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Technical Note

Characterization of Volatile Organic Pollutant Emissions from Smoldering Mosquito Coils Containing Various Atomic Hydrogen/Carbon Ratios

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ABSTRACT

Volatile organic compounds (VOCs) emitted from smoldering mosquito coils containing various atomic hydrogen/carbon (H/C) ratios in different relative humidity were examined. The concentrations of individual VOCs were determined using a preconcentrator-gas chromatography/mass spectrometer (GC/MS) and a flame ion detector (FID). Up to 72 VOCs were found. It is noteworthy that benzene and 1,3-butadiene, human carcinogenic substances, were emitted by all tested mosquito coils during burning. The five types of mosquito coils studied had atomic H/C ratios ranging from 1.23 to 1.57, yielding total VOC emission rates and factors of 7,295.72–14,308.17 µg hr^{-1} and 3,192.78–6,835.03 µg g^{-1}, respectively. VOC emissions were significantly influenced by the carbon, hydrogen and oxygen contents of coils. According to the analyses of VOC emissions, mosquito coils containing the lowest H/C ratio, a low oxygen content, with suitable additives such as $CaCO_3$ are recommended for minimizing total VOC emission.

Keywords: Mosquito coils; Smoldering; Volatile organic compounds; Emission.

INTRODUCTION

Around the world, burning mosquito coils indoors is a common and efficient method for keeping mosquitoes away (WHO, 2005). Every year, approximately 50 billion mosquito coils are burned with two billion people being exposed to the smoke generated (Zhang *et al.*, 2010). Mosquito coils are commonly made up of fillers, such as wood dust and coconut, dyes, binders, active ingredients, such as metofluthrin, allethrin, esbiothrin and prallethrin, as well as other additives (Chang and Lin, 1998; Lukwa and Chandiwana, 1998; Liu *et al.*, 2003).

Evidence of inflammation and metaplasia of epithelial cells, focal deciliation of the tracheal epithelium, and intercellular fibrosis of lungs have been observed in animals exposed to mosquito coil smoke (Liu *et al.*, 1987; Liu and Sun, 1988; Liu *et al.*, 1989). Epidemiological studies have also revealed that asthma and persistent wheezing in children are associated with exposure to mosquito coil smoke (Koo and Ho, 1994). In view of these findings, mosquito coil smoke indeed poses adverse health risks, constituting a notable public health issue.

Mosquito coil burning, which involves incomplete biomass combustion, is a significant source of indoor air pollution. Toxic smoke comprising gases and particles is produced during the combustion process. The gaseous phase contains carbon monoxide, VOCs, allethrin, and carbonyl compounds (Chang and Lin, 1998; Liu and Sun, 1988; Liu *et al.*, 2003; Lee and Wang, 2006; Zhang *et al.*, 2010). Particles generated are mainly of size smaller than 1 µm (including nanoparticles) and are the typical respirable dusts (Chang and Lin, 1998; Liu *et al.*, 2003). Liu *et al.* (2014) found that essential oil-based mosquito repellent products can generate ultrafine particles and ozone. Not only are these particles more likely to reach and accumulate in human lung alveoli tissue, they also contain a higher concentration of particulates constituting a larger surface area to which harmful substances, such as heavy metals (Cd, Zn and Pb), aldehyde, allethrin, and polycyclic aromatic hydrocarbons (PAHs) can attach (Liu *et al.*, 1987; Chang and Lin, 1998; Liu *et al.*, 2003; Lee and Wang, 2006; Zhang *et al.*, 2010; Dubey *et al.*, 2014; Yang *et al.*, 2015). Harmful health effects may be related to the high concentration of particulates and chemicals attached to the particles.

The mechanisms of mosquito coil burning are similar to those of burning incense sticks. Our previous studies found that volatile organic pollutants formed from burning

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incense sticks are strongly associated with their carbon and hydrogen contents (Yang *et al.*, 2007a, b). Furthermore, in Taiwan, the climate is usually warm and humid, most previous reports do not go into detail as to the effects of environmental conditions (airflow rate and relative humidity) and chemical compositions of mosquito coil on the emissions of volatile organic pollutants. Hence, this research investigated how different chemical compositions of mosquito coil and environmental conditions affect volatile organic pollutant emission. The results thus obtained can provide useful references for selecting the coils with various chemical compositions and the environmental conditions that can minimize VOC emission so as to alleviate indoor air-pollution problems caused by burning mosquito coils.

METHODS

Mosquito Coil Selection

Five types of mosquito coils, same as those used in our previous research, were selected and denoted as A, B, C, D and E, respectively (Yang *et al.*, 2015). According to the information supplied by the manufacturers, mosquito coils A, B, and C contained 0.015 wt% of metofluthrin while coils D and E contained 0.25 wt% and 0.30 wt% of allethrin, respectively. Coils B–D were made in Taiwan while coils A and E were imported from Indonesia and Malaysia, respectively. However, the basic materials used for making these mosquito coils were unknown except for coil E. As shown on the packaging label, the materials used for making coil E included charcoal with sandalwood odor. The mosquito coils were stored in a cabinet with relative humidity of 50 ± 2% and temperature of 25 ± 1°C for 10 days prior to use. The contents of carbon, hydrogen, nitrogen and oxygen in the coils were analyzed using an elemental analyzer (Flash 2000 Analyzer, Thermo, Italy) at the Precision Instrumentation Center, National Taiwan University.

Burning Mosquito Coil Test System

The coil combustion test system used in this work consists of an air-cleaning train, a mosquito coil smoke generation unit (a steel cylinder with an external diameter of 12 cm and a height of 14 cm), a sampling chamber (a steel cylinder with an external diameter of 10 cm and a height of 59 cm), and an airflow unit with humidity and temperature controllers (Fig. 1) (Yang *et al.*, 2015). The relative humidity and temperature of airflow A were measured using a

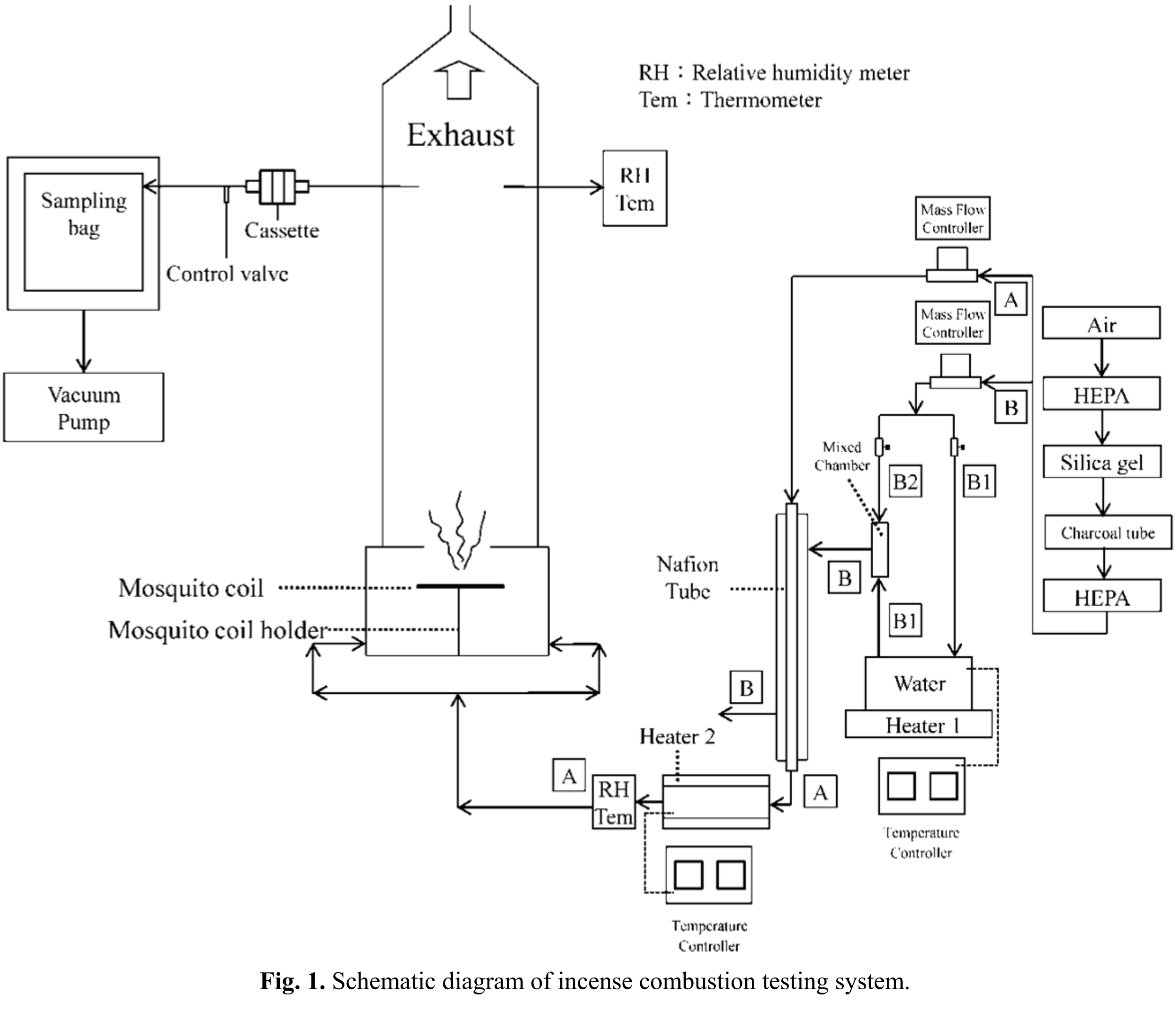

Fig. 1. Schematic diagram of incense combustion testing system.

hygrometer (Hygro Palm 3, Rotronic AG, Switzerland) before it was introduced into the combustion chamber. The air sampling point was at 40 cm from the bottom of the sampling chamber and 1 cm from the wall. The coils were individually ignited and burned at an air flow rate of 8 L min^{-1}. The temperature (28.5 ± 1.5°C) and relative humidity (49.7 ± 0.9%) of the airflow within the configuration and the temperature (24.1 ± 1.9°C) and relative humidity (66.9 ± 2.7%) of the reaction chamber were maintained at steady conditions. Mosquito coil **C** was smoldered at airflow rates of 4, 8 and 12 L min^{-1}, respectively, in the smoldering chamber (circle tube) corresponding to air surface velocities of 0.005, 0.010 and 0.015 m s^{-1} based on calculation of the cross-section area of the circular tube. During the experiment, the airflow and chamber were maintained at a constant temperature (29.4 ± 2.4°C) and relative humidity (50.1 ± 1.6%), and temperature (25.3 ± 2.0°C) and relative humidity (65.5 ± 2.6%). Finally, mosquito coil **C** was burned in the chamber with 49.0 and 94.3% relative humidity at a flow rate of 8 L min^{-1}. The airflow within this configuration was maintained at a constant temperature (28.8 ± 1.7°C) and chamber temperature (25.1 ± 2.6°C).

Volatile Organic Pollutant Sampling Procedure

Each experiment was performed at least in triplicate to ensure experimental reliability. The sampling proceeded as follows. An accurately weighed mosquito coil was placed in the combustion unit. The exit airflow rate was maintained constant. A dynamic equilibrium was achieved in approximately 4 min. according to the results calculated using a single-compartment mass balance model (Fan and Zhang, 2001). After the mosquito coil had been burned for 10 min., particulates and gaseous volatile organic pollutants were collected at a flow rate of 0.5 L min^{-1} for 5 min. by a sampling train, which comprises a two-piece filter cassette (255-2050LF, SKC, USA) with a 37-mm quartz membrane filter (7201, Pall, USA), and a negative pressure vacuum sampling chamber (Vac-U-Chamber, SKC, USA) with a 3-liter tedlar® sample bag (232-03, SKC, USA) connected in series. After sampling, the mosquito coil was burned for an additional min, at which point the coil was extinguished using a stream of nitrogen gas. The remaining mosquito coil was weighed for determining the amount of coil being consumed.

Analysis of Volatile Organic Pollutants

The analytical systems used for verification of peak identities and routine sample analysis are a preconcentrator (7150 Headspace Preconcentrator, ENTECH, USA) with a gas chromatography/mass spectrometer (GC/MS, 6890 GC and 5975 MS, Hewlett Packard, USA) and a preconcentrator (7100 Preconcentrator, ENTECH, USA) with a gas chromatography/flame ion detector (GC/FID; HP6850, Hewlett Packard, USA), respectively. The operating conditions of the preconcentrator-GC/FID are described in detail below. The VOC preconcentrator is composed mainly of the following three modules. Module 1 is made up of a small glass sphere with a trap temperature of –150°C, a desorbed temperature of 10°C, and a bake temperature of 150°C. Module 2 comprises Tenax Ta adsorbent, with a trap temp of –30°C, a desorbed temperature of 180°C, and a baking temperature of 190°C. Module 3 is the condenser focus module, composed of a small curved glass pipe with a trap temperature of –160°C; when the valve temperature is set at 100°C, the desorbed temperature is about 60–70°C. The analysis conditions of the GC/FID are as follows: (a) Capillary column: DB-1, with a column inner diameter of 0.32 mm, a length of 60 m, and a film thickness of 1 µm; (b) Oven temperature: the initial temperature of 35°C maintained for 5 min., then raised at 6 °C min^{-1} to reach 120°C, further increased at 10 °C min^{-1} to 210°C and finally maintained at 210°C for 3 min; with total analysis time of 31.17 min.; (c) Injection port temperature: 250°C; (d) Detector temperature: 250°C, and (e) Column flow rate: 1.4 mL min^{-1}.

The peaks of the bromochloromethane and 1,4-difluorobenzene of internal standards in the chromatography of preconcentrator-GC/FID analyses could not be resolved from the ethyl acetate and cyclohexane of standards, respectively. Therefore, only two internal standards were used for quantitative analysis in this study. The standards of 64 VOCs (Linde Spectra Environmental Gases, USA) were analyzed at the range of 8.94–633.87 ng to establish the standard calibration curves for determining the concentrations of VOCs. The correlation coefficients of the calibration curves were greater than 0.995, except that the correlation coefficients of 1,4-dichlorobenzene, 1,2,4-trichlorobenzene, naphthalene and 1,1,2,3,4,4-hexachloro-1,3-butadiene were greater than 0.971 (Table 1). The detection limits were determined using a threefold standard deviation of seven measurements of the lowest concentration of a calibration curve. The detection limit range was 18.4–575.4 µg m^{-3}. The blank of the combustion test system was used in each experiment. The background values for the blank of the combustion test system were subtracted from all samples.

Emission Rates and Factors of Individual Pollutants

According to the mass balance principle, the emission rate and emission factor of volatile organic pollutants can be derived from formula (1):

$$V \times \frac{dCi}{dt} = R \times E_f - Q \times Ci \tag{1}$$

where V : chamber volume (m^3),

C_i : gaseous pollutant concentration (µg m^{-3}),

R : mosquito coil burn rate (g hr^{-1}),

E_f : emission factor (µg g^{-1}), and

Q : tested gas flow volume (m^3 hr^{-1}).

Upon ignition of the mosquito coil, the volatile organic pollutant concentration reaches a dynamic equilibrium within a short period of time. At this point, $dCi/dt = 0$ in formula (1), which can be rewritten as:

$$E_f = \frac{Q \times C}{R} \tag{2}$$

where C: gaseous pollutant concentration (µg m^{-3}) inside the testing chamber at equilibrium.

Table 1. Correlation coefficients and method detection limits for individual compounds.

I.D.	Compounds	r[c]	MDL	Remark	I.D.	Compounds	r	MDL	Remark
1	Propylene	0.999	20.3		33	Bromodichloromethane	0.996	70.3	
2	Dichlorodifluoromethane	0.999	47.8		34	1,4-Dioxane	0.999	20.4	Merge
3	Methane, chloro-	0.999	19.4		35	Trichloroethylene			
4	Dichlorotetrafluoroethane	0.999	58.8		36	Methyl Methacrylate	0.999	77.1	
5	Ethene, chloro-	0.999	25.6		37	Heptane	0.999	38.7	
6	1,3-Butadiene	0.999	22.2		38	1-Propene, 1,3-dichloro-, (Z)-	0.999	46.8	Merge
7	Methane, bromo-	0.999	35.8		39	Methyl Isobutyl Ketone			
8	Ethyl chloride	0.999	32.7		40	1-Propene, 1,3-dichloro-, (E)-	0.999	37.3	
9	Ethanol	0.999	18.4		41	Ethane, 1,1,2-trichloro-	0.999	46.9	
10	Acetone	0.999	77.6		42	Toluene	0.999	35.5	
11	Trichloromonofluoromethane	0.999	52.8	Merge	43	Methyl Butyl Ketone	0.998	51.3	
12	Isopropyl Alcohol				44	Dibromochloromethane	0.999	76.8	
13	Ethene, 1,1-dichloro-	0.999	52.1		45	Ethane, 1,2-dibromo-	0.999	68.7	
14	Methylene Chloride	0.999	66.4		46	Tetrachloroethylene	0.999	44.6	
15	Ethane, 1,1,2-trichloro-1,2,2-trifluoro(Freon 113)	0.999	237.4	Merge	A3	Benzene-d5-, chloro-	-	-	
16	Carbon disulfide				47	Benzene, chloro-	0.999	84.1	
17	Ethene, 1,2-dichloro-, (E)-	0.999	69.9		48	Ethylbenzene	0.998	63.0	
18	Ethane, 1,1-dichloro-	0.999	54.9		49	p-xylene	0.998	318.8	Merge
19	Methyl Tert Butyl Ether	0.999	160.8		50	m-xylene			
20	Vinyl Acetate	0.999	175.7		51	Bromoform	0.999	185.4	
21	Methyl Ethyl Ketone	0.999	54.7		52	Styrene	0.999	42.5	
22	Ethene, 1,2-dichloro-, (Z)-	0.999	52.9		53	Ethane, 1,1,2,2-tetrachloro-	0.999	299.2	Merge
A1[b]	Bromochloromethane	-	-	Not isolated	54	o-xylene			
23	Ethyl Acetate				A4	Benzene, 1-bromo-3-fluoro-	-	-	
24	Hexane	0.999	47.7		55	4-Ethyltoluene	0.998	199.7	
25	Chloroform	0.999	63.6		56	Benzene, 1,3,5-trimethyl-	0.998	251.8	
26	Tetrahydrofuran	0.999	105.9		57	Benzene, 1,2,4-trimethyl-	0.996	254.8	
27	Ethane, 1,2-dichloro-	0.999	46.8		58	Benzyl Chloride	0.995	168.3	Merge
28	Ethane, 1,1,1-trichloro-	0.999	68.4		59	Benzene, 1,3-dichloro-			
29	Benzene	0.999	35.5		60	Benzene, 1,2-dichloro-	0.996	93.9	
30	Carbon Tetrachloride	0.999	93.2		61	Benzene, 1,4-dichloro-	0.994	166.0	
31	Cyclohexane	-	-	Not isolated	62	Benzene, 1,2,4-trichloro-	0.971	217.9	
A2	Benzene, 1,4-difluoro-				63	Naphthalene	0.973	139.5	
32	Propane, 1,2-dichloro-	0.999	49.9		64	1,3-Butadiene, 1,1,2,3,4,4-hexachloro-	0.993	575.4	

[a] Unit is μg m^{-3}.

[b] A1, A2, A3 and A4 were internal standard.

[c] Correlation coefficients of the calibration curves.

The above formula can also be employed to calculate the concentrations of volatile organic compounds, with ppb (v/v at 25°C, 1 atm.) converted into µg m^{-3} according to the proper molecular weight of each compound with the ideal gas law.

RESULTS AND DISCUSSION

Chemical Compositions of Mosquito Coil

The contents of carbon, hydrogen, nitrogen and oxygen in mosquito coils A–E were 41.25–51.43, 4.24–5.71, 0.25–0.85 and 25.28–41.93%, respectively, as shown in Table 2. The contents of other elements ranged from 8.50 to 28.81%. The atomic hydrogen/carbon (H/C) ratios were in the range of 1.23–1.57.

Identification of Volatile Organic Pollutants

The VOCs emitted from burning five kinds of mosquito coil were identified and quantified using preconcentrator-GC/FID (GC/MS) analysis. A total of 67, 72, 41, 52 and 62 VOCs were identified in the smoke from smoldering mosquito coils A–E, respectively by comparing the mass spectra with the NIST (National of Standards and Technology). According to the carcinogen classification by the International Agency for Research on Cancer (IARC), the results showed that burning mosquito coils produced some chemicals harmful to human health, such as benzene and 1,3-butadiene (category 1, human carcinogens); acetaldehyde, furan, methyl chloride, ethylbenzene and styrene (category 2B, possible human carcinogens); propylene, toluene, xylene, furfural and *d*-limonene (category 3, not classifiable) (IARC, 2015). On the other hand, 14 VOCs (propylene, methyl chloride, 1,3-butadiene, ethanol, acetone, methyl ethyl ketone, hexane, benzene, heptane, toluene, ethylbenzene, *m*/*p*-xylene and styrene) were further quantified by comparing the preconcentrator-GC/FID chromatogram of the sampled mosquito coils with the preconcentrator-GC/FID chromatogram of real standard materials (Table 3). However, the quantifications of methylene chloride and *o*-xylene by comparing with a real standard sample failed because of the inseparable peaks containing *o*-xylene and 1,1,2,2,-tetrachloroethane (preconcentrator-GC/FID chromatography) and methylene chloride and an unknown peak (GC/FID chromatography). The presence of methyl chloride, benzene, toluene, ethylbenzene, *p*/*m*-xylene, styrene and *o*-xylene were confirmed in this work, which was consistent with the findings of Lee *et al.* (2006). In addition, more adverse health compounds, such as 1,3-butadiene, furan, furfural, propylene, were found in this study.

Effect of Chemical Compositions of Mosquito Coil on Volatile Organic Pollutant Emissions

The total concentration of all organic compounds (sum of propylene, methyl chloride, 1,3-butadiene, ethanol, acetone, methyl ethyl ketone, hexane, benzene, heptane, toluene, ethylbenzene, *m*/*p*-xylene and styrene) released from the burning of the five sampled mosquito coils ranged from 15,199.42 to 29,808.70 µg m^{-3}. Among the organic pollutants, propylene was dominant (28.2–38.1%), followed by acetone (21.4–29.3%), methyl chloride (8.1–21.2%), benzene (7.4–10.1%), 1,3-butadiene (4.3–7.4%), toluene (4.5–5.5%), methyl ethyl ketone (4.1–6.2%), and others (2.9–4.2 %).

Table 3 shows that the emission rates of all VOCs during smoldering of the five mosquito coils studied ranged from 7,295.72 to 14,308.17 µg hr^{-1}. The range of VOC emission rate of coils A–E are as follows: benzene (718.01–1,307.46 µg hr^{-1}, category 1), 1, 3-butadiene (311.38–1,061.92 µg hr^{-1}, category 1), ethylbenzene (N.D.–57.09 µg hr^{-1}, category 2B), styrene (21.46–129.03 µg hr^{-1}, category 2B), toluene (377.96–717.99 µg hr^{-1}, category 3), propylene (2,779.01–4,383.49 µg hr^{-1}, category 3) and *m*/*p*-xylene (87.79–155.62 µg hr^{-1}, category 3). In summary, the total emission rates of detrimental VOCs during smoldering of mosquito coils in decreasing order is D > B > E > A > C.

On the other hand, the VOC emission factors from smoldering coils A–E ranged from 3,192.78 to 6,835.03 µg g^{-1} (Table 4) and were as follows: benzene (314.23–587.33 µg g^{-1}, category 1), 1,3-butadiene (136.23–506.97 µg g^{-1}, category 1), ethylbenzene (N.D.–27.29 µg g^{-1}, category 2B), and styrene (9.19–61.76 µg g^{-1}, category 2B), toluene (165.42–345.14 µg g^{-1}, category 3), propylene (1,215.85–2,102.12 µg g^{-1}, category 3) and *m*/*p*-xylene (39.29–80.36 µg g^{-1}, category 3). Lee *et al.* (2006) reported that the emission factors of benzene and toluene during mosquito coil burning were 55.4–232.2 and 58.5–728.7 µg g^{-1}, respectively. Higher benzene emission observed in the current findings might be attributed to the variations in coil chemical composition or different experimental conditions. However, the toluene emission factors in this work were within the same range as reported by Lee *et al.* (2006).

It is worthy to note that the emission factors of total acetone and methyl ethyl ketone for coils A, B and D were higher than those for coils C and E. A possible explanation is that coils A, B and D had similar oxygen contents, ranging

Table 2. Weight percentages of carbon, hydrogen, nitrogen and oxygen in mosquito coils

Mosquito Coil Type	Element Content (%)					Atomic Ratio
	C	H	N	O	Other	H/C
A	43.55 ± 0.61[a]	5.56 ± 0.01	0.85 ± 0.07	41.52 ± 0.51	8.52 ± 0.03	1.53 ± 0.02
B	42.04 ± 0.06	5.45 ± 0.07	0.25 ± 0.003	41.60 ± 0.45	10.67 ± 0.58	1.55 ± 0.02
C	41.25 ± 0.06	4.24 ± 0.001	0.42 ± 0.001	25.28 ± 0.33	28.81 ± 0.40	1.23 ± 0.001
D	43.54 ± 0.02	5.71 ± 0.03	0.32 ± 0.01	41.93 ± 0.35	8.50 ± 0.41	1.57 ± 0.008
E	51.43 ± 0.13	5.33 ± 0.05	0.37 ± 0.001	30.00 ± 0.13	12.87 ± 0.31	1.24 ± 0.008

[a] Mean ± standard deviation. Sample size = 2.

[b] The contents listed were from the work of Yang *et al.* (2015).

Table 3. VOC emission rates of mosquito coils studied.

Compounds	Mosquito Coil Type										IARC
	A		B		C		D		E		
	Mean	S.D.[a]	Mean	S.D.	Mean	S.D.	Mean	S.D.	Mean	S.D.	
Propylene	3971.58	531.79	4383.49	416.65	2779.01	117.33	4284.76	264.50	3650.75	263.64	3
Methane, chloro-	994.78	67.20	2198.74	262.57	1155.39	26.28	2347.11	125.51	2744.62	198.63	
1,3-Butadiene	907.98	148.40	995.76	34.61	311.38	15.70	1061.92	96.38	588.84	43.42	1
Ethanol	27.68	5.43	99.54	22.03	16.02[e]	N.D.	83.23	20.87	125.12	19.97	
Acetone	3591.75	312.95	3602.76	389.38	1564.60	101.87	3586.16	236.52	3162.19	233.02	
Methyl Ethyl Ketone	761.63	50.42	705.91	87.91	295.88	27.48	696.90	59.40	611.10[d]	36.14	
Hexane	41.86	9.13	30.67	5.75	31.12	6.36	32.06[d]	0.74	45.88	6.71	
Benzene	1032.66	105.84	1029.09	64.10	718.01	29.65	1155.62	67.77	1307.46	67.37	1
Heptane	28.53	2.40	23.45	1.59	20.62	2.30	25.49	2.41	28.84	1.61	
Toluene	669.21	40.80	623.12	33.95	377.96	14.42	717.99	39.81	634.11	38.54	3
Ethylbenzene	40.66	0.65	39.99	1.36	N.D.[c]	N.D.	57.09	5.88	44.74	1.92	2B
m/p-xylene	155.62	3.03	131.45	3.85	87.79[e]	N.D.	141.51	8.92	130.99	2.21	3
Styrene	33.98	8.70	59.77	2.48	21.46[e]	N.D.	129.03	20.11	74.00	6.31	2B
Total	12257.91	1221.27	13923.74	1258.11	7295.72	271.44	14308.17	853.75	12944.97	515.04	

[a] Sample size = 3. S.D.: Standard deviation.

[b] The unit of individual volatile organic compound emission rate is μg hr^{-1}.

[c] N.D.: Not detectable.

[d] Only occurred twice.

[e] Only occurred once.

Table 4. VOC emission factors of mosquito coils studied.

Compounds	Mosquito Coil Type										IARC
	A		B		C		D		E		
	Mean	S.D.[a]	Mean	S.D.	Mean	S.D.	Mean	S.D.	Mean	S.D.	
Propylene	2046.57	227.67	2102.12	173.72	1215.85	26.42	2046.51	67.25	1639.98	107.34	3
Methane, chloro-	512.99	20.42	1054.00	110.21	505.71	11.00	1121.26	29.24	1232.70	74.77	
1,3-Butadiene	467.38	63.73	477.80	13.53	136.23	4.41	506.97	31.98	264.54	18.38	1
Ethanol	14.24	2.44	47.94	11.78	6.86[e]	N.D.	39.85	10.50	56.16	8.32	
Acetone	1851.50	110.12	1727.03	158.28	684.77	42.81	1713.35	84.17	1420.15	85.77	
Methyl Ethyl Ketone	392.76	15.11	338.26	35.81	129.51	11.98	332.90	23.01	276.66[d]	16.60	
Hexane	21.52	4.16	14.67	2.41	13.59	2.49	15.11[d]	0.003	20.59	2.73	
Benzene	532.17	39.85	493.47	19.76	314.23	11.30	552.12	21.61	587.33	23.72	1
Heptane	14.71	0.84	11.24	0.55	9.01	0.81	12.17	0.86	12.96	0.55	
Toluene	345.14	11.66	298.81	8.55	165.42	5.86	343.11	14.90	284.82	13.55	3
Ethylbenzene	20.99	0.28	19.19	0.15	N.D.[c]	N.D.	27.29	2.87	20.10	0.61	2B
m/p-xylene	80.36	2.81	63.13	3.58	39.29[e]	N.D.	67.65	4.34	58.86	0.98	3
Styrene	17.47	4.03	28.68	1.25	9.19[e]	N.D.	61.76	10.40	33.25	2.73	2B
Total	6317.80	459.68	6676.34	501.98	3192.78	89.84	6835.03	239.18	5815.87	185.71	

[a] Sample size = 3. S.D.: Standard deviation.

[b] The unit of individual volatile organic compound emission factor is $\mu g\ g^{-1}$

[c] N.D.: Not detectable.

[d] Only occurred twice.

[e] Only occurred once.

from 41–42%, while coils C and E had relatively lower oxygen contents, 25% and 30%, respectively. Hence, oxygen content of the coil is the key determinant of emission factors of volatile oxygenated organic compounds.

The total VOC emission factors for during smoldering of coils A–E were 6,317.80 ± 459.68, 6,676.34 ± 501.98, 3,192.78 ± 89.84, 6,835.03 ± 239.18 and 5,815.87 ± 185.71 µg g^{-1}, respectively; showing significant variations (ANOVA, $p < 0.05$) and were in the following order C < E < A < B < D. The total VOC emission factors for coils A, B and D were higher than that of the others owing to their higher H/C ratios (Yang *et al.*, 2007b). Sawada *et al.* (2000) found that the materials previously treated at higher carbonization temperature resulted in lower atomic H/C ratio of entrained carbonized wood because volatile compounds and pyrolysis products were already released as smoke during carbonization. Comparing between coils C and E with similar atomic H/C ratios (1.23 and 1.24, respectively) revealed that the total VOC emission factor for coil C was substantially lower than that for coil E because coil C had the lowest oxygen content and greater amounts of other elements in additional to C, H, N, and O (Tab1e 1). Coils containing similar H/C ratios with lower oxygen content can reduce the formation of volatile oxygenated organic compounds (such as acetone and methyl ethyl ketone). On the other hand, coils with similar H/C ratios and lower oxygen and carbon contents also contain greater amount of heteroatom(s) and/or $CaCO_3$, which could lead to the consumption of hydrogens and reduce the possibility for the formation of CHCH fragments available for VOC formation during smoldering. Therefore, mosquito coil C with the lowest H/C ratio, lowest oxygen and highest heteroatom contents has the lowest total VOC emission factor.

Effect of Airflow Rates on Volatile Organic Pollutant Emissions

Coil C was chosen for studying the effect of airflow because of the lowest oxygen content and greater amounts of other elements in additional to C, H, N, and O. The range of concentrations, emission rates and factors of total VOC released during burning mosquito coil C at airflow rates of 4, 8 and 12 L min^{-1} were 13,180.16–26,580.72 µg m^{-3}, 6,379.37–9,489.72 µg hr^{-1} and 3,121.43–3,818.80 µg g^{-1}, respectively. The total VOC emission rates for coil C at airflow rates of 4, 8 and 12 L min^{-1} were 6,379.37 ± 209.70, 7,295.72 ± 271.44 and 9,489.72 ± 438.74 µg hr^{-1}, respectively (Table 5). The total VOC emission rates differ significantly with the variation in airflow rates (ANOVA, $p < 0.05$). When the airflow rate increases between 4 and 12 L min^{-1}, the total VOC emission rates increase, however, the total VOC concentrations decrease (Table 5).

The all (total) VOC emission factors do not differ significantly between 4 and 8 L min^{-1} (t-test, $p > 0.05$). However, the emission factors of 1,3-butadiene, acetone, methyl ethyl ketone, benzene, toluene and total VOC show significant variations (t-test, $p < 0.05$) between 8 and 12 L min^{-1}, except for propylene, methyl chloride, ethanol, hexane, heptane, ethylbenzene, *m*/*p*-xylene and styrene compounds. The increase the proportion of the 1,3-butadiene, acetone, methyl ethyl ketone, benzene, toluene and total VOC emission factors from 8 to 12 L min^{-1} range from 18.04–34.41%. This analytical result is similar to a previous study reported by Yang *et al.* (2005) indicated when the higher air flow rate with a less combustion efficiency, the VOC emission factors were more higher. Hence, these data from smoldering mosquito coil C with various airflow rates indicate environmental combustion was controlled at low airflow rates and produced a low emission rates and factors for total volatile organic pollutants.

Effect of Environmental Relative Humidity on Volatile Organic Pollutant Emissions

The range of concentrations, emission rates and factors of total VOC released during burning coil **C** at a relative humidity of 49.0 and 94.3% were 13,923.13–15,199.42 µg m^{-3}, 6,683.10–7,295.72 µg hr^{-1} and 3,192.78–3,634.99 µg g^{-1}, respectively. The emission rates of propylene, methyl chloride, 1,3-butadiene, acetone, methyl ethyl ketone, hexane, benzene, heptane, toluene and total VOC decreasing upon increasing the relative humidity (from 49.0 to 94.3%), except for ethanol, ethylbenzene, and styrene compounds (Table 6). On the other hand, the emission factors of propylene, methyl chloride, 1,3-butadiene, acetone, benzene, toluene and total VOC show significant variations (t-test, $p < 0.05$) between 49.0 and 94.3% and a positive influence (from 49.0 to 94.3%), except for ethanol, methyl ethyl ketone, hexane, heptane, ethylbenzene, *m*/*p*-xylene and styrene compounds (Table 6). This analytical result is similar to a previous study reported by Manoukian *et al.* (2016) investigate the effect of relative humidity on VOC/SVOC emission from burning incense. They have found that benzene, toluene, acetone and diethylhexyl phthalate showed a positive influence when relative humidity increase from 30 to 70%, the other analyzed compounds (chrysene, benzo[*a*]anthracene and benzo[*a*]pyrene) has not been found significant variations.

CONCLUSIONS

This study confirms that the carbon, hydrogen and oxygen contents of mosquito coil were the critical determinants of emission factors of toxic volatile organic pollutants. Benzene and 1,3-butadiene (category 1, confirmed human carcinogens); acetaldehyde, furan, methyl chloride, ethylbenzene and styrene (category 2B, possible human carcinogens); propylene, toluene, furfural, xylene and *d*-limonene (category 3, not classifiable as to its carcinogenicity to humans) were detected from burning mosquito coils. Smoldering coil as a mosquito repellent indoor releases toxic volatile organic pollutants, posing health risks to people exposed to the smoke; and the poorer the ventilation, the more severe the potential harmful health risks.

Thus, in consideration of volatile organic pollutant emissions, individuals should avoid burning mosquito coils indoors, especially in the presence of the sick, the elderly, children, infants, and pregnant women who are more vulnerable to the detrimental impacts of VOCs. Exposure to various VOCs leads to the adverse health effects including

Table 5. Effect of air flow rates on VOC emission rates and factors from mosquito coil C.

Compounds	Airflow Rate (L min^{-1})											
	Emission Rate						Emission Factor					
	4		8		12		4		8		12	
	Mean	S.D.[a]	Mean	S.D.	Mean	S.D.	Mean	S.D.	Mean	S.D.	Mean	S.D.
Propylene	2220.17	133.48	2779.01	117.33	3407.05	153.86	1087.04	99.06	1215.85	26.42	1371.98	78.06
Methane, chloro-	1061.73	13.84	1155.39	26.28	1477.33	166.31	519.39	26.02	505.71	11.00	594.09	57.95
1,3-Butadiene	220.00	31.70	311.38	15.70	455.06	27.48	107.92	18.71	136.23	4.41	183.10	8.82
Ethanol	24.20[e]	N.D.	16.02[e]	N.D.	31.25	11.09	11.35[e]	N.D.	6.86[e]	N.D.	12.58	4.43
Acetone	1460.34	18.35	1564.60	101.87	2124.74	83.01	714.13	26.77	684.77	42.81	855.07	23.82
Methyl Ethyl Ketone	281.32	9.28	295.88	27.48	416.36	19.41	137.49	3.44	129.51	11.98	167.52	4.82
Hexane	25.24	1.84	31.12	6.36	38.95	2.18	12.36	1.29	13.59	2.49	15.69	1.09
Benzene	657.98	26.74	718.01	29.65	921.82	42.93	321.88	20.22	314.23	11.30	370.91	11.43
Heptane	17.36	0.37	20.62	2.30	N.D.	N.D.	8.49	0.40	9.01	0.81	N.D.	N.D.
Toluene	331.58	6.82	377.96	14.42	509.02	23.76	162.16	7.07	165.42	5.86	204.80	5.58
Ethylbenzene	17.15	0.32	N.D.[c]	N.D.	N.D.	N.D.	8.39	0.41	N.D.	N.D.	N.D.	N.D.
m/p-xylene	64.41	0.94	87.79[e]	N.D.	117.89[d]	2.73	31.51	1.58	39.29[e]	N.D.	46.94[d]	1.61
Styrene	14.03	5.84	21.46[e]	N.D.	44.31[d]	6.75	6.88	2.93	9.19[e]	N.D.	17.66[d]	2.88
Total	6379.37	209.70	7295.72	271.44	9489.72	438.74	3121.43	196.98	3192.78	89.84	3818.80	133.84

[a] Sample size = 3. S.D.: Standard deviation.

[b] The unit of individual volatile organic compound emission rate and factor are µg hr^{-1} and µg g^{-1}, respectively.

[c] N.D.: Not detectable.

[d] Only occurred twice.

[e] Only occurred once.

Table 6. Effect of relative humidity on VOC emission rates and factors from mosquito coil C.

Compounds	Relative Humidity (%)							
	Emission Rate				Emission Factor			
	49.0		94.3		49.0		94.3	
	Mean	S.D.[a]	Mean	S.D.	Mean	S.D.	Mean	S.D.
Propylene	2779.01	117.33	2464.31	60.31	1215.85	26.42	1342.25	57.41
Methane, chloro-	1155.39	26.28	1110.57	72.40	505.71	11.00	603.79	3.49
1,3-Butadiene	311.38	15.70	286.48	27.80	136.23	4.41	155.57	6.87
Ethanol	16.02[e]	N.D.	65.78[e]	N.D.	6.86[e]	N.D.	33.64[e]	N.D.
Acetone	1564.60	101.87	1438.32	126.34	684.77	42.81	781.26	23.18
Methyl Ethyl Ketone	295.88	27.48	277.56	36.46	129.51	11.98	150.47	10.61
Hexane	31.12	6.36	24.85[d]	1.53	13.59	2.49	13.06[d]	0.29
Benzene	718.01	29.65	645.09	25.24	314.23	11.30	351.17	11.65
Heptane	20.62	2.30	19.84[e]	N.D.	9.01	0.81	10.15[e]	N.D.
Toluene	377.96	14.42	339.04	12.15	165.42	5.86	184.59	6.78
Ethylbenzene	N.D.[c]	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
m/p-xylene	87.79[e]	N.D.	79.93[d]	4.74	39.29[e]	N.D.	44.86[d]	0.25
Styrene	21.46[e]	N.D.	23.33	2.17	9.19[e]	N.D.	12.68	0.50
Total	7295.72	271.44	6683.10	375.21	3192.78	89.84	3634.99	49.59

[a] Sample size = 3. S.D.: Standard deviation.

[b] The unit of individual volatile organic compound emission rate and factor are μg hr^{-1} and μg g^{-1}, respectively.

[c] N.D.: Not detectable.

[d] Only occurred twice.

[e] Only occurred once.

irritation of eyes and skin, asthma in young children (Rumchev *et al.*, 2004), airway inflammation (Koren *et al.*, 1992) and obstruction (Harving *et al.*, 1991) and cancer. Furthermore, the sick, elderly, children and infants have more weak immune defenses and airway functions, they are more less ability to eliminate toxic VOCs than health adult. Alternatively, mosquito coils containing the lowest H/C ratio, a low oxygen level, with suitable additives such as $CaCO_3$ should be chosen to minimize the total VOC emission.

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