

Characteristics of emissions of air pollutants from mosquito coils and candles burning in a large environmental chamber

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Abstract

The objective of this study was to characterize the emissions of air pollutants from mosquito coils and candles burning in a large environmental test chamber. The target pollutants included particulate matters (PM₁₀, PM_{2.5}), carbon monoxide (CO), nitrogen oxides (NO_x), methane (CH₄), non-methane hydrocarbons (NMHC), volatile organic compounds (VOCs) and carbonyl compounds. The average PM₁₀ concentrations for all tested mosquito coils exceeded Excellent and Good Classes objectives specified by *Indoor Air Quality Objectives for Office Buildings and Public Places* (IAQO) [HKEPD, 2003. *Guidance Notes for the Management of Indoor Air Quality in Offices and Public Places*. Indoor Air Quality Management Group, The Government of the Hong Kong Special Administrative Region]. The emission factors (mg g⁻¹ mosquito coil) of mosquito coils combustion were: PM_{2.5}, 20.3–47.8; PM₁₀, 15.9–50.8; CO, 74.6–89.1; NO, 0.1–0.5; NO₂, n.d.–0.1; NO_x, 0.1–0.5; CH₄, n.d.–4.7; NMHC, 0.1–5.7. Formaldehyde and acetaldehyde were the most abundant carbonyls species in the coil smoke. The average concentrations of formaldehyde and benzene of all tested mosquito coils exceeded Good Class of IAQO. Nitrogen oxides were the most abundant gas pollutants relating to candle burning among all target air pollutants. The candle made of gel (CAN 4) would emit more air pollutants than the paraffin candles (CAN 1, 2 and 3) and beeswax candle (CAN 5). Among five candles tested, CAN 5, the one made of beeswax, generated relatively smaller amount of air pollutants. It was noted that the concentrations of most VOCs from candles combustion were below the detection limit. © 2005 Elsevier Ltd. All rights reserved.

Keywords: Environmental chamber; Emission factor; Candle; Mosquito coil; VOCs

1. Introduction

Burning of mosquito coils and candles will release a variety of toxic chemicals. A mosquito coil is spiral-shaped, the smoke of which is generally used for a mosquito repellent and insecticide in the tropical and subtropical areas (Li et al., 1993). The

combustion of mosquito coils could generate particles containing heavy metals such as Cd, Zn and Pb, allethrin, and other organic compounds e.g. phenol and *O*-cresol (Liu et al., 1987). Liu and Sun (1988) investigated emissions of organic compounds from mosquito coil smoke by using gas chromatography–mass spectrometry (GC–MS). The major identified volatile organic compounds constituted allethrin, phenol, benzene, toluene and xylene, as well as aromatic and aliphatic hydrocarbons. Liu et al. (2003) also indicated that burning one

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mosquito coil would release the same amount of $PM_{2.5}$ mass as burning 75–137 cigarettes.

When candles are burned, they emit trace of organic chemicals, including acetaldehyde, formaldehyde, acrolein and naphthalene (Lau et al., 1997). Some candles also emit lead which is the primary constituent of public health concern in candle emissions (USEPA, 2001). Metal was originally put in wicks to keep the wick standing straight when the surrounding wax begins to melt. It is possible for consumers to unknowingly purchase candles containing lead wick cores and repeatedly expose themselves to harmful amounts of lead and organic chemicals through regular candle-burning (USEPA, 2001). Burning several candles exceeded the USEPA's 10^{-6} increased risk for cancer for acetaldehyde and formaldehyde, and exceeded the Reference Concentration (R_{fC}) for acrolein.

The objectives of this project were to characterize emissions of air pollutants from the burning of mosquito coils and candles using a large environmental chamber; to compare air pollutant emissions from different types of mosquito coils and candles, which are popular on Hong Kong market; to qualify and quantify emissions from burning of mosquito coils and candles with respect to particulate matter ($PM_{2.5}$ and PM_{10}), volatile organic compounds (VOCs), carbonyl compounds, carbon monoxide (CO), nitrogen oxides (NO_x), methane (CH_4) and non-methane hydrocarbons (NMHC). The proper usage of mosquito coils and candles will lead to

reducing the health risks of consumers. In addition, the project will provide a useful database for further risk assessment or health-related study as well as regulatory agencies to set up guidelines or standards for mosquito coils and candles combustion.

2. Methodology

2.1. Materials

Five types of mosquito coils and five types of candles, which are commonly used in households in Hong Kong, were tested in this study. The material selection was based on production quantity of the materials and extent of use in a home. The appearances (shape, length, cross-sectional area and color) of these mosquito coils are quite similar. The length for mosquito coil burning was measured before the experiment. Mosquito coil was cut in certain length in order to facilitate the burning time to approximately 1 h. The general information of selected mosquito coils is shown in Table 1(a).

Selected candles are manufactured in China and Australia. They have different appearances (shape, length, diameter, colour, fragrance) and ingredients. Most of the candles tested were scented candles for home decorating and for producing fragrance. Selected candles are made of beeswax, paraffin and gel. Beeswax is formed from a liquid extruded from the wax glands of a honeybee; paraffin is a by-product from the oil-refining process and gel

Table 1

| ID No. | Country of origin | Shape | Length of combustion part (cm) | Color | Avg. burning time (min) | Mass (g) | |
|--|-------------------|--------|--------------------------------|----------------|-------------------------|-------------------------|-------------|
| <i>(a). General information of tested mosquito coils</i> | | | | | | | |
| MC 1 | Hong Kong | Spiral | 15 | Dark green | 60 | 2.49 | |
| MC 2 | Hong Kong | Spiral | 15 | Dark green | 60 | 2.20 | |
| MC 3 | Hong Kong | Spiral | 15 | Dark green | 60 | 2.37 | |
| MC 4 | Hong Kong | Spiral | 15 | Dark green | 60 | 2.02 | |
| MC 5 | Hong Kong | Spiral | 15 | Dark green | 60 | 2.02 | |
| ID No. | Country of origin | Color | Size | Fragrance | Wax | Avg. burning time (min) | Weight (kg) |
| <i>(b). General information of tested candles</i> | | | | | | | |
| CAN 1 | China | White | 7.5 cm H × 5 cm D | French vanilla | Paraffin | 120 | 0.1272 |
| CAN 2 | China | White | 7.5 cm H × 5 cm D | None | Paraffin | 185 | 0.1263 |
| CAN 3 | China | Red | 7.5 cm H × 5 cm D | Rose | Paraffin | 120 | 0.1239 |
| CAN 4 | China | Purple | Container | Iris | Gel | 91 | 0.1296 |
| CAN 5 | Australia | White | 14 cm H × 2.3 cm D | Nectar | Beeswax | 91 | 0.0364 |

candles use liquids such as mineral oil, terpene-type chemicals, or modified hydrocarbons as their primary fuel. The general information of selected candles is shown in Table 1(b).

2.2. Chamber experiments

The controlled experiments were conducted in a stainless-steel environmental test chamber ($3.2\text{ m} \times 3.2\text{ m} \times 2.5\text{ m}$ with 18.26 m^3 effective volume) maintained at a controlled environmental conditions: temperature $23 \pm 0.5\text{ }^\circ\text{C}$, RH $50 \pm 5\%$ and air exchange rate $0.5 \pm 0.02\text{ h}^{-1}$. The air exchange rate of $0.5 \pm 0.02\text{ h}^{-1}$ was to simulate the typical natural ventilation conditions. The mixing and leakage of the large environmental chamber were evaluated by using the same method described in Lee et al., 2003. The variation of air leakage rate was 0.02 h^{-1} which was found to be acceptable because it was $<1\%$ of the air exchange rate (ECA-IAQ, 1989). A portable Q-Trak monitor (model 8550, TSI Inc.) was put in the chamber to monitor that the local temperature and RH. Fig. 1 shows the schematic diagram of the experimental set-up. The chamber was purged by blower air, which was passed through a clean air system with activated charcoal particle filters and high-efficiency particulate air (HEPA) filters. Mixing fans were installed at the ceiling of the chamber to ensure adequate air mixing. Air samples were drawn from the chamber via Teflon tubing into a

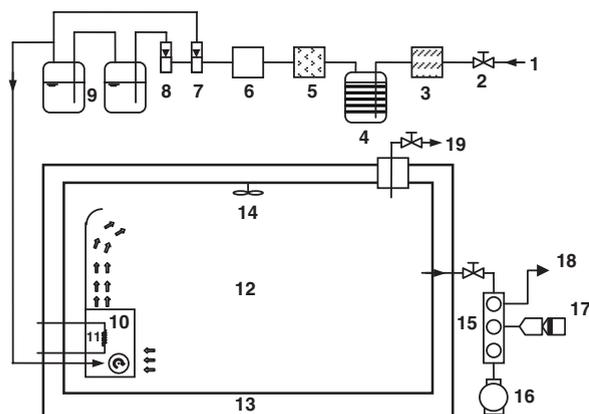


Fig. 1. Schematic diagram of the experimental set-up. (1) Inlet, (2) valve, (3) blower, (4) active charcoal filters, (5) HEPA filters, (6) mass flow controllers, (7) flow controller dry air, (8) flow controller wet air, (9) humidifier, (10) rotating cylinder, (11) heating unit, (12) large environmental test chamber, (13) insulation, (14) mixing fan, (15) sampling manifold, (16) canister, (17) ozone scrubber & DNPH cartridge, (18) A Teflon tubing connecting to gas analyzers, (19) outlet.

pre-evacuating canister for VOC sampling. For carbonyl sampling, a pump located outside the chamber drew the chamber air to pass through sampling cartridges. A Teflon tubing connected the chamber with a CO analyzer, a NO–NO₂–NO_x analyzer and a CH₄–NMHC analyzer.

Prior to the experiments, the chamber was conditioned for about 4 h at tested environmental conditions. The background air samples were collected and the concentrations do not exceed $10\text{ }\mu\text{g m}^{-3}$ of TVOC and $2\text{ }\mu\text{g m}^{-3}$ of any individual VOC (USEPA, 1999).

2.3. Sampling methods and analysis

Two Dust-Trak monitors (model 8520, TSI Inc.) were used to measure PM₁₀ and PM_{2.5} concentrations simultaneously in the chamber. GFC Ambient CO Analyzer (Model 48, Thermo Environmental Instruments Inc.), Chemiluminescence NO–NO₂–NO_x Analyzer (Model 42C, Thermo Environmental Instruments Inc.) and Direct Methane, Non-Methane Hydrocarbon Analyzer (Model 55C, Thermo Environmental Instruments Inc.) were used to quantify CO, NO_x, CH₄ and NMHC concentrations, respectively. A five-point calibration and a zero check were performed daily for each instrument with standard gas at known concentrations.

For VOCs sampling, grab samples were taken before and after burning (30 min after the tested specimens have extinguished). Time-integrated samples were taken during burning. The flow rates were controlled by the mass flow controllers (model FC4101CV-G, Autoflow Inc., CA) and ranged from 21.6 to 66.7 ml min^{-1} (depend on the burning time). After sampling, the canisters were shipped to the laboratory and analyzed within three hours. Prior to sampling, the canisters were cleaned by sequential evacuating and pressurizing with humidified zero air. Blank sample (10%) canisters filled with standard gas containing target organic compounds of known concentrations were conducted during sample collection and storage with other sampling canisters for GC/MS analysis. Detailed description of the analysis can be found in Lee et al. (2002). A total of 38 VOCs species were identified by the gas chromatograph/mass selective detector (GC/MSD) system (model 6890/5973, Hewlett Packard) and 10 species were selected in this study. The analytical procedures were performed according to the US EPA method TO-14 (USEPA, 1998b).

Carbonyls were collected by a silica gel cartridge impregnated with acidified 2,4-dinitrophenylhydrazine (Waters Sep-Pak DNPH-silica). An ozone scrubber was connected before the DNPH-silica cartridge in order to prevent interference from ozone. Time-integrated samples were taken with the flow rate of approximately 1000 ml min^{-1} and no breakthrough was found. The sampling period was 1 h before burning and 30 min after burning. The sampling period during burning depended on the burning time of each tested specimens. Flow rate was measured at the start and end of each sample collection period using a soap-bubble flow meter (Gilian Gilibrator 2, Enviro-Equipment, Inc., NC). The carbonyls in the cartridges were then measured using reverse-phase high-performance liquid chromatography (HPLC). The HPLC system consisted of a dual wavelength absorbance detector (Waters 2487) operating at 360 nm with a binary pump (Waters 1525) and an in-line degasser. A Nova-Pak (Waters) C_{18} reverse phase column ($150 \text{ mm} \times 3.9 \text{ mm}$) with a particle size of 4 mm and pore size of 60 \AA was used to separate the hydrazones. Three blank cartridges were analyzed from each batch of 50 cartridges to account for the cartridge background concentration. Background air samples were collected to ensure background formaldehyde levels do not exceed $2 \mu\text{g m}^{-3}$ (USEPA, 1999). A total of 10 carbonyl compounds were identified in this study with a detection limit of 0.2 ppbv. All analysis procedures were according to the US EPA method TO-11 (USEPA, 1998a).

The mosquito coils and candles tested were weighed using a micro-balance with an accuracy of 0.01 mg. The detection mechanism, ranges and limits for each analytical method are summarized in

Table 2. Duplicate samples were collected in order to ensure the data consistency and accuracy.

2.4. Modelling of emissions

The emission rates and emission factors for the mosquito coils were determined by using a single-compartment mass balance model (Fan and Zhang, 2001). The concentration of the pollutant in the chamber could be described with the following equations:

$$C = P(1 - e^{-kt})/Vk \quad (\text{when } 0 \leq t \leq T), \quad (1)$$

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

where C is the pollutant concentration (mg m^{-3}) in the chamber, P is the emission rate (mg h^{-1}), V is the volume of the chamber (m^3), C_{\max} is the maximum pollutant concentration (mg m^{-3}) in the chamber, i.e., the concentration at the time when the mosquito coils were extinguished, k is the pollutant removal rate (h^{-1}), t is time (h): $t = 0$ when the mosquito coils were ignited; $t = T$ when the mosquito coils were extinguished. The value of k is the regression slope of the plot of $\ln(C)$ versus t , derived from Eq. (2). The value of P/Vk is the regression slope of the plot of C versus $(1 - e^{-kt})$. Then, P can be obtained because V is known. Background concentrations of all the measured pollutants were subtracted in determination of k and P/Vk using the above equations. The burning rate B (g h^{-1}) was obtained from the amount (g) of mosquito coils burnt and the duration (h) of burning, the emission factor E_f (mg g^{-1}) was calculated using the following equation:

$$E_f = P/B. \quad (3)$$

Table 2
Detection mechanism, ranges and limits of the analytical methods

| Air parameter | Detectable mechanism | Range | Minimum detection limit |
|---|---|-------------------------------------|---|
| Carbon monoxide (CO) | Non-dispersive infra-red (NDIR) | 0–50 ppm | 0.1 ppm |
| Nitrogen oxides (NO_x) | Chemiluminescence | 0–1000 ppb | 0.40 ppb |
| Methane (CH_4) and non-methane hydrocarbons (NMHC) | Flame ionization detection (FID) | 0–200 ppm (non-methane hydrocarbon) | 0.02 ppm methane 0.05 ppm NMHC as propane |
| VOCs | Gas chromatography mass selective detector (GC/MSD) | 0–5000 $\mu\text{g m}^{-3}$ | 0.20 ppb |
| Carbonyls | High performance liquid chromatography (HPLC) | 0–5000 $\mu\text{g m}^{-3}$ | 0.20 ppb |

3. Results and discussion

3.1. Particulate matter

Table 3 lists out the emission rates and emission factors of PM_{2.5} and PM₁₀ during the combustion of the mosquito coils. The value of linear regression R^2 ranged from 0.9523 to 0.9996. The emission rates and factors varied considerably across different mosquito coils. The emission factors ranged from 21.5 to 47.8 mg g⁻¹ mc for PM_{2.5} and from 15.9 to 50.8 mg g⁻¹ mc for PM₁₀. MC 2 had both the highest emission rate and emission factor for PM_{2.5} and PM₁₀. Liu et al. (2003) found that the ultrafine and fine particles dominated the counts of particles emitted by coil combustion, and the emission factor of PM_{2.5} were from 32.7 to 70.1 mg h⁻¹, which was close to the findings in this study.

Table 3
Emission rates and factors for the 5 tested mosquito coils

| Sample ID | Emission rate (mg h ⁻¹) | Emission factor (mg g ⁻¹ mc) |
|-------------------|-------------------------------------|---|
| PM _{2.5} | | |
| MC 1 | 72.1 | 28.9 |
| MC 2 | 109.8 | 47.8 |
| MC 3 | 48.0 | 20.3 |
| MC 4 | 88.5 | 43.9 |
| MC 5 | 43.4 | 21.5 |
| PM ₁₀ | | |
| MC 1 | 79.0 | 31.7 |
| MC 2 | 112.1 | 50.8 |
| MC 3 | 40.1 | 16.9 |
| MC 4 | 70.1 | 34.8 |
| MC 5 | 32.0 | 15.9 |

Table 4
Ranges of PM_{2.5} and PM₁₀ concentrations during mosquito coils and candles burning

| | PM _{2.5} ranges (mg m ⁻³) | PM _{2.5} average conc. (mg m ⁻³) | PM ₁₀ ranges (mg m ⁻³) | PM ₁₀ average conc. (mg m ⁻³) | PM _{2.5} /PM ₁₀ ratio (%) |
|-------|--|---|---|--|---|
| MC 1 | 0.81–6.83 | 3.92 | 0.94–7.72 | 4.55 | 86.15 |
| MC 2 | 0.02–10.05 | 4.95 | 0.02–10.53 | 5.21 | 95.01 |
| MC 3 | 0.15–4.36 | 2.39 | 0.39–4.47 | 2.61 | 91.57 |
| MC 4 | 0.01–8.44 | 5.75 | 0.01–9.04 | 6.34 | 90.69 |
| MC 5 | 0.04–6.11 | 4.61 | 0.04–7.75 | 5.31 | 86.82 |
| CAN 1 | 0.015–0.032 | 0.021 | 0.017–0.035 | 0.023 | 91.30 |
| CAN 2 | 0.013–0.049 | 0.044 | 0.019–0.052 | 0.049 | 89.80 |
| CAN 3 | 0.013–0.025 | 0.019 | 0.015–0.028 | 0.022 | 86.36 |
| CAN 4 | 0.019–0.063 | 0.043 | 0.025–0.086 | 0.054 | 79.63 |
| CAN 5 | 0.012–0.044 | 0.031 | 0.019–0.061 | 0.04 | 77.50 |

Table 4 shows the ranges of PM_{2.5} and PM₁₀ concentrations during mosquito coils and candles burning. The average concentrations were taken from ignition to extinguishing, no background concentrations are included. The PM_{2.5} concentrations of mosquito coils ranged from 0.01 to 10.05 mg m⁻³, and PM₁₀ ranged from 0.01 to 10.53 mg m⁻³. The average PM concentration for MC 4 (i.e. 5.75 mg m⁻³ for PM_{2.5} and 6.34 mg m⁻³ for PM₁₀) was the highest. The average PM₁₀ concentrations for all tested mosquito coils exceeded the Hong Kong IAQO 8-h-average Good Class concentration of PM₁₀ (0.18 mg m⁻³). The PM_{2.5}/PM₁₀ ratios of the mosquito coils burning were around 90%, which suggests that the majority particulate matter emitted was in the PM_{2.5} size fraction.

The range and average concentrations of PM_{2.5} and PM₁₀ for tested candles are shown in Table 4. Most of the peak values were observed immediately after the candles were extinguished. The PM_{2.5} and PM₁₀ concentrations of tested candles were relatively lower than the tested mosquito coils. CAN 4, which was made of gel, had the highest average PM_{2.5} and PM₁₀ concentrations (0.043 and 0.054 mg m⁻³, respectively). The measured PM₁₀ concentrations of all tested candles satisfied the Good Class of IAQO.

3.2. Criteria gas pollutants (CO, NO, NO₂, NO_x, CH₄, and NMHC)

The emission rates and factors of tested mosquito coils for 6 criteria gas pollutants (CO, NO, NO₂, NO_x, CH₄, and NMHC) are summarized in Table 5. The CO emission rates and factors ranged from

150.4 to 220.0 mg h⁻¹ and 74.6 to 89.1 mg g⁻¹ mc, respectively. High CO concentration was measured from mosquito coils because the coils are made purposely to have very inefficient combustion

Table 5
Emission rates and factors of mosquito coils

| | MC 1 | MC 2 | MC 3 | MC 4 | MC 5 |
|---|-------|-------|-------|-------|-------|
| Emission rate (mg h ⁻¹) | | | | | |
| CO | 220.0 | 196.5 | 182.6 | 150.4 | 167.2 |
| NO | 0.3 | 1.1 | 0.8 | 0.3 | 0.2 |
| NO ₂ | 0.2 | n.d. | n.d. | n.d. | n.d. |
| NO _x | 0.6 | 1.1 | 0.8 | 0.3 | 0.2 |
| CH ₄ | 11.9 | 9.1 | 11.2 | n.d. | n.d. |
| NMHC | 12.2 | 12.5 | 13.3 | 0.1 | 1.1 |
| Emission factor (mg g ⁻¹ mc) | | | | | |
| CO | 88.3 | 89.1 | 77.1 | 74.6 | 82.8 |
| NO | 0.1 | 0.5 | 0.3 | 0.2 | 0.1 |
| NO ₂ | 0.1 | n.d. | n.d. | n.d. | n.d. |
| NO _x | 0.2 | 0.5 | 0.3 | 0.2 | 0.1 |
| CH ₄ | 4.6 | 4.1 | 4.7 | n.d. | n.d. |
| NMHC | 4.9 | 5.7 | 5.6 | 0.1 | 0.5 |

Note: n.d.: detected but not quantified (The change of concentrations is too small to quantify the emission rate or factor).

(smoldering effect) (Zhang et al., 2000). Large amount of incomplete combustion product, therefore, would be emitted from mosquito coil burning.

The NO emission rates and factors varied among tested mosquito coils. The highest emission rate occurred on MC 2 (1.1 mg h⁻¹), which was nearly five times of that of MC 5 (0.2 mg h⁻¹). MC 2 also had the highest NO emission factor (0.5 mg g⁻¹ mc), which was approximately five times of that of MC 1 (0.1 mg g⁻¹ mc). Except MC 1, there was no obvious NO₂ emission from the mosquito coils. The amount of NO₂ emitted from MC 1 was small, the emission rate and factor were 0.2 mg h⁻¹ and 0.1 mg g⁻¹ mc, respectively. The emission rates and factors of CH₄ and NMHC for MC 1, 2, and 3 were close.

Burning candles also emitted certain air pollutants, including CO, NO, NO₂, NO_x, CH₄, and NMHC. However, the changes of concentrations were too small to quantify the emission rate or emission factor. The maximum, average and minimum concentrations of the air pollutants emitted from five candles are shown in Fig. 2. The minimum concentrations were usually measured at the beginning of the experiment. The peak values were usually measured immediately after the candle

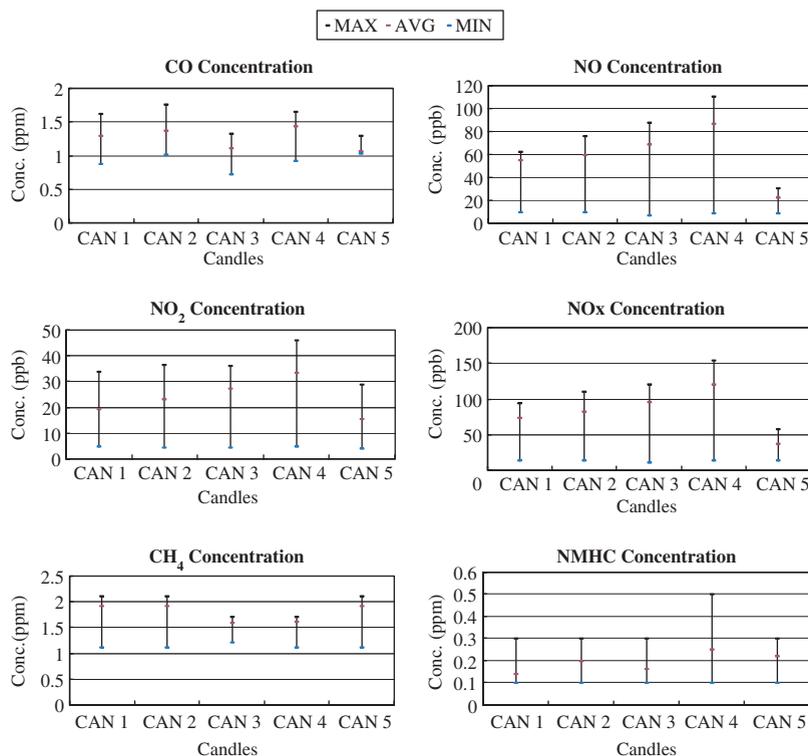


Fig. 2. Concentrations of gas pollutants of five tested candles during burning. Note: MAX: maximum concentration during burning, AVG: average concentration during burning, MIN: minimum concentration during burning.

extinguishing. It can be seen that CO, CH₄ and NMHC concentrations did not increase significantly due to the burning of candles. However, the concentrations of NO and NO₂ were observed to increase significantly. The peak NO concentrations did not vary significantly across the first four candles, however, for CAN 5, the peak NO concentration was relatively lower. The peak concentrations of NO₂ were similar among all tested candles. CAN 4 had the highest NO₂ concentration (45.7 ppb), while CAN 5 had the lowest concentration (28.7 ppb). The recommended 8-h average CO and NO₂ Good Class concentrations are 8.7 ppm and 80 ppb, respectively. All the CO and NO₂ concentrations emitted from candles burning were below the IAQO.

CAN 4, the one made of gel, would emit more air pollutants than the paraffin candles (CAN 1, 2, and 3) and beeswax candle (CAN 5). Because of the softness, gel wax was only usable in a container, which would cause the incompleteness of combustion. Among five candles tested, CAN 5, made of beeswax, generated relatively smaller amount of air pollutants. The main reason is that beeswax is an animal-origin wax with natural color and scent and burns cleaner. However, the other two materials, paraffin and gel, both are by-products of oil refining process. In addition, they both contain waste petroleum, which has been proved to be an air pollutant.

3.3. Carbonyl compounds

Fig. 3 illustrates the concentrations of 10 carbonyl compounds identified for five tested mosquito coils during and after combustion. Formaldehyde and acetaldehyde were the most abundant carbonyl species in the coil smoke. The concentrations of formaldehyde, acetone, acrolein, propionaldehyde, and valeraldehyde varied significantly among tested mosquito coils. The variation is likely due to the difference in smoldering materials used in making mosquito coil. The concentrations of formaldehyde, acetone, acrolein, and propionaldehyde reach the peaks during the coil burning. However, the concentrations of acetaldehyde, crotonaldehyde, methacrolein, valeraldehyde, and m-tolualdehyde reach the peaks shortly after burning. The formaldehyde concentration recommended by IAQO Good Class is 100 $\mu\text{g m}^{-3}$. However, the average formaldehyde concentrations of all tested mosquito coils exceeded this level. Especially for MC 1, 4, and

5, the peak concentrations were more than two times of the specified criteria.

The carbonyl compounds concentrations emitted by the mosquito coils were normalized by the mosquito coils mass. The emission factors are presented in Fig. 4. The variations were quite similar among tested mosquito coils, except acetone and acrolein. For acetone, the emission factors of MC 4 and MC 5 were substantially higher than other three mosquito coils. The acetone emission factor of MC 5 approached 600 $\mu\text{g m}^{-3}$, which was five times more than that of MC 2. The emission factor of acrolein of MC 1 reached 700 $\mu\text{g m}^{-3}$, which was nearly four times of that of MC 5. The carbonyl concentrations found in this study are less than that found by Chang and Lin's study (1998). Two mosquito coils were tested and the results showed that burning 1 g of mosquito coil generated 2025.0 μg formaldehyde, 2030.9 μg acetaldehyde, and 1423.7 μg acrolein, while burning the other one produced 3922.7 μg formaldehyde, 2147.8 μg acetaldehyde, and 1747.1 μg acrolein.

3.4. Volatile organic compounds

Most VOCs concentrations measured from candles combustion were below the detection limit. Several VOCs emitted from coil smoke were identified, including benzene, toluene, methylene chloride, with relatively high concentrations. The identification of these compounds indicated that the mosquito coil smoke contains large amounts of aromatic compounds, some of which are known to cause adverse health effects.

Table 6 shows the temporal changes of VOCs concentrations during and after burning of tested mosquito coils and the emission factors normalized by the mosquito coil mass. Benzene, methylene chloride, and toluene were the most abundant VOCs species from mosquito coils burning. The concentrations of methylene chloride and benzene reached the peak values once the mosquito coils were extinguished. However, for MC 1, 2, and 3, the concentrations of toluene during burning were higher than the concentrations after burning.

The concentrations of benzene, methylene chloride, and toluene varied significantly among tested mosquito coils. At after-burning stage, MC 2 had the highest methylene chloride concentration (109.6 $\mu\text{g m}^{-3}$), which was two times of that of MC 1 (52.3 $\mu\text{g m}^{-3}$). MC 1 had the highest benzene concentration (59.5 $\mu\text{g m}^{-3}$) after burning while MC

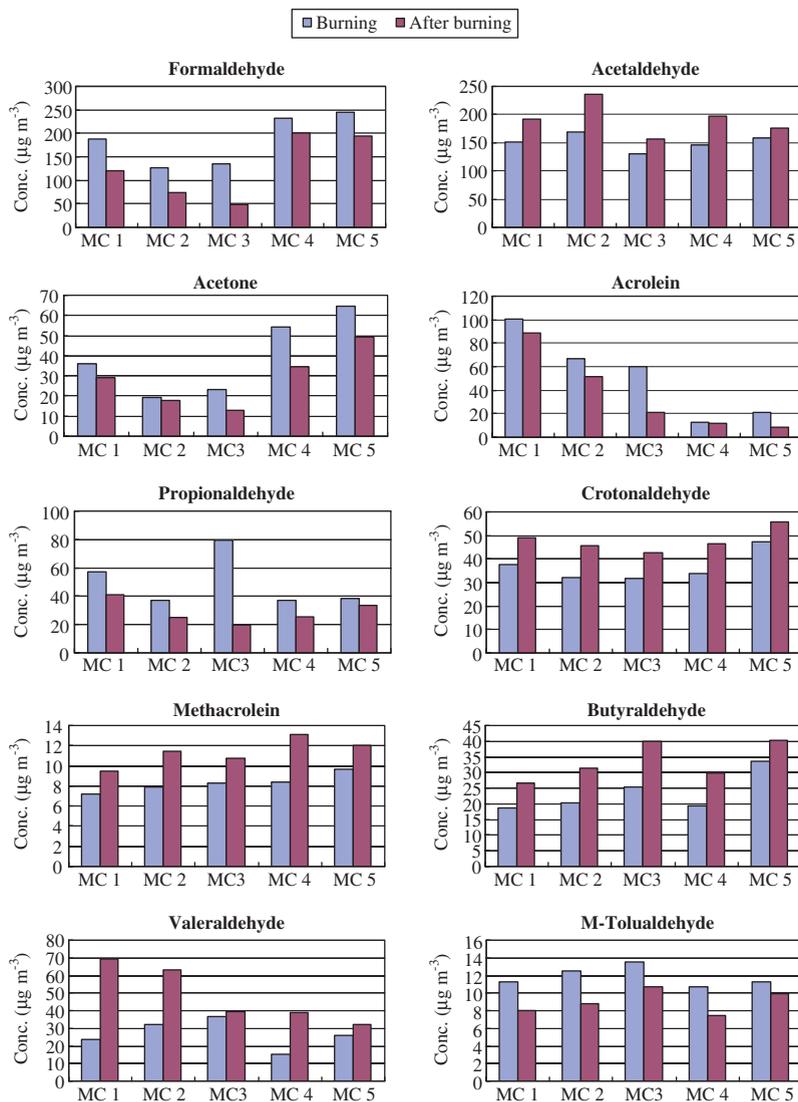


Fig. 3. Carbonyl compounds concentrations of mosquito coils burning.

3 had the lowest ($24.7 \mu\text{g m}^{-3}$). For toluene, the highest concentration occurred on burning stage of MC 2, which reached $88.1 \mu\text{g m}^{-3}$. However, the lowest toluene concentration occurred on MC 3 ($7.6 \mu\text{g m}^{-3}$).

The recommended benzene, chloroform, and toluene levels according to IAQO Good Class are 16.1, 163, and $1092 \mu\text{g m}^{-3}$, respectively. As shown in Table 6, the concentration levels of chloroform and toluene all complied with these criteria very well. However, the benzene concentrations of all tested mosquito coils exceeded the specified criteria significantly, especially after the mosquito coil burning.

The average concentrations of other individual volatile organic compounds, such as chloroform, 1,2-dichloroethane, *cis*-1,2-dichloroethene, ethylbenzene, *m,p*-xylene, also increased after the mosquito coils were ignited. However, the concentrations were not significant when compared to benzene, methylene chloride, and toluene.

4. Conclusions

Five types of mosquito coils and five types of candles were tested in a large environmental chamber. The emission factors ($\text{mg g}^{-1} \text{mc}$) of mosquito coils combustion were: $\text{PM}_{2.5}$, 20.3–47.8;

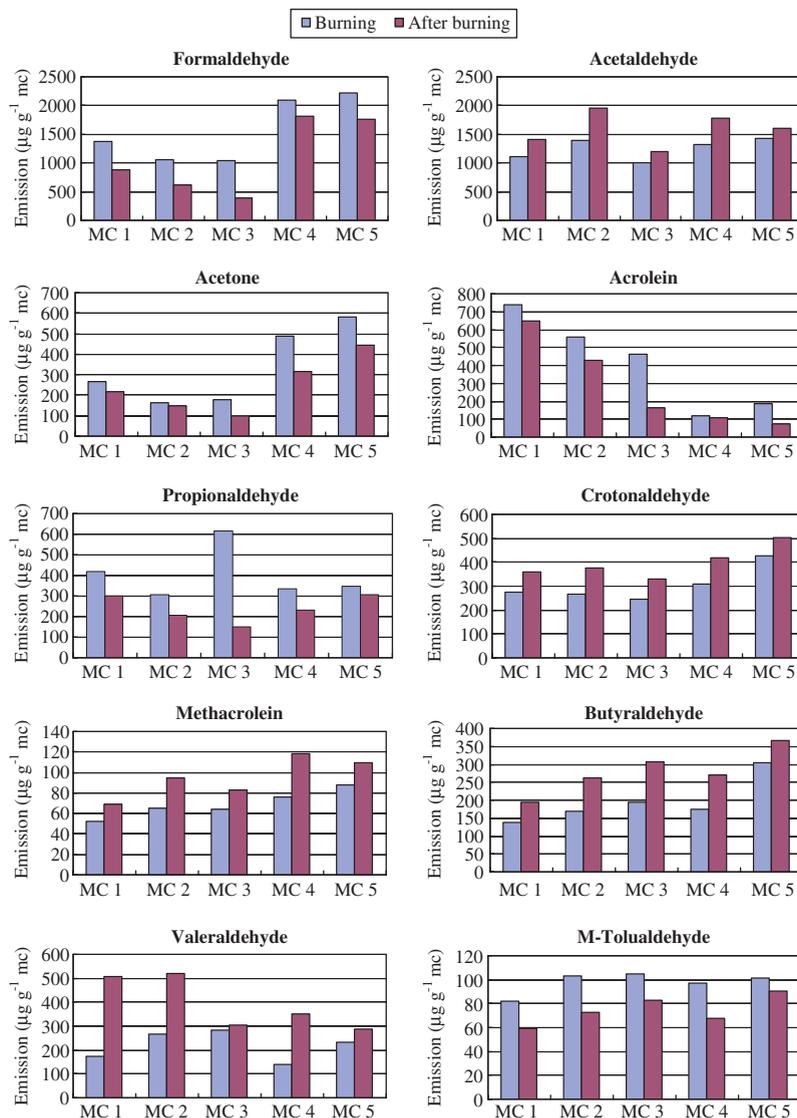


Fig. 4. Emission factors of carbonyl compounds of mosquito coils burning.

PM_{10} , 15.9–50.8; CO, 74.6–89.1; NO, 0.1–0.5; NO_2 , n.d.–0.1; NO_x , 0.1–0.5; CH_4 , n.d.–4.7; NMHC, 0.1–5.7. The average PM_{10} concentrations of all tested mosquito coils significantly exceeded the Good Class of IAQO level. CO was the major gas pollutant and resulted from the smoldering effect of mosquito coils. Formaldehyde and acetaldehyde were the major carbonyl compounds identified in the coil smoke. The average formaldehyde concentrations of all tested mosquito coils exceeded IAQO Good Class level. Especially for MC 1, 4, and 5, the peak concentrations were more than two times of the specified criteria. Benzene, methylene chloride,

and toluene were the most abundant VOCs species from mosquito coils burning. The concentrations of these three VOCs varied significantly among tested mosquito coils. The benzene concentrations of all tested mosquito coils exceeded IAQO Good Glass level significantly, especially after the mosquito coil burning. Mosquito coils burning would emit large amount of benzene, which has adverse health effect to human beings. $\text{PM}_{2.5}$ and PM_{10} concentrations of tested candles were relatively lower than the tested mosquito coils. The measured PM_{10} concentrations of all tested candles satisfied the Good Class of IAQO. Nitrogen oxides were the most abundant gas

Table 6
Temporal changes of VOCs concentrations and emission factors for five types of mosquito coils

| | MC 1 | | MC 2 | | MC 3 | | MC 4 | | MC 5 | |
|---------------------------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| | a | b | a | b | a | b | a | b | a | b |
| VOCs ($\mu\text{g m}^{-3}$) | | | | | | | | | | |
| Methylene chloride | 28.8 | 52.3 | 64.5 | 109.6 | 55.0 | 87.9 | 54.4 | 58.2 | 63.3 | 60.9 |
| <i>cis</i> -1,2-Dichloroethene | 7.9 | 22.0 | n.d. | n.d. | 12.1 | 11.3 | 11.1 | 23.8 | 18.6 | 1.0 |
| Chloroform | 4.0 | 6.8 | 4.2 | 5.8 | n.d. | n.d. | 4.7 | 7.9 | 5.5 | 6.4 |
| 1,2-Dichloroethane | 1.3 | 5.9 | 1.1 | 7.7 | n.d. | n.d. | n.d. | 2.0 | n.d. | 4.2 |
| Benzene | 13.8 | 59.5 | 22.3 | 57.2 | 7.2 | 24.7 | 25.6 | 39.1 | 19.8 | 40.0 |
| Toluene | 40.1 | 27.0 | 88.1 | 62.4 | 7.6 | 15.7 | 8.7 | 17.2 | 17.7 | 24.7 |
| Ethylbenzene | 4.4 | 3.2 | 8.2 | 7.6 | n.d. | n.d. | n.d. | n.d. | n.d. | 0.9 |
| <i>m,p</i> -Xylene | 1.9 | 2.4 | 4.5 | 5.4 | n.d. | 0.6 | n.d. | n.d. | 1.2 | 1.9 |
| Styrene | n.d. | 2.1 | n.d. | 6.4 | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. |
| <i>O</i> -Xylene | 0.9 | 0.9 | 1.7 | 2.0 | n.d. | n.d. | n.d. | n.d. | n.d. | 0.4 |
| VOCs ($\mu\text{g g}^{-1}$ mc) | | | | | | | | | | |
| Methylene chloride | 211.0 | 383.2 | 533.5 | 906.8 | 424.4 | 678.3 | 492.5 | 526.7 | 572.5 | 550.5 |
| <i>cis</i> -1,2-Dichloroethene | 57.9 | 161.3 | n.d. | n.d. | 93.6 | 86.8 | 100.4 | 215.7 | 168.2 | 9.4 |
| Chloroform | 29.4 | 49.7 | 34.4 | 47.9 | n.d. | n.d. | 42.6 | 71.3 | 50.1 | 58.2 |
| 1,2-Dichloroethane | 9.7 | 42.8 | 8.9 | 64.1 | n.d. | n.d. | n.d. | 18.3 | n.d. | 37.6 |
| Benzene | 100.9 | 435.8 | 184.8 | 473.7 | 55.4 | 190.3 | 232.2 | 354.0 | 178.7 | 361.6 |
| Toluene | 294.0 | 198.0 | 728.7 | 516.7 | 58.5 | 120.9 | 78.4 | 155.7 | 160.4 | 223.4 |
| Ethylbenzene | 4.4 | 3.2 | 8.2 | 7.6 | n.d. | n.d. | n.d. | n.d. | n.d. | 8.4 |
| <i>m,p</i> -Xylene | 1.9 | 2.4 | 4.5 | 5.4 | n.d. | 0.6 | n.d. | n.d. | 10.7 | 17.5 |
| Styrene | n.d. | 2.1 | n.d. | 6.4 | n.d. | n.d. | n.d. | n.d. | n.d. | n.d. |
| <i>O</i> -Xylene | 0.9 | 0.9 | 1.7 | 2.0 | n.d. | n.d. | n.d. | n.d. | n.d. | 4.0 |

Note:

- (1) a: Average value during time between ignition and completion of mosquito coil burning.
- (2) b: Average value after completion of mosquito coil burning.
- (3) n.d.: Not detected.

pollutants relating to burning of candles among all target air pollutants. The concentrations of most VOCs from candles combustion were below the detection limit. CAN 4, the one made of gel, would emit more air pollutants than the paraffin candles (CAN 1, 2, and 3) and beeswax candle (CAN 5). Among five candles tested, CAN 5, made of beeswax, generated relatively smaller amount of air pollutants. It was recommended to burn the candles and mosquito coils in a room with good ventilation and comprehensive information should be labeled on the packaging.

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References

- Chang, J.Y., Lin, J.M., 1998. Aliphatic aldehydes and allethrin in mosquito coil smoke. *Chemosphere* 36 (3), 617–624.
- ECA-IAQ (European Collaborative Action Indoor Air Quality and Its Impact on Man), 1989. Strategy for Sampling Volatile Organic Compounds (VOCs) in Indoor Air. Report No. 14. EUR 12617 EN. Luxembourg, Office for Official Publications of the European Community.
- Fan, C.W., Zhang, J.J., 2001. Characterization of emissions from portable household combustion devices: particle size distributions, emission rates and factors, and potential exposures. *Atmospheric Environment* 35 (7), 1281–1290.
- HKEPD, 2003. Guidance Notes for the Management of Indoor Air Quality in Offices and Public Places. Indoor Air Quality Management Group, The Government of the Hong Kong Special Administrative Region.
- Lau, C., Fiedler, H., Hutzinger, O., Schwind, K.H., Hosseinpour, J., 1997. Levels of selected organic compounds in materials for candle production and human exposure to candle emissions. *Chemosphere* 34 (5–7), 1623–1630.
- Lee, S.C., Li, W.M., Ao, C.H., 2002. Investigation of indoor air quality at residential homes in Hong Kong—case study. *Atmospheric Environment* 36, 225–237.

- Lee, S.C., Kwok, N.H., Guo, H., Hung, W.T., 2003. The effect of wet film thickness on VOC emissions from a finishing varnish. *The Science of the Total Environment* 302, 75–84.
- Li, C.S., Lin, W.H., Jenq, F.T., 1993. Characterization of outdoor submicron particles and selected combustion sources of indoor particles. *Atmospheric Environment* 27B (4), 413–424.
- Liu, W.K., Sun, S.E., 1988. Ultrastructure changes of tracheal epithelium and alveolar macrophages of rats exposed to mosquito coil smoke. *Toxicology Letter* 41, 145–157.
- Liu, W.K., Wong, M.H., Mui, Y.L., 1987. Toxic effects of mosquito coil (a mosquito repellent) smoke on rats. *Toxicology Letter* 39, 223–230.
- Liu, W.L., Zhang, J.F., Jamal, H.H., Juliana, J., Zailina, H., Bernard, D.G., 2003. Mosquito coil emissions and health implications. *Environmental Health Perspectives* 111, 1454–1460.
- USEPA, 1998a. Compendium method TO-11, the determination of formaldehyde in ambient air using adsorbent cartridge. Quality Assurance Division, Environmental Monitoring Systems Laboratory, Research Triangle Park, North Carolina.
- USEPA, 1998b. Compendium Method TO-14, The Determination of Volatile Organic Compounds (VOCs) in Ambient Air using Summa Passivated Canister Sampling and Gas Chromatographic Analysis. Quality Assurance Division, Environmental Monitoring Systems Laboratory, Research Triangle Park, North Carolina.
- USEPA, 1999. Large Chamber Test Protocol for Measuring Emissions of VOCs and Aldehydes. Research Triangle Park, North Carolina.
- USEPA, 2001. Candles and incense as potential sources of indoor air pollution: market analysis and literature review. Prepared by National Risk Management, Research Triangle Park, USEPA-600/R-01-001.
- Zhang, J., Smith, K.R., Ma, Y., Ye, S., Jiang, F., Qi, W., Liu, P., Khalil, M.A.K., Rasmussen, R.A., Thorneloe, S.A., 2000. Greenhouse gases and other pollutants from household stoves in China: a database for emission factors. *Atmospheric Environment* 34, 4537–4549.