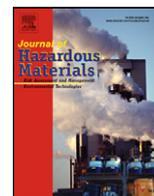




Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat



Assessing hazardous risks of human exposure to temple airborne polycyclic aromatic hydrocarbons

Kuo-Chih Chiang, Chia-Pin Chio, Yu-Hui Chiang, Chung-Min Liao*

Department of Bioenvironmental Systems Engineering, National Taiwan University, Taipei 10617, Taiwan, ROC

ARTICLE INFO

Article history:

Received 4 June 2007
Received in revised form 19 July 2008
Accepted 25 November 2008
Available online xxx

Keywords:

Incense burning
Polycyclic aromatic hydrocarbons
Risk assessment
Indoor air quality
Temple

ABSTRACT

We proposed an integrated probabilistic risk assessment framework based on reported data to quantify human health risks of temple goers/workers to airborne polycyclic aromatic hydrocarbons (PAHs) from incense burning in typical Taiwanese temples. The framework probabilistically integrates exposure, human respiratory tract, and incremental lifetime cancer risk (ILCR) models to quantitatively estimate size-dependent PAHs exposure in human lung regions and cancer risks for temple goers (moderate and high exposures) and temple workers (extreme exposure). Our results show that the ILCRs are greater than the acceptable level of 10^{-6} for extreme and high exposure groups through inhalation route. The result also indicates that the higher ILCRs (10^{-6} to 10^{-4}) are found in ingestion and dermal contact routes for temple goers/workers. For personal extreme exposure to carcinogenic PAH in the temple, 95% probability total ILCR (TILCR) (9.87×10^{-4} to 1.13×10^{-3}) is much greater than the range of 10^{-6} to 10^{-4} , indicating high potential health risk to temple workers. For temple goers with high and moderate exposure groups, however, the 95% probability TILCRs were estimated from 6.44×10^{-5} to 7.50×10^{-5} and 5.75×10^{-6} to 6.99×10^{-6} , respectively. This study successfully offers a scientific basis for risk analysis due to incense burning to enhance broad risk management strategies for temple indoor air quality.

© 2008 Elsevier B.V. All rights reserved.

1. Introduction

Burning incense to worship deities is a daily religious ritual in most Buddhist and Taoist temples in Taiwan. It is also a part of the daily routine of about 50% of families in Taiwan [1]. Approximated 1.5 million frequent temple goers visit more than 14,500 temples across the Taiwan region and subject to burn incense inside the temples (<http://www.moi.gov.tw/stat/>). Incense burning is found to be the significant sources of large amount of particulate matters (PMs) and carcinogenic polycyclic aromatic hydrocarbons (PAHs) [2–10]. Due to the nature of its long, slow, and incomplete combustion process, this practice produces non-stop smoke. Many studies indicated that peoples exposed to such smoke caused significant potential risk for the occurrence of acute irritative symptoms, especially for throat and upper respiratory tract irritation [4,9–10].

To obtain a better estimate of the health risk associated with incense burning in the temples, the mass concentrations of PAHs and individual compound could be compared to present regulatory standards that are established to protect public or workers health with adequate safety margin. The target annual mean values of B[a]P of 0.7 to 1.3 ng m^{-3} established by European countries [11

and the WHO risk estimate for PAHs in air based on lung cancer in coke-oven workers had led to a health-based guideline value of 0.1 ng m^{-3} B[a]P for ambient air [12]. Fang et al. [13], Lin et al. [2], Chen [7] and Chuang [8] have reported the relevant measurements of PAHs in the selected Taiwanese temples and these measurements were sufficiently high to cause concern for personal exposure to carcinogenic PAHs in temples, implicating that temple PAHs exposure may result in the potential cancer risk. In fact, our previous researches [14–15] had provided evidence that 90% probabilities of B[a]P- and B[a]P-based total incremental lifetime cancer risks (ILCRs) for human in temple were larger than 10^{-6} , implicating that exposure to smoke emitted from heavy incense burning may promote lung cancer risk. Hence, developing the useful knowledge for protecting human health for temple goers/workers is necessary.

To further understand and identify individuals who are “at risk” of carcinogenic effects by PAHs exposure during religious practices, we argue that, by understanding the linkage between human respiratory tract dynamics and probabilistic risk analysis, we can provide a cancer risk estimates and scientific based risk methodology to enhance broad risk management for temple indoor air quality issues. Here we intended to develop an integrated risk assessment framework, including probabilistic exposure model, human respiratory tract (HRT) model, and incremental lifetime cancer risk (ILCR) model, to quantify temple goers/workers exposed to airborne PAHs in temples. Specifically, the objectives of this study are 3-fold:

* Corresponding author. Tel.: +886 2 2363 4512; fax: +886 2 2362 6433.
E-mail addresses: cmliao@ntu.edu.tw, cmliao@ccms.ntu.edu.tw (C.-M. Liao).

Nomenclature

AB	dermal adsorption fraction
ABS _{GI}	gastrointestinal absorption
AF _d	dermal adherence rate
AI	the alveolar-interstitial region in HRT
AT _c	averaging time for cancer effects
B[a]P	Benzo[a]pyrene
B[a]P _{eq}	B[a]p toxic equivalent
bb	the bronchiolar region in HRT
BB	the bronchial region in HRT
BW	body weight
C _a	air concentration
C _{pa}	p-PAH concentration
cd _{fs}	cumulative density functions
cf	conversion factor
CSF	cancer slope factor
CSF _i	cancer slope factor for each exposure route <i>i</i>
ED	exposure duration
ET ₁	the nasal passage in HRT
ET ₂	the pharynx in HRT
EV	event frequency
HRT	human respiratory tract
ILCR	incremental lifetime cancer risk
IR _{inh}	inhalation rate
IR _{ing}	ingestion rate
IR _a	air inhalation rate
IR _p	particle ingestion rate
LADD	lifetime average daily dose
LN	lognormal distribution
MMDs	mass median diameters
PAHs	polycyclic aromatic hydrocarbons
pdfs	probability density functions
PEF	potency equivalency factor
PMS	particulate matters
p-PAH	particle-bound PAH
Q	breathing rate
RT	residence time
SA	dermal surface exposure
TILCR	total ILCR
U	uniform distribution
UFPs	ultrafine particles
VF _{adv}	visiting frequency advice
VF	visiting frequency

(i) to conduct a probabilistic lifetime cancer risk assessment of personal multi-routes exposure to PAHs for temple goers/workers; (ii) to estimate the PAHs mass concentrations, size distribution, and daily dose for different HRT regions; and (iii) to recommend a visiting frequency advice to temple goers and a suggested incense burning amount of different types of commonly used incense.

2. Materials and methods**2.1. Study population**

A Taiwanese temple that we employed to study is a typical famous Buddhist–Taoist combined temple. The average numbers of temple goers are nearly 3000–5000 d⁻¹. We delineated exposure populations into three subgroups: temple goers with moderate and high exposure levels and temple workers with extreme exposure level. Temple goers visiting the temple on the 1st and 15th days of each lunar month based on the Chinese Lunar calendar and on the major religious festivals (e.g., Duan Wu Jie and Zhong Yuan Jie)

are defined as the moderate exposure level subgroup, whereas high exposure level subgroup is designated as the daily temple goers for 10–60 min incense burning exposure. On the other hand, temple workers who exposed to daily basis incense burning for 8–12 h are defined as the extreme exposure level subgroup. The ages of three subgroups range from 20 to 70 yrs.

2.2. Reanalyze the published PAHs data

We quantitatively reanalyzed the particle size distribution, total-PAH, particle-bound PAH (p-PAH), and individual PAH (particulate and gas phase) concentrations, and size-dependent PAHs concentrations of temple incense burning from published data. Thanks to Fang et al. [13], Lin et al. [2], and Chuang [8] who have provided the remarkable dataset related to existed PAHs in Taiwanese temples. The PAHs data give us the opportunity to test all theoretical considerations of temple PAHs exposure effects and quantify its strength. Fang et al. [13] selected a famous Taiwanese temple Tzu Yun Yen located at Ching Shui town in central Taiwan as the study site. The sampling time was from 9:00 am to 7:00 pm daily and sampling periods were from August 2001 to January 2002. Lin et al. [2] selected a Taiwanese temple located at the suburban area of Tainan city in southern Taiwan. Sampling was conducted from 9:00 am to 5:00 pm and from 9:00 am to 9:00 am the next day, respectively, for 3 sequential days during March 1996. Moreover, the reported measurements also included the relationship between incense compositions and PAH emissions for three representative types of incense of aloe wood, Taiwan yellow, and Taiwan black incense that all burned in the temple on a daily basis. Chuang [8] selected a famous old Taiwanese temple Lung Shan Temple located at Wan Hua District in Taipei. Sampling was conducted from 6:30 am to 9:30 pm during Zhong Yuan Jie and Li Dou Ceremony. We have also adopted the research from Li and Ro [16] for PAH concentrations measured in five incense-burning homes and in 14 mixed residential homes (including one smoking household, five incense-burning households, and eight households without incense burning or smoking) in Taipei region for a comparison study.

We used the individual compound potency equivalency factor (PEF) relative to B[a]P based on a PEF scheme developed by Collins et al. [17] to estimate multi-component PAH exposure. Table 1 lists

Table 1
Potency equivalency factor (PEF) for PAHs relative to B[a]P used in this study.

Name with abbreviation	PEF
<i>Reference</i>	
Benzo[a]pyrene (B[a]P)	1 [18]
<i>PAHs</i>	
Acenaphthene (Acp)	0.001 [18]
Acenaphthylene (AcPy)	0.001 [18]
Anthracene (Ant)	0.01 [18]
Benzo[a]anthracene (B[a]A)	0.1 [18]
Benzo[b]chrycene (B[b]C)	NA
Benzo[b]fluoranthene (B[b]FT)	0.1 [18]
Benzo[k]fluoranthene (B[k]FT)	0.1 [18]
Benzo[e]pyrene (B[e]P)	0.01 [19]
Benzo[g,h,i]perylene (B[g,h,i]P)	0.01 [18]
Chrysene (CHR)	0.01 [18]
Coronene (COR)	0.001 [19]
Cyclopenta[c,d]pyrene (C[c,d]P)	0.1 [19]
Dibenzo[a,h]anthracene (DB[a,h]A)	1 [19]
Fluoranthene (FL)	0.001 [18]
Fluorene (Flu)	0.001 [18]
Indeno[1,2,3-c,d]pyrene (In[c,d]P)	0.1 [18]
Naphthalene (Nap)	0.001 [18]
Perylene (PER)	0.001 [19]
Phenanthrene (PA)	0.001 [18]
Pyrene (Pyr)	0.001 [18]

Table 2
Mathematical models used in the present study.

Models	
Probabilistic exposure model ^{a,b,c}	
LADD _{inh}	$= C_a \cdot IR_a \cdot RT \cdot VF \cdot ED \cdot cf, \quad (1)$
LADD _{ing}	$= \frac{C_{pa} \cdot IR_p \cdot RT \cdot VF \cdot ED \cdot cf}{BW \cdot AT_c}, \quad (2)$
LADD _{dermal}	$= \frac{C_{pa} \cdot AB \cdot SA \cdot EV \cdot AF_d \cdot RT \cdot VF \cdot ED \cdot cf}{BW \cdot AT_c}, \quad (3)$
HRT model ^d	
$\frac{dC(k,t)}{dt}$	$= [L]\{C(k,t)\} + [B]\{u(k,t)\}, \quad (4)$
ILCR model ^e	
ILCR _i	$= LADD_i \cdot \left(CSF_i \cdot \left(\frac{BW}{70 \text{ kg}} \right)^{1/3} \right), \quad (5)$
TILCR	$= \sum_i^3 ILCR_i, \quad (6)$

^a LADD_{inh} is the lifetime average daily dose associated with inhalation (mg kg⁻¹ d⁻¹), C_a is the air concentration (ng m⁻³), IR_a is the air inhalation rate (m³ d⁻¹), RT is the residence time (min time⁻¹), VF is the visiting frequency (time yr⁻¹), ED is the exposure duration (yr), cf is the conversion factor (1440⁻¹ × 10⁻⁶), BW is the body weight (kg), and AT_c is the averaging time for carcinogens (d) (365 d yr⁻¹ × 70 yr = 25550 d).

^b LADD_{ing} is the lifetime average daily dose associated with ingestion (mg kg⁻¹ d⁻¹), C_{pa} is the p-PAH concentration (μg g⁻¹), and IR_p is the particle ingestion rate (mg d⁻¹).

^c LADD_{dermal} is the lifetime average daily dose associated with dermal contact (mg kg⁻¹ d⁻¹), AB is the dermal adsorption fraction (dimensionless), SA is the dermal surface area exposed (cm²), EV is the event frequency (event d⁻¹), and AF_d is the particle-to-skin adherence factor (mg cm⁻² event⁻¹).

^d Eq. (4) is the general matrix form of presenting HRT model [21–23] where {C(k,t)} = {C₁(k,t), C₂(k,t), C₃(k,t), C₄(k,t), C₅(k,t)}^T is the state variable vector of PAH concentration in lung regions ET₁, BB, bb, and AI (more explanations in text), respectively (μg cm⁻³); {u(k,t)} = {C₁(k,t), 0, 0, 0, 0}^T represents an input vector of indoor PAH concentration (ng cm⁻³); [L] and [B] = diag{Q/V₁, 0, 0, 0} are the state matrix (s⁻¹) and the constant input matrix (s⁻¹), respectively; Q is the breathing rate (cm³ h⁻¹) and V₁ is the volume of ET₁ compartment (cm³).

^e ILCR_i is the incremental lifetime cancer risk for each exposure route *i* (–), LADD_i is the lifetime average intake dose for each exposure route *i* (mg kg⁻¹ d⁻¹), and CSF_i is the cancer slope factor for each exposure route *i* (mg kg⁻¹ d⁻¹)⁻¹.

the PAHs and the PEFs used in the calculation associated with the cancer evidence of individual PAH compound classified by Nisbet and LaGoy [18] and Malcom and Dobson [19].

2.3. Mathematical models

Exposure is expressed in terms of a lifetime average daily dose (LADD) and is calculated separately for each element and for each exposure pathway. Specifically, the doses contact through inhalation and ingestion of particles and absorb through the skin have been documented by U.S. EPA [20]. LADD for incidental inhalation, ingestion and dermal contact pathways are listed in Table 2. We treated C_a, IR_a, C_{pa}, IR_p, AB, SA, AF_d, RT, and BW in Eqs. (1)–(3) probabilistically.

We divided human respiratory tract (HRT) into five major compartments from the suggestion of ICRP66 [21]: (i) the nasal passage (ET₁), comprising the anterior nose and the posterior nasal passages; (ii) the pharynx (ET₂), comprising the larynx and mouth; (iii) the bronchial region (BB), comprising the airway from the trachea, main bronchi, and intrapulmonary bronchi; (iv) the bronchiolar region (bb), comprising the bronchioles and terminal bronchioles; and (v) the alveolar-interstitial region (AI), comprising the airway from the respiratory bronchioli through the alveolar sacs. Followed by the principle of mass balance, the dynamic equations of inspiratory oral cavity varying with particle size range *k* and time *t* to each regional compartment are given a by a state-space realization form of a linear dynamic representation (Table 2, Eq. (4)) [22,23]. The reference values for anatomical and physiological parameters, including volumes, breathing rates, transfer coefficients, and clearance rate, are taken from the ICRP66 [21]. More details for HRT model developments and constructions have been described in elsewhere [22,23].

The incremental lifetime cancer risk (ILCR) is estimated as the incremental probability of an individual developing cancer over a lifetime as a result of exposure to a potential carcinogen. We used the linear low-dose carcinogenic risk equation to reflect each of exposure routes of inhalation, ingestion, and dermal contact (Table 2, Eq. (5)). The cancer slope factor (CSF), which is used to estimate the risk potential (Table 2, Eq. (5)) in the present study, for each exposure routes are normalized to account for extrapolation to a different body weight from standard of 70 kg. The CSFs for B[a]P inhalation and ingestion exposure were 12 and 3.9 (mg kg⁻¹ d⁻¹)⁻¹, respectively. Those values were adopted from OEHHA [24] and Neal and Rigdon [25]. For exposure to B[a]P by dermal contact pathway, the potencies were estimated to be 37.47 and 23.5 (mg kg⁻¹ d⁻¹)⁻¹, based on incidence of skin tumors in mice [26] and a gastroin-

Table 3
Exposure parameters considered as point estimates and random variables for temple goers/workers.

Parameter	Symbol	Exposure groups		
		Temple goers with moderate exposure	Temple goers with high exposure level	Temple workers (extreme exposure)
Point estimate				
Visiting frequency (time yr ⁻¹) ^a	VF	33	365	365
Exposure duration (yr)	ED	1–50	1–50	1–50
Event frequency (event d ⁻¹)	EV	1	1	1
Averaging time for cancer effects (d)	AT _c	25550	25550	25550
Random variable				
Residence time (min time ⁻¹) [4]	RT	U(10, 60) ^b	U(10, 60)	U(480, 720)
Inhalation rate (m ³ d ⁻¹) [30]	IR _{inh}		LN(18.16, 1.65) ^c	
Ingestion rate (mg d ⁻¹) [38,39]	IR _{ing}		LN(26.95, 1.88)	
Dermal surface exposure (cm ²) [31]	SA		LN(4320.11, 1.04)	
Dermal adherence rate (mg cm ⁻² event ⁻¹) [31]	AF _d		LN(0.02, 2.67)	
Dermal adsorption fraction (unitless) [31]	AB		LN(0.13, 1.26)	
Body weight (kg) [40]	BW		LN(59.19, 1.06)	

^a Yearly estimated values: We assumed that moderate exposed groups visit the temple on the 1st and 15th days and on the major religious festivals for each lunar month based on the Chinese Lunar calendar and high and extreme exposed groups are daily temple goers and temple workers, respectively.

^b U denotes uniform distribution.

^c LN denotes lognormal distribution.

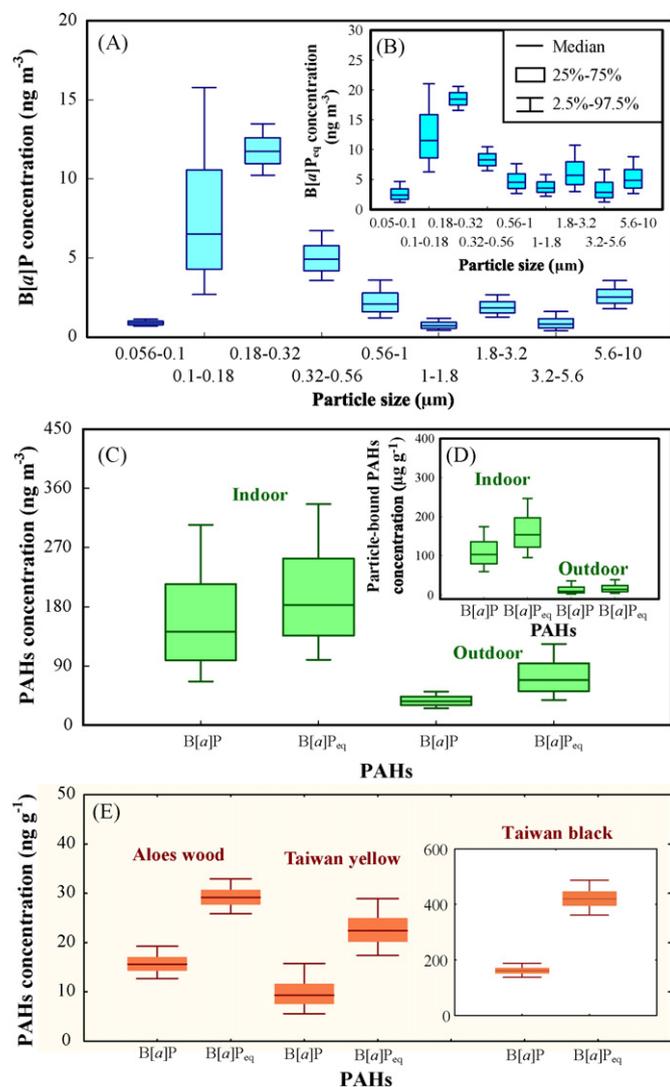


Fig. 1. Box and whisker plots of (A) B[a]P- and (B) B[a]P_{eq}-based concentrations for different size ranges in the north temple, (C) total and (D) particle-bound B[a]P- and B[a]P_{eq}-based concentrations in indoor/outdoor of the south temple, and (E) B[a]P- and B[a]P_{eq}-based concentrations for three representative types of incense of Taiwan yellow, Taiwan black, and aloe wood.

Table 4
B[a]P- and B[a]P_{eq}-based lifetime average daily dose for temple workers (extreme) and temple goers with high and moderate exposure levels.

Exposure route	Lifetime average daily dose (mg kg ⁻¹ d ⁻¹)			
	50th Percentile		95th Percentile	
	B[a]P	B[a]P _{eq}	B[a]P	B[a]P _{eq}
Temple workers				
Extreme exposure level				
Inhalation	4.94 × 10 ⁻⁶	7.58 × 10 ⁻⁶	1.27 × 10 ⁻⁵	1.95 × 10 ⁻⁵
Ingestion	1.06 × 10 ⁻⁵	1.29 × 10 ⁻⁵	3.59 × 10 ⁻⁵	4.25 × 10 ⁻⁵
Dermal contact	4.29 × 10 ⁻⁶	5.45 × 10 ⁻⁶	2.67 × 10 ⁻⁵	3.05 × 10 ⁻⁵
Overall	2.32 × 10 ⁻⁵	3.05 × 10 ⁻⁵	5.94 × 10 ⁻⁵	7.23 × 10 ⁻⁵
Temple goers				
High exposure level				
Inhalation	2.63 × 10 ⁻⁷	4.00 × 10 ⁻⁷	9.07 × 10 ⁻⁷	1.31 × 10 ⁻⁶
Ingestion	5.53 × 10 ⁻⁷	6.96 × 10 ⁻⁷	2.40 × 10 ⁻⁶	2.84 × 10 ⁻⁶
Dermal contact	2.27 × 10 ⁻⁷	2.81 × 10 ⁻⁷	1.63 × 10 ⁻⁶	1.88 × 10 ⁻⁶
Overall	1.23 × 10 ⁻⁶	1.62 × 10 ⁻⁶	4.15 × 10 ⁻⁶	5.03 × 10 ⁻⁶
Moderate exposure level				
Inhalation	2.38 × 10 ⁻⁸	3.61 × 10 ⁻⁸	8.19 × 10 ⁻⁸	1.24 × 10 ⁻⁷
Ingestion	4.99 × 10 ⁻⁸	6.44 × 10 ⁻⁸	2.22 × 10 ⁻⁷	2.62 × 10 ⁻⁷
Dermal contact	2.02 × 10 ⁻⁸	2.55 × 10 ⁻⁸	1.40 × 10 ⁻⁷	1.73 × 10 ⁻⁷
Overall	1.11 × 10 ⁻⁷	1.48 × 10 ⁻⁷	3.74 × 10 ⁻⁷	4.70 × 10 ⁻⁷

testinal absorption (ABS_{GI}) factor of 0.31, respectively [27,28]. We averaged those two CSF values and resulted in the arithmetic mean $30.5 \text{ (mg kg}^{-1} \text{ d}^{-1})^{-1}$.

Cancer risks from various exposure routes are assumed to be additive, as long as the risks are for the same individuals and time period. The total ILCR (TILCR) is the sum of risks associated with each exposure route (Table 2, Eq. (6)). The lower end of the range of acceptable risk distribution (TILCR) is defined by a single constraint on the 95th percentile of risk distribution that must be equal or lower than 10^{-6} for carcinogens.

A Monte Carlo simulation is performed using Crystal Ball software (Version 2000.2, Decisioneering, Inc., Denver, CO, USA) to quantify the uncertainty and its impact on the estimation of expected risk. A sensitivity analysis by using Spearman rank correlations is performed to determine which probability density functions have the greatest effect on the risk estimates.

Current literature was reviewed to develop probability distributions for the random variables appearing in the risk models adopted (Table 3). Having no site-specific data on population body weight, a second-order distribution was chosen for this parameter with the population age distribution in the Taiwan region to estimate the population body weight followed a lognormal distribution as a function of age [29]. Probability distributions chosen for the inhalation, ingestion, and dermal adherence rates and for the skin surface area are based on the body weight distribution probability. The PAHs inhalation rates are estimated from USEPA [30] suggested inhalation rates for various activities combined with the specific activity patterns (rest, sedentary, light, and moderate activities) of the average household. The exposed skin surface areas of PAHs are given by the specific exposed skin surface area for difference seasons (summer and winter) [31]. In our probabilistic exposure assessment, not only the point estimate values (e.g., VF , ED , EF , and AT_C) but also the proposed random variables (e.g., RT , IR_{inh} , IR_{ing} , SA , AF_d , AB , and BW) are considered (Table 3).

3. Results

3.1. Quantitative temple PAHs concentrations

The size distributions of B[a]P and B[a]P_{eq} are similarly followed a bimodal in the northern Taiwan temple (data reanalyzed from [8]) (Fig. 1A and B). Two peaks of the average mass distributions were found at 0.18–0.32 and 1.8–3.2 μm for B[a]P and B[a]P_{eq}, respec-

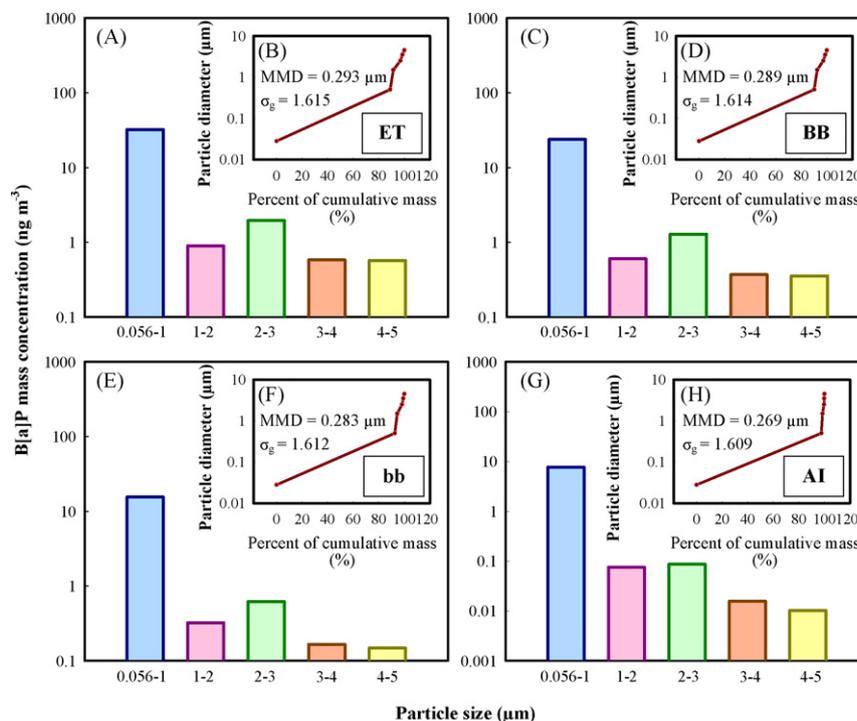


Fig. 2. Steady-state concentration distribution and size distribution patterns of B[a]P in different HRT regions: (A, B) ET, (C, D) BB, (E, F) bb, and (G, H) AI.

tively. The concentrations of B[a]P and B[a]P_{eq} in fine particles (aerodynamic diameter <1 μm) are nearly 1 order of magnitude higher than that in coarse particles (1 < aerodynamic diameter <10 μm), indicating that fine particles are the major contribution of B[a]P and B[a]P_{eq} concentrations in temple. In the southern Taiwan temple (data reanalyzed from [2]), the median B[a]P- and B[a]P_{eq}-based concentrations are 142.45 and 182.88; and 36.28 and 68.95 ng m⁻³ in indoor and outdoor, respectively (Fig. 1C), whereas the median particle-bound B[a]P- and B[a]P_{eq}-based concentrations are estimated to be 102.61 and 153.40; and 10.14 and 14.36 ng m⁻³ for indoor and outdoor, respectively (Fig. 1D). The median B[a]P- and B[a]P_{eq}-based concentrations for three types incense are estimated to be 9.36 and 22.44; 160.92 and 420.04; and 15.64 and 29.19 ng g⁻¹, respectively, for Taiwan yellow, Taiwan black, and aloe wood (Fig. 1E), implicating that Taiwan black incense may be a significant contribution factor to PAH sources.

3.2. Estimates of lifetime average daily dose (LADD)

LADD estimates of PAHs for temple goers/workers and three exposure routes are shown in Table 4. For temple workers, the median and 95th percentile of LADDs of B[a]P and B[a]P_{eq} for overall routes have orders of 10⁻⁵ mg kg⁻¹ d⁻¹, indicating high potential exposure risk; whereas for temple goers, the median and 95th percentiles of LADDs of B[a]P and B[a]P_{eq} for overall routes have orders of 10⁻⁶ and 10⁻⁷ mg kg⁻¹ d⁻¹, respectively. Our results also show that ingestion exposure has higher LADD estimates of PAHs than those of inhalation and dermal for all three exposure groups (Table 4).

3.3. PAHs in human respiratory tract

The steady-state B[a]P mass concentrations are estimated to be 0.57–32.23, 0.35–23.84, 0.15–15.50, and 0.01–7.69 ng m⁻³ in ET, BB, bb, and AI regions, respectively (Fig. 2A, C, E, G). The mass median diameters (MMDs) are calculated to be 0.293, 0.289, 0.283,

and 0.269 μm in ET, BB, bb, and AI regions, respectively (Fig. 2B, D, F, H). Temple goers with 35 min of daily exposure (high exposure group), the average daily doses of B[a]P and B[a]P_{eq} are estimated to be 0.002–0.020 and 0.008–0.32 μg d⁻¹; 0.016–0.227 and 0.077–0.360 μg d⁻¹; and 0.033–2.479 and 0.135–3.927 μg d⁻¹, respectively, in the BB, bb, and AI regions (Fig. 3A). Temple workers with 10 h of daily exposure (extreme exposure group), the estimated average daily doses of B[a]P and B[a]P_{eq} are 0.03–0.35 and 0.14–0.55 μg d⁻¹; 0.27–3.98 and 1.34–6.30 μg d⁻¹; and 0.57–43.38, and 2.36–68.72 μg d⁻¹, respectively, in the BB, bb, and AI regions (Fig. 3B). The similar results indicate that the average daily doses of B[a]P and B[a]P_{eq} obtained from fine fraction depositing to the AI region are significantly higher than those to the BB and bb regions.

3.4. Risk estimates

The 95% probability lung cancer risks (10⁻⁶ to 10⁻⁵) are greater than the USEPA acceptable level of 10⁻⁶ for temple workers and temple goers with high exposure level through inhalation route (Fig. 4A and B). Our result indicates that the higher ILCRs (10⁻⁶ to 10⁻⁴) are found in ingestion and dermal contact routes for temple goers/workers (Fig. 4). Notably, a large proportion of the risk comes from above two exposure routes for which they are assumed to occur in the temples.

The probability density functions (pdfs) of TILCRs for temple goers/workers (geometric standard deviations of lognormal distribution range from 1.95 to 2.42) are tended to be skewed (Fig. 5A, C, E). Percentile predictions of TILCRs personal exposure of temple goers/workers could be determined from cumulative density functions (cdfs) corresponding to pdfs. Under most regulatory program, an ILCR between 10⁻⁶ and 10⁻⁴ indicates potential risk; moreover, larger than 10⁻⁴ indicates high potential health risk. All 95% probabilities of B[a]P- and B[a]P_{eq}-based TILCRs are large than 10⁻⁶, indicating unacceptable probability distributions for temple goers/workers (Fig. 5). For personal extreme exposure to carcinogenic PAH in the temple, 95% probability TILCR (9.87 × 10⁻⁴ to

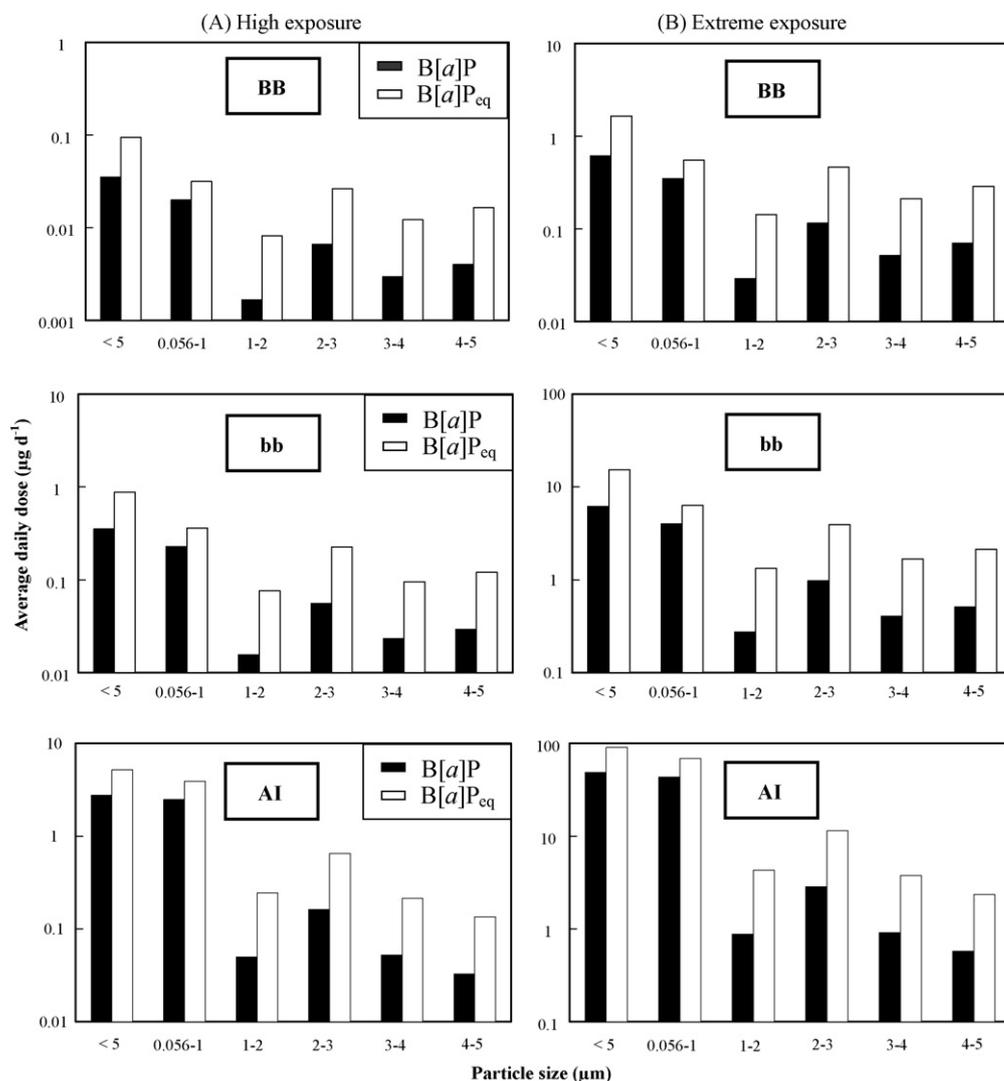


Fig. 3. Average daily dose to different size ranges of B[a]P and B[a]P_{eq} in different HRT regions: BB, bb, and AI for (A) temple goers with high exposure and (B) temple workers.

1.13×10^{-3}) is much greater than the range of 10^{-6} to 10^{-4} , indicating high potential health risk to temple workers; whereas for high and moderate exposed groups, 95% probability TILCRs range from 6.44×10^{-5} to 7.50×10^{-5} and 5.75×10^{-6} to 6.99×10^{-6} , respectively (Fig. 5B, D, F), indicating that health risk is alarming to temple goers.

3.5. Sensitivity analysis

For temple goers/workers, the most contributions to variance in risk are 47.2 and 78.0% in air inhalation rate (IR_a); 54.7 and 78.1% in particle ingestion rate (IR_p); and 72.6 and 86.1% in particle-to-skin adherence factor (AF_d), respectively, for inhalation, ingestion, and dermal contact routes (Fig. 6). Moreover, residence time (RT) (16.9–42.1%) and B[a]P concentration ($C_{B[a]P}$) (6–13%) play a more sensitive variable for temple goers, and $C_{B[a]P}$ (6.7–17.6%) also does for temple workers, in three exposure routes.

The present result indicates that for all exposure routes, the exposures of 365 yr^{-1} for temple goers yield ILCRs higher than 10^{-6} . The yielded risk values would be approximately 12-fold times of risk estimates for 33 yr^{-1} . We also calculated a risk-based visiting frequency advice (VF_{adv}) (yr^{-1}) and a suggested amount of incense burning associated with unit occupied volume of temple based on a maximum acceptable individual lifetime risk level of 10^{-6} . Our

results indicate that the B[a]P_{eq}- and B[a]P-based median VF_{adv} for temple goers are estimated to be 312.84 and 462.59 yr^{-1} ; 48.15 and 60.01 yr^{-1} ; 43.75 and 59.94 yr^{-1} ; and 18.08 and 22.67 yr^{-1} , respectively, for inhalation, ingestion, dermal contact, and overall routes. The suggested median incense burning amount for three representative incense types of Taiwan yellow, Taiwan black, and aloe wood range from 0.31 to 0.74 , 0.02 to 0.04 , and 0.24 to 0.44 g m^{-3} ; 5.84 to 13.73 , 0.31 to 0.81 , and 4.49 to 8.16 g m^{-3} ; and 64.58 to 150.39 , 3.49 to 8.95 , and 48.87 to 92.33 g m^{-3} , respectively, for temple workers and temple goers with high and moderate exposure levels, based on B[a]P_{eq}- and B[a]P-based calculation (Fig. 7).

4. Discussion

We have developed an integrated probabilistic risk assessment framework based on limited reported data to quantify exposure risk of temple goers/workers to airborne polycyclic aromatic hydrocarbons (PAHs) in typical Taiwanese temples. For temple workers, the median and 95th percentile of LADDs of B[a]P and B[a]P_{eq} for overall routes have orders of $10^{-5} \text{ mg kg}^{-1} \text{ d}^{-1}$, indicating high potential exposure risk. The MMDs of B[a]P are calculated to be 0.293 , 0.289 , 0.283 , and $0.269 \mu\text{m}$, respectively, in ET, BB, bb, and AI, indicating that fine particles are apt to exist in the AI region. The average daily doses of B[a]P and B[a]P_{eq} obtained from fine fraction depositing

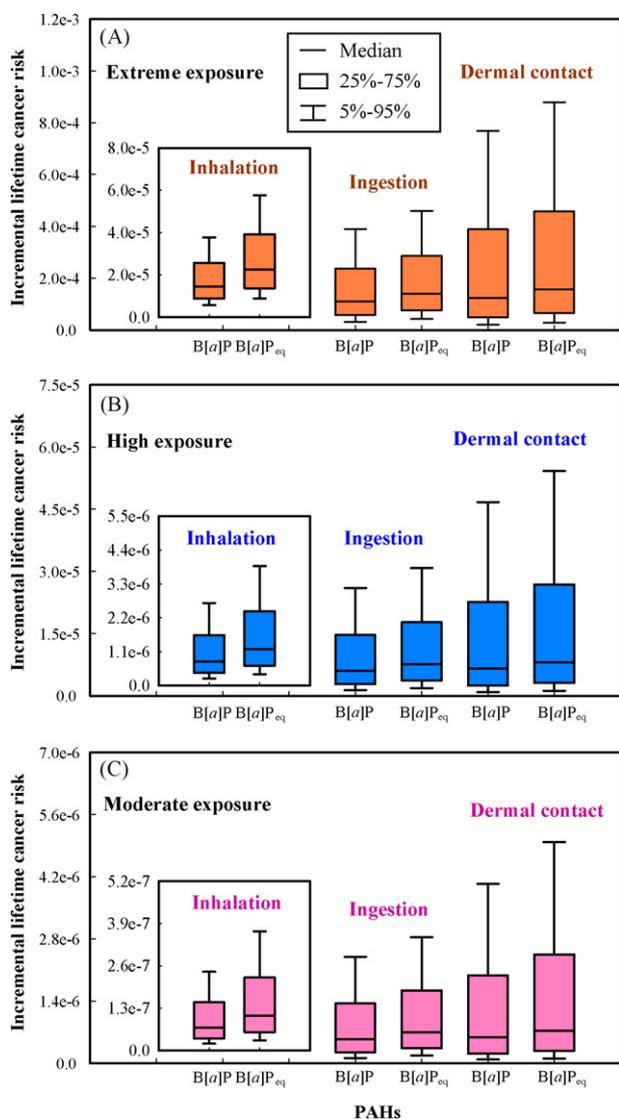


Fig. 4. Box and whisker plots of B[a]P- and B[a]P_{eq}-based inhalation, ingestion, and dermal contact incremental lifetime cancer risks for (A) temple workers and temple goers with (B) high, and (C) moderate exposure in the temples.

to the AI region are significantly higher than those to the BB and bb regions. Incremental risks of lung cancer are greater than the USEPA acceptable level of 10^{-6} for temple workers and temple goers with high exposure through inhalation route. Our finding also indicates that the higher ILCRs (10^{-6} to 10^{-4}) are found in ingestion and dermal contact route for temple goers/workers. For temple workers exposed to carcinogenic PAH, 95% probability TILCRs (9.87×10^{-4} to 1.13×10^{-3}) lie outside the range of 10^{-6} to 10^{-4} , indicating high potential health risk; whereas for temple goers, 95% probability TILCRs range from 6.44×10^{-5} to 7.50×10^{-5} and 5.75×10^{-6} to 6.99×10^{-6} , respectively, indicating that potential health risk is alarming.

The suggested visiting frequencies for temple goers are recommended to be 300, 50, 40, and 20 yr^{-1} , respectively, based on inhalation, ingestion, dermal contact, and overall routes considerations. The recommended maximum incense burning amount for three representative incense types of Taiwan yellow, Taiwan black, and aloe wood are suggested to be 0.74, 0.04, and 0.44 g m^{-3} ; 13.73, 0.81, and 8.16 g m^{-3} ; and 150.39, 8.95, and 92.33 g m^{-3} , respectively, for temple workers and temple goers with high and moderate exposures.

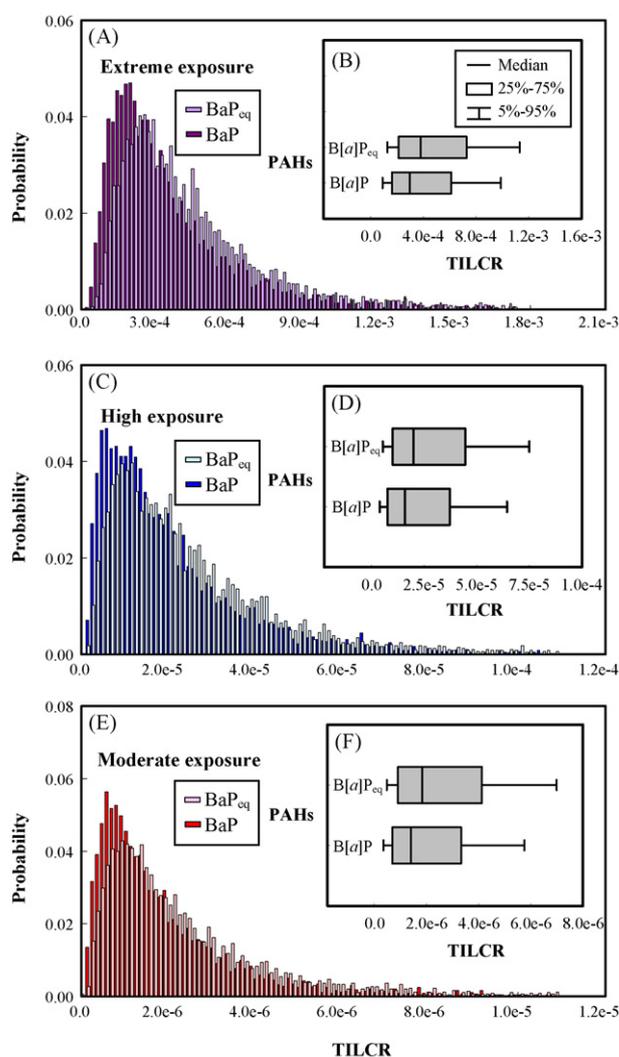


Fig. 5. Predicted probability density functions and box and whisker plots of B[a]P- and B[a]P_{eq}-based total incremental lifetime cancer risks (TILCRs) for (A, B) temple workers and temple goers with high (C, D), and moderate (E, F) exposures in the temples.

Generally, variables in the numerator of the exposure models (Eqs. (1)–(3)) (IR_a , IR_p , AF_d , RT , etc.) will tend to be positively correlated with risk, whereas variables in the denominator (body weight (BW)) will tend to be negatively correlated with risk. Because temple workers spend much longer time in the temples, the variables with the greatest effect on risk are IR_a , IR_p , and AF_d , followed by $C_{B[a]P}$. For temple goers/workers, there are several risk management options, for example (i) to reduce the residence time; (ii) to visit temples with a better ventilation conditions, fewer censers, and a lower required number of joss sticks; (iii) to avoid worshipping on the 1st and 15th days and major religious festivals in each lunar month; (iv) to wear masks; and (v) to maintain the cleanness of the censers.

In fact, the population groups have different susceptibility for disease. Therefore, the potential risk induced from incense burning for specific subgroups (child, adolescent, and adult) should be taken into account in the future work, especially for child. In light of this aspect, different contact rates (CR , e.g., $CR = IR_{inh}$ through inhalation pathway), exposure durations (ED), and subgroup body weights (BW) should be taken into account in the estimates of lifetime average daily dose (LADD). A paradigm for consequent exposure group

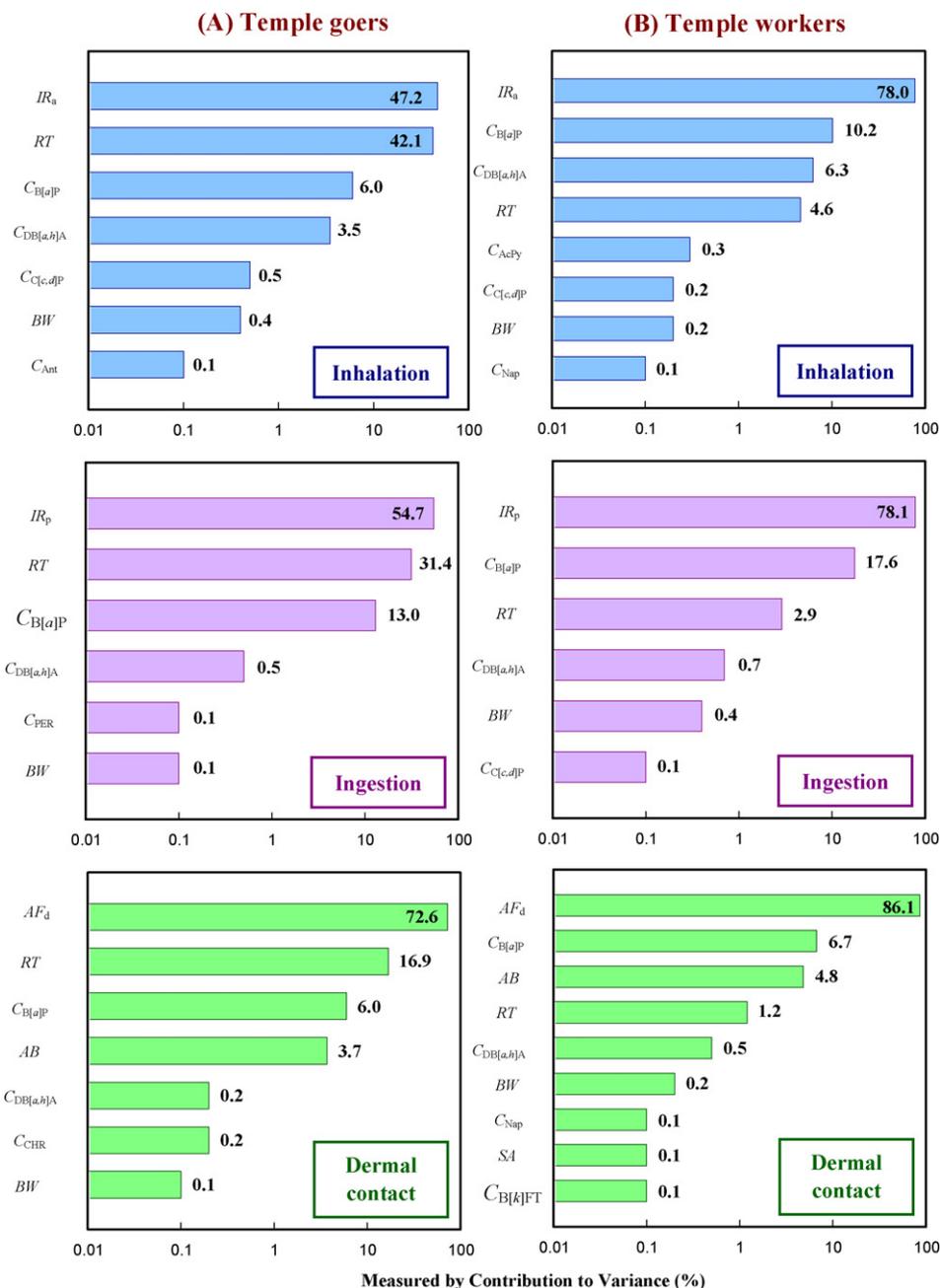


Fig. 6. Sensitivity analysis of inhalation, ingestion, and dermal contact cancer risk models for (A) temple goers and (B) temple workers.

in residential home, the $LADD_{inh}$ should be revised as,

$$LADD_{inh} = \frac{C_a \cdot RT \cdot VF \cdot cf}{AT_c} \cdot \left(\frac{CR_{child} \cdot ED_{child}}{BW_{child}} + \frac{CR_{adolescent} \cdot ED_{adolescent}}{BW_{adolescent}} + \frac{CR_{adult} \cdot ED_{adult}}{BW_{adult}} \right),$$

where the parameters of C_a , RT , VF , cf , and AT_c have been defined in Table 3.

The differences in CSF_i for different routes of exposure are due in part to the different treatment procedures and to assumptions adopted on animal physiological parameters (e.g., surface area scaling and route-to-route extrapolation factors). The one of the used dermal CSF_i is based on a gastrointestinal absorption (ABS_{GI}) factor of 0.31 [28]. It is consistent with the following conditions: (i) the critical study upon which the toxicity is based on employing an

administered dose (e.g., delivery in diet or by gavages) in its study design and (ii) adjustment of the oral toxicity value is significant only when the ABS_{GI} is <50% [31].

Kameda et al. [32] and Liao and Chiang [14] have evaluated the actual human exposure by HRT model and ILCRs exposed to carcinogenic PAHs, the former estimated the $B[a]P_{eq}$ -based ILCR from ambient using WHO unit risk, whereas the later used an integrate framework which combine U.S. EPA protocol and probabilistic risk assessment model to assess the $B[a]P_{eq}$ -based ILCR. Recently, the resident home and church were investigated for human exposed to airborne pollutants from indoor activities [33–36], showing that the PAHs and/or PMs in smaller size fraction have significant potential toxicities, especially for ultrafine particles (UFPs). Ott and Siemann [37] monitored the p-PAH emitted from different activity indoors by concentration and active surface methods simultaneously. In the future, the mass concentration of exposure PAHs maybe replaced

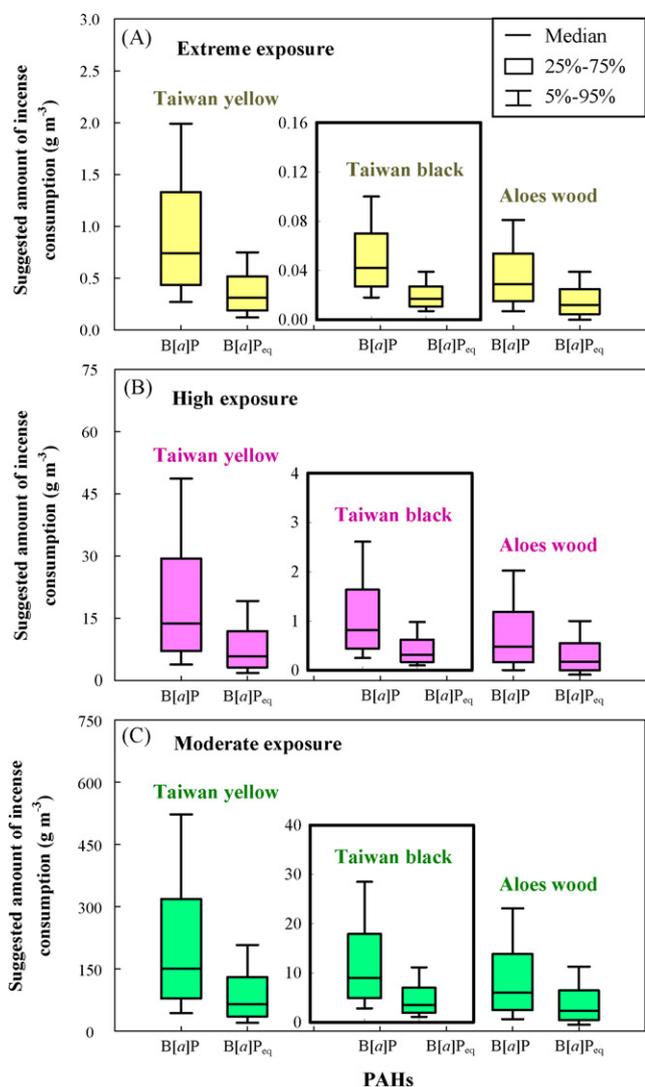


Fig. 7. Box and whisker plots of estimated suggested amount of incense consumption for three types of incense of Taiwan yellow, Taiwan black, and aloe wood for (A) temple workers and temple goers with (B) high, and (C) moderate exposures.

by number concentration and surface area of UFPs, those can be as ideal dosimetry for toxic exposure. Based on our results and literature evidence, Taiwan regulatory agencies should pay more attention to study UFP number concentrations and carcinogenic compositions in indoor environments and to establish the related criteria of indoor air quality.

We recognize limitations in each of our data sources, particularly the inherent problem of uncertainty and variability of the data. The strength of these results rests on the consistent agreement of mathematical models and public and regulatory authority's guideline values. Our analysis may provide a wider context for the interpretation of regional PAHs-induced cancer risk estimates that produced diverging and controversial outcomes, which has economic and policy implications. Although more complex models may be necessary to answer specific questions regarding risk or particular management strategies, our simple model captures the essential risk analysis methodology and its flexible enough to integrate effects occurring at varying subpopulation scales.

In conclusion, our proposed probabilistic risk assessment framework provides a template for integrating the temple PAHs data, human respiratory tract model, and risk modeling techniques to accurately estimate the safe temple visiting frequency guideline associated with the safe incense burning amounts. We are confi-

dent that our model can be used to assess the cumulative cancer risk easily for any temple exposure scenario. At the same time, the framework also encourages risk managers to establish the appropriate safe airborne PAHs guideline or to quantify rigorous health risk estimates for temple goers/workers.

References

- [1] H.H. Yang, R.C. Jung, Y.F. Wang, L.T. Hsieh, Polycyclic aromatic hydrocarbon emissions from joss paper furnaces, *Atm. Environ.* 39 (2005) 3305–3312.
- [2] T.C. Lin, F.H. Chang, J.H. Hsieh, H.R. Chao, M.R. Chao, Characteristics of polycyclic aromatic hydrocarbons and total suspended particulate in indoor and outdoor atmosphere of a Taiwanese temple, *J. Hazard. Mater.* 95 (2002) 1–12.
- [3] G.C. Fang, C.N. Chang, C.C. Chu, Y.S. Wu, P.P.C. Fu, S.C. Chang, I.L. Yang, Fine (PM_{2.5}), coarse (PM_{2.5-10}), and metallic elements of suspended particulates for incense burning at Tzu Yun Yen temple in central Taiwan, *Chemosphere* 51 (2003) 983–991.
- [4] S.C.C. Lung, M.C. Kao, S.C. Hu, Contribution of incense burning to indoor PM₁₀ and particle-bound polycyclic aromatic hydrocarbons under two ventilation conditions, *Indoor Air* 13 (2003) 194–199.
- [5] S.C.C. Lung, K.J. Guo, P.Y. Chen, P.F. Tsai, P.C. Chen, Participants' exposure to PM_{2.5} and gaseous/particulate polycyclic aromatic hydrocarbons during the Ma-tsu Goddess parade, *J. Expo. Anal. Environ. Epidemiol.* 14 (2004) 536–543.
- [6] C.C. Tonne, R.M. Whyatt, D.E. Camann, F.P. Perera, P.L. Kinney, Predictors of personal polycyclic aromatic hydrocarbon exposures among pregnant minority women in New York City, *Environ. Health Perspect.* 112 (2004) 754–759.
- [7] H.W. Chen, Environmental exposure to nitrated polycyclic aromatic hydrocarbons in the Taiwanese temple, *B. Environ. Contam. Toxicol.* 74 (2005) 399–406.
- [8] H.C. Chuang, Effects of temple particles on inflammatory reaction and endothelial dysfunction by human coronary artery endothelial cell, Master thesis, National Taiwan University, Taipei, Taiwan ROC, 2005.
- [9] C.K. Ho, W.R. Tseng, C.Y. Yang, Adverse respiratory and irritant health effects in temple workers in Taiwan, *J. Toxicol. Environ. Health Part A* 68 (2005) 1465–1470.
- [10] J. Pauluhn, Mosquito coil smoke inhalation toxicity. Part I: validation of test approach and acute inhalation toxicity, *J. Appl. Toxicol.* 26 (2006) 269–278.
- [11] P.P. Ballesta, E. De Salgar, D. Kotzias, State of the art of the PAHs' analysis in ambient air, *Fresenius. Environ. Bull.* 8 (1999) 499–505.
- [12] C.E. Boström, P. Gerde, A. Hanberg, B. Jernstrom, C. Johansson, T. Kyrklund, A. Rannug, M. Tornqvist, K. Victorin, R. Westerholm, Cancer risk assessment, indicators, and guidelines for polycyclic aromatic hydrocarbons in the ambient air, *Environ. Health Perspect.* 110 (2002) 451–489.
- [13] G.C. Fang, C.C. Chu, Y.S. Wu, P.P.C. Fu, Emission characters of particulate concentrations and dry deposition studies for incense burning at a Taiwanese temple, *Toxicol. Ind. Health* 18 (2002) 183–190.
- [14] C.M. Liao, K.C. Chiang, Probabilistic risk assessment for personal exposure to carcinogenic polycyclic aromatic hydrocarbons in Taiwanese temples, *Chemosphere* 63 (2006) 1610–1619.
- [15] K.C. Chiang, C.M. Liao, Heavy increase burning in temples promotes exposure risk from airborne PMs and Carcinogenic PAHs, *Sci. Total Environ.* 372 (2006) 64–75.
- [16] C.S. Li, Y.S. Ro, Indoor characteristics of polycyclic aromatic hydrocarbons in the urban atmosphere of Taipei, *Atm. Environ.* 34 (2000) 611–620.
- [17] J.F. Collins, J.P. Brown, G.V. Alexeeff, A.G. Salmon, Potency equivalency factors for some polycyclic aromatic hydrocarbons and polycyclic aromatic hydrocarbon derivatives, *Regul. Toxicol. Pharm.* 28 (1998) 45–54.
- [18] I.C.T. Nisbet, P.K. LaGoy, Toxic equivalency factors (TEFs) for polycyclic aromatic hydrocarbons (PAHs), *Regul. Toxicol. Pharm.* 16 (1992) 290–300.
- [19] H.M. Malcolm, S. Dobson, The Calculation of an Environmental Assessment Level (EAL) for Atmospheric PAHs Using Relative Potencies, Department of the Environment, London, UK, 1994.
- [20] U.S. EPA, Risk Assessment Guidance for Superfund: Human Health Evaluation Manual Part A (Interim Final), EPA/540/1-89/002, Office of Emergency and Remedial Response, Washington, DC, 1989.
- [21] ICRP, Human respiratory tract model for radiological protection, a report of a task group of the international commission on radiological protection, ICRP Publication No. 66. Pergamon Press, New York, 1994.
- [22] C.M. Liao, J.W. Chen, S.J. Huang, Size-dependent PM₁₀ indoor/outdoor/personal relationships for a wind-induced naturally ventilated airspace, *Atm. Environ.* 37 (2003) 3065–3075.
- [23] J.W. Chen, C.M. Liao, S.C. Chen, Compartmental human respiratory tract modeling of airborne dust exposure from feeding in swine buildings, *J. Air Waste Manage. Assoc.* 54 (2004) 331–341.
- [24] OEHHA, Air Toxics Hot Spots Program Risk Assessment Guidelines, Part II: Technical Support Document For Describing Available Cancer Potency Factors, California Environmental Protection Agency, Sacramento, California, 2005.
- [25] J. Neal, R.H. Rigdon, Gastric tumors in mice fed benzo[a]pyrene: a quantitative study, in: *Texas Reports on Biology Medicine*, Vol. 25, 1967, pp. 553–557.
- [26] U. Mohr, D. Schmahl, L. Tomatis, Air Pollution and Cancer in Man, IARC Publication No. 16, World Health Organization, France, 1977.
- [27] M. Hussain, J. Rae, A. Gilman, P. Kauss, Lifetime health risk assessment from exposure of recreational users to polycyclic aromatic hydrocarbons, *Arch. Environ. Contam. Toxicol.* 35 (1998) 527–531.

- [28] RAIS, Toxicity profiles: benzo[a]pyrene. Risk Assessment Information System (RAIS). Available from the website (<http://risk.lsd.ornl.gov/index.shtml>), 2006.
- [29] D.E. Burmaster, E.A.C. Crouch, Lognormal distributions for body weight as a function of age for males and females in the United States, 1976–1980, *Risk Anal.* 17 (1997) 499–505.
- [30] U.S. EPA, Exposure Factors Handbook, EPA/600/P-95/002F, National Center for Environmental Assessment, Washington, DC, 1997.
- [31] U.S. EPA, Risk Assessment Guidance for Superfund: Human Health Evaluation Manual (Part E, Supplemental Guidance for Dermal Risk Assessment), EPA/540/R/99/005, Office of Superfund Remediation and Technology Innovation, Washington, DC, 2004.
- [32] Y. Kameda, J. Shirai, T. Komai, J. Nakanishi, S. Masunaga, Atmospheric polycyclic hydrocarbons: size distribution, estimation of their risk and their deposition to the human respiratory tract, *Sci. Total Environ.* 340 (2005) 71–80.
- [33] S. Weber, Exposure of churchgoers to airborne particles, *Environ. Sci. Technol.* 40 (2006) 5251–5256.
- [34] T. Hussein, T. Glytsos, J. Ondráček, P. Dohányosová, V. Zdimal, K. Hämeri, M. Lazaridis, J. Smolík, M. Kulmala, Particle size characterization and emission rates during indoor activity in a house, *Atm. Environ.* 40 (2006) 4285–4307.
- [35] L. Wallace, Indoor sources of ultrafine and accumulation mode particles: size distributions, size-resolved concentrations, and source strengths, *Aerosol Sci. Technol.* 40 (2006) 348–360.
- [36] S.W. See, Y.H. Wang, R. Balasubramanian, Contrasting reactive oxygen species and transition metal concentrations in combustion aerosols, *Environ. Res.* 103 (2007) 317–324.
- [37] W.R. Ott, H.C. Siegmann, Using multiple continuous fine particle monitors to characterize tobacco, incense, candle, cooking, wood burning, and vehicular sources in indoor, outdoor, and in-transit settings, *Atm. Environ.* 40 (2006) 821–843.
- [38] U.S. EPA, Guidance Manual for the Integrated Exposure Uptake Biokinetic Model for Lead in Children, EPA/540/E-93/081, Office of Solid Waste and Emergency Response, Washington, DC, 1994.
- [39] U.S. EPA, Recommendations of the Technical Review Workgroup for Lead for an Approach to Assessing Risks Associated with Adult Exposures to Lead in Soil, EPA-540-R-03-001, United States Environmental Protection Agency, Washington, DC, 1996.
- [40] DOH (Department of Health, Taiwan, ROC), Available from the website (<http://www.doh.gov.tw/cht/index.aspx#>), 2006.