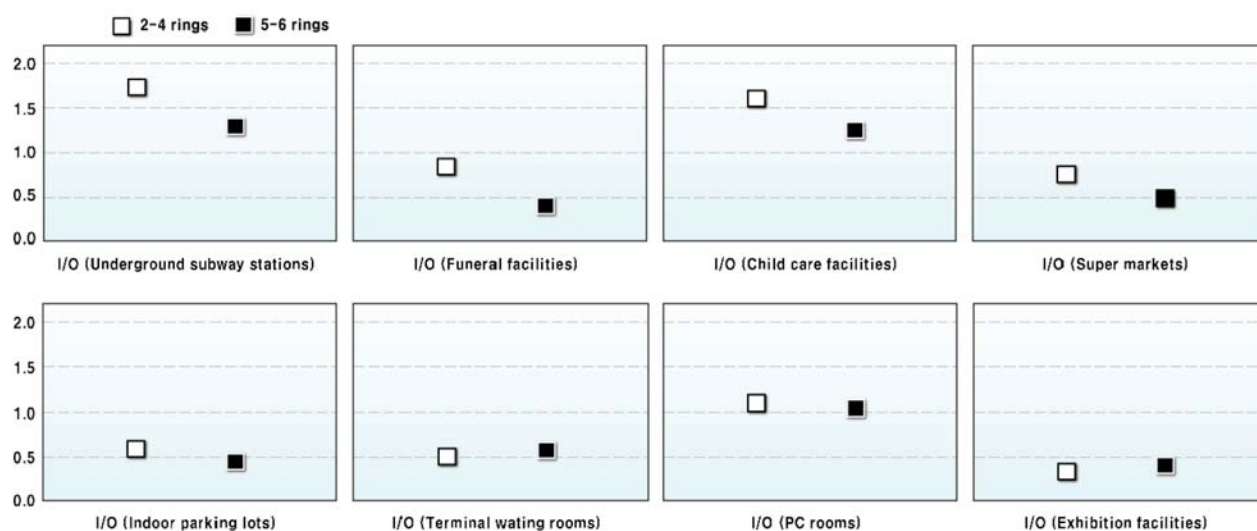


Fig. 2. Ratio of I/O (indoor/outdoor) PAHs concentrations in various public facilities.

(g,h,i)perylene ( $3.01 \text{ ng/m}^3$ ), 3-methylcholanthrene ( $2.35 \text{ ng/m}^3$ ), and indeno[1,2,3-cd]pyrene ( $2.25 \text{ ng/m}^3$ ), respectively.

Further, concentrations of PAHs with more than five rings, including benzo(b+j)fluoranthene, benzo[k]fluoranthene, and benzo[e]pyrene, were found to be high in internet cafes and indoor parking lots. Benzo[a]pyrene, which is known as a carcinogen, was highest in internet cafes ( $2.34 \text{ ng/m}^3$ ), followed by indoor parking lots ( $2.34 \text{ ng/m}^3$ ), terminal waiting rooms ( $2.07 \text{ ng/m}^3$ ), funeral halls ( $1.63 \text{ ng/m}^3$ ), child care facilities ( $1.56 \text{ ng/m}^3$ ), super markets ( $1.40 \text{ ng/m}^3$ ), exhibition facilities ( $1.11 \text{ ng/m}^3$ ), and underground subway stations ( $0.93 \text{ ng/m}^3$ ), respectively.

According to the World Health Organization (WHO) guidelines for indoor air quality (IAQ) regarding selected pollutants, the suggested level of benzo(a)pyrene is  $1.0 \text{ ng/m}^3$ . The average concentration of this carcinogen in our study was higher than the suggested WHO level in all facilities except subway stations. Benzo[a]pyrene is a representative carcinogenic marker of PAHs, and is known to account for 51-65% of total PAHs (Ohura *et al.*, 2004). It was previously reported that benzo[a]pyrene ranges from  $1.45$  to  $4.1 \text{ ng/m}^3$  in pubs, restaurants, and discotheques. These amounts which also exceeded the WHO recommendations (Harrison *et al.*, 2009; Zhu and Wang, 2003).

In general, PAHs are present as either a vapor or a particulate phase, and the composition of this vapor or particulate phase can differ depending upon physical factors (temperature, humidity), the characteristics of indoor spaces, and the season (Zhu *et al.*, 2009). Substances with two to four benzene rings were pre-

sent mostly in a vapor phase, while those with five to six rings were typically found in a particulate phase (Fig. 1). As a reference, when the relative distribution was confirmed with the exclusion of acenaphthene (which is the most common substance in both the vapor and particulate phases), vapor phase substances were found to account for 66% of individual PAHs with two to four benzene rings, while particulate phase substances accounted for 62% of those with five to six rings. It was difficult to confirm this distribution trend since acenaphthene is relatively common as compared to other substances.

It is known that representative indoor emission sources for PAHs are smoking, cooking, and heating (Ohura *et al.*, 2002; Liu *et al.*, 2001). Smoking is a definite emission source, whereas cooking (for visitors) contributes partially to PAHs in internet cafes in the study. Considering the conditions in internet cafes (lack of ventilation) and the season when this study was performed, contributions from outdoor air and heating would be minimal. Ohura *et al.* (2004) similarly reported that the summer/winter (S/W) ratios of PAHs and associated particles were low.

Although the statistical power could be relatively low due to the small sample numbers, it was found that the average indoor-outdoor ratios of PAHs were all higher than 1 in all eight facilities, indicating the presence of indoor pollutants. The average in these facilities was highest in subway stations, followed by daycare centers and internet cafes, respectively (Fig. 2). Individual substances with two to four rings showed high I/O ratios compared to substances with five to six rings, with the exception of terminal waiting



rooms and exhibition facilities, (the average I/O ratios of the eight facilities were 0.93 and 0.74 for substances with two to four rings and five to six rings, respectively) (Fig. 2). In this study, ratios are calculated in terms of I/O and it is expected that in the absence of indoor sources, the ratios between I/O will be less than or equal to 1 (Mannino and Orecchio, 2008). A few traditional studies showed that PAH distributions found in indoor environment were higher than those found in outdoor environment (Chuang *et al.*, 1992; Wilson *et al.*, 1989). In recent, Zhu *et al.* (2009) showed similar results in that the average I/O ratio of substances with two to four rings in homes (including the living room, bedroom and kitchen) was higher than 1, while the average I/O ratios of substances with five to six rings was lower than 1 in the winter season. But, little study has been conducted that indoor sources may contribute to pollution with PAHs of 2 or 3 rings, whereas larger PAH molecules mainly existed from outdoor sources (Ohura *et al.*, 2004; Naumova *et al.*, 2002).

One of the limitations of the present study is that we did not evaluate seasonal factors for the results of indoor PAHs measurement. Nonetheless, Ohura *et al.* (2004) showed identical results in that S/W ratios were lower for PAHs associated with particles. However, it would be interesting to measure PAHs along in terms of the higher use of heating in combination with lower ventilation in the winter season. In addition, Ohura *et al.* (2002) assessed the Summer/Winter (S/W) ratios were lower for PAH associated with particle, which was consistent with the data reported in study. Further studies of these data might include a more detailed evaluation that optimizes the research between sampling time and seasonal variation, as well as the influence of outdoor exposure.

### 3.3 Health Risk Assessment

The results of risk assessment from the exposure scenario for each public facility in terms of PAHs (suggested by four different TEF-adjusted methods) are summarized in Table 3 and Fig. 3. Of the individual PAHs, the risk from seven carcinogenic PAHs was assessed according to the TEFs of the US EPA from 1993. These seven carcinogenic PAHs are benz[a]anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, indeno(1,2,3-cd)pyrene, and dibenz(a,h)anthracene. Of the 24 carcinogenic substances investigated in the study, these substances accounted for 9.67% of the total substances. Under a general exposure scenario, the estimated levels were  $2.32E-05$  in internet cafes and  $4.80E-07$  in funeral halls. Under the worst-case scenario (24-hr exposure), the estimated level was  $10^{-4}$  in all facilities.

Risk was assessed using the TEF values of 21 indi-

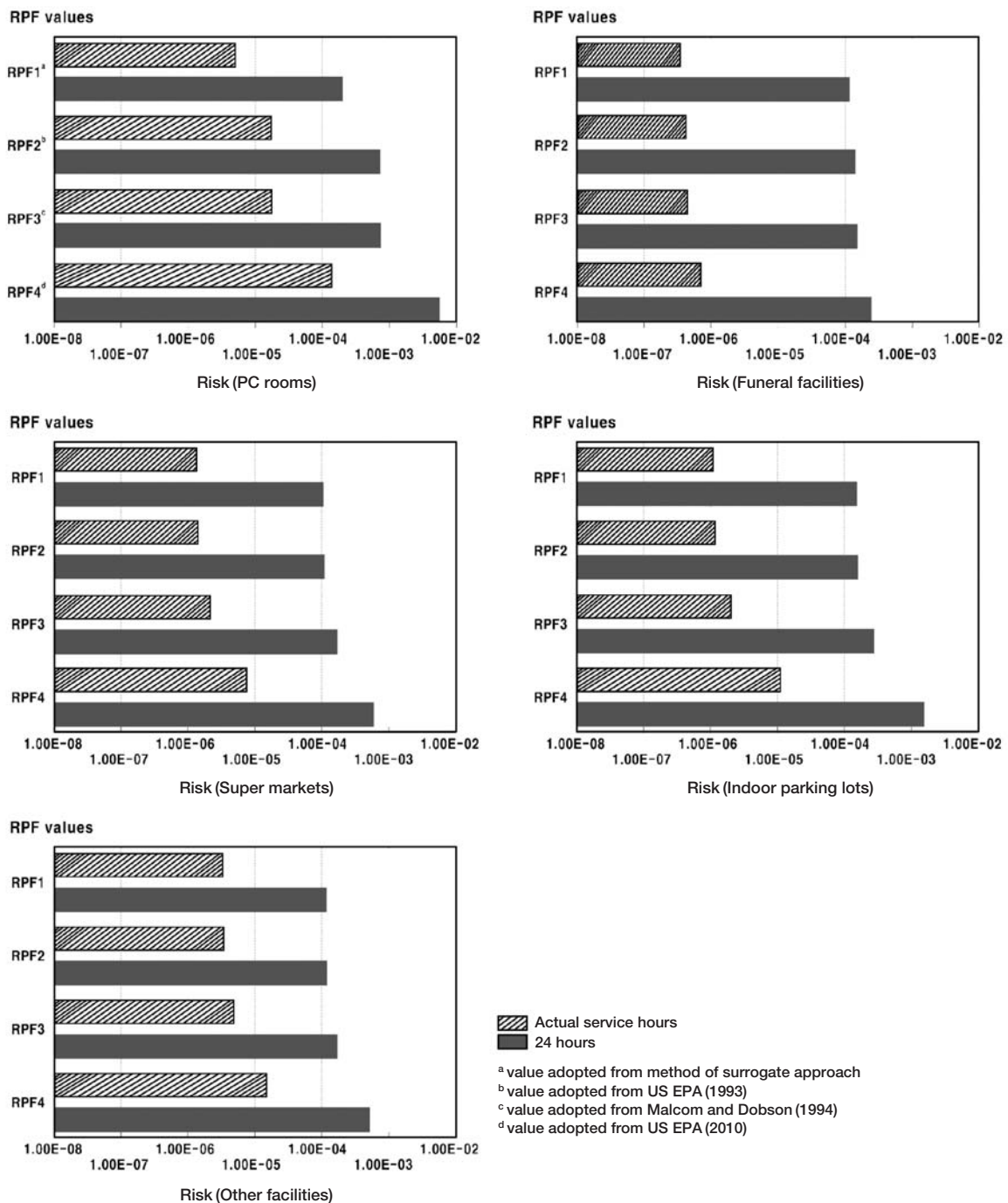
**Table 3.** Relative risk comparison in various public facilities by applying different relative potency factor (RPF).

	Internet cafes			Funeralhalls			Super markets			Indoor parking lots			Other facilities		
	RPF 1 <sup>a</sup>	RPF 2 <sup>b</sup>	RPF 3 <sup>c</sup>	RPF 1	RPF 2	RPF 3	RPF 1	RPF 2	RPF 3	RPF 1	RPF 2	RPF 3	RPF 1	RPF 2	RPF 3
Risk values (per persons)	2.32E-05	2.40E-05	1.78E-04	4.80E-07	5.14E-07	8.12E-07	1.70E-06	2.69E-06	9.43E-06	1.42E-06	2.52E-06	1.42E-05	4.47E-06	6.25E-06	1.91E-05
LADD (ng/kg-day)	7.62E-02	7.87E-02	5.86E-01	1.58E-03	1.69E-03	2.67E-03	5.59E-03	8.83E-03	3.10E-02	4.66E-03	8.27E-03	4.66E-02	1.47E-02	2.05E-02	6.28E-02
Risk values (per persons)	9.44E-04	9.75E-04	7.25E-03	1.75E-04	1.87E-04	2.96E-04	1.36E-04	2.14E-04	7.51E-04	2.00E-04	3.55E-04	2.00E-03	1.55E-04	2.17E-04	6.63E-04
LADD (ng/kg-day)	3.10E+00	3.20E+00	2.83E+01	5.74E-01	6.16E-01	9.72E-01	4.45E-01	7.03E-01	2.47E+00	6.57E-01	1.17E+00	6.57E+00	5.09E-01	7.11E-01	2.18E+00

<sup>a</sup>value adopted from US EPA (1993)

<sup>b</sup>value adopted from Malcom and Dobson (1994)

<sup>c</sup>value adopted from US EPA (2010)



**Fig. 3.** Results of risk assessment in various public facilities by applying different RPF.

vidual PAHs, as suggested by Malcolm & Dobson (1994). Under a general exposure scenario, it was estimated that risk was highest in internet cafes ( $2.40E-05$ ), while other facilities were estimated to be  $10^{-7}$ - $10^{-6}$ . Under the worst-case scenario (24-hr exposure), the estimated level was  $10^{-4}$  in all facilities.

Bostron *et al.*(2002) reported that lifetime lung cancer risks were  $1.9 \times 10^{-4}$  in summer and  $0.9 \times 10^{-3}$  in winter when assessed using the TEF values of Malcolm & Dobson’s (1994); both estimations are higher than the health-based guideline level of  $1 \times 10^{-5}$ .

A risk assessment of 11 PAHs was also estimated

utilizing the TEFs of the US EPA (2010). A notable difference was found compared to conventional TEFs in that the TEF values of benzo(a) pyrene, dibenzo(a,h) anthracene, and dibenzo(a,l)pyrene, the most potent carcinogenic substances, were estimated to be 1, 10, and 30, respectively. If these TEFs values were applied to the risk assessment, the general exposure scenario was estimated to be  $1.78E-04$ ; under the worst exposure scenario (24-hr exposure), the estimated value was  $10^{-3}$  in internet cafes and indoor parking lots, while it was  $10^{-4}$  in all other facilities. Smoking will be prohibited in internet cafes starting in 2013 in Korea; thus, the additional risk due to smoking should decrease.

According to the WHO recommendations, the excess unit risk of PAH exposure over a lifetime (70 years) is estimated to be  $8.7 \times 10^{-5} (\text{ng}/\text{m}^3)^{-1}$  (Ohura *et al.*, 2004). Therefore, given the TEFs values of the US EPA, most facilities will experience higher levels than the unit risk level suggested by the WHO under the general exposure scenario of public facilities usage.

In summary, the benzo(a)pyrene indicator method can be utilized in order to manage and set standards for PAHs in foreign countries, while the risk assessment method for the seven types of carcinogens and TEFs can be used in studies regarding the risk assessment of each media in various spaces (Froehner *et al.*, 2011; Wang *et al.*, 2011; Bai *et al.*, 2009; Zhu *et al.*, 2009; Chen and Liao, 2006). To the best of our knowledge, this is the first study to adopt the TEF values of the US EPA for the comparison of each TEF. In order to effectively manage indoor PAHs, it has been suggested that focus be placed on benzo(a)pyrene toxicity and on standard management for health protection via health risk assessment based on research of actual conditions. In the European Union, benzo(a) pyrene standard levels have been recommended for both indoor and outdoor settings. Therefore, a standard should be set regarding non-regulated indoor PAHs through additional investigations based on various perspectives. One of the limitations of HRAs is that sensitive groups, applied for age-dependent adjustment factors (ADAFs), were not appropriately evaluated, even though most of the facilities investigated in the study were for infants, children, and adolescents.

#### 4. CONCLUSIONS

Of the eight investigated public facilities, the  $\text{PM}_{2.5}$  concentration (which is mostly from smoking) was  $110.0 \mu\text{g}/\text{m}^3$  on average in internet cafes (PC-rooms), which is higher than the standard ( $100 \mu\text{g}/\text{m}^3$ ) defined by the Public Facilities Indoor Air Quality Management Act. The ratio of PAHs between the vapor phase

and the particulate phase was similar as that reported in previous studies. That is, low-molecular-weight PAHs (two and three rings) were mainly distributed in the vapor phase in the atmosphere, while multi-ringed PAHs (five rings or more) were usually present as particles. However, since the present study was carried out between May and October and measured one sample from each facility (for 24 hrs), it is expected that the contributions from indoor pollutants and the characteristics of public facilities were more significant than seasonal factors. Seven of the investigated facilities exhibited an average concentration of benzo(a) pyrene, which is a representative carcinogenic PAHs, that was higher than the WHO recommendation of  $1.0 \text{ ng}/\text{m}^3$ , with the exception being subway stations.

With the exception of the EPA's TEFs method, we determined that the benzo(a)pyrene excess cancer risk was  $10^{-7}$ - $10^{-6}$  under the general exposure scenario using the HRA method. Similarly, the TEF evaluation showed a  $10^{-7}$ - $10^{-6}$  excess cancer risk for seven types of cancers for each facility. Under the general exposure scenario, internet cafes were the facilities with the most risk, with a  $10^{-5}$  level, which was mainly due to smoking. Furthermore, when the TEF values of the US EPA were applied, the risk was higher than  $1 \times 10^{-6}$ , which is the cutoff limit suggested by the WHO.

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