

Mosquito Coil Emissions and Health Implications

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Burning mosquito coils indoors generates smoke that can control mosquitoes effectively. This practice is currently used in numerous households in Asia, Africa, and South America. However, the smoke may contain pollutants of health concern. We conducted the present study to characterize the emissions from four common brands of mosquito coils from China and two common brands from Malaysia. We used mass balance equations to determine emission rates of fine particles (particulate matter < 2.5 μm in diameter; $\text{PM}_{2.5}$), polycyclic aromatic hydrocarbons (PAHs), aldehydes, and ketones. Having applied these measured emission rates to predict indoor concentrations under realistic room conditions, we found that pollutant concentrations resulting from burning mosquito coils could substantially exceed health-based air quality standards or guidelines. Under the same combustion conditions, the tested Malaysian mosquito coils generated more measured pollutants than did the tested Chinese mosquito coils. We also identified a large suite of volatile organic compounds, including carcinogens and suspected carcinogens, in the coil smoke. In a set of experiments conducted in a room, we examined the size distribution of particulate matter contained in the coil smoke and found that the particles were ultrafine and fine. The findings from the present study suggest that exposure to the smoke of mosquito coils similar to the tested ones can pose significant acute and chronic health risks. For example, burning one mosquito coil would release the same amount of $\text{PM}_{2.5}$ mass as burning 75–137 cigarettes. The emission of formaldehyde from burning one coil can be as high as that released from burning 51 cigarettes. **Key words:** aldehydes, mosquito coil, PAHs, particulate matter, smoke. *Environ Health Perspect* 111:1454–1460 (2003). doi:10.1289/ehp.6286 available via <http://dx.doi.org/> [Online 30 April 2003]

Mosquito coil is widely known as an efficient mosquito repellent. The major active ingredients of the mosquito coil are pyrethrins, accounting for about 0.3–0.4% of coil mass (Lukwa and Chandiwana 1998). When a mosquito coil is burned, the insecticides evaporate with the smoke, which prevents the mosquito from entering the room. Pyrethrins are of low chronic toxicity to humans and low reproductive toxicity in animals, although headache, nausea, and dizziness were observed in male sprayers exposed to 0.01–1.98 $\mu\text{g}/\text{m}^3$ pyrethrins for 0.5–5 hr (Zhang et al. 1991). No carcinogenic and mutagenic effects have been found for these insecticides (Ecobichon 1995). The lowest lethal oral dose of pyrethrum is 750 mg/kg for children and 1,000 mg/kg for adults [Occupational Health Services (OHS) 1987]. The remaining components of mosquito coil are organic fillers, binders, dyes, and other additives capable of smoldering well. The combustion of the remaining materials generates large amounts of submicrometer particles and gaseous pollutants. These submicrometer particles can reach the lower respiratory tract and may be coated with a wide range of organic compounds, some of which are carcinogens or suspected carcinogens, such as polycyclic aromatic hydrocarbons (PAHs) generated through incomplete combustion of biomass (mosquito coil base materials). Researchers have found that the gas phase of

mosquito coil smoke contains some carbonyl compounds with properties that can produce strong irritating effects on the upper respiratory tract—for example, formaldehyde and acetaldehyde (Chang and Lin 1998). Because coil consumers usually use mosquito coils for at least several months every year, cumulative effects from long-term exposure to the coil smoke may also be a concern.

Epidemiologic studies have shown that long-term exposure to mosquito coil smoke can induce asthma and persistent wheeze in children (Azizi and Henry 1991; Fagbule and Ekanem 1994; Koo and Ho 1994). Toxicologic effects of mosquito coil smoke on rats include focal deciliation of the tracheal epithelium, metaplasia of epithelial cells, and morphologic alteration of the alveolar macrophages (Liu and Sun 1988; Liu and Wong 1987). For example, when a group of 30 female albino rats were exposed to mosquito-coil smoke in a 22.5- m^3 chamber for 8 hr per day, 6 days per week, for 6 months, these rats lost typical ruffled membranes of their alveolar macrophages. In addition, the levels of total protein and lecithin and the activities of lactate dehydrogenase, acid phosphatase, and β -glucuronidase in the lung-lavage fluid of the rats were significantly higher than those in a control group that was exposed to air for the same exposure duration (Liu et al. 1989).

Despite the fact that mosquito coil smoke may have many potential adverse health effects, large populations in developing countries still use mosquito coils in their daily lives. In previous studies of various aspects of mosquito coil smoke, emissions of irritating and carcinogenic compounds and other pollutants have not been quantified, which precludes the use of emission rate data to predict pollutant concentrations in households and to quantify health risks. Data are also lacking for comparing emissions from different types of mosquito coils. To make informative recommendations to consumers as to which types of mosquito coil have lower emissions of health-damaging pollutants, it is necessary to perform tests of coil emissions in a systemic manner. In the present study, we selected for comparison six brands of mosquito coils popularly used in China and Malaysia. We measured emission rates of a variety of pollutants of health concern, present in both the particulate phase and the gas phase of the coil smoke. We also determined particle size distribution of the coil smoke and identified a suite of volatile organic compounds (VOCs), including human carcinogens and suspected carcinogens. Finally, using the emission rates determined in the present study and typical room parameters, we predicted room concentrations and compared these with reference risk values.

Materials and Methods

Experimental apparatus. The experimental apparatus is shown in Figure 1, the core of which is a polyvinyl chloride chamber with a volume of 0.15 m^3 . The incoming air, after passing through a series of filters impregnated with activated carbon (charcoal filters), was introduced into the chamber through the inlet valve at a constant flow rate of 5.59 L/min.

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The chamber was under a slightly positive pressure to prevent the infiltration of air from outside by controlling the flow rate of exiting air via the sampling ports and the outlet valve. During each experimental run, a lit mosquito coil on the metal stand provided within the coil packet was placed inside the chamber. Air samples were drawn out of the chamber at 10 mL/min through Teflon tubing and diluted with zero-grade air (Air Products & Chemicals, Inc., Edison, NJ, USA) 80× in a flask to match the measurement range of an eight-channel optical particle counter. Additional sampling lines on the top of the chamber were used to collect samples for measuring the mass of particulate matter of 2.5 μm in diameter ($\text{PM}_{2.5}$) and for identifying gas-phase organic compounds.

Tested mosquito coils included two brands purchased from Malaysia (coils M1 and M2) and four brands from China (coils C1–C4). Based on the information on the product labels, all six tested brands had similar contents of active ingredients (pyrethrins). However, it is our understanding that the base material used in making the Chinese coils was mainly sawdust, whereas materials used in making the Malaysian coils were coconut husks/shells.

Measurement of particle number and mass concentration. We used the eight-channel optical particle counter (LASAIR model 1002 unit; Particle Measuring Systems, Inc., Boulder, CO, USA) to measure number concentrations of particulate matter from 0.1 to 10 μm in diameter with eight size ranges: 0.1–0.2 μm , 0.2–0.3 μm , 0.3–0.4 μm , 0.4–0.5 μm , 0.5–0.7 μm , 0.7–1.0 μm , 1.0–2.0 μm , and > 2 μm . This particle counter was operated for 3 hr and 10 min

during each coil test, which included a 10-min background monitoring before initiating the coil smoke, a 2-hr coil burning period, and a 1-hr postburning period. A similar set of experiments were conducted in a room with a volume of 32 m^3 (4 × 3 × 2.5 m) and an air exchange rate of about 1/hr (with window half open) to examine whether there were differences in particle size distributions between the room measurements and the chamber measurements. An ultrafine particle counter (model 3007; TSI Inc., St. Paul, MN, USA) was used in addition to the eight-channel optical particle counter to monitor particles with diameters as small as 0.01 μm in the room experiments.

A personal $\text{PM}_{2.5}$ sampling head (Personal Environmental Monitor model 200; MSP Co., Minneapolis, MN, USA) with a 25-mm Teflon filter (Pall Co., Ann Arbor, MI, USA) was located in the middle of the chamber, as shown in Figure 1, to collect $\text{PM}_{2.5}$ mass. The samples were collected using an SKC pump (model 224-PCXR4; SKC Inc., Eighty Four, PA, USA) providing a sampling flow rate of 0.8 L/min. Each of these $\text{PM}_{2.5}$ samples was collected for 10 min after the coil had been steadily burned for 1.5 hr. The particle mass on the filters was determined gravimetrically.

Measurement of PAHs. The $\text{PM}_{2.5}$ filters, after mass concentrations had been determined, were extracted with 150 mL dichloromethane individually for 16 hr at 75°C using a Soxhlet apparatus (Farant and Garipey 1998; Sanderson and Farant 2000). The extracts were evaporated to near dryness and redissolved in 1 mL acetonitrile (ACN). The extracts were analyzed using a high-performance liquid chromatographic (HPLC) system (Waters 600E; Waters Corp., Milford, MA, USA)

with a fluorescence detector (Waters 470). The HPLC column used was a Supelcosil LC-PAH column (4.6 × 250 mm) (Supelco, Inc., Bellefonte, PA, USA) under controlled temperature at 30°C. The mobile phase used was as follows: solution A = 50% ACN and 50% water; solution B = 100% ACN. The gradient program was 100% A for 20 min, linear gradient from 100% A to 100% B in 20 min, 100% B for 15 min, then from 100% B back to 100% A in 10 min, and held at 100% A for 10 min. The mobile phase flow rate was 1 mL/min. The injection volume was 20 μL . The fluorescence detector program was started at an excitation wavelength of 270 nm and an emission wavelength of 350 nm; at 34 min, the wavelength was changed to excitation at 250 nm and emission at 400 nm and held for 13.5 min; then it was changed to excitation at 280 nm and emission at 425 nm for another 22.5 min. PAH concentrations were determined through calibration curves prepared using certified standard solutions of PAHs purchased commercially (Supelco, Inc.).

Measurement of carbonyl compounds. U.S. Environmental Protection Agency (U.S. EPA) method TO-11A was used for collecting and analyzing carbonyl compounds (U.S. EPA 1999a). Carbonyl compounds (aldehydes and ketones) in the coil smoke were collected using 2,4-dinitrophenyl hydrazine (DNPH)-coated C_{18} cartridges at a flow rate of 0.07 L/min for 10 min for each test. The samples were slowly eluted with 4 mL ACN immediately after the sampling, and the extracts were analyzed using an HPLC system with a reverse-phase Nova-Pak C_{18} column (3.9 × 150 mm; Waters Corp.). The mobile phase gradient program used was as follows: 100% of solvent A (water/ACN/tetrahydrofuran 60/30/10), hold for 5 min, then program to 100% solvent B (ACN/water 60/40) in 28 min, and hold at 100% B for 10 min, and then program back to 100% A in 5 min. The flow rate of the mobile phase was kept constant at 1 mL/min. The sample injection volume was 20 μL . The ultraviolet detector was set at 365 nm. Carbonyl compound concentrations were determined through calibration curves prepared using certified standard solutions of DNPH-carbonyl derivatives purchased commercially (Supelco, Inc., and Accustandards Inc., New Haven, CT, USA).

Identification of VOCs. VOC samples were collected with stainless steel traps (0.5 cm inner diameter × 8.8 cm; Perkin Elmer Inc., Shelton, CT, USA) packed with 0.25 g Tenax TA (Supelco, Inc.) at a flow rate of 0.1 L/min for 2 min during each test (Xu and Weisel 2003). A glass fiber filter was placed in front of the Tenax trap to remove particles. After sample collection, the samples were desorbed from the trap at 250°C using an automated thermal desorption system

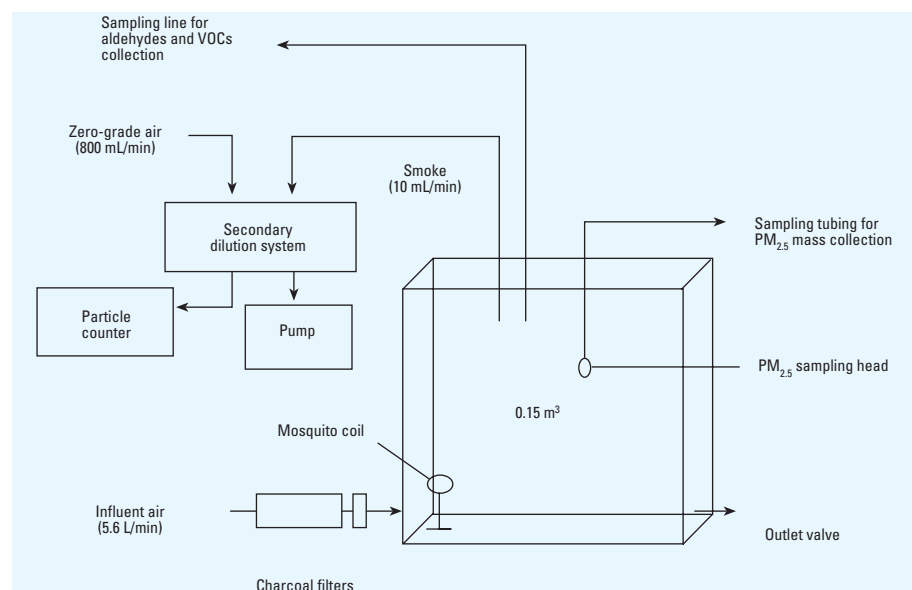


Figure 1. Apparatus and setup for the chamber experiments.

(ATD-400; Perkin-Elmer Inc., Wellesley, MA, USA) and transferred to a capillary gas chromatograph (GC; DB-5 capillary column, 60 m × 0.25 mm inner diameter, 1 μm film thickness, HP 5890; Hewlett-Packard, Wilmington, DE, USA) equipped with a mass spectrometer (MS; HP 5971A; Hewlett-Packard) system for compound identifications. The GC temperature program was set at 35°C for 10 min, gradually increased to 250°C in 20 min, then held at 250°C for another 10 min.

Measurement of air exchange rate. Ventilation rate, expressed as air exchange rate, during each experiment, equals the volumetric flow rate of the incoming air divided by the chamber volume. In our experiment, the volumetric flow rate of the incoming air was controlled using a needle valve of a flow meter. The theoretical air exchange rate in this small chamber should be 2.24/hr. However, this value may not be exactly reached because of the variation of the incoming air. The air exchange rates were thus also determined experimentally by spiking sulfur hexafluoride (SF₆) gas into the chamber after the coil had been extinguished for 1 hr to avoid the interference of high-level particles in the smoke. A real-time SF₆ monitor (type 1302; INNOVA Air Tech Instruments, Ballerup, Denmark) was used to measure SF₆ concentrations in the chamber every 3 min for 30 min. The regression slope of the plot of logarithm of concentration against time was the air exchange rate. The experimentally measured air exchange rates were later used to calculate the emission rates of gas-phase compounds. The results show that the experimental determined air

exchange values were about 10% lower than the theoretical value.

Determination of emission rates. A single-compartment mass balance model was used to describe the whole combustion process and to determine the emission rate. Basic assumptions for this model were as follows: *a*) background concentration is zero; *b*) pollutant concentrations are homogeneous within the chamber; and *c*) emission rate and decay rate of the pollutants remain constant throughout the entire period of concern. The relationship between the pollutant concentration C (mg/m³) and the emission rate P (mg/hr) can be expressed as

$$C = \frac{P}{Vk} (1 - e^{-kt}) \quad (\text{when } 0 \leq t \leq T); \quad [1]$$

$$C = C_{\max} (e^{-k(t-T)}) \quad (\text{when } t \geq T), \quad [2]$$

where k is the total removal rate of pollutant (hr⁻¹), t is the time (hr)—the coil burning was started at $t = 0$ and extinguished at $t = T$; V is the volume of the chamber (m³), and C_{\max} is the maximum pollutant concentration at the time (T) when coil was extinguished.

When the concentration of the pollutant stabilized in the chamber, that is, had reached a steady state, Equation 1 can be simplified as

$$C = \frac{P}{Vk}. \quad [3]$$

Therefore, the emission rate P could be easily obtained using steady-state concentration (C),

pollutant removal rate (k), and the volume of the chamber (V).

Assuming the removal of the gas-phase compounds in the chamber is caused only by ventilation, the removal rate of gas-phase compounds is equal to the air exchange rate in the chamber. However, particle removal is controlled by a number of processes, such as deposition, coagulation, diffusion, and so forth. Equation 2 was used to determine particle total removal rate k . A real-time particle mass concentration in the chamber during the postburning period is needed for the calculation. We converted the real-time particle number concentration (number per liter) collected by the eight-channel optical particle counter after the coil was extinguished to mass concentration (milligrams per cubic meter) by assuming all particles have a spherical shape and unit density. For calculation convenience, the geometric mean of the two end points of each size range was used as the diameter for all particles in this size range. The linear regression slope of the plot of $\ln(C)$ against t would be the total removal rate of particulate matter in the chamber.

Results and Discussion

Particle behaviors and size distribution.

Number concentration of fine particles at different size ranges as a function of time in one experiment is illustrated in Figure 2. We did not include particles larger than 1 μm in Figure 2 because the concentration of large particles remained the same as the background level. This is typical for concentration profiles of particles in a well-mixed chamber. After the coil was lit, the concentrations of particles with

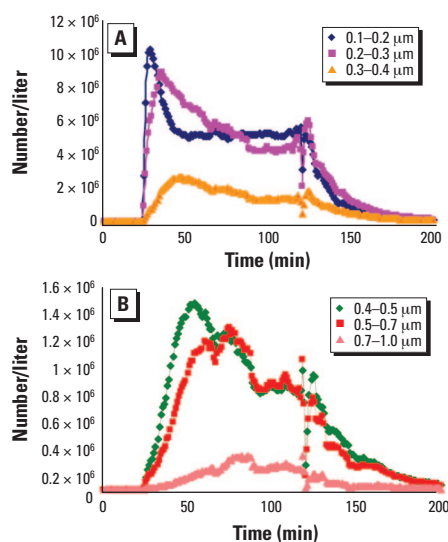


Figure 2. Fine particle number concentration profile in the chamber as a function of time for C1 during the whole experimental process. (A) Particles with size range of 0.1–0.4 μm. (B) Particles with size range of 0.4–1 μm. Particles larger than 1 μm were not observed.

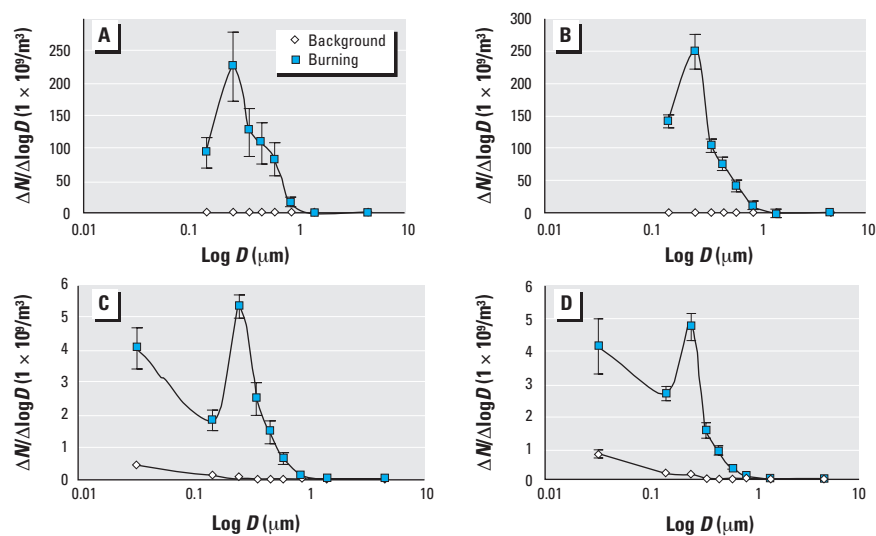


Figure 3. Normalized particle size distributions in the small chamber (A, B) and the room (C, D) for M2 (A, C) and C2 (B, D). The plots reflect averaged concentrations with standard deviations of the last half-hour of emissions before the coil was extinguished. Only particles smaller than 0.1 μm were measured in the room. ΔN represents the concentrations of particles within a specified size interval, and $\Delta \log D$ is the difference in the logarithms of the largest and smallest particle sizes of that interval. The logarithm of the midpoint of a ΔD ($\log D$) is plotted for convenience.

diameters < 0.3 μm increased quickly to a peak level in only a few minutes, then decreased in the first half hour and stabilized at a level of approximately 50% of the peak level until the coil was extinguished. The concentrations of particles > 0.3 μm in diameter increased more gradually and stabilized in the first hour of burning. After the coil burned out, the particle number concentration decreased exponentially. Because burning time (2 hr) was long enough to allow the concentration of pollutants to reach a steady state, the steady-state burning process can be described using the single-compartment model.

The complex processes of formation and removal of particles in the chamber are governed by a number of mechanisms, including diffusion, gravitational deposition, convection, impaction, and coagulation. Diffusion is the primary mechanism for small particles, whereas gravitational force is usually the dominant process controlling the removal of large particles. Convection and impaction cause deposition of particles in all directions. Coagulation is a process whereby small particles collide with one another through diffusion to form larger particles. The rate of simple monodisperse coagulation can be represented as $dN/dt = -KN^2$ (Hinds 1982), where N is particle number concentration and K is the coagulation coefficient. Based on this equation, the coagulation rate is proportional to the square of the number concentration of particles. This may explain in part the concentration changes observed during the first half hour: The concentration of particles < 0.3 μm increased dramatically in the first few minutes of coil burning; then the coagulation process became dominant and the particle concentration decreased to a stable level.

The particle size distributions of the combustion of M2 and C2 in both the small chamber and the room study are shown in Figure 3. The plots reflected an averaged concentration of the last half-hour's emission before the coil was extinguished. The plots presented the normalized distribution with $\Delta N/\Delta \log D$ versus $\log D$, where ΔN is the concentration of particles within a specified size interval and $\Delta \log D$ is the difference in the logarithms of the largest and smallest particle sizes of that interval (Reist 1984). The logarithm of the midpoint of a ΔD ($\log D$) was plotted for convenience. Particles > 2.0 μm were not observed.

In the small chamber study, the highest normalized concentration fell in the size range of 0.2–0.3 μm . In the room study, we were able to monitor ultrafine particles in addition to monitoring fine particles in the eight size ranges. A bimodal size distribution curve was observed with peaks at both 0.01–0.1 μm and 0.2–0.3 μm , which means the ultrafine and fine particles dominate the counts of particles emitted by coil combustion. No particles > 1 μm in diameter were generated during coil burning. The room was measured to examine whether particle size distributions can be “artificially” affected by the nature of the small chamber (high surface-to-volume ratio). No measurable effects were found on particles > 0.1 μm . Ultrafine particles were not analyzed in the chamber experiments.

Emission rates. Table 1 summarizes the measured coil weight, burn rate, air exchange rate, $\text{PM}_{2.5}$ concentration in the chamber, $\text{PM}_{2.5}$ removal rate, and $\text{PM}_{2.5}$ emission rate. The data were based on an average of five experiments for each brand of coil tested. As shown in Table 1, all experiments were conducted

under a constant air exchange rate of $\sim 2/\text{hr}$, close to the theoretical value, 2.24/hr. The chamber concentrations of $\text{PM}_{2.5}$ mass generated from the combustion of the two brands of Malaysian coils, 363 ± 12 and 365 ± 19 mg/m^3 , were much higher than from any Chinese brands tested, the highest concentration among which was 246 ± 23 mg/m^3 . The emission rates varied largely across different brands of coils, from 51.1 ± 7 to 117 ± 14 mg/hr . The particulate matter emission rates of the Malaysian mosquito coils were markedly higher than those of the Chinese coils. However, burn rates of different coils were similar, ranging from 1.5 to 2.0 g/hr . Different contents of organic fillers (base materials) used for smoldering could be the main reason of the difference in emission rates. The difference in base materials was reflected by the fact that the tested Chinese coils had a longer burning time, ranging from 9 to 11 hr, compared with the Malaysian ones, which can burn for only 7 hr.

High concentrations of PAHs were observed in the particulate phase of mosquito coil smoke (Table 2). The table shows that the emission rates for low-molecular-weight PAHs were higher than those for heavier ones. However, some heavy PAHs are suspected human carcinogens. Benz[*a*]anthracene, benzo[*a*]pyrene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, dibenz[*a,h*]anthracene, and indeno[1,2,3-*cd*]pyrene are classified by the U.S. EPA as probable human carcinogens (U.S. EPA 1994). Even at trace levels, long-term exposure to these compounds could increase cancer risk. Benzo[*a*]pyrene, benzo[*b*]fluoranthene, and benzo[*k*]fluoranthene were detected in the particulate phase of mosquito coil smoke.

Table 1. Summary of coil weight, burn rate, air exchange rate in the chamber, $\text{PM}_{2.5}$ removal rates, concentrations, and emission rates for different mosquito coils.

Brand ^a	W^b (g)	B^c (g/hr)	A^d (hr^{-1})	K^e (hr^{-1})	$C(\text{PM}_{2.5})^f$ (mg/m^3)	$P(\text{PM}_{2.5})^g$ (mg/hr)
M1	12.7 \pm 0.5	1.67 \pm 0.04	2.04 \pm 0.05	2.14 \pm 0.20	363 \pm 12	117 \pm 14
M2	11.1 \pm 0.8	1.70 \pm 0.05	1.99 \pm 0.09	2.09 \pm 0.14	365 \pm 19	115 \pm 12
C1	20.4 \pm 1.2	1.81 \pm 0.05	2.02 \pm 0.06	2.22 \pm 0.18	189 \pm 9	63 \pm 3
C2	13.3 \pm 0.6	1.48 \pm 0.03	1.97 \pm 0.11	2.33 \pm 0.37	246 \pm 23	85 \pm 9
C3	15.6 \pm 1.0	1.56 \pm 0.05	1.98 \pm 0.10	2.10 \pm 0.20	162 \pm 15	51 \pm 7
C4	18.5 \pm 0.9	2.05 \pm 0.05	2.09 \pm 0.11	2.18 \pm 0.16	215 \pm 24	71 \pm 13

Data expressed as mean \pm SD.

^aNumber of experiments ($n = 5$ for all). ^bCoil weight. ^cBurn rate. ^dAir exchange rate. ^eParticle removal rate. ^f $\text{PM}_{2.5}$ concentration in the chamber. ^gEmission rate of $\text{PM}_{2.5}$.

Table 2. Emission rates of particulate phase PAHs in coil smoke ($\mu\text{g}/\text{hr}$).

Compound ^a	M1	M2	C1	C2	C3	C4
Acenaphthene	17.4 \pm 8.6	ND	ND	ND	ND	ND
Fluorene	20.6 \pm 0.4	16.7 \pm 0.2	0.903 \pm 0.102	15.6 \pm 0.6	1.46 \pm 0.08	2.49 \pm 0.04
Phenanthrene	29.0 \pm 0.5	20.3 \pm 0.7	1.20 \pm 0.04	20.6 \pm 0.8	3.95 \pm 0.35	5.99 \pm 0.07
Anthracene	0.988 \pm 0.13	1.04 \pm 0.04	0.350 \pm 0.079	0.146 \pm 0.052	0.416 \pm 0.078	1.21 \pm 0.13
Fluoranthene	94.5 \pm 1.1	18.0 \pm 0.4	ND	4.26 \pm 0.58	1.74 \pm 0.15	2.38 \pm 0.10
Pyrene	9.37 \pm 0.21	14.2 \pm 1.4	0.917 \pm 0.093	1.90 \pm 0.05	0.959 \pm 0.031	3.78 \pm 0.11
Benzo[<i>b</i>]fluoranthene	ND	0.14 \pm 0.008	0.043 \pm 0.012	0.225 \pm 0.046	0.076 \pm 0.029	0.158 \pm 0.045
Benzo[<i>k</i>]fluoranthene	ND	0.028 \pm 0.004	0.009 \pm 0.003	0.044 \pm 0.011	0.011 \pm 0.003	0.034 \pm 0.006
Benzo[<i>a</i>]pyrene	0.825 \pm 0.048	0.237 \pm 0.026	0.053 \pm 0.016	0.322 \pm 0.039	0.109 \pm 0.023	0.300 \pm 0.061
Benzo[<i>ghi</i>]perylene	3.14 \pm 0.24	0.095 \pm 0.013	ND	ND	ND	ND

ND, not detected. Data are expressed as mean \pm SD.

^aNumber of experiments ($n = 3$ for all).

Table 3 lists the emission rates of carbonyl compounds identified in the mosquito coil smoke. The results were based on three experiments for each tested brand. The standard deviation of the means of carbonyl compounds emission rates was less than 10% for most of the compounds, suggesting that the experiments had good reproducibility. Among all the carbonyl detected, formaldehyde and acetaldehyde had the highest emission rates and together represented as much as 55% of the total carbonyl compounds emitted from the coil combustion. Acrolein, glyoxal, and methyl-glyoxal, known for their high reactivity, strong irritation effects, and suspected carcinogenic effects, were also detected in the coil smoke in relatively high concentrations. Among all the six tested brands, the two Malaysian coils emitted markedly higher levels of aldehydes than did the Chinese coils. The difference is likely due to different smoldering materials used in making the coils, because the burning patterns for all the tested coils were very similar.

Several VOCs in the coil smoke were identified through the GC/MS analysis, including benzene, toluene, ethylbenzene, *p,m,o*-xylene, and styrene, in relatively high concentrations. According to the GC/MS library, furan, 1,3-pentadiene, 2-burene, isoprene, cyclopentene, 3-hexyne, 2,4-hexadiene, 1,3-cyclohexadiene, cyclohexene, and 1-heptene were tentatively identified. The identification of these compounds indicates that the mosquito coil smoke contains large amounts of aromatic

compounds, alkenes, and furans, some of which are known to cause adverse health effects.

Mosquito coil smoke versus environmental tobacco smoke. Emission factor—presented as microgram of pollutant emitted per gram of coil burned—can be obtained from the emission rate divided by the burn rate. Table 4 lists a comparison of the emission factors of some pollutants found in the mosquito coil smoke with those found in environmental tobacco smoke (ETS) (Daisey et al. 1998; Klepeis et al. 1999). Incorporating the emission factors, coil weight, and cigarette weight (0.55 g/cigarette, excluding the filter), we derived ETS equivalents for all the tested mosquito coils. The results, shown in Table 4, indicate that PM_{2.5} mass released from burning one mosquito coil would be equivalent to PM_{2.5} mass released as ETS from burning 75 (coil C3) to 137 cigarettes (coil C2). The emission of formaldehyde from one tested Malaysian coil can be as high as that from burning 51 cigarettes. However, total PAHs appeared to be substantially lower in mosquito coil smoke than in ETS.

Potential exposure and health risk. The simplified single-compartment mass balance model, Equation 3, was used to estimate pollutant concentrations in room conditions. Mosquito coils are usually used overnight on a daily basis to control mosquitoes in tropical areas and seasonally in subtropical and temperate areas. Each tested Malaysian coil burned 7–8 hr. Although each tested Chinese coil burned for up to 11 hr, people may usually burn coils no more than 8 hr each night.

Hence, we set the average combustion time (*t*) for one coil as 8 hr and ignored any exposure beyond this coil burning period. We assume the air exchange rate of the bedroom is between 2 and 5/hr and the volume of the bedroom (*V*) is in the range of 50–100 m³. The total pollutant removal rate was considered as the same as the air exchange rate, given that air exchange values used here are significantly higher than possible surface deposition rate.

Using the above assumptions, we calculated PM_{2.5} concentrations resulting from burning M1 under varying room volume and air exchange rate values (Figure 4). When volume is 50 m³ (5 × 5 × 2.5 m) and the air exchange rate is 2/hr, the concentration will be 1.17 mg/m³. Even this highest estimate is not unrealistic given that many rooms are smaller and air exchange rate is not substantially higher than 2. [The recommended typical air exchange rate for the United States is 0.45/hr (U.S. EPA 1999b).] Because health-based standards are given as 24-hr average values, we calculated 24-hr average concentrations for several measured pollutants. In our calculation, we assumed that room concentrations during the other 16 hr without mosquito coil use are zero. This apparently resulted in conservative estimates as other sources (e.g., cooking, outdoor penetration, smoking) may also contribute to the 24-hr averages. The lower bound and higher bound of our conservative estimates are shown in Table 5, where the lower-bound values are associated with air exchange rate of 5/hr and room volume of 100 m³, and the higher-bound

Table 3. Emission rates of carbonyl compounds in the gas phase of coil smoke (mg/hr).

Compound ^a	M1	M2	C1	C2	C3	C4
Formaldehyde	7.52 ± 0.61	5.66 ± 0.20	2.21 ± 0.08	1.43 ± 0.07	0.454 ± 0.020	1.31 ± 0.14
Acetaldehyde	2.47 ± 0.19	2.76 ± 0.46	1.00 ± 0.09	2.31 ± 0.21	1.60 ± 0.44	1.66 ± 0.07
Acetone	1.30 ± 0.07	1.48 ± 0.13	0.462 ± 0.009	1.19 ± 0.18	0.880 ± 0.218	0.921 ± 0.072
Acrolein	1.56 ± 0.15	1.22 ± 0.087	0.165 ± 0.022	0.399 ± 0.068	0.0852 ± 0.0059	0.200 ± 0.055
Propanaldehyde	0.852 ± 0.096	0.813 ± 0.083	0.239 ± 0.014	0.507 ± 0.056	0.270 ± 0.052	0.280 ± 0.043
Crotonaldehyde	0.506 ± 0.026	0.459 ± 0.040	0.069 ± 0.119	0.298 ± 0.042	0.321 ± 0.073	0.211 ± 0.009
2-Butanone	2.00 ± 0.34	ND	0.638 ± 0.059	0.723 ± 0.062	0.623 ± 0.173	0.338 ± 0.038
Glyoxal	0.210 ± 0.01	0.352 ± 0.169	0.135 ± 0.092	0.135 ± 0.058	ND	0.051 ± 0.012
<i>o</i> -Tolualdehyde	0.272 ± 0.050	0.209 ± 0.042	0.110 ± 0.095	ND	ND	ND
4-Methyl-2-pentanone	0.590 ± 0.217	1.09 ± 0.59	0.463 ± 0.058	0.590 ± 0.050	0.354 ± 0.049	0.252 ± 0.040
Methylglyoxal	1.20 ± 0.03	1.34 ± 0.30	0.724 ± 0.186	0.667 ± 0.090	0.402 ± 0.102	0.416 ± 0.036

ND, not detected. Data are expressed as mean ± SD.

^aNumber of experiments (*n* = 3 for all).

Table 4. Emission factors of mosquito coils and emission ETS equivalents.

Brand	PM _{2.5}		Formaldehyde		Acetaldehyde		Total PAH ^a	
	Ef (mg/g) ^b	ETS equivalent ^c	Ef (mg/g) ^b	ETS equivalent ^c	Ef (mg/g) ^b	ETS equivalent ^c	Ef (μg/g) ^b	ETS equivalent ^c
M1	70.4 ± 7.3	131	4.54 ± 0.29	51	1.49 ± 0.12	10	64.6	4
M2	68.6 ± 7.7	112	3.38 ± 0.20	33	1.65 ± 0.29	10	19.2	1
C1	34.5 ± 2.9	103	1.21 ± 0.06	22	0.552 ± 0.070	6	0.6	0
C2	57.3 ± 5.1	137	0.967 ± 0.037	14	1.56 ± 0.13	14	4.6	0
C3	32.8 ± 4.9	75	0.291 ± 0.006	4	1.04 ± 0.32	9	1.9	0
C4	34.6 ± 6.0	94	0.639 ± 0.058	11	0.810 ± 0.029	8	3.2	0
ETS	12.4 ± 1.3 ^d	—	2.04 ± 0.41 ^d	—	3.34 ± 0.53 ^d	—	340 ^e	—

Data are expressed as mean ± SD.

^aTotal PAH mass was obtained as the sum of all PAHs quantified in the coil smoke. ^bEmission factor: microgram of pollutant emitted per gram of mosquito coil or per gram of cigarette.

^cThe number of cigarettes needed to produce the same amount of pollutant emitted from burning one mosquito coil. ^dETS data were from Daisey et al. (1998). ^eETS data were from Klepeis et al. (1999).

values are associated with air exchange rate of 2/hr and room volume of 50 m³. Reference exposure levels for aldehydes derived by the Office of Environmental Health Hazard Assessment (OEHHA 2000), and those for PM_{2.5} derived by the U.S. National Ambient Air Quality Standard (NAAQS) (U.S. EPA 2003a), are used for comparison. Our conservative estimates indicate that PM_{2.5} concentrations can be six times higher than the allowable NAAQS for 24-hr average PM_{2.5} concentration. In general, for all the tested Malaysian coils, even the lower-bound estimates are higher than the reference concentrations, except for acetaldehyde. Burning mosquito coils such as the tested Chinese brands may also produce 24-hr-averaged indoor concentrations exceeding the standards or reference values if the condition of the bedroom is not favorable for pollutant removal—for example, if the ventilation is low and/or room volume is small. For example, formaldehyde level can be higher than the reference chronic inhalation concentration, 3 µg/m³. If the coil is placed close to the bed, the exposure to the coil smoke may be even higher. The OEHHA has shown that exposure to acrolein at 0.19 µg/m³ for 1 hr can induce irritation to the eye and respiratory system. Almost all of the estimated concentrations are

higher than this level (Table 5), suggesting that exposure to mosquito coil smoke may cause significant acute health effects. In addition, formaldehyde and acetaldehyde are classified by the U.S. EPA as probable human carcinogens (U.S. EPA 2003b).

The estimates shown above are derived based on the assumption that pollutant concentration in the room is homogeneous. In reality, concentrations actually inhaled by room occupants may be higher than the estimated concentrations because the room air may not necessarily be well mixed and the source (coil) may be placed in close proximity to the breathing zone (the bed level during sleeping). In houses using mosquito coils, children usually sleep in small rooms. To prevent them from excessive mosquito biting, the windows of their rooms are often closed during sleeping hours. Thus, the predicted indoor concentrations above are likely to be very conservative and underestimate actual concentrations in children's rooms. Unfortunately, children are substantially more susceptible to air pollution exposure and thus can be more readily affected than adults (Azizi and Henry 1991; Fagbule and Ekanem 1994).

The pollutants measured in the mosquito coil smoke, as reported in this article, were generated from incomplete combustion of base materials (biomass). Because the coils are made purposely to have very inefficient combustion (smoldering effect), large amounts of products of incomplete combustion are expected from the burning mosquito coils (Zhang et al. 2000). The findings from this study can be indirectly supported by previous studies of other types of biomass (e.g., wood, crop residue, cow dung), burning of which produces the same types of pollutants (Ezzati and Kammen 2001; McCracken and Smith 1998; Smith et al. 2000; Zhang and Smith 1996). The existing literature provides strong evidence that indoor biomass smoke from cooking and heating is a risk factor for acute respiratory infections (ARIs) and chronic obstructive pulmonary disease (COPD). Evidence from 13 studies in developing countries indicate that young children living in homes burning biomass fuels

experience two to three times higher risk of serious ARIs than do unexposed children, after adjustment for potential confounders, including socioeconomic status (Smith et al. 2000). An evaluation of eight studies in developing countries indicates that women cooking over biomass fires for many years have two to four times more risk of COPD than those unexposed, after adjustment for potential confounding factors (Bruce et al. 2000). A recent World Health Organization report estimated that indoor smoke from solid fuels (mainly biomass, also coal) ranked as one of the top 10 risk factors for the global burden of disease in 2000, accounting for 1.6 million premature deaths each year (WHO 2002). Among all environmental risk factors, it ranked second only to poor water/sanitation/hygiene. Comparing the indoor pollutant concentrations predicted in this article with those resulting from biomass fuel combustion, we estimate that exposure to mosquito coil smoke may pose comparable or higher respiratory health risks in people who use mosquito coils for a large fraction of their lifetimes. However, it is more complicated to evaluate health risks of mosquito coils given the obvious benefit associated with mosquito coil use—prevention of malaria and other mosquito-borne diseases.

Conclusions

In this study, a comprehensive characterization of emissions was carried out for six brands of mosquito coils commonly used in China and Malaysia. The pollutants characterized included fine and ultrafine particles, PAHs, VOCs, and aldehydes, with high irritation or suspected carcinogenic effects. We found that all particles emitted from burning mosquito coils were fine, < 1 µm in diameter. Most particles were in the size ranges of 0.01–0.1 µm and 0.2–0.3 µm. In general, the pollutant emissions from the two tested Malaysian brands were substantially higher than those from the four tested Chinese brands. After comparing health-based standards and guidelines, we suggest that exposure to the mosquito coil smoke poses both acute and chronic health risks. Before smoke-generating mosquito coils can be ultimately replaced with nonsmoke mosquito controlling methods, switching from a more polluting type (brand) to a “cleaner” type (brand) may bring substantial reductions in exposure and respiratory health risks.

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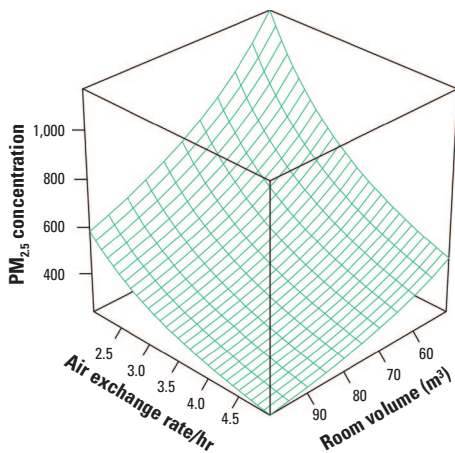


Figure 4. Estimated PM_{2.5} concentrations, during burning M1, as a function of room volume and air exchange rate.

Table 5. The ranges of 24-hr average concentrations, estimated assuming that mosquito coils are used for 8 hr and that room concentrations during the other 16 hr are zero.

Brand	PM _{2.5} (µg/m ³)	Formaldehyde (µg/m ³)	Acetaldehyde (µg/m ³)	Acrolein (µg/m ³)
M1	77.7–388 ^a	5.01–25.0	1.64–8.22	1.04–5.21
M2	76.5–383	3.77–18.8	1.84–9.20	0.81–4.06
C1	41.9–209	1.47–7.36	0.67–3.34	0.11–0.55
C2	56.7–284	0.96–4.78	1.54–7.71	0.27–1.33
C3	34.0–170	0.30–1.51	1.07–5.34	0.06–0.28
C4	47.3–236	0.87–4.37	1.11–5.53	0.13–0.67
Reference	65 ^b	3 ^c	9 ^c	0.19 ^d

^aThe lower bound value represents exposure in a room with an air exchange rate of 5/hr and room volume of 100 m³; the higher bound value represents exposure in a room with an air exchange rate of 2/hr and room volume of 50 m³. ^bU.S. NAAQS for 24-hr average PM_{2.5} concentration. ^cChronic reference exposure levels developed by OEHHA. ^dAcute reference exposure level for acrolein developed by OEHHA.

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