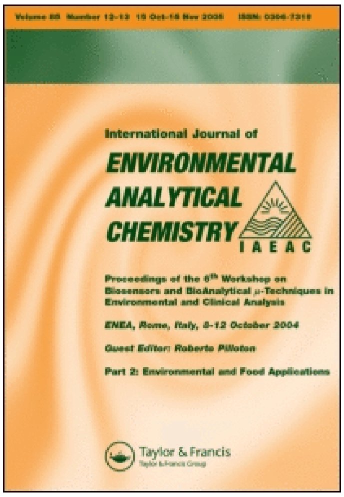

International Journal of Environmental Analytical Chemistry
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713640455

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Wan K. Jo[a]; Jong H. Lee[b]
[a] Department of Environmental Engineering, Kyungpook National University, Daegu 702-701, Korea [b] Department of Environmental Management, Kumho PetroChemical Co., Chun-Nam 555-280, S. Korea

To cite this Article Jo, Wan K. and Lee, Jong H.(2009) 'Determination of emission characteristics of volatile compounds from mosquito repellents (MRs) by headspace and chamber methods, and indoor levels related to use of MRs', International Journal of Environmental Analytical Chemistry, 89: 15, 1125 — 1137
To link to this Article: DOI: 10.1080/03067310902777934
URL: http://dx.doi.org/10.1080/03067310902777934

Intern. J. Environ. Anal. Chem.
Vol. 89, No. 15, 20 December 2009, 1125–1137

Determination of emission characteristics of volatile compounds from mosquito repellents (MRs) by headspace and chamber methods, and indoor levels related to use of MRs

Wan K. Jo[a]* and Jong H. Lee[b]

[a]*Department of Environmental Engineering, Kyungpook National University, 1370 Sankeok-dong, Buk-gu, Daegu 702-701, Korea;* [b]*Department of Environmental Management, Kumho PetroChemical Co., Yeosoo, Chun-Nam 555-280, S. Korea*

(*Received 30 September 2008; final version received 24 January 2009*)

Although the use of mosquito repellents (MRs) may lead to several adverse health effects, very limited information is available on exposure to their emissions. As such, the present study evaluated the emission characteristics of volatile pollutants from three different types of MRs (coil, mat, and liquid-vaporiser types) by headspace and/or chamber methods and measured apartment bedroom levels associated with the use of the MRs. By means of a headspace test, 18 to 22 compounds were tentatively identified from mat and liquid MRs. Six toxic/hazardous compounds identified in this headspace test were chosen as target compounds for a further chamber-emission test. An empirical equation was well fitted with time-series concentration levels in a small-scale chamber for all test MRs. Based on the chamber study, the coil MR exhibited the highest emission rate for the target compounds, with the exception of naphthalene, followed by liquid and mat MRs. It is noteworthy that naphthalene was emitted from the liquid MR only at a high emission rate ($2.4\,\mathrm{mg\,h^{-1}}$). A comparison of apartment bedroom levels with predicted values, which were estimated by using the calculated emission data and the one-compartment mass balance equation, suggested that predictions obtained from the chamber study were similar to, or larger than, the measured bedroom levels. Furthermore, the apartment bedroom measurements suggested that the use of coil MR could significantly elevate the indoor levels of four compounds (benzene, ethyl benzene, and m, p-xylene) by up to 1.8 to 1.9 times, while the use of liquid MR could significantly increase the indoor levels of naphthalene by up to 4.2 times. The current findings can provide valuable information for the estimation of the population inhalation exposure to these emissions in indoor environments, and enhance the selection of safer MRs.

Keywords: liquid mosquito repellent; mat mosquito repellent; coil mosquito repellent; apartment bedroom

1. Introduction

The use of mosquito repellents (MRs) helps reduce mosquito bites that may carry various diseases. When MRs are applied, insecticides evaporate, preventing mosquitoes from entering indoor environments. However, in contrast to any benefits, the use of MRs may

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*Corresponding author. Email: wkjo@knu.ac.kr

ISSN 0306–7319 print/ISSN 1029–0397 online

DOI: 10.1080/03067310902777934
http://www.informaworld.com

actually cause several adverse health effects. Certain coil-type MRs release a variety of toxic compounds such as volatile organic compounds (VOC), carbonyl compounds, fine particles, polycyclic aromatic hydrocarbons, and pyrethroids such as allethrin, deltamethrin, esbiothrin, prallethrin and transfluthrin [1–4]. These chemicals may be carcinogens, sensitisers, possible reproductive toxins, or neurotoxins [5,6]. A previous toxicological study has reported that rats exposed to mosquito coil smoke could induce focal deciliation of the tracheal epithelium, metaplasia of epithelial cells, and morphologic alteration of the alveolar macrophages [7]. Moreover, certain epidemiological studies have suggested that long-term exposure to mosquito coil smoke could cause asthma and persistent wheezing in children [8,9].

Many residents of Asia, South America, and Africa employ MRs in their homes or workplaces [2]. According to the marketing department of one of the largest MR manufacturers in South Korea, the four most popular types of MRs include electronic mats, liquid-vaporisers, coils, and liquid-sprays. The MR mat works by means of heating a mat saturated with MR. The MR liquid evaporates through a vaporiser or is sprayed into the air. Nevertheless, very limited information is available regarding exposure to emissions from these MRs. A few studies have reported on the emission characteristics or exposure levels of coil MRs [1–4]. However, to the best of the collective level at the time of the current study, this information is unavailable regarding liquid or mat MRs in any scientific literature.

Exposure data is needed in order to comprehensively link environmental exposure to health effects. Consequently, the present study evaluated the emissions of pollutants from three different types of MRs (coil, mat, and liquid-vaporiser types) that are currently available in Korea and measured the indoor exposure levels associated with the use of the MRs. This study focuses on volatile compounds, since these are expected to be emitted from all three types of MRs. Emission characteristics were determined by using a headspace analysis and a small-scale environmental chamber. The headspace measurement method was utilised regarding the semiquantitative determination of volatile components that are emitted from two types of MRs (mat and liquid types). However, the headspace method was not applied regarding the coil-type MR, since emissions from this MR are directly associated with burning. For six VOCs, which were chosen from the compounds detected in the headspace gas phase on the basis of selection criteria, a small environmental test chamber was used in order to estimate the emission rates of the mat and liquid types of MRs. This environmental chamber analysis was also applied for the characterisation of emissions from a coil-type MR. In addition, indoor levels regarding the selected compounds were measured in an apartment bedroom, while the MRs were utilised, in comparing them with those estimated based on the chamber study. These results can be employed as scientific exposure data regarding risk assessment associated with the use of MRs in many Asian, South American, and African countries.

2. Experimental

2.1 *Selection of MRs*

A marketing survey regarding the distribution of MRs was conducted in three of the largest supermarkets in Korea. There were 11 different brands of MRs found on the shelves of the supermarkets: liquid-vaporiser (four); mat (four); and coil (three). The term 'liquid' represents 'liquid-vaporiser'. Three products (two liquid MRs and one mat MR) were selected for the headspace test on the basis of sales figures for the previous year. One of the two liquid MRs, having higher emission levels as determined by means of a headspace test, was utilised for the chamber test, along with the mat MR used in the headspace test as well as one coil MR. All products were purchased from one of the three supermarket companies within one year of being manufactured.

2.2 *Headspace tests*

A headspace analysis was conducted in order to determine the existence of volatile chemical components emitted from the three selected liquid and mat MRs. This was accomplished by applying the method employed by the USEPA [10]. Sample materials occupied approximately 40% of the total volume of the clean 40 mL glass bottles fitted with Teflon-septum top screw-caps. The glass bottles were placed in a water bath at 60°C for 90 minutes in order to allow for the evolution of volatile organic pollutants, if any, from each of the materials. One ml of the gaseous sample was then drawn into a 10 mL pressure lock syringe and transferred to a gas chromatograph (HP 5890II) and mass spectrometer (HP MSD5973) (GC/MS) system. A 30 m fused silica capillary column (internal diameter 0.32 mm; film thickness 1 μm) (Agilent Technologies, HP-5) was used in order to separate the target analytes. The GC oven was initially programmed at 40°C for 5 minutes, subsequently increased at a rate of 5°C minute^{-1} up to 250°C, and held for 5 minutes. It is noteworthy that compounds were 'tentatively' identified by using a Wiley mass spectral library. A spectral search quality of 90% was employed for the criterion of the compound selections. Neither the compounds with a spectral search quality of less than 90%, nor those detected in insignificant amounts (chromatographic peak area $<10^4$), were included in the current paper. The peak area of 'total volatile organic compounds (TVOC)' (integrated chromatographic peak areas of analytes, converted to toluene-equivalent response), for analytes in the headspace gas phase of each product, was determined for this experiment. Although there exist substantial variations in the total-ion-current chromatogram response of VOCs with different chemical functionality, toluene can be used as the reference compound in regards to a semi-quantitative measurement of TVOC [11].

2.3 *Measurements of selected VOCs in a small-scale chamber and in an apartment bedroom*

2.3.1 *Small-scale chamber*

The concentrations of six VOCs, which were chosen based on compounds detected in the headspace gas phase, on the basis of abundance as well as their own toxic effects [5,6], were measured in an electropolished stainless steel (SS) chamber (40 × 25 × 50 cm^3) as well as in an apartment bedroom with or without using the three types of MRs (coil, liquid, and mat products). These selected compounds were known or suspected carcinogens, such as benzene where exposure can cause leukemia, and others have been associated with acute effects [5,6]. The chamber tests were performed by applying the procedures utilised in other studies [12–14]. The chamber top functions as an entry way, whereas sealing is accomplished by means of using a silicon gasket. A number of 1/4 in. air inlet and outlet holes were drilled through the SS walls for instrument connections and sampling ports.

The holes were plugged with Swagelok valves or Teflon tape. Clean air for the chamber was supplied from a zero-grade air cylinder. Air leakage from the chamber was determined by measuring the flow rate at the chamber outlet and comparing this with the supply airflow rate. The chamber temperature ranged between 20 and 23°C. The relative humidity (RH) was measured at between 20 and 26% regarding the coil and the mat MR tests, and between 22 and 46% in regards to the liquid MR test, just prior to the chamber inlet and outlet by using a humidity meter (Thermo Recorder TR-72S, T & D Co.). The air in the chamber was mixed by a metal fan (14 cm diameter). Accuracy of the sampling determined by the simultaneous collection of samples at two different ports was within the limits allowed for the measurement ratings (less than 15%). The outlet-flow rate was within 5% of the inlet-flow rate. Since the velocity near the surface of the test products can alter the mass transfer coefficient, products were tested for emissions under a typical indoor velocity (5–10 cm s^{-1}) [15]. The ventilation rate was 1 ± 0.05 ACH. This was computed by dividing flow rate (0.83 L min^{-1}) determined at the chamber outlet, via the digital bubble flow meter, by chamber volume (50 L). This ACH value was same as that suggested by CDHS [11]. The ACH value (±0.05) represents a standard deviation (precision) calculated as regards the outlet-flow rates, which were measured at the beginning and the end of an experimental procedure. The chamber does not incorporate any form of recirculation. Any background concentrations in the empty chamber were either undetected or trace amounts, considered to be insignificant for the purpose of this study.

Chamber tests started within 10 minutes after the full-size test products were placed in the chamber. Since full-size samples were tested regarding all three types of MRs, the amount of MR samples tested was not normalised in relation to the sample mass. Air samples were collected at average elapsed times of 5, 25, 45, 65, 85, 105, 180, 240, and 300 minutes after test initiation, by passing air through adsorbent tubes containing Tenax-TA. The tubes were connected to calibrated constant-flow pumps for 10 min sampling periods. The sampling pump was calibrated by means of a digital flow meter, prior to and following the collection of each sample. The average of these two rates was used as the sample-flow rate for all volume calculations. The flow rate was nominally adjusted to 250 mL min^{-1}. The sampling flow rate did not exceed 50% of the inlet flow rate (as suggested by the CDHS [11]).

2.3.2 *Time dependency of concentration*

Initially, the general mass transfer governing equation for chamber air VOC concentration was used as follows [16–19]:

$$V(\mathrm{d}C_t/\mathrm{d}t) = QC_{in} - D \times A(\mathrm{d}C/\mathrm{d}X) - QC_{out} \tag{1}$$

C_t represents the concentration as a function of time (t, min), C_{in} and C_{out} represent the inlet and outlet concentrations, respectively, C represents VOC concentration in MRs, Q represents volumetric flow rate ($m^3\,s^{-1}$), D represents VOC diffusion coefficient in MRs ($m^2\,s^{-1}$), A represents the surface area of MRs (m^2), and X represents the direction of VOC diffusion in MRs. Following key assumptions were made to simplify the analysis: (1) Unlike previous studies [16–19], the present study supplied heat by electricity as regards the mat and liquid MRs or by burning coil-type MRs during the course of the experiment. As such, it is assumed that heat was constantly supplied to the MRs, (2) the experiment was stopped before the MRs were used up. As such, it is assumed that the VOC was continuously generated during the course of the experiment, and that the VOC generation rate in the chamber was constant, and (3) adsorption is insignificant on the surface of chamber constructed by smooth and chemically-inert SS. Accordingly, the solution of Equation (1) is simplified as follows:

$$V(\mathrm{d}C_t/\mathrm{d}t) = -QC_t + G \tag{2}$$

where G represents the constant generation rate (mg min^{-1}). The solution of Equation (2) is as follows:

$$C_t = A(1 - \mathrm{e}^{-Bt}) \tag{3}$$

where C_t represents the concentration as a function of time (t, min), A $(=G/Q)$ represents the linear parameter (mg m^{-3}), and B $(=Q/V)$ represents the rate parameter (min^{-1}). This simple empirical equation was tried to describe the time dependency of concentrations in the chamber.

2.3.3 *Calculation of emission rates*

Assuming no net loss of target compounds from air due to other effects such as adsorption on the inner surface of the chamber or chemical reactions, the emission rates were calculated based on the observed concentrations by utilising the following equation:

$$C_{\mathrm{eq}} - C_{\mathrm{o}} = S/Va \tag{4}$$

where C_{eq} represents the equilibrium concentration (mg m^{-3}), C_{o} represents the background concentration in the chamber, S represents the source emission rate (mg h^{-1}), V represents the volume of the chamber (m^3) and a represents ACH (h^{-1}). In the present study, C_{eq}, which represents the quasi-steady-state concentration, was calculated using Equation (3). Since the background concentrations in the empty chamber were either undetected or were trace amounts, C_{o} was ignored in the process of making calculations. The chamber volume employed for the calculations was 0.05 m^3.

2.3.4 *Apartment bedroom*

Indoor levels of the six selected compounds were measured during the summer season in an apartment bedroom with a volume of 35 m^3, equipped with a bedroom air conditioner (AC) attached to a wall, with or without using the three different MRs. Mothballs and other consumer products were removed from the bedroom in order to avoid potential confounding factors. Bedroom furniture included a single bed and a wardrobe. The floor was composed of linoleum. At the beginning of all experiments, the bedroom windows and doors were left open for a minimum of one hour in order to equilibrate the interior levels to the ambient levels. In order to attain a worse exposure condition, the windows and doors were closed, the AC was switched on, and the temperature level was set to 20°C. With or without placing a MR in the centre of the floor, a 8 hr bedroom air sample was collected by using a Tenax adsorbent tube at a breathing zone close to the bed. Generally, one coil-type MR burns slowly and lasts 8 or more hours, one mat-type MR is utilised for a night without changing to a fresh one, and one liquid-type MR can be utilised for several nights. As such, the air sampling time was set to 8 hr. Prior to, and after measuring the indoor VOC levels, the air exchange rate was estimated by means of the tracer CO_2 gas decay method [20]. During the VOC or CO_2 measurement period, no occupant remained in the bedroom. This entire experimental procedure regarding each MR was repeated three times.

2.3.5 *Sampling and analysis*

The VOCs collected on the Tenax TA trap were analysed by means of coupling a thermal desorption system (TDS, Tekmar Model Aerotrap 6000) to a GC (Hewlett Packard 6890) with a flame-ionisation detector. A 60 m fused silica capillary column (internal diameter 0.32 mm; film thickness 1 μm) (Supelco Co. SPB-5) was used to separate the target analytes. The adsorbent trap was thermally desorbed at 250°C for 10 min, and the target compounds were cryofocused at −120°C in a cryo trap (15.2 cm-length, 0.32 cm-o.d. tube packed with glass beads). The cold trap was rapidly heated to 250°C, then the contents were flushed into the Cryofocusing Module (CM) of the TDS and cooled to −120°C in order to refocus the target compounds. The CM was then heated to 225°C and flushed in order to transfer the target compounds to the GC. The initial oven temperature was set at 35°C for five min and ramped at 4°C min^{-1} to 200°C for five min.

2.3.5 *Quality control programme*

The quality control programme regarding the measurements included laboratory and field blank Tenax traps and spiked samples. At the beginning of the day, the laboratory and field blank Tenax traps were analysed in order to check for any contamination during the sampling and analytical procedures. In order to check the quantitative response, a known standard was directly injected into the trap and the target compounds were transferred to the GC. When the quantitative response differed by more than ±15% from that predicted by means of a specified calibration equation, a new calibration equation was determined. The precisions and method detection limits of the target compounds, determined by seven repeated analyses of one of the calibration standards, were within 15% and ranged from 0.5 to 2.3 $\mu g\,m^{-3}$, respectively. Meanwhile, since m-xylene and p-xylene were co-eluted, the combined concentrations of the two compounds were reported.

3. Results and discussion

3.1 *Emissions composition*

Table 1 outlines the chemical composition and the peak areas of TVOCs, which were determined from the headspace gas phase of the three different MRs. This table includes the compounds that satisfied the cut-off point regarding reporting compounds, as described earlier. Respectively, 18, 22, and 22 compounds were tentatively identified in a liquid MR (Liq_1), another liquid MR (Liq_2), and a mat MR (Mat). Although *N*,*N*-diethyl-3 methylbenzamide (DEET), picaridin, and pyrethrins are commonly used in MRs [2,21], these compounds were not detectable in any samples, most likely due to their low vapour pressure. Benzene, toluene, ethyl benzene, and m,p-xylene (BTEX), which were identified in all three products, are listed as toxic air contaminants, hazardous air pollutants, or are noted on California's Proposition 65 list [6].

Table 1. Abundance of constituents of the emissions from three different type of MR, as determined by means of a headspace analysis, according to the type of MR[a].

	% of Peak area		
Compound	Liq_1	Liq_2	Mat
Benzene	1.7	1.8	0.6
Bis(trimethylsilyl)acetylene	8	18	9.0
Carbon tetrachloride	ND	ND	0.1
Chlorobenzene	ND	ND	0.1
Chloroform	0.1	0.2	ND
Cyclohexane	ND	ND	2.0
Decadien	ND	3.0	ND
Decanal	3.2	4.0	3.6
Decane	ND	3.1	3.2
Dodecane	5.4	2.7	32
Ethyl benzene	12	5.1	0.1
4-Ethyltoluene	0.1	0.1	0.1
Heptadecane	ND	ND	2.3
Hexadecane	ND	14	20
Hexamethylcyclotrisiloxane	6.6	2.0	ND
Isobornyl thiocyanoaetate	ND	ND	9.5
Limonene	4.4	ND	6.5
2-Methyltetrahydrofuran	ND	6.6	ND
Naphthalene	31	11	ND
Nonanal	ND	3.7	ND
Pentadecane	5.1	6.2	3.6
Pentan-1,3-dioldiisobutyrat	5.1	3.4	2.5
Styrene	0.1	0.1	0.1
Γ-Terpinene	4.0	ND	ND
Tetramethylsilane	ND	2.6	ND
Toluene	4.7	5.0	2.2
1,3,5-Trimethylbenzene	0.1	0.1	ND
1,2,4-Trimethylbenzene	0.1	0.1	0.2
Undecane	ND	ND	2.1
M,p-Xylene	8.3	7.2	0.2
TVOCs (peak area)	1,740,856	532,332	248,781

[a] ND: not detected.

Although the peak areas of individual VOC or TVOCs may not be an absolute tool for emissions comparison purposes, they were represented in order to tentatively compare the emission rates of different MRs. On the basis of the peak area of TVOCs, Liq_1 exhibited the highest emissions of volatile compounds, followed by Liq_2 and Mat. Meanwhile, on the basis of the peak area, naphthalene had the highest emission rate among the volatile pollutants detected from Liq_1 (31%) and the third highest for Liq_2 (11%), whereas it was not detected in the mat MR. Since the inhalation of naphthalene has caused cancer in animals through testing, this compound has been classified as being possibly carcinogenic to humans [5,22]. Unfortunately, there is no comparative report regarding present findings available for emissions from liquid MRs. Six toxic/hazardous compounds (BTEX and naphthalene) identified in this headspace test were chosen as target compounds for a further chamber-emission test.

3.2 *Emission concentrations and rates*

Chamber tests were conducted in order to quantitatively characterise the emissions of six volatile compounds, chosen from compounds detected in the headspace gas phase on the basis of selection criteria, from the three different types of MR. Concentration-time profiles were developed after the introduction of the sample products into the chamber. Figure 1 illustrates the measured and predicted (dotted curve) time-series concentrations of

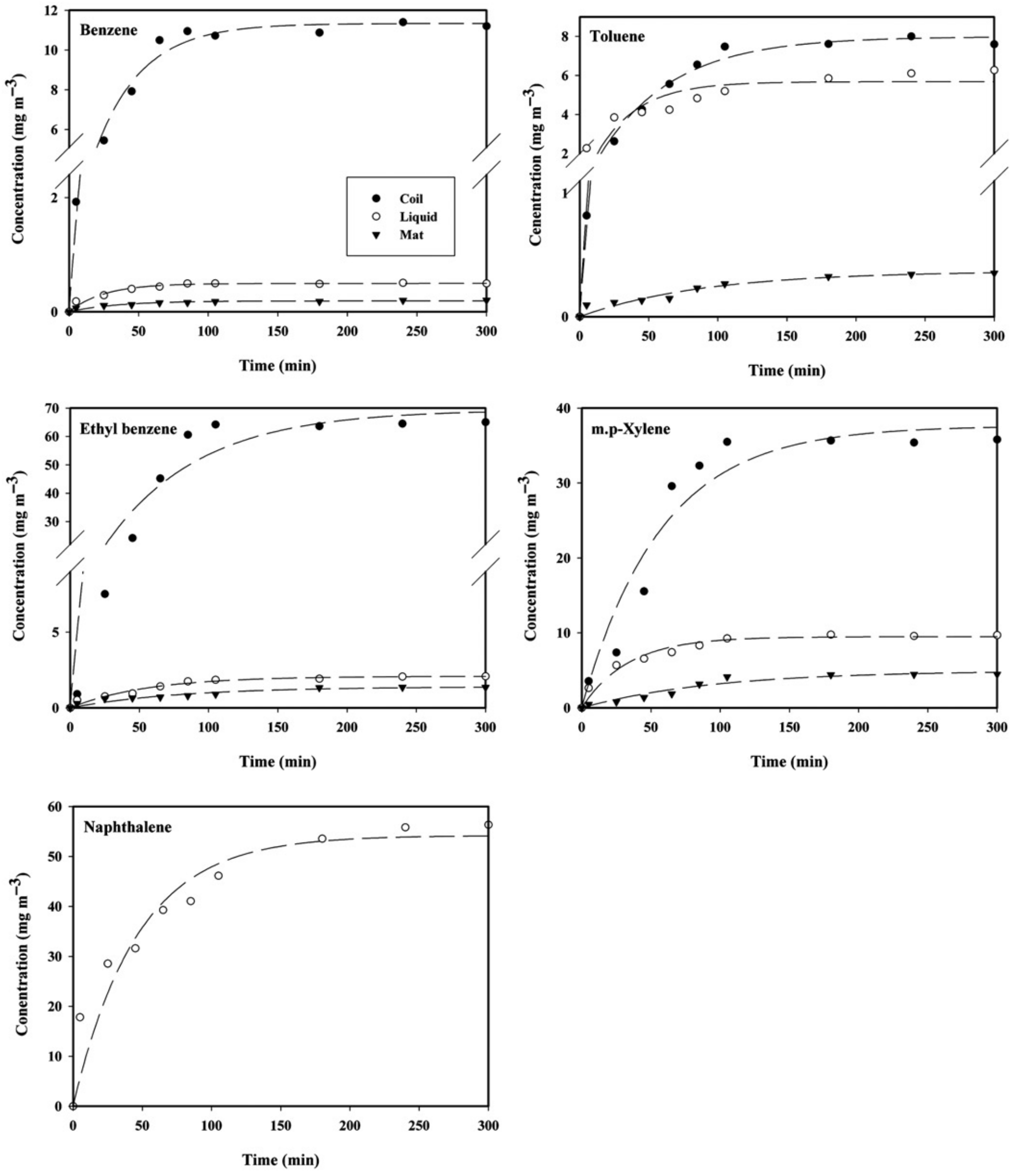

Figure 1. Time-series concentrations of six volatile pollutants emitted from three different types of MR. The dotted curves represent the empirical equation.

six target compounds, emitted from three MRs, under the conditions of ACH = 1. Most quasi-steady-state conditions were obtained between 85 and 105 minutes after the initiation of the chamber tests. The quasi-steady-state concentration levels varied according to the type of MRs or compounds. The liquid MR emitted all six target VOCs, whereas the coil and the mat MRs emitted five VOCs, with the exception of naphthalene. As presented in Table 2, most correlation coefficients (R^2) are close to the maximum possible value of unity. This suggests that the exponential, empirical model was well fitted with the time-series concentrations in the chamber, and that the empirical model was appropriate for testing emission purposes.

The emission rates of the six target VOCs, obtained from three products under the conditions of ACH = 1, are presented in Table 2. Since full-size samples were tested, a normalisation to sample mass was unnecessary for all three types of MR. As with respect to the emission concentrations, the coil MR exhibited the highest emission rate for all target compounds, with the exception of naphthalene, followed by the liquid MR (Liq_1) and mat MR. For example, the benzene emission rate for coil MR was 54 times higher than that of the mat MR and 18 times higher than that of the liquid MR. These emission rates are comparable to those of other indoor sources [23]. The benzene emission rate was between 0.01 and 0.54 mg h^{-1} regarding MRs, while it was 0.93 mg h^{-1} regarding the use of washer/drier. The toluene emission rate was between 0.01 and 0.35 mg h^{-1} regarding MRs, and it was 1.51 mg h^{-1} regarding closet storage chemicals. The ethyl benzene emission rate was between 0.05 and 2.3 mg h^{-1} regarding MRs, while it was 0.302 mg h^{-1} regarding glue. The xylene emission rate was between 0.16 and 1.7 mg h^{-1} regarding MRs, and it was 0.897 mg h^{-1} regarding glue. In regards to the current study, naphthalene was only emitted from the liquid MR at a high emission rate (2.4 mg h^{-1}).

Table 2. Empirical equation parameters and emission rates for six target compounds determined under the conditions of ACH = 1, according to the type of MR[a].

Compound	MR type	R^2	A, linear parameter ($mg\,m^{-3}$)	B, rate parameter (min^{-1})	Emission rate ($mg\,h^{-1}$)
Benzene	Coil	0.98	11	0.03	0.54
	Liq_1	0.96	0.5	0.04	0.03
	Mat	0.96	0.19	0.03	0.01
Toluene	Coil	0.99	8.0	0.02	0.35
	Liq_1	0.88	5.7	0.03	0.27
	Mat	0.93	3.7	0.01	0.01
Ethyl benzene	Coil	0.92	69	0.01	2.3
	Liquid	0.96	2.1	0.02	0.09
	Mat	0.93	1.4	0.01	0.05
M,p-Xylene	Coil	0.94	37	0.02	1.7
	Liq_1	0.97	9.5	0.03	0.45
	Mat	0.95	4.9	0.01	0.16
Naphthalene	Coil	NA	NA	NA	NA
	Liq_1	0.92	54	0.02	2.4
	Mat	NA	NA	NA	NA

[a] NA: not applicable.

This emission rate was much higher than that associated with the storage of mothballs indoors (0.675 mg h^{-1}) [23].

3.3 *VOC levels in apartment bedroom*

Potential indoor levels estimated by using the calculated emission data and one-compartment mass balance equation (Equation 4) were compared with measured levels in the apartment bedroom. The mean indoor VOC levels measured without placing any MR in the apartment bedroom were employed as C_o. The mean ACH value (0.75 h^{-1}) was used for a. The V was 35 m^3. Some predictions were in agreement with the measured values, whereas others were not (Table 3). Generally, the estimated concentrations were similar to, or larger than, the measured concentrations. In regards to benzene from the liquid and mat MRs, as well as toluene from all MRs, the ratios of estimated and measured values were close to 1. In contrast, regarding benzene, ethylbenzene, and m,p-xylene from coil MR, the ratios were 2.5, 9.4, and 2.8, respectively. In addition, the ratio for naphthalene from liquid MR was 12. This discrepancy might have been due to the combined effects of several parameters, such as sink effects and mixing efficiency. Sink effects would result in an underestimation of the predictions, while the other factor (mixing efficiency) would result in overestimation. The emission rates were determined by using an electropolished SS chamber, in order to minimise the influence of adsorption into the chamber walls. However, in actual apartment bedrooms, VOCs emitted from MRs can be adsorbed onto the surface of furniture such as beds and closets. In addition, building

Table 3. Indoor concentrations (μg m^{-3}) measured in an apartment bedroom and estimated from chamber study according to the type of MR[a].

Compound	MR type	Measured[b]	Estimated	Ratio[c]
Benzene	Coil	11	27	2.5
	Liquid	6.6	6.5	1.0
	Mat	6.3	6.9	1.1
Toluene	Coil	41	49	1.2
	Liquid	32	44	1.4
	Mat	34	31	0.9
Ethyl benezene	Coil	9.9	93	9.4
	Liquid	5.4	8.1	1.5
	Mat	5.1	6.4	1.3
m,p-Xylene	Coil	29	80	2.8
	Liquid	16	30	1.9
	Mat	12	20	1.6
Naphthalene	Coil	1.7	NA	NA
	Liquid	7.6	93	12
	Mat	1.5	NA	NA

[a] NA: not applicable.
[b] Mean concentrations measured while a MR was employed.
[c] Ratio of estimated concentrations to measured concentrations.

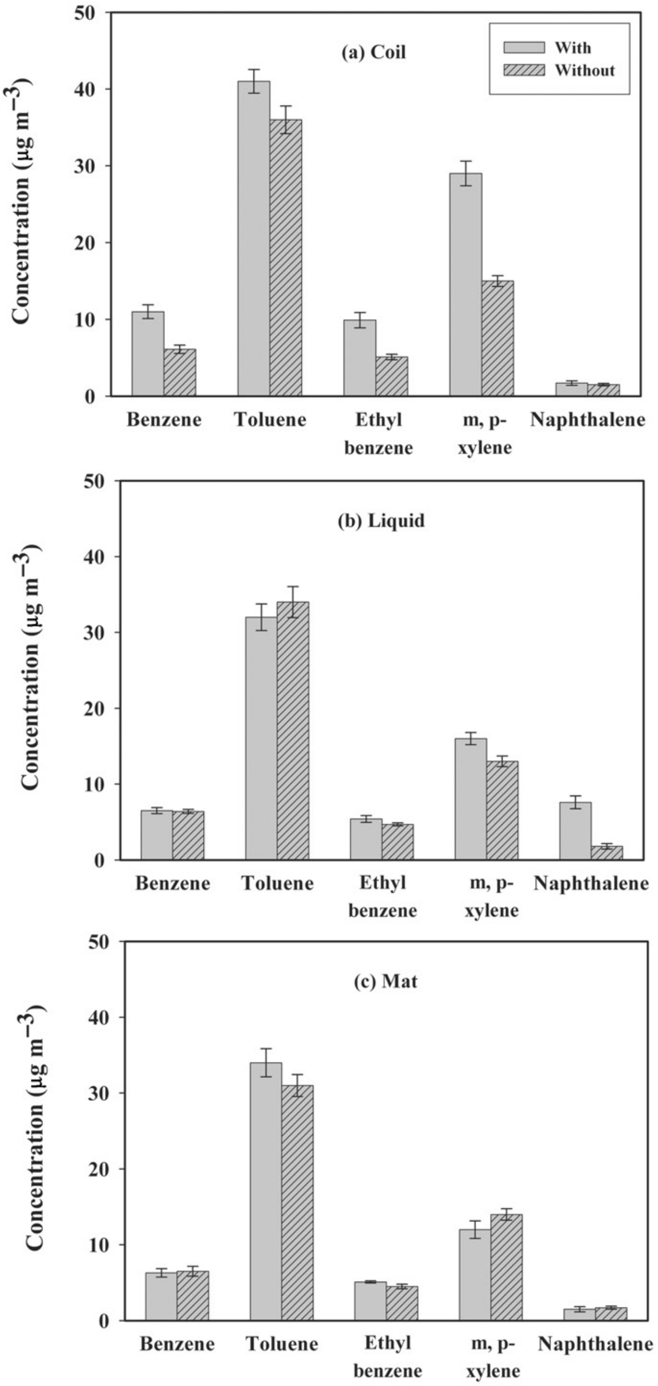

Figure 2. Mean concentrations (μg m^{-3}) and standard errors of selected compounds measured in an apartment bedroom with or without three types of MRs: (a) coil-type, (b) liquid-type, and (c) mat-type MRs. Mean ACHs and standard deviation (h^{-1}): coil type-with, 0.9 (0.3); coil type-without, 0.8 (0.5); liquid type-with, 0.7 (0.5); liquid type-without, 0.9 (0.4); mat type-with, 0.7 (0.4); and mat type-without, 0.5 (0.3).

finishing materials such as walls, floors and ceilings can also adsorb VOCs [24–26]. Indoor chemistry would be insignificant as regards the discrepancy of measured and estimated data for BTEX, since these compounds are inert [27]. Meanwhile, one assumption was made for the predictions that pollutants emitted from MRs are well mixed quickly in bedrooms. In fact, the air in the chamber was mixed by means of a metal fan, thereby causing homogeneous pollutant concentration, whereas the air in the apartment bedroom was not mixed with any fan. As such, the bedroom air could not be completely mixed. This could result in an underestimation of the predicted values, since actual bedroom measurements were conducted near the MRs. Consequently, it was suggested that, in regards to the ratios of predicted values to measured bedroom levels that were greater than 1, the effects of surface adsorption would outweigh the effect of mixing efficiency. As regard the ratios that were close to 1 (e.g. toluene for mat-type MR), the reason is unclear. Consequently, further studies are recommended in order to verify this assertion, since all the necessary information for assumptions made in the present study was not collected.

Figure 2 illustrates the comparison of the indoor levels of six target compounds measured in an apartment bedroom, with and without MRs. Analysis of variance test performed with the criterion of $p < 0.05$ suggested that the use of coil MRs could significantly elevate the indoor levels of four compounds (benzene, ethyl benzene, and m, p-xylene) by up to 1.8 to 1.9 times, while the use of liquid MRs could significantly increase the indoor levels of naphthalene by up to 4.2 times. However, the indoor levels of toluene regarding the coil MR, BTEX regarding the liquid MR, and all target compounds regarding the mat MR, which exhibited relatively low emission rates, were not significantly different between those with, and without using, these MRs. This result suggests that although those compounds were emitted from MRs, their respective emission strengths were not strong enough to significantly elevate indoor levels. Consequently, it is further suggested that adverse health effects reported by previous toxicological and epidemiological studies associated with the use of MRs [7–9] was less linked to VOCs as compared to other MR emission compositions. Meanwhile, the ventilation rates would not influence the difference of the indoor levels between the two measurement conditions, since the ventilation rates measured when the MRs were used (between 0.7 and $0.9\,h^{-1}$) were similar to those measured when no MRs were used (between 0.5 and $0.9\,h^{-1}$) (Figure 2). The present findings will assist consumers in selecting safer MR products, along with the knowledge of the effect of MRs in preventing mosquito bites.

Acknowledgements

We would like to thank the reviewers for their thoughtful corrections and valuable suggestions. This work was supported by the Korea Science and Engineering Foundation (KOSEF) grant funded by the Korea government (MOST) (No. R01-2006-000-10851-0).

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