Bull. Environ. Contam. Toxicol. (1997) 59:868-874

Bulletin of Environmental Contamination and Toxicology

Vapor Phase and Particulate Bound Polycyclic Aromatic Hydrocarbons in the Smoke of Mosquito Coils

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Received: 23 May 1997/Accepted: 15 September 1997

Mosquito coils, despite of having been categorized as a source of indoor air pollutant, are still widely used for fumigating mosquitos in Taiwan. Several epidemiological studies have shown that smoke from the mosquito coils has harmful effects on health. For instance, smoke from burning mosquito coils increases the prevalence rate of asthma and persistent wheezing among children between the ages of seven and twelve (Azizi and Henry, 1991). Furthermore, burning mosquito coils is found to be a significant risk factors for epidermoid carcinoma and adenocarcinoma in lung (Chen et al., 1990). Some animal studies also demonstrate the pernicious effects of burning mosquito coils. One study (Liu and Wong, 1987b; Liu and Sun, 1988; Liu et al., 1989) finds that smoke from burning mosquito coils causes morphological, biochemical and cellular changes in the respiratory system of rats, while another study (Moorthy and Murthy, 1994) shows that it raises the frequency of chromatid exchange and chromosomal aberration in rodents. The Ames Salmonella test with TA98 identifies the mutagenic effects of particulate aerosol in mosquito coil smoke (Lofroth et al., 1991).

The active ingredients in mosquito coils usually are pyrethrium or synthetic pyrethroid. In addition, it has other constituents such as organic fillers, binder, fungicides and other additives in the coils. The filler make up 99 % of a mosquito coil. Burning the mosquito coil releases about 70 % of its originally containing Cd, Cr and Pb, and about 99% of allethrin(Liu et al., 1987a). The GC/MS qualitatively identifies sixteen compounds including allethrin, benzene, toluene, xylene, o-tert-butylphenol, eicosyne and several polycyclic aromatic hydrocarbons (PAHs) in mosquito coil smoke.

This study also attempts to identify the concentrations of PAHs, both vapor phase and particulate bound, in the smoke from burning two prevalent brands of mosquito coils in Taiwan, because PAHs are related to the excess occurrence of lung cancers (Mumford et al., 1989; Verma et al., 1992; Seto et al., 1993), and the carcinogenic effects of some PAHs such as benzo(a)pyrene and benzo(b)fluoranthene are demonstrated in animal studies (Sittig, 1991).

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MATERIALS AND METHODS

Two brands of mosquito coils, Er-Yi and Bai-Gon were used in this study. A section of mosquito coil (about 2 cm in length and 12 gram in weight) was fixed in a holder inside a burning chamber, which was constructed by polypropylene. The dimension of the burning chamber was 6 x 48 x844 cm^3 (146.31 L).

Purified air was led into the burning chamber for 60 minutes to purge any possible contaminants, before the piece of mosquito coil was ignited. The purified air was conditioned to simulate the atmosphere with temperature of 20 + 2°C and relative humidity of 50±2%, through a flow temperature humidity control system (HCS-301, Miller Nelson Research, U.S.A.). At this stage, a blank sample was collected. Next, the inlet and outlet of the chamber were closed, and the piece of mosquito coil was ignited by a nickle-chrome wire controlled by a voltage-regulator. The samples were collected immediately after burning ceased.

A 2-pieces close-face opaque cassette holding a PTFE membrane filter (37mm, 2µm, SKC, U.S.A.), incorporated with a XAD-2 Sorbent Sample Tube (50/100 mg, 8x 110 mm, SKC, U.S.A.) was used to collect the particulate bound PAHs and the vapor phase PAHs, as described in the NIOSH method 5506 (NIOSH, 1994). Sampling was carried out at flow rate of 1 L/min for 20 minutes.

The PTFE membrane filter was put into an amber vial (4 mL, Alltech, U.S.A.) containing 2 mL acetonitrile, let stand for 1 hr prior to ultrasonic extraction for 30 minutes. Next, the extract was filtered through a syringe filter (PTFE, 13mm, 0.45µm, Gelman, U.S.A.) for removing the particulates.

The two sections of XAD-2 in a sorbent sample tube were separately put into two amber vials each containing 2 mL dichloromethane (GR grade, Merck, Germany). They were let stand at room temperature for 1 hr, then ultrasonically extracted for 1 hr. Dichloromethane was evaporated by gently blowing nitrogen into the vial and the residue dissolved in 1 mL acetonitrile.

A high performance liquid chromatograph (PU-980, Jasco, Japan) equipped with fluorescence detector (FP-920, Jasco, Japan) was employed to determine 14 PAHs. The column was EnviroSep-PP (125 X 3.2 mm, Phenomenex, U.S.A.). The mobile phase was acetonitrile in water using a programmed gradient elution at flow rate of 1 mL/min. The initial composition of acetonitrile was 40%. It changed linearly to 100% in 24 minutes, then was held constant for 4 minutes. Next, it decreased to 40% and was held for 7 minutes.

PAHs peaks were identified based on U.V. spectral analysis and the retention time of authentic PAHs relative to p-terphenyl (Fluka, Switzerland) which was added as an internal standard.

RESULTS AND DISCUSSION

Table 1 shows that vapor phase and particulate bound PAHs were present in both brands of coils. The three vapor phase PAHs were naphthalene, fluorene and anthracene, among which naphthalene made up about 89-97 % of the total vapor PAHs, depending on which brand of coil was tested. Similarly, naphthalene also made up the largest portion of particulate bound PAHs, among them naphthalene made up about 34-40%. Table 1 also shows that PAHs with molecular weight exceeding 202.26, such as fluoranthene and pyrene, were only present in particulate.

Table 2 shows the concentration of PAHs from burning one gram of mosquito coil in the unventilated chamber. It indicates that the concentration of particulate bound PAHs were higher compared to vapor PAHs, although they only made 44-46% of total determined PAHs. Since some latter groups of PAHs, except benzo (g, h, i) perylene, are suspected to be carcinogenic (Sittig, 1991), it is plausible that the particulate bound PAHs might have more negative effects on health compared to the vapor phase PAHs.

In order to apply the data to real life situations, it is essential to calculate the concentration of PAHs with a dynamic equation $C = G(1 - e^{-Qt/v}) / Q$, where C is concentration; G, generation rate; Q, ventilation rate; V, volume of room; and t, period from the start of burning. To calculate the concentration of either vapor PAHs or particulate bound PAHs after the burning ceased, a different equation $Ce = Co\, e^{-Qt/v}$ is used. Each mosquito coil weights about twelve grams, and needs about six hours for it to bum completely according to these data collected by burning twelve grams cf coil inside the sealed chamber. If a mosquito coil is burning in a 40 m^3 room with air exchange rate less than l/hr, then the estimated concentrations of PAHs from burning coils in the room at various ventilation rates are displayed in Table 3. For particulate bound PAHs, the concentration is presented separately for PAHs with molecular weight less or equal to 202.26 and for those greater than 202.26. As shown in Table 3, the concentration for all PAHs increases with the time of burning up to six hours and starts to decrease afterwards. In comparing both brands, the Bai-Gon mosquito coil seem to have a much higher concentration of PAHs compared to Er-Yi. This is particular true for particulate bound PAHs with molecular weight greater than 202.26. The Bai-Gon coil emits at least twice as much of these PAHs than the other brand. The highest concentration is about 0.5 $\mu g/m^3$ for total particulate bound PAHs with molecular weight exceeding 202.26. If the PAHs having carcinogenic effect are the non-threshold chemicals, then the particulate bound PAHs might play a partial role in the excess cancer risk arising from exposure to the smoke of mosquito coil.

This paper shows that mosquito coils do emit health threatening, and possibly carcinogenic PAHs. Furthermore, the amount of carcinogenic PAHs emitted varied greatly between brands. To reduce the risk of cancer, it is recommended to avoid using mosquito coils.

Table 1. Yield of PAHs from burning one gram of mosquito coil

Name of PAH	Yield of PAH, μg / g burning material			
	Er-Yi mosquito coil		Bai-Gon mosquito coil	
	Vapor PAH	Particulate-bound PAH	Vapor PAH	Particulate-bound PAH
Naphthalene	20.98±1.25*	7.35±3.84	30.45±20.55	9.23±5.99
Acenaphthene	—	—	—	—
Fluorene	0.62±0.19**	5.24±2.07	3.93±5.04**	4.48±1.49
Phenanthrene	—	2.63±1.11	—	4.87±1.33
Anthracene	0.07±0.01	0.44±0.15	0.10±0.06	0.94±0.32
Fluoranthene	—	0.74±0.31	—	2.13±0.20
Pyrene	—	0.74±0.02**	—	2.31±0.40
Benz(a)anthracene	—	—	—	—
Chrysene	—	0.19±0.06	—	1.20±0.23
Benzo(b)fluoranthene	—	0.43±0.20	—	0.43±0.06
Benzo(k)fluoranthene	—	0.06±0.01	—	0.15±0.01
Benzo(a)pyrene	—	0.18±0.02	—	0.48±0.03
Dibenz(a,h)anthrene	—	—	—	—
Benzo(g,h,i)perylene	—	0.29±0.11	—	0.96±0.64

* Mean ± Standard deviation; Sample size = 3

** The data only occurred twice in triplicate samples.

— Designates non-detectable

Table 2. Concentration of PAHs from burning one gram of mosquito coil in the unventilated chamber

Name of PAH	Concentration of PAH			
	Er-Yi mosquito coil		Bai-Gon mosquito coil	
	Vapor PAH (ppb / g_b)	Particulate-bound PAH (μg / $g_p g_b$ ***)	Vapor PAH (ppb / g_b)	Particulate-bound PAH (μg / $g_p g_b$)
Naphthalene	26.69±1.59 *	399.57±183.72	42.72±26.24	915.28±665.10
Acenaphthene	—	—	—	—
Fluorene	0.61±0.19**	299.76±143.76	3.87±4.96 **	418.80±148.81
Phenanthrene	—	141.43±27.81	—	464.60±176.92
Anthracene	0.06±0.01	23.82±4.29	0.09±0.06	90.71±41.23
Fluoranthene	—	39.64±8.16	—	201.56±44.25
Pyrene	—	47.08±3.57 **	—	219.73±60.24
Benz(a)anthracene	—	—	—	—
Chrysene	—	10.60±4.59	—	111.96±24.62
Benzo(b)fluoranthene	—	23.95±12.42	—	40.57±8.10
Benzo(k)fluoranthene	—	3.54±1.14	—	14.36±2.26
Benzo(a)pyrene	—	10.35±1.29	—	45.49±8.80
Dibenz(a,h)anthrene	—	—	—	—
Benzo(g,h,i)perylene	—	16.77±7.71	—	94.51±72.43

* Mean±Standard deviation; Sample size=3

** The data only occurred twice in triplicate sample.

*** g_p : gram of particulate; g_b : gram of burning material.

— designates non-detectable

Table 3. The estimated concentrations of PAHs from burning a mosquito coil in a 40 m^3 room at various ventilation rates

Brand of moisquito coil	Time sequence of burning, hr	Estimated concentration, μg / m^3					
		Vapor phase		Particulate phase			
		PAH with MW=202.26		PAH with MW≦202.26		PAH with MW>202.26	
		Q=10	Q=20	Q=10	Q=20	Q=10	Q=20
Er-Yi	0	0.00	0.00	0.00	0.00	0.00	0.00
	1	0.88	0.79	0.69	0.62	0.05	0.04
	2	1.57	1.26	1.24	0.99	0.09	0.07
	3	2.10	1.55	1.66	1.22	0.12	0.09
	4	2.52	1.73	2.00	1.36	0.14	0.09
	5	2.85	1.83	2.24	1.44	0.16	0.10
	6	3.10	1.90	2.44	1.49	0.17	0.10
	7	2.42	1.15	1.90	0.91	0.13	0.06
	8	1.89	0.70	1.48	0.55	0.10	0.04
Bai-Gon	0	0.00	0.00	0.00	0.00	0.00	0.00
	1	1.37	1.22	0.95	0.85	0.13	0.11
	2	2.44	1.96	1.70	1.36	0.23	0.18
	3	3.27	2.41	2.28	1.68	0.31	0.23
	4	3.92	2.68	2.73	1.86	0.37	0.25
	5	4.43	2.85	3.08	1.98	0.41	0.27
	6	4.82	2.95	3.35	2.05	0.45	0.28
	7	3.76	1.79	2.61	1.24	0.35	0.17
	8	2.92	1.08	2.03	0.75	0.27	0.10

The MW designates molecular weight, and Q indicates ventilation rate in m^3 / hr .

Acknowledgments. This study was partially supported by funding (NSC 85-2621-B-002-0211-Z) from the National Science Council of the Republic of China.

REFERENCES

Azizi BHO, Henry RL (1991) The effects of indoor environmental factors on respiratory illness in primary school children in Kuala Lumpur. Int J Epidemiol 20:144-150

Chen CJ, Wu HY, Chuang YC, Chang AS, Luh KT, Chao HH, Chen KY, Chen SG, Lai GM, Huang HH, Lee HH (1990) Epidemiologic characteristics and multiple risk factors of lung cancer in Taiwan. Anticancer Res 10:971-976

Liu WK, Ng TB, Wong CC (1989) Biochemical and cellular changes in bronchoalveolar lavaged samples from rats after inhalation of mosquito coil smoke. Toxicol Lett 45:121-132

Liu WK, Sun SE (1988) Ultrastructural changes of tracheal epithelium and alveolar macrophages of rats exposed to mosquito coil smoke. Toxicol Lett 41:145-157

Liu WK, Wong MH, Mui YL (1987a) Toxic effects of mosquito coil (a mosquito repellent) smoke on rats I: Properties of the mosquito coil and its smoke. Toxicol Lett 39:223-230

Liu WK, Wong MH (1987b) Toxic effects of mosquito coil (a mosquito repellent) smoke on rats II: Morphological changes of the respiratory system. Toxicol Lett 39:231-239

Lofroth G, Stensman C, Brandhorst-Satzkorn M (1991) Indoor sources of mutagenic aerosol particulate matter: smoking, cooking and incense burning. Mutat Res 261:21-28

Moorthy MV, Murthy PB (1994) Analysis of sister chromatid exchange, micronucleus and chromosomal aberration frequencies in rodents exposed to mosquito coil smoke by inhalation route. Toxicol Lett 70:357-362

Mumford RS, Chapman RS, Harris DB, He XZ, Cao YL, Xian YL, Li XM (1989) Indoor air exposure to coal and wood combustion emission associated with a high lung cancer rate in Xuan Wei, China Environ Int 15:315-320

National Institute for Occupational Safety and Health (1994) Polycyclic aromatic hydrocarbons by HPLC (method 5506). In: Eller PM (ed) NIOSH Manual of Analytical Methods. Division of Physical Sciences and Engineering , Cincinnati, Ohio

Seto H, Ohkubo T, Kanoh T, Koike M, Nakamura K, Kawahara Y (1993) Determination of polycyclic aromatic hydrocarbons in the lung. Arch Environ Contam Toxicol 24:498-503

Sittig M (1991) Handbook of toxic and hazardous chemicals and carcinogens. Noyes Publications, Park Ridge, NJ

Verma DK, Julian JA, Roberts RS, Muir DC, Jadon N, Shaw DS (1992) Polycyclic aromatic hydrocarbons (PAHs): a possible cause of lung cancer mortality among nickel/copper smelter and refinery workers. Am Ind Hyg Assoc J 53:317-324