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Rates of Polycyclic Aromatic Hydrocarbon Emissions from Incense

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Incense burning has been associated with many cultures and religions in the world. Only since the 1960s, however, have researchers suspected that incense smoke might be a contributing factor to high incidence of certain cancers (Shanmugaratnam et al. 1967; MacLennan et al. 1977; Lowengart et al. 1987). These studies prompted the search for carcinogenic and other toxic constituents in incense smoke. Schoental and Gibbard (1967) first identified several carcinogenic polycyclic aromatic hydrocarbons (PAHs) in incense smoke condensates, including 3,4-benzopyrene [i.e., benzo(a)pyrene]. Sato et al. (1980) found that incense smoke condensates had mutagenic activity in *Salmonella typhimurium* strains TA98 and TA100. Rasmussen (1987) reported mutagenic activity in *Salmonella typhimurium* TA100 and TA104 for both particulate and vapor phase extracts from incense smoke. He also found a weak response in *Salmonella typhimurium* TA98 for particulate extracts, suggesting the presence of frameshift mutagens, such as polycyclic hydrocarbons, in the particulate extracts. A study conducted by Chen and Lee (1996) showed that incense smoke condensates were mutagenic and/or genotoxic in the Ames test, the SOS chromotest, and the SCE/CHO (Sister Chromatid Exchange/Chinese Hamster Ovary) assays and that the genotoxicity of certain incense smoke condensates in mammalian cells seemed to be higher than that of tobacco smoke condensates.

Estimation of human exposure to incense smoke and associated health risk requires field measurements of pollution levels and/or emissions data. A series of field studies have demonstrated that incense burning is associated with elevated air-borne PAHs concentrations in churches (Huynh et al. 1991), temples (Chao et al. 1997), and homes (Li and Ro 2000). Emissions data for incense burning are still rare, however. Endo et al. (2000) reported the emission rates (in ng/stick) of seven particle-bound PAHs from four brands of common incense and three of mosquito repellent incense. More recently, Lung and Hua (2003) reported the emission rates (in μg/min) and emission factors (in μg/g incense) for 13 particle-bound PAHs from two types of incense sticks. Little information is available in the literature about the emissions of the total (i.e., gas- and particle-phase PAHs combined) from incense burning. This paper attempts to fill this data gap by

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reporting the emission rates of total PAHs for 10 types of incense. Comparison between the emission rates of total and particle-bound PAHs demonstrates that the majority of PAHs masses emitted from incense burning exists in the gas-phase.

MATERIALS AND METHODS

Ten types of incense were collected from local stores. Their descriptions are provided in Table 1.

Table 1. Incense tested.

Incense ID	Incense Form	Origin	Average burn rate (g/min)	Average burn time (min)
A	Stick	North America	0.038	140
B	Stick	Asia	0.176	26
C	Stick	Asia	0.274	38
D [1]	Joss Stick	Asia	0.141	23
E [1]	Joss Stick	Asia	0.144	25
F	Cone	Asia	0.537 [2]	20
G	Smudge bundle	Central America	0.255 [2]	180
H	Rope incense	Asia	0.348	25
I	Powder	North America	0.174	39
J	Rocks	North America	0.164	36

[1] Incense D and E are of same brand with different scents.

[2] Burn rate varied considerably over time.

The emissions tests were conducted in a cylindrical, borosilicate glass chamber with an inside diameter of 28.6 cm and internal volume of about 46 L (Wasson et al. 2002). The chamber was designed to maximize the collection efficiency of particulate matter and to minimize losses of particulate to the surfaces of the chamber by using an inverted funnel as a hood over the combustion source. This hood can be adjusted for testing sources of different heights. During the burning of incense samples, a sampling pump with an in-line orifice plate drew unconditioned laboratory air through a high efficiency particulate air (HEPA) filter into the chamber through a hole in the bottom, located directly beneath the incense sample under test. The sample stream was then pulled through a glass funnel and directed through a collection system consisting of a Particle Measuring Systems Laser Aerosol Spectrometer (LAS-X) particle counter, a cyclone with cut-point of 2.5 μm aerodynamic diameter, quartz fiber filter, and XAD-2® sorbent module. The filter/XAD-2 assembly allowed capture of both gas- and particle-phase PAHs. For analyses of particle-phase PAHs, only the quartz filter was used. The glass funnel/hood apparatus was positioned within an inch (2.54 cm) of the tip of each incense sample. Chamber exhaust was directed through a laboratory fume hood to prevent laboratory contamination. Individual PAHs were either not detected or below the detection limit in field blanks and chamber blanks.

Prior to sampling, quartz fiber filters were cleaned by extracting for 16 hours with dichloromethane in a Soxlet extractor. Following extraction, filters were dried in a vacuum oven at 120 °C, cooled, then transferred to a desiccator in a climate controlled weigh room where they were equilibrated for at least 24 hours prior to weighing on a five place (10 µg) balance. XAD-2 sorbent was prepared by extracting in Type II water for 8 hours, methyl alcohol for 22 hours, dichloromethane for 22 hours, followed by a final 22 hour dichloromethane extraction. After drying, clean 10 cm long by 4 cm ID Teflon® sampling cartridges were packed sequentially with a hexane-cleaned polyurethane foam plug, clean glass wool, about 20 g of cleaned XAD-2, and a clean glass wool plug.

The sampling flow rate was 14.6 L/min for the filter/XAD-2 assembly and 15.8 L/min for filters only. The sampling duration ranged from 1 to 8 minutes for filter/XAD-2 and 5 to 20 min for filters only. Following each incense test, the chamber funnel, cyclone, and filter pack were triple-rinsed with dichloromethane. The rinse from each test was included with the corresponding filter and XAD-2 sorbent module for extraction and analyses. Prior to sample extraction, filters were equilibrated in a climate controlled weigh room for 24 hours before final weights were obtained on a five place (10 µg) balance. Following gravimetric analyses, filters were combined with the corresponding XAD-2 sorbent and dichloromethane rinse for Soxlet extraction in dichloromethane. After extraction by Method TO-13A (U.S. EPA 1999), samples were concentrated to 1.0 mL before analysis by gas chromatograph with a mass selective detector (GC/MS) in selective ion monitoring (SIM) mode. Sample analysis and quantification were performed according to Method TO-13A except that the SIM mode was employed to gain higher instrument sensitivity.

RESULTS AND DISCUSSION

The emission rates of the total PAHs (i.e., gas- and particle-phase combined) are presented in Table 2. These values can be easily converted to emission factors (in ng per g incense burned) by using the incense burn rates in Table 1. Two- and three-ring PAHs were most abundant in all the samples tested. Naphthalene alone accounts for 38 to 68% of the total PAH mass. Other most abundant PAHs include phenanthrene, acenaphthylene, fluoranthene, and anthracene. Most four- and five-ring compounds were also detectable. Coronene – a six-ring PAH – was not found in any of the smoke samples. Other rarely detected PAHs include fluorene, perylene, dibenz(a,h)anthracene. The emission rates of the total PAHs ranged from 458 to 13957 ng/min with a mean of 3266 ng/min.

Nine out of 10 incense samples produced detectible amounts of benzo(a)pyrene; five of them had an emission rate greater than 10 ng/min. Samples J and G, which

Table 2. Emission rates of total PAHs (gas- and particle-phase combined) in ng/min. [1]

Incense ID	A [2]	B	C	D [2]	E	F	G	H	I	J
Naphthalene	1203	489	462	276	258	**1777**	2470	1052	1880	6800
Acenaphthylene	233	94.7	73.2	65.9	59.1	224	417	111	228	604
Acenaphthene	*20.1*	*5.97*	*6.03*	*6.00*	*4.92*	27.9	269	*23.8*	*29.6*	163
Fluorene [3]	12.3	BDL	BDL	BDL	BDL	BDL	360	BDL	BDL	BDL
Benzo(b)fluoranthene [4]	*13.7*	*2.89*	*2.05*	*3.56*	*2.742*	*10.2*	49.0	*6.49*	29.4	235
Perylene [4]	BDL	BDL	BDL	*2.621*	BDL	BDL	BDL	BDL	*3.11*	BDL
Indeno(1,2,3-cd)pyrene [4]	*4.90*	*1.13*	BDL	*1.34*	*1.43*	BDL	23.2	*2.38*	7.08	179
Dibenz(a,h)anthracene	BDL	BDL	BDL	BDL	BDL	BDL	*15.5*	BDL	8.25	362
Benzo(ghi)perylene [4]	*4.83*	*1.01*	BDL	*1.19*	*0.96*	*5.00*	21.2	*1.72*	11.5	275
Coronene [4]	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL	BDL
Phenanthrene	222	74.4	55.1	85.5	67.2	703	1480	193	491	2590
Anthracene [5]	64.5	17.1	*9.07*	18.6	14.1	138	502	46.3	85.9	685
Fluoranthene	113	26.4	17.2	25.8	23.0	188	376	51.1	185	750
Pyrene	*44.2*	*9.82*	*7.10*	*9.18*	*9.42*	*49.4*	175	*22.0*	*26.8*	207
Benzo(a)anthracene	*25.9*	*7.00*	*4.67*	*8.27*	*6.87*	*29.2*	141	*19.5*	*18.6*	426
Chrysene	*15.0*	*4.11*	2.76	*5.42*	*4.13*	*34.3*	*86.7*	*19.4*	59.3	292
Benzo(k)fluoranthene	*3.37*	*1.02*	0.72	*1.22*	*1.13*	*3.04*	*15.8*	*2.06*	*5.22*	105
Benzo(e)pyrene	*7.44*	*1.80*	BDL	*1.64*	*1.73*	*8.07*	*29.4*	*3.14*	*23.4*	*36.2*
Benzo(a)pyrene [5]	*13.1*	*3.27*	BDL	*3.07*	*2.92*	*13.2*	*54.3*	*3.88*	*10.7*	247
All PAHs	1995	739	640	514	458	3210	6483	1559	3103	13957

[1] Values in bold font are above the calibration range; those in italics are above the estimated detection limit but below the calibration range; BDL = below detection limit. [2] Values in these columns are averages of duplicate tests. [3] Values for fluorene are probably under reported because of interference by an unknown substance. [4] Values for these compounds are probably under reported because of their low response factors. [5] Values for these compounds should be considered semi-quantitative because of low spike recoveries.

Table 3. Contents of particle-bound PAHs and their emission rates. [1]

Incense ID	A		B	
$PM_{2.5}$ emission rate (mg/min)	4.11		0.26	
	Content in $PM_{2.5}$ (ng/mg)	Emission rate (ng/min)	Content in $PM_{2.5}$ (ng/mg)	Emission rate (ng/min)
Naphthalene	2.58	10.6	3.04	0.78
Acenaphthylene	15.0	61.6	30.3	7.74
Acenaphthene	1.89	7.77	BDL	ND
Fluorene	BDL	ND	BDL	ND
Benzo(b)fluoranthene	2.91	12.0	38.0	9.71
Perylene	*0.28*	*1.16*	BDL	ND
Indeno(1,2,3-cd)pyrene	*1.03*	*4.23*	14.3	3.66
Dibenz(a,h)anthracene	BDL	ND	*4.24*	*1.08*
Benzo(ghi)perylene	*0.80*	*3.29*	8.04	2.05
Coronene	BDL	ND	BDL	ND
Phenanthrene	61.3	252	386	98.7
Anthracene	13.4	55.0	93.1	23.8
Fluoranthene	24.0	98.7	256	65.5
Pyrene	8.17	33.5	92.3	23.6
Benzo(a)anthracene	6.22	25.6	84.8	21.7
Chrysene	*3.24*	*13.3*	46.2	11.8
Benzo(k)fluoranthene	*1.65*	*6.78*	*11.4*	*2.90*
Benzo(e)pyrene	*1.52*	*6.25*	*15.9*	*4.06*
Benzo(a)pyrene	*2.88*	*11.8*	40.2	10.3
All particle-bound PAHs	147	603	1124	287

[1] Values in italics are above detection limit but below calibration range; BDL = below detection limit; ND = not detected.

had the largest emission rates for total PAHs, were also the largest emitters of benzo(a)pyrene.

There is no correlation between the incense burn rate and PAH emission rate. In terms of incense forms, rock (sample J) and smudge bundle (sample G) had the highest emission rates. Two joss stick incense (D and E) and one stick incense (C) had the lowest emission rates.

To estimate the partition of incense PAHs between gas- and particle-phases, incense A and B were also tested by sampling a larger volume of exhaust air (69 and 317 L, respectively) with quartz filters only. The contents of particle-bound PAHs and their emission rates are presented in Table 3. It should be pointed out that the analytical method used in this work has lower sensitivity to PAHs with higher molecular weights. Since the contents of particle-bound PAHs in these filter samples were fairly low, the results presented in Table 3 should be considered as semi-quantitative. Comparison of the data in Tables 2 and 3 shows that the percentage of particle-bound PAHs in the total PAHs was 30.2% for incense A and 38.9% for incense B. We can therefore conclude that the majority of PAH mass emitted from incense exists in the gas phase and that human exposure to PAHs in incense smoke can be underestimated if one considers only particle-bound PAHs. As expected, two- and three-ring PAHs exist almost exclusively in the gas-phase. Among all particle-bound PAHs detected, phenanthrene had the highest emission rate, followed by fluoranthene, anthracene, pyrene, and benzo(a)anthracene. In general, these results are in agreement with those reported by Endo et al. (2000) and Lung and Hua (2003). The emission rates for benzo(a)pyrene are similar for incense A and incense B. For these two samples of incense, emission rates are generally comparable for all of the particle-bound PAHs. Note that, although the PAH emission rates are comparable, PAHs are significantly more concentrated in incense B particles. In other words, although incense B produces less particulate matter with aerodynamic diameters less than 2.5 μm ($PM_{2.5}$), it emits similar amounts of particle-bound PAHs because its concentrations of PAHs are higher in the particle phase.

With the emission rates presented above, the indoor PAH concentrations can be predicted by using indoor air quality (IAQ) models. Figure 1 shows the simulation results with a simple (i.e., no surface adsorption and desorption) IAQ model for benzo(a)pyrene by using typical emission rates from this work. There is little doubt that incense burning is a significant source of indoor PAHs especially when burning multiple pieces of incense at the same time and/or burning incense multiple times per day.

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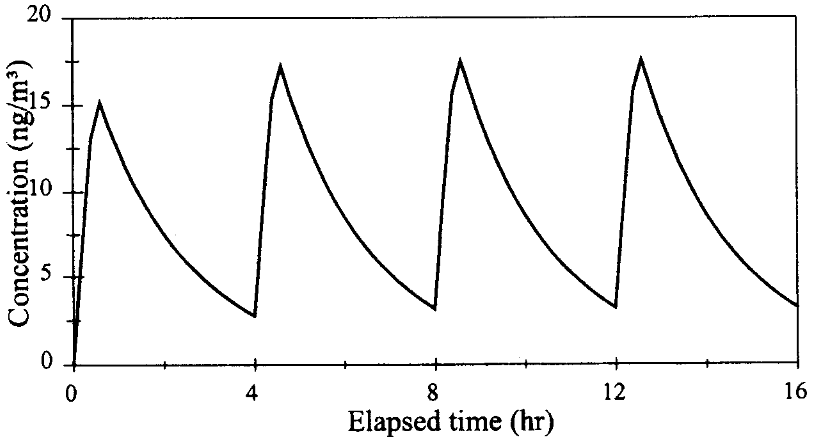

Figure 1. Predicted benzo(a)pyrene concentration when burning incense four times during the day and three pieces each time. Parameters used were: benzo(a)pyrene emission rate = 600 ng/hr for each piece of incense, room volume = 50 m³, air change rate = 0.5 hr^{-1}, and incense burning time = 0.5 hr.

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