

Assessment of Organic Compounds Emission from Consumer Products in an Environmental Chamber System

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Abstract

In order to present the scientific information of organic emissions from consumer products available, the current study examined the emissions composition for three different types of consumer products being used in Korea in an environmental chamber: air freshener (AF), mosquito repellents (MSQR), and moth repellents (MTHR). Emission rates were evaluated by determining emission rates of the target compound from the selected products using an electropolished stainless steel (SS) test chamber ($40 \times 25 \times 50 \text{ cm}^3$). A time-dependent empirical relationship developed in this study agreed well with the test results. As same with the emission concentrations, MSQR exhibited the highest emission rate for all target compounds except for limonene and naphthalene. MTHR ($9,200 \mu\text{g h}^{-1}$) showed the highest emission rate of naphthalene followed by MSQR ($8,300 \mu\text{g h}^{-1}$). Moreover, the concentrations in residential bedroom conditions for target compounds emitted from three types of consumer products were estimated. This estimation suggests that the uses of consumer products can elevate indoor levels of target compounds. In particular, any types of the consumer products may increase the indoor level of m, p-xylene.

Key words : Air freshener, Mosquito repellent, Moth repellent, Empirical relationship, Emission rate

1. INTRODUCTION

Even though consumer products are employed to provide perceived benefits of human life such as promotion of hygiene and aesthetics (Nilsen *et al.*, 2002; Rusin *et al.*, 1998), the emissions composition from them has potentially been associated with health risks for building inhabitants. Previous studies have implicated these consumer products as sources of indoor air pollutants (Su *et al.*, 2007; Singer *et*

al., 2006; CDHS, 2004). These compounds have been shown to cause symptoms similar to those forming the Sick Building Syndrome, a group of symptoms including sleepiness, irritability, inability to concentrate, and other health hazards (Grande *et al.*, 2007; ATSDR, 2005; OEHHA, 2003). A major cause of these health effects for building occupants is the inhalation exposure to consumer-product constituents and the secondary pollutants produced by the reaction of unsaturated organic constituents with oxidants (Nazaroff and Weschler, 2004; Atkinson and Arey, 2003; Finlayson-Pitts and Pitts Jr., 2000). The inhalation exposure of indoor air pollutants associated with the use of the consumer products

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include the inhalation of nonvolatile components as well as volatile components (Nazaroff and Weschler, 2004). However, the present study focused on the organic compounds emitted directly from consumer products.

The chemical composition emitted from the products has been surveyed by several research groups (Habib *et al.*, 2006; Liu *et al.*, 2004; Wainman *et al.*, 2000), but investigations have focused on Western Europe and the USA. Very limited information is available about the emissions composition for the consumer products available in other countries. Chemical components and their proportions emitted from consumer products are suspected to be different among manufacturers in different countries.

Three types of consumer products are widely employed in homes: air fresheners (AFs), mosquito repellents (MSQRs), and moth repellents (MTHR). The major commercial use of naphthalene is in the manufacture of polyvinyl chloride plastics (ATSDR, 2005). Its major consumer use is in moth repellents (MTHR). Air fresheners (AFs) are widely used in homes and non-industrial work places to supply pleasant odors and to counteract offensive odors emitted from a variety of indoor sources (EHANS, 2004). However, AFs emit significant amounts of variety compounds when applied indoors (Singer *et al.*, 2006; Zhu *et al.*, 2001; Salthammer, 1999). MSQRs release a variety of other toxic compounds such as carbonyl compounds, fine particles, and polycyclic aromatic hydrocarbons (Lee and Wang, 2006; Liu *et al.*, 2003; Liu and Sun, 1998). Consequently, the current study examined the emissions composition for three different types of consumer products (AF, MSQR, and MTHR) sold in Korea, in order to provide scientific information about the emissions composition. This examination was established using an environmental chamber.

2. METHODS

A compact group of six target compounds were selected for chamber tests, and are presented in Table 1. The target compounds were selected on the basis

Table 1. Method detection limits for target compounds selected for chamber test.

Compounds	Detection limit ($\mu\text{g m}^{-3}$)
Benzene	0.8
Ethylbenzene	1.2
Limonene	0.2
Naphthalene	1.1
Toluene	0.5
m, p-Xylene	2.3

Table 2. Information on three consumer products.

Product	Phase	Full-size capacity	Odor
AF	Gel	210 g	Jasmine
MSQR	Liquid	45 mL	—
MTHR	Solid	27 g	—

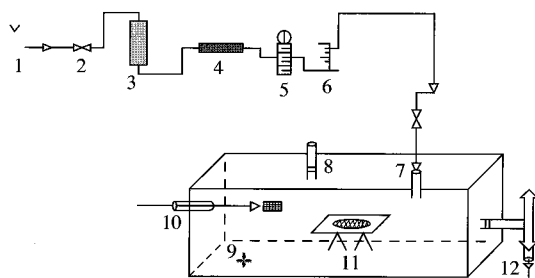


Fig. 1. Schematic diagram of experimental set-up. 1) air cylinder; 2) valve; 3) active charcoal; 4) fine filter; 5) mass flow controller; 6) flow controller (dry air); 7) inlet port; 8) sampling port; 9) fan; 10) temperature and RH monitoring; 11) test specimen and sample; and 12) sampling port.

of their own toxic effects (OEHHA, 2003; USEPA, 2003; IARC, 2002). Three products were chosen from each of three different types of consumer (AF, MSQR, and MTHR) for the chamber tests (Table 2). An electropolished stainless steel (SS) test chamber ($40 \times 25 \times 50 \text{ cm}^3$) was employed to determine emission rates of the target compound from the selected products (Fig. 1). The top of the chamber acts as a door, the sealing being made with a silicon gasket. A number of 1/4 in. holes were drilled through the SS walls for air inlet and outlet, instrument connections and sampling ports. The holes were

stoppered with Swagelok valves or Teflon tape. The clean air for the chamber is supplied from a zero-grade air cylinder. The air leakage of the chamber was determined by measuring the flow rate at the chamber outlet and comparing this to supply airflow rate. The chamber temperature ranged between 19 and 24°C. Relative humidity (RH) was measured between 20 and 26% for the tests of MOSR and MOTR and between 19 and 54% for AFs just prior to the chamber inlet and outlet using a humidity meter (Thermo Recorder TR-72S, T & D Co.). It is noted that since temperature and RH are important parameters for emission of volatile compounds (Colombo *et al.*, 1990), the variation of these parameters may influence the emission characteristics during the course of experiments. The air in the chamber was mixed by a metal fan (14-cm diameter). Homogeneity within the chamber was tested by the simultaneous collection of samples at two different ports (numbers 8 and 12 in Fig. 1). The dispersion of the results obtained from three trials was within the precision of the measurements (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 affect the mass transfer coefficient, the products were tested for emissions under a typical indoor velocity (5–10 cm s⁻¹) (ASTM, 1998). The ventilation rate was 1 ± 0.05 ACH. This was tested by means of a digital bubble flow meter and the amount is comparable to that suggested by the ASTM (1998). The background concentrations in the empty chamber were either undetected or just trace amounts.

Chamber tests started within 10 minutes after the placement of the full-size test products in the chamber (Fig. 1). The products were half-opened. Chamber-air samples were collected at average elapsed times of 5, 25, 45, 65, 85, 105, 180, 240, and 300 minutes after initiating the test. Air samples were collected by passing air through adsorbent tubes, containing Tenax-TA (Lu *et al.*, 2005; Reisen and Arey, 2005). The tubes are connected to calibrated constant flow pumps for 10-min sampling periods. The sampling pump was calibrated by 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 in all volume calculations. The sample-flow rate was nominally adjusted to 150 to 350 cm³/min, depending on the ventilation rates. The sampling flow rate did not exceed 50% of the inlet flow rate, as suggested by the ASTM (1998).

The compounds collected on the Tenax TA trap were analyzed by coupling a thermal desorption system (TDS, Tekmar Model Aerotrap 6000) to a GC (Hewlett Packard 6890) with a flame ionization detector. A 60-m long fused silica capillary column (internal diameter 0.32 mm; film thickness 1 µm) (Supelco Co. SPB-5) was used for the separation of the target analytes. The GC oven was programmed from 35°C to 220°C at 4°C min⁻¹. The adsorbent trap was thermally desorbed at 250°C for 10 min, and the target compounds cryofocused at -120°C on a cryo trap (15.2-cm-long, 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 to refocus the target compounds. The CM was then heated to 225°C and flushed 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.

The quality control program for 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 analyzed to check for any contamination during the sampling and analytical procedures. To check the quantitative response, a known standard was directly injected into a trap and the target compounds transferred to the GC. When the quantitative response differed by more than ± 15% from that predicted by 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 µg m⁻³ (Table 1), respectively. Meanwhile, since m-xylene and p-xylene were coeluted, the summed concentrations of the two compounds were reported.

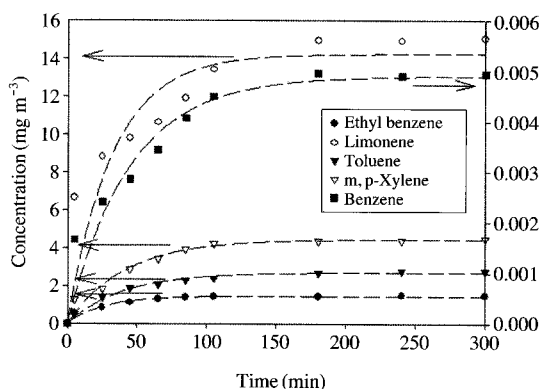


Fig. 2. Time-series concentrations of six compounds emitted from air freshener. The dotted curves represent the empirical relationship.

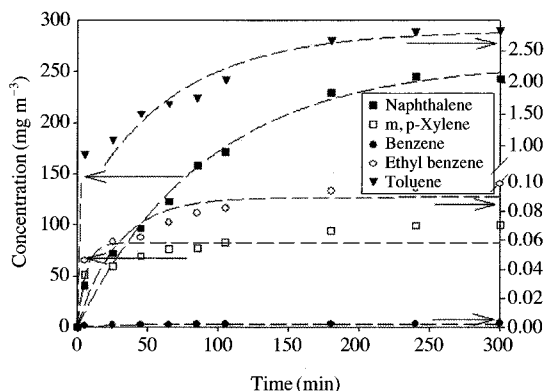


Fig. 4. Time-series concentrations of six compounds emitted from moth repellent. The dotted curves represent the empirical relationship.

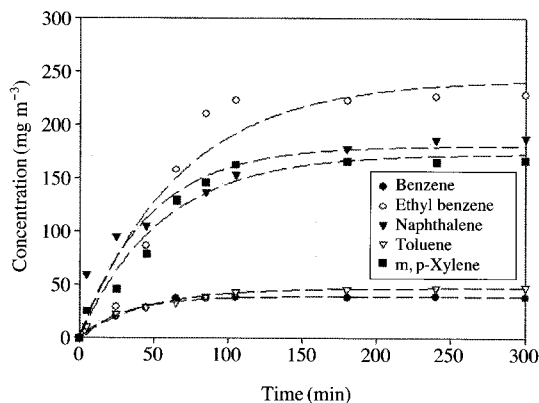


Fig. 3. Time-series concentrations of six compounds emitted from mosquito repellent. The dotted curves represent the empirical relationship.

3. RESULTS AND DISCUSSION

3.1 Concentration-time profile

The time-series emission concentrations were measured after the introduction of the sample products into the chamber, once for each product. The following empirical equation was used to describe the time dependence of concentrations in the chamber:

$$C_t = A(1 - e^{-Bt}) \quad (1)$$

where C_t is the concentration as a function of time (t , min), A is the linear parameter (mg m^{-3}), and B is the rate parameter (min^{-1}). Figs. 2 through 4 present the measured and the predicted (dotted curve) time-series concentrations of the target compounds emitted from the test products. The difference between the measured and the predicted values would be due to unidentified experimental errors. All three types of consumer products were found to emit benzene, ethylbenzene, toluene, and *m, p*-xylene. These compounds were also emitted from various consumer products employed in other countries (Lee and Wang, 2006; Zhu *et al.*, 2001). Limonene was emitted from AF only, while naphthalene was emitted from MSQR and MTHR. The empirical relationship parameters for these figures are presented in Table 3. Most quasi-steady-state conditions were achieved between 100 and 150 minutes after initiating the chamber tests. To determine quasi-steady-state emission conditions, the procedure included the determination of the time profiles of the naphthalene concentration, which were developed after the introduction of the sample products into the chamber. The quasi-steady-state concentration levels varied according to the type of products or compounds. For AF, correlation coefficients (R^2) were between 0.85 (for limonene) and 0.99 (for toluene). The linear parameter, A , ranged from $5.0 \mu\text{g m}^{-3}$ (for benzene) to $14,000 \mu\text{g m}^{-3}$ (for limonene), while

Table 3. Empirical relationship parameters and emission rates for target compounds.

Compound	Product type	R ²	A, linear parameter (µg m ⁻³)	B, rate parameter (min ⁻¹)	Emission rate (µg h ⁻¹)
Benzene	AF	0.93	5.0	0.002	0.055
	MSQR	0.99	39000	0.03	1900
	MTHR	0.84	2.0	0.03	0.098
Ethylbenzene	AF	0.95	1500	0.004	26
	MSQR	0.93	243000	0.02	11000
	MTHR	0.82	90	0.007	2.6
Limonene	AF	0.85	14000	0.03	680
	MSQR	NA	NA	NA	NA
	MTHR	NA	NA	NA	NA
Naphthalene	AF	NA	NA	NA	NA
	MSQR	0.92	181000	0.02	8300
	MTHR	0.98	259000	0.01	9200
Toluene	AF	0.99	2700	0.003	42
	MSQR	0.98	47000	0.02	2200
	MTHR	0.92	2800	0.02	130
m, p-Xylene	AF	0.97	4500	0.002	5000
	MSQR	0.97	173000	0.02	7900
	MTHR	0.82	83000	0.2	4200

Note. NA, not available; The criterion for significance in the values of two parameters (A and B) was $p < 0.05$; The statistical analyses were performed using the SigmaPlot program (Version 8.0) on a personal computer.

the rate parameter, B, from 0.002 min^{-1} (for benzene and m, p-xylene) to 0.03 min^{-1} (for limonene). For MSQR, R² were between 0.92 (for naphthalene) and 0.99 (for benzene). A values ranged from $39,000 \text{ µg m}^{-3}$ (for benzene) to $243,000 \text{ µg m}^{-3}$ (for ethylbenzene), while for B values from 0.02 min^{-1} (for ethylbenzene, naphthalene, toluene, and m, p-xylene) to 0.03 min^{-1} (for benzene). For MTHR, R² were between 0.82 (for ethylbenzene and m, p-xylene) and 0.98 (for naphthalene). A values ranged from 2.0 µg m^{-3} (for benzene) to $259,000 \text{ µg m}^{-3}$ (for naphthalene), while for B values from 0.007 min^{-1} (for ethylbenzene) to 0.2 min^{-1} (for ethylbenzene). A values are closely related to the quasi-steady state concentrations. The high values of R² suggest that the exponential, empirical relationship fitted well with the time-series concentrations in the chamber, and then that the empirical relationship was appropriate for testing emissions.

3.2 Emission rates of compounds

Assuming perfect mixing within the chamber and no net loss of target compounds from air due to

other effects such as adsorption on the inner surface of the chamber and chemical reactions, the emission rates were calculated from the observed concentrations by employing the following equation (Wallace *et al.*, 1987):

$$C_{\text{eq}} - C_o = S/Va \quad (2)$$

where C_{eq} is the equilibrium concentration (µg m^{-3}), C_o is the background concentration in the chamber, S is the source emission rate (µg h^{-1}), V is the volume of the chamber (m^3) and a is the ACH (h^{-1}). In the present study, C_{eq} , which is the quasi-steady-state concentration, was calculated using Eq. (1). Since the background concentrations in the empty chamber were either undetected or trace amounts, C_o was ignored. The V was 0.05 m^3 . The emission rates of six target compounds obtained from three types of products are presented in Table 3. As same with the emission concentrations, MSQR exhibited the highest emission rate for all target compounds except for limonene and naphthalene. MTHR ($9,200 \text{ µg h}^{-1}$) showed the highest emission rate of naphthalene followed by MSQR ($8,300 \text{ µg h}^{-1}$). The emission

rates of consumer products might depend upon the combined effects of the mass transfer effect, which increases the emission rates under high ACHs, and the ventilation effect, which decreases the emission concentrations under high ACHs (Zhu *et al.*, 1999; ASTM, 1998).

The household inhalation exposure levels of target compounds emitted from three types of consumer products were evaluated by estimating the concentrations in residential bedroom conditions. This estimation was done by using the one-compartment mass balance model (eq. 2). Liu *et al.* (2003) assumed that the volume of the bedroom is in the range of 50~100 m³. As such, the present study used the median value of this range: the volume of bedroom (V) as 75 m³. A typical ventilation rate, 1 ACH, was employed (ASTM, 1998). The source emission rates (S) are presented in Table 3. The indoor air concentrations reported by previous studies (Reisen and Arey, 2005; Chuang *et al.*, 1999; Jo and Moon, 1999) are employed as typical room background levels (Table 4).

Table 4 exhibits the estimated concentrations associated with the use of three types of consumer products and the ratios of the estimated concentrations to background concentrations. For AF, the ratios for limonene (5.0) and m, p-xylene (5.8) exceeded 1, whereas those for three compounds (benzene, ethylbenzene and toluene) were similar to 1. For MSQR, the ratios exceeded 1 for all target compounds, ranging from 1.8 for toluene to 71 for naphthalene. For MTHR, the ratios for m, p-xylene (5.0) and naphthalene (78) exceeded 1, whereas those for three compounds (benzene, ethylbenzene and toluene) were similar to 1. This finding suggests that the uses of consumer products can elevate indoor levels of limonene and m, p-xylene for AF, m, p-xylene (5.0) and naphthalene for MSQR, and m, p-xylene (5.0) and naphthalene for MTHR. Moreover, the use of any types of the consumer products can elevate indoor levels of m, p-xylene.

It is noted that certain assumptions employed for these calculations likely underestimate actual room concentrations. One is that pollutants emitted from consumer products are well mixed quickly in rooms,

Table 4. Estimated room concentrations and their ratios to background concentrations according to the type of products.

Compound	Product type	Estimated concentration ($\mu\text{g m}^{-3}$)	Ratio
Benzene	AF	5.8	1.0
	MSQR	31	5.4
	MTHR	5.8	1.0
Ethylbenzene	AF	5.2	1.1
	MSQR	152	31
	MTHR	4.9	1.0
Limonene	AF	11	5.0
	MSQR	NA	NA
	MTHR	NA	NA
Naphthalene	AF	NA	NA
	MSQR	113	71
	MTHR	124	78
Toluene	AF	36	1.0
	MSQR	64	1.8
	MTHR	37	1.0
m, p-Xylene	AF	81	5.8
	MSQR	119	8.5
	MTHR	70	5.0

Note. Ratio of the estimated concentration to background concentration; background concentration: mean concentrations for benzene ($5.8 \mu\text{g m}^{-3}$), ethylbenzene ($4.9 \mu\text{g m}^{-3}$), toluene ($35 \mu\text{g m}^{-3}$), and m, p-xylene ($14 \mu\text{g m}^{-3}$) were reported by Jo and Moon (1999), who measured them in apartments in a Korean city during winter, without using any MRs; Chuang *et al.* (1999) measured the mean indoor concentration of $2.2 \mu\text{g m}^{-3}$ at low-income homes in North Carolina, without excluding smokers; Reisen and Arey (2005) measured the naphthalene of 0.2 to $1.6 \mu\text{g m}^{-3}$ in Los Angeles and Riverside during January 2003 (the maximum value is used for this table); NA, not available.

thereby making pollutant concentration homogeneous in rooms. However, the room air may not completely be mixed. Certain consumer products like MSQR are generally placed near the bed during sleeping in order to effectively prevent from mosquito biting, actual concentrations inhaled by the sleepers may be higher than the estimated concentrations. Moreover, in certain homes employing consumer products, the windows of rooms are closed when air conditioners are operated. These conditions will result in underestimating of actual concentrations. In contrast, other parameters such as mixing between rooms, homogeneous and heterogeneous transformation, and sorptive interactions on

surfaces may overestimate actual concentrations.

4. CONCLUSIONS AND SUMMARY

The present study evaluated the emissions composition for three different types of consumer products (AF, MSQR, and MTHR) using an environmental chamber. A compact group of six target compounds were selected on the basis of their own toxic effects. Empirical relationship fitted well with the time-series concentrations in the chamber, thereby suggesting that the relationship would efficiently be employed for testing emissions. As same with the emission concentrations, MSQR exhibited the highest emission rate for all target compounds except for limonene and naphthalene. MTHR showed the highest emission rate of naphthalene followed by MSQR. It was suggested that the uses of consumer products can elevate indoor levels of limonene and m, p-xylene for AFs, m, p-xylene and naphthalene for MSQR, and m, p-xylene and naphthalene for MTHR. In addition, the use of any types of the consumer products can elevate indoor levels of m, p-xylene.

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