

Characteristics of Polycyclic Aromatic Hydrocarbon Emissions of Particles of Various Sizes from Smoldering Incense

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Abstract Release of polycyclic aromatic hydrocarbons (PAHs) in particles of various sizes from smoldering incenses was determined. Among the three types of incense investigated, yielding the total PAH emission rate and factor ranges for PM_{0.25} were 2,139.7–6,595.6 ng/h and 1,762.2–8,094.9 ng/g, respectively. The PM_{0.25}/PM_{2.5} ratio of total PAH emission factors and rates from smoldering three incenses was greater than 0.92. This study shows that total particle PAH emission rates and factors were mainly <0.25 μm . Furthermore, the total toxic equivalency emission rates and factors of PAHs for PM_{0.25} were 241.3–469.7 and 198.8–576.2 ng/g from the three smoldering incenses. The benzo[a]pyrene accounted for 65.2%–68.0% of the total toxic equivalency emission factor of PM_{2.5} for the three incenses. Experimental results clearly indicate that the PAH emission rates and factors were influenced significantly by incense composition, including carbon and hydrogen content. The study concludes that smoldering incense with low atomic hydrogen/carbon ratios minimized the production of total polycyclic aromatic hydrocarbons of both PM_{2.5} and PM_{0.25}.

Keywords Incense · Smoldering · Polycyclic aromatic hydrocarbons · Emission rate

Although a common religious ritual in Chinese society, burning incense sticks generates air pollutants that are correlated with lung cancer incidence (MacLennan et al. 1977), childhood leukemia (Lowengard et al. 1987) and brain tumors (Preston-Martin et al. 1982). Additionally, the Ames test has proven the mutagenic effect of incense smoke (Rasmussen 1987; Sato et al. 1980). Incense smolders producing smoke containing both gas and particles. The gas phase consists of carbon monoxide, carbon dioxide, formaldehyde, volatile organic compounds (VOCs) and other compounds (Schoental and Gibbard 1967; Chang et al. 1997; Lee and Wang 2004; Yang et al. 2007; Liou et al. 2008). The particles are generally <1 μm in size (Yang et al. 2005, 2006) with embedded organic compounds such as polycyclic aromatic hydrocarbons (PAHs) (Lin and Lee 1998). The adverse health effects caused by incense smoke may be related to an extremely high number of fine particles and organic compounds, which can enhance each other's effects and easily penetrate the respiratory system, deep into alveolar regions. The characteristics of incense smoke may vary with incense ingredients (carbon, hydrogen, and nitrogen) and combustion conditions (i.e., airflow, temperature, humidity, and oxygen content). This study elucidates the effects of burning various incenses with various chemical compositions (atomic hydrogen/carbon (H/C) ratios) on yields of PAHs with various particle sizes during combustion in an attempt to prevent indoor contamination caused by burning incense.

Materials and Methods

Three types of incense comprised of various incense powders were tested in this study: standard-smoke Taiwanese Black Signaloes Incense (incense A; traditional

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incense); low-smoke Japanese Wild Rose Incense (incense B); and, very-low-smoke Japanese Cherry Blossom Incense (incense C). The powder in incense A was lignaloes. Incense B was made of an unknown natural wood powder, rose essential oil, and charcoal. Incense C was made of Meng Zong bamboo charcoal, plant essential oils, and cherry tree bark. An elemental analyzer (Elementar Vario EL-III; Elementar Analysen System, GmbH, Germany) was used to measure the carbon, hydrogen, and nitrogen content in these incenses.

A joss stick was ignited and inserted into an incense holder at the bottom of the smoldering chamber, where a purified compressor air supplied air that was regulated by a mass flow controller (Brooks, USA) (Fig. 1). The test flow rate was set at 11 L/min, such that particle emissions reached a steady state (Yang et al. 2007). The generated gases and aerosol pollutants were introduced into a 4.7-L test chamber (inner diameter, 10 cm; height, 60 cm). Smoke sampling was conducted at the airstream exit of the test chamber. A Sioutas Personal Cascade Impactor (SKC, USA) with 25 and 37-mm quartz membrane filters (Pall, USA) at a sampling flow rate of 9 L/min for 15 min was utilized to collect particulates in five size ranges: >2.5 , 1.0–2.5, 0.50–1.0, 0.25–0.5, and <0.25 μm (PM_{0.25}). Therefore, the PM_{2.5} is the sum of particle mass collected on the latter four stages. Before and after burning tests, joss sticks were weighed to determine net mass loss, which was used to calculate the incense burning rate. Each tested incense was analyzed three times and three representative samples were obtained.

The extraction procedure was as follows. Each sample was extracted using 40 mL dichloromethane (UN1539; Merck, Germany) in an ultrasonic bath (DC400H; Delta, Taiwan) for 15 min, followed by a 5-min rest period. This extraction procedure was repeated three times to ensure a complete extraction. Extracted solutions were filtered through a pre-cleaned Pasteur pipette filled with solvent-rinsed glass wool (Sigma-Aldrich, USA), cleaned with anhydrous Na_2SO_4 (12–60 mesh; Mallinckrodt Baker, Inc., USA), and concentrated in a rotary evaporator with a thermostatic bath (Eyela, Japan) at $T = 35^\circ\text{C}$. Final volume was roughly 2 mL. This 2 mL of solution was dried to 0.5 mL under a weak nitrogen flow; 0.1 mL internal standards (Naphthalene-D₈, Acenaphthene-D₁₀, Phenanthrene-D₁₀, Chrysene-D₁₂, Perylene-D₁₂) (Supelco, USA) was then added. Finally, solution volume was adjusted to 1 mL by adding pure dichloromethane.

The 16 organic PAH pollutants produced by smoldering incense were identified and quantified by gas chromatography/mass spectrometry (GCMS) (GCMS-QP2010; Shimadzu, Japan). The GC/MS operational conditions were as follows. Injection mode was splitless with an injection volume of 1 μL . Injection port temperature was 280°C ; ion

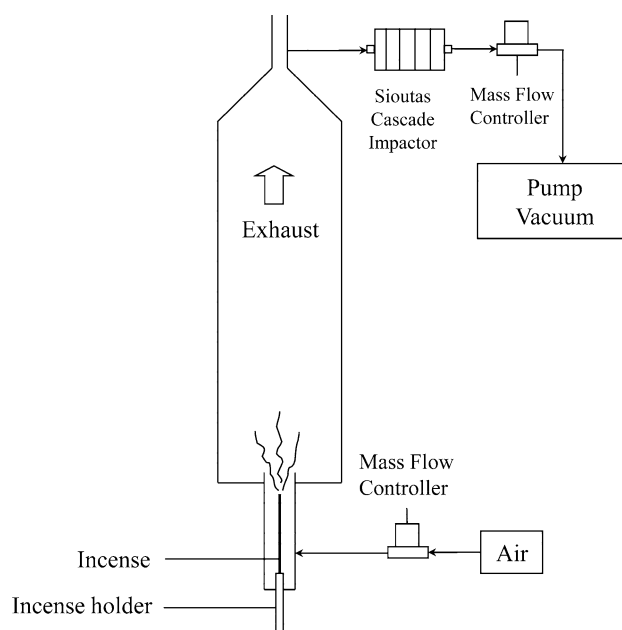


Fig. 1 A schematic diagram of the incense combustion test system

source temperature was 230°C ; and interface temperature was 290°C . The capillary column (DB-5MS; Agilent J&W, USA) had an inner diameter of 0.25 mm, length of 30 m, and film thickness of 0.25 μm . The linear velocity for the carrier gas, He, was 36.7 cm/sec. The initial temperature of the oven was maintained at 70°C for 3 min, and then increased at $10^\circ\text{C}/\text{min}$ to 300°C . Last, the oven was maintained at 300°C for 20 min. Total analytical time was 46 min. Analysis was conducted in selective ion monitoring (SIM) mode. The quantification ion and confirmation ion for SIM mode were the values used in the study by Orecchio (2007).

The standards of the 16 organic PAH compounds (Naphthalene (Nap), Acenaphthylene (AcPy), Acenaphthene (AcP), Fluorene (Flu), Phenanthrene (Phen), Anthracene (Ant), Fluoranthene (FL), Pyrene (Pyr), Benzo[a]anthracene (BaA), Chrysene (Chr), Benzo[b]fluoranthene (BbF), Benzo[k]fluoranthene (BkF), Benzo[a]pyrene (BaP), Indeno[1,2,3-cd]pyrene (INP), Dibenzo[a,h]anthracene (DBA), and Benzo[g,h,i]perylene (Bghip)) (AccuStandard, USA) with 5 internal standards were analyzed in the 5–800 ng/mL range to establish standard calibration curves to determine the concentrations of the 16 PAHs collected. All correlation coefficients for calibration curves were >0.998 . Method detection limits were 0.82–4.77 ng/m³, determined by threefold standard deviation of seven measurements of the lowest concentration of a calibration curve. Blank filters were analyzed for each test in order to estimate the potential contamination. The background value of a blank filter was subtracted for all samples. An appropriate amount of PAHs was added to investigate the

extraction efficiency. After sample extraction, the PAHs were analyzed by GC/MS to determine recovery of the 16 PAHs. Mean recovery for all compounds was 72.7%–107.1%.

The emission rates and emission factors of each pollutant were calculated, based on the conservation of mass, as following:

$$V \times \frac{dC_i}{dt} = R \times E_f - Q \times C_i \quad (1)$$

where V (m^3) is test chamber volume; C_i (ng/m^3) is the concentration of each pollutant at a given time; R (g/h) is the incense burning rate; E_f (ng/g) is the emission factor of each pollutant, and Q (11 L/min) is airflow rate. The $dC_i/dt = 0$ as generation and removed is in dynamic equilibrium, Eq. 1 can be rewritten as follows:

$$E_f = \frac{Q \times C}{R} \quad (2)$$

In Eq. 2, emission rate (ng/h) was calculated by multiplying flow rate by the equilibrium concentration of pollutants. The emission factor, E_f , is the specific emission rate normalized to the incense burning rate.

Results and Discussion

The carbon, hydrogen, and nitrogen weight percentages of standard-smoke (incense A), low-smoke (incense B) and very-low-smoke (incense C) incenses were as follows: 43.91%, 5.90%, and 0.44%; 47.60%, 4.98%, and 0.17%; 54.14%, 4.03%, and 0.27%, respectively. The atomic H/C ratio of incense A, B, and C was 1.61, 1.25, and 0.89, respectively. The atomic H/C ratio of very-low-smoke incense was lower than that of standard-smoke and low-smoke incense. The burning rates of incense A, B, and C were 0.81 ± 0.02 (average \pm S.D., $n = 3$), 1.04 ± 0.02 ($n = 3$), and 1.21 ± 0.005 ($n = 3$) g/h, respectively. The burning rate of very-low-smoke incense was faster than that of standard-smoke and low-smoke incense.

The PM_{0.25}/PM_{2.5} ratio of mass emission factors from smoldering standard-smoke (incense A), low-smoke (incense B) and very-low-smoke (incense C) incense was 0.85 ± 0.01 ($n = 3$), 0.78 ± 0.01 ($n = 3$) and 0.73 ± 0.01 ($n = 3$), respectively (Fig. 2). This analytical result indicates that particle sizes in incense smoke were mainly $<0.25 \mu m$. The geometric mean diameter (GMD) of particles in incense smoke from high to low was $0.22 \mu m$ for very-low-smoke, $0.19 \mu m$ for low-smoke, and $0.16 \mu m$ for standard-smoke incense. The main reason is that smoldering very-low-smoke and low-smoke incense sticks produced more mass of particles in the range of $1\text{--}2.5 \mu m$ than standard-smoke incense. Furthermore, the PM_{2.5}

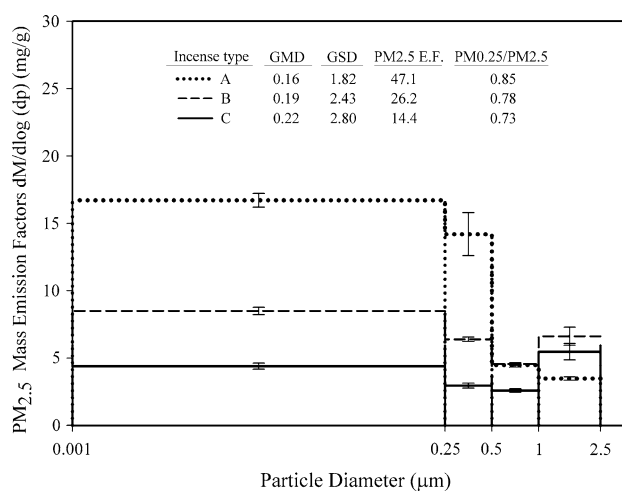


Fig. 2 Aerosol emission factor versus particle size from smoldering incenses (Bar: mean \pm standard deviation)

mass emission factors for standard-smoke (A), low-smoke (B) and very-low-smoke (C) incense smoldering were 47.1 ± 1.45 ($n = 3$), 26.2 ± 0.41 ($n = 3$), and 14.4 ± 0.71 ($n = 3$) mg/g, respectively. Thus, PM_{2.5} mass emission factors during smoldering was in the order of very-low-smoke $<$ low-smoke $<$ standard-smoke incense. Analytical results demonstrate that smoldering incense with a low atomic H/C ratio minimized the PM_{2.5} mass emission.

Except for Nap and Acp in very-low-smoke incense, the PM_{0.25}/PM_{2.5} ratio of individual PAH emission factors from smoldering standard-smoke (A), low-smoke (B) and very-low-smoke (C) incense was 0.81–1.00, 0.92–1.00, and 0.84–1.00, respectively (Table 1). The percentages of two- (Nap), three- (AcPy, Acp, Flu, Phen, Ant), four- (FL, Pyr, BaA, Chr), five- (BbF, BkF, BaP, DBA), and six-ring PAHs (INP, BghiP) in PM_{0.25} for three incenses were 0%–0.5%, 11.1%–29.9%, 52.9%–62.7%, 12.8%–20.1%, and 4.0%–6.1%, respectively. These analytical results indicate that four-ring PAHs predominated in PM_{0.25}. The major species of PM_{0.25} were FL and Pyr for all three incenses. Additionally, the range of total PAH content per unit mass of PM_{0.25} from smoldering standard-smoke (incense A), low-smoke (incense B) and very-low-smoke (incense C) incense was 167.9–206.3 $\mu g/g$. The total PAH content per unit mass of PM_{0.25} was higher than that of particles in the range 0.25–0.5, 0.5–1 and 1–2.5 μm for the three incenses (Table 1). These experimental results demonstrate that PM_{0.25} has higher health risks than particles in the range 0.25–0.5, 0.5–1 and 1–2.5 μm . Furthermore, measurement results obtained by Lung and Hu (2003) for total PAH content per unit mass of total particles were 963 and 618 $\mu g/g$ for two incenses. This experimental result in this study was significantly different in those identified by Lung and Hu (2003), likely because of differences in the combustion conditions.

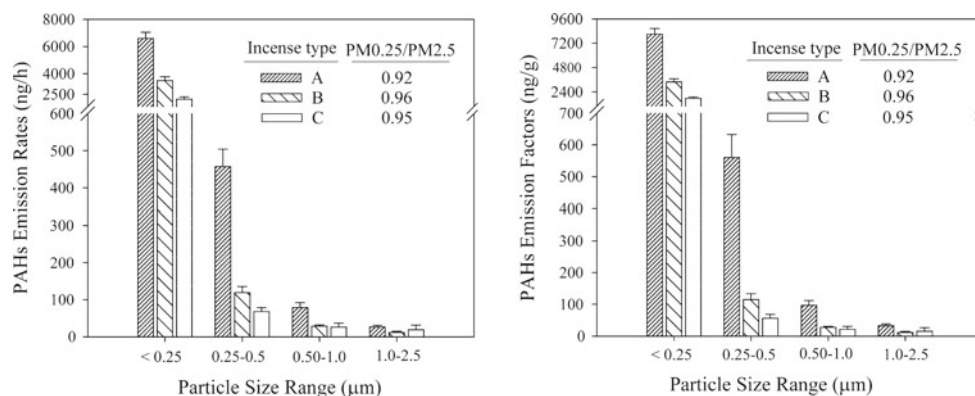
Table 1 The PAH emission factors and total PAH content per unit mass of particles versus particle size from burning various incense sticks

Chemicals	Incense type											
	A				B				C			
	Particle size range (μm)				Particle size range (μm)				Particle size range (μm)			
	<0.25	0.25–05	0.50–1.0	1.0–2.5	<0.25	0.25–05	0.50–1.0	1.0–2.5	<0.25	0.25–05	0.50–1.0	1.0–2.5
Nap	37.3 ± 9.5 ^a	ND	2.0 ^c	ND ^d	7.4 ± 1.6	ND	ND	ND	ND	ND	ND	2.5 ^c
AcPy	99.9 ± 8.7	ND	ND	ND	27.9 ± 1.2	ND	ND	ND	3.5 ± 1.4 ^b	ND	ND	ND
AcP	127.4 ± 4.1	ND	ND	ND	56.1 ± 5.4	ND	ND	ND	ND	ND	ND	ND
Flu	296.4 ± 12.5	28.4 ± 3.1	4.5 ± 0.4	2.1 ± 0.3	162.4 ± 13.9	7.0 ± 1.0	2.0 ± 0.4	0.9 ± 0.4 ^b	55.2 ± 5.2	1.2 ± 0.2	0.9 ^c	ND
Phen	1,543.1 ± 175.9	32.4 ± 9.0	9.0 ± 4.5	4.1 ± 0.8	510.8 ± 180.2	1.4 ^c	1.8 ± 0.8	1.2 ± 0.4 ^b	107.3 ± 27.4	ND	1.3 ^c	2.7 ± 2.1 ^b
Ant	353.6 ± 45.6	16.3 ± 3.7	ND	ND	108.3 ± 40.9	ND	ND	ND	29.5 ± 6.6	ND	ND	ND
FL	1,315.5 ± 105.7	122.7 ± 14.1	16.6 ± 3.5	13.0 ± 6.4	546.2 ± 32.1	18.1 ± 2.4	5.4 ± 0.4	3.7 ± 1.1	242.1 ± 30.6	5.3 ± 1.7	6.1 ± 8.1	1.9 ± 0.4
Pyr	1,621.1 ± 115.4	132.8 ± 10.9	14.1 ± 2.8	4.3 ± 0.6	661.6 ± 27.2	20.0 ± 2.1	3.6 ± 0.4	1.2 ± 0.8	370.1 ± 43.3	6.1 ± 1.4	ND	ND
BaA	603.6 ± 58.6	53.5 ± 3.0	8.5 ± 0.4	3.2 ± 0.6	306.1 ± 3.4	17.5 ± 1.5	4.4 ± 0.8	2.5 ± 0.3	219.8 ± 11.2	9.1 ± 1.0	1.6 ± 0.2	1.5 ± 0.3
Chr	743.6 ± 61.7	58.2 ± 6.4	ND	ND	351.0 ± 2.6	14.9 ± 4.9	ND	ND	273.8 ± 14.5	8.6 ± 3.6	5.2 ^c	ND
BbF	504.0 ± 48.9	48.3 ± 3.4	13.3 ± 2.5	4.0 ± 0.7	255.7 ± 1.7	16.0 ± 1.3	4.7 ± 0.5	2.4 ± 0.3	178.9 ± 9.2	10.7 ± 1.4	2.7 ± 0.5	3.2 ± 1.8
BkF	111.4 ± 25.8	8.6 ± 1.5	4.2 ± 0.1	ND	41.5 ± 0.7	2.8 ± 0.8	0.5 ± 0.2	ND	27.7 ± 3.1	ND	2.9 ^c	ND
BaP	379.0 ± 23.9	31.5 ± 2.8	6.1 ± 2.1	4.9 ^c	173.8 ± 4.9	9.5 ± 0.1	3.2 ± 0.3	ND	135.6 ± 6.2	8.2 ± 0.8	3.0 ^c	4.0 ^c
INP	191.1 ± 22.5	16.5 ± 0.8	5.6 ± 1.6	ND	86.1 ± 2.8	5.3 ± 0.8	1.8 ± 0.3	ND	49.0 ± 1.8	3.1 ± 0.7	4.6 ± 5.8	3.3 ± 2.0 ^b
DBA	38.8 ± 4.8	ND	9.1 ± 4.0	ND	16.6 ± 0.7	ND	ND	ND	11.3 ± 0.7	0.3 ^c	0.5 ^c	3.7 ^c
BghiP	129.1 ± 14.5	11.1 ± 0.4	6.4 ± 2.0	ND	73.8 ± 2.0	3.4 ± 0.6	ND	ND	57.7 ± 3.1	3.6 ± 0.4	4.3 ^c	4.2 ^c
ΣPAHs-EF (ng/g)	8,094.9 ± 563.7	560.3 ± 72.5	96.6 ± 15.3	32.4 ± 5.8	3,385.2 ± 311.2	115.0 ± 18.5	27.4 ± 3.1	11.2 ± 3.2	1,762.2 ± 136.6	55.9 ± 12.4	21.1 ± 9.3	15.5 ± 10.7
ΣPAHs/PM (μg/g)	206.3 ± 13.2	134.9 ± 9.1	76.4 ± 9.6	29.3 ± 3.9	167.9 ± 11.6	61.6 ± 7.5	21.2 ± 2.0	4.9 ± 1.4	168.5 ± 3.2	66.0 ± 7.5	30.3 ± 11.3	9.3 ± 5.8

^a Sample size = 3; Mean ± SD, ^b Only occurred twice, ^c Only occurred once, ^d ND not determined, possibly present at low level

ΣPAHs-EF is total PAH emission factors, ΣPAHs/PM is total PAH contents per unit mass of particulate matter

Fig. 3 The PAH emission rate and factors versus particle size from burning various incense sticks (Bar: mean \pm SD)



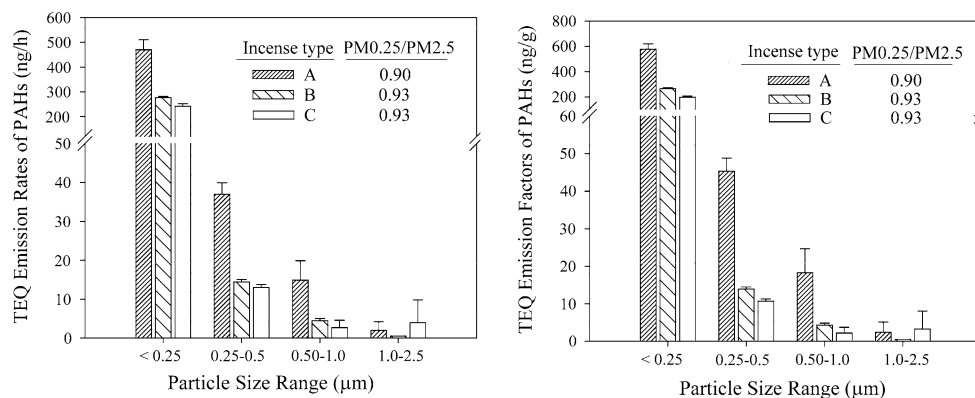
The PM_{0.25}/PM_{2.5} ratio of total PAH emission rates from smoldering A, B, and C incense was 0.92 ± 0.009 ($n = 3$), 0.96 ± 0.003 ($n = 3$) and 0.95 ± 0.012 ($n = 3$), respectively. The PM_{0.25}/PM_{2.5} ratio of total PAH emission factors for A, B, and C incense was the same as that of total PAH emission rates (Fig. 3). This analytical result indicates that total particle PAH emission were generally <0.25 μm in size (>90%). Furthermore, total PAH emission rates of PM_{0.25} for standard-smoke (incense A), low-smoke (incense B) and very-low-smoke (incense C) incense were $6,595.6 \pm 453.5$ ($n = 3$), $3,501.7 \pm 275.6$ ($n = 3$), and $2,139.7 \pm 160.9$ ($n = 3$) ng/h, respectively. The total PAH emission factors of PM_{0.25} for standard-smoke (incense A), low-smoke (incense B) and very-low-smoke (incense C) incense were $8,094.9 \pm 563.7$ ($n = 3$), $3,385.2 \pm 311.2$ ($n = 3$), and $1,762.2 \pm 136.6$ ($n = 3$) ng/g, respectively. Thus, the total PAH emission rate and factors of PM_{0.25} for smoldering incense were as follows: very-low-smoke incense < low-smoke incense < standard-smoke incense.

The Benzo[a]pyrene equivalent (BaP_{eq}) emission rates (or factors) of individual PAHs are the toxic equivalent factors (TEFs) of individual PAHs (Nisbet and LaGoy 1992) multiplied by analytical results for individual PAHs emission rates (or factors). The sum of all 16 BaP_{eq} emission rates (or factors) comprised the toxic equivalency

(TEQ) emission rates (or factors). The PM_{0.25}/PM_{2.5} ratio of the TEQ emission rates of PAHs from smoldering A, B and C incense were 0.90 ± 0.009 ($n = 3$), 0.93 ± 0.001 ($n = 3$), and 0.93 ± 0.015 ($n = 3$), respectively. The PM_{0.25}/PM_{2.5} ratio of TEQ emission factors for A, B and C incense were the same as that of the TEQ emission rates (Fig. 4). Hence, total toxicity of particle PAHs generated by smoldering incense was <0.25 μm in size. Furthermore, the TEQ emission rates and factors of PAHs for PM_{0.25} were 241.3–469.7 ng/h and 198.8–576.2 ng/g from the three smoldering incenses. This study also determined that BaP accounted for 65.2%–68.0% of the TEQ emission factor of PM_{2.5} for the three incenses. The TEQ emission rates and factors of PAHs for PM_{0.25} during incense smoldering were as follows: very-low-smoke incense < low-smoke incense < standard-smoke incense.

Finally, based on the atomic H/C ratios of tested incenses, we suggest that incenses with low atomic H/C ratios will reduce the amounts of particulates, total PAHs, and TEQ emissions for both PM_{2.5} and PM_{0.25}. Additionally, since particle PAH emissions during smoldering incense were mainly <0.25 μm, which can easily deposit deep into lungs with high deposition rates, such that the adverse effects of PAHs on human lungs may be enhanced. Hence, those who wish to continue burning incense to pay respect to ancestors or gods should use smoldering incense

Fig. 4 Total toxic equivalency emission rate and factor of PAHs versus particle size from burning various incense sticks (Bar: mean \pm SD)



with high carbon and low hydrogen content to minimize production of total PAH emission factors of both PM_{2.5} and PM_{0.25}. Analytical results can help solve indoor air-pollution problems caused by burning incense.

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