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Publisher's version / Version de l'éditeur:

<https://doi.org/10.1016/j.atmosenv.2011.12.060>

Atmospheric Environment, 50, pp. 79-87, 2012-04-01

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NRCC-54578

A version of this document is published in :

Atmospheric Environment, 50, pp. 79–87, April-01-12, DOI:
doi.org/10.1016/j.atmosenv.2011.12.060

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Household materials as emission sources of naphthalene in Canadian homes and their contribution to indoor air

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Abstract

The objective of this study was to identify household materials that may contribute to the indoor naphthalene concentration in Canadian homes. Ninety-nine household materials including building materials, furnishings, and consumer products were tested. These materials included well-known naphthalene-containing products such as mothballs as well as building and consumer products where naphthalene could have been either added as part of a liquid formulation or used as a chemical intermediate in the manufacture of solid materials and product components. A fast screening method was used to determine the naphthalene concentration in a micro-scale test chamber. The tested materials were ranked based on the naphthalene emission strength combined with the amount of products typically used in homes. As expected, the results showed that mothballs, which had the highest emission factor, are one of the predominant sources. Interestingly, vinyl and wooden furniture with high emission factors and painted walls and ceiling with large surface areas were found to be important sources with the source strength even larger than those of mothballs when maximum emission factors were assumed for these building materials and furnishings. This suggests that some building materials and furnishings could be significant contributors to indoor naphthalene concentrations. This study shows that selecting materials with lower naphthalene emission factors could be one of many ways to reduce the indoor naphthalene concentration.

Keywords

Naphthalene, Household materials, Emission source, Canadian homes, Exposure, Micro-scale chamber

1. Introduction

The toxic effects of naphthalene have been recognized as a public health concern. Naphthalene is classified as a possible human carcinogen by the International Agency for Research on Cancer (IARC, 2002). In Canada, naphthalene is recognized as a toxic substance in the context of a known or potential harmful effect on the environment and human health under the Canadian Environmental Act (Government of Canada, 2010). The primary source of public exposure to naphthalene was identified as indoor air (Government of Canada, 2010). The main health concerns of exposure to naphthalene are respiratory tract lesions, including respiratory tract carcinogenicity indicated in animal studies, and haemolytic anaemia in humans (Wilson et al., 1996; Morris and Buckpitt, 2009; WHO, 2010).

Indoor naphthalene concentrations in homes have been reported in several studies (Jia et al., 2008a; Lu et al., 2005). Zhu et al. measured indoor volatile organic compound (VOC) concentrations of 75 residences in Ottawa, Canada and reported a median naphthalene concentration of $0.39 \mu\text{g m}^{-3}$ with a maximum of $144.44 \mu\text{g m}^{-3}$ (Zhu et al., 2005). Similar concentrations were observed in homes in Quebec, Canada (maximum of $23.02 \mu\text{g m}^{-3}$), according to a study involving 96 dwellings (H  roux et al., 2008). Recent studies conducted in two Canadian cities of Halifax (Wheeler et al.,

2011) and Edmonton (Kindzierski et al., 2011) showed a maximum concentration of 24.16 and 11.7 $\mu\text{g m}^{-3}$, respectively. The maximum indoor concentrations reported in the four Canadian studies are above the US EPA's inhalation reference concentration of 3 $\mu\text{g m}^{-3}$ (US EPA, 1998). Although several studies have identified potential emission sources affecting indoor concentrations such as tobacco smoke (Charles et al., 2007; Nazaroff and Singer, 2004), combustion of wood (Gustafson et al., 2008; Kakareka et al. 2005) and attached garage (Jia et al., 2008b; Héroux et al., 2008), there is little information that can lead to quantification of these sources (Jia and Batterman, 2010).

Naphthalene is used in the manufacture of various building materials and consumer products. The major usage of naphthalene is to produce phthalic anhydride, naphthalene sulfonate and moth repellents (Kälin and Funada, 2009). Phthalic anhydride and naphthalene sulfonate are primarily used in the production of plasticizers, synthetic tanning agents, dyes and rubber formulations that are employed in the manufacture of end-use products for a wide range of applications in the building and consumer product industries. Unlike moth preventatives which contain pure naphthalene, various household products may contain raw materials derived from the naphthalene production of naphthalene sulfonates and phthalic anhydride. Because of the physical-chemical properties of naphthalene, there is a potential for evaporation or sublimation when it is used in products and materials. However, no systematic effort has been given to quantify the naphthalene emissions from household products.

Due to the lack of information to characterize controllable sources of naphthalene in indoor air, Health Canada announced in 2008 that it would undertake further

investigation of household products (Government of Canada, 2008). This study, funded by Health Canada, was conducted to identify household materials available in Canada that may contribute to the naphthalene concentrations in Canadian homes. Ninety-nine household materials including building materials, furnishings and consumer products were tested. The tests involved naphthalene-containing products as well as other building and consumer products where naphthalene was believed to be unintentionally included. A fast screening method was used to determine the naphthalene concentrations in a micro-scale test chamber. The tested materials were ranked based on the naphthalene emission factor combined with the amount of products typically used indoors in a residence. The more comprehensive analysis of naphthalene emissions from household materials is expected to contribute to understanding the main naphthalene sources that exist in homes and aiding in finding ways to control the sources and to reduce the health risks caused by naphthalene in indoor air.

2. Methods

2.1. Selection of test materials

Test materials were organized into 17 product categories (Table 1) which were further organized into 38 subcategories (Tables 4-7). Fifty-five building materials and furnishings, and 44 consumer products were selected for the testing. For the selection of test materials, the individual types of products were identified based on a literature review and hazardous ingredient information available on Material Safety Data Sheets (MSDS). The focus of the search was to find products that were reported to emit naphthalene through material emission tests in literature, products that were reported to

contain naphthalene (CAS number 91-20-3) or naphthenic distillate solvent extract (CAS number 64742-88-7) using the information on MSDS, and products believed to contain other chemical substances (e.g., materials used in furniture) where naphthalene could have been used as a chemical intermediate, such as naphthalene sulfonate or phthalic anhydride, but would not need to be reported on MSDS. After the search, several retail outlets were visited to check the availability of the identified products and to identify any additional products that may contain naphthalene. The final list and the number of test products were determined, and then the products were purchased from local retail outlets in Ottawa, Canada in December 2010.

[Table 1 near here]

2.2. Fast screening method using a micro-chamber system

A fast screening method was used to determine the relative abundance of naphthalene emissions from the 99 selected materials. Experiments were carried out in a micro-scale chamber using the Microchamber/Thermal Extractor (μ -CTE, MARKES International). The six chambers of the μ -CTE system were placed in an adjustable heated block and supplied simultaneously with clean air via a gas restrictor. Clean air for the chambers was supplied using an Aadco Model 737 Pure Air Generator. The clean air entered at the top of each chamber and the exhaust flow was collected through an adsorption tube attached to the exhaust port.

The μ -CTE system uses chambers with a small volume and, therefore, facilitates rapid equilibration which enables the reduction of testing time and cost compared to

conventional emission testing methods, e.g., ASTM D5116, D6007, D7143, and DIN EN ISO 16000-9 and -10. The micro-chamber measurements using a μ -CTE system were reported to demonstrate fair prospect on the suitable tool for rapid and meaningful screening of material emissions and to offer enhanced sensitivity for semi-volatile organic compounds (SVOCs) as a result of reduced sink effects (Uhde and Salthammer, 2006). This method was considered to be suitable for detecting naphthalene, since naphthalene is classified as a SVOC with a vapor pressure of 0.087 mmHg at 25°C, which is just below the 0.1 mmHg cut-off often used to define VOCs (US EPA, 1999).

2.3. Material preparation and measurement procedures

Depending on the type of materials being tested, two different testing modes were used and specimens were prepared accordingly. The surface emission testing mode (cell mode) was used to measure naphthalene emitted only from the top surface of solid building materials eliminating the interference from the edges and rear surface. If the cell mode was not applicable due to the nature of the specimen, e.g., irregular shape, the bulk emission testing mode (bulk mode) was used, which enabled the entire specimen to be exposed to the chamber air. In this mode, clean air passed around the specimen with naphthalene being swept from the chamber and collected onto the sampling tube.

For the surface emission testing, the original solid specimen was cut into a smaller piece that could fit into a Tedlar bag to prevent any cross-contamination as soon as it was purchased from a retail outlet. The Tedlar bag was sealed until the beginning of a test. The specimen was further cut into a circular shape with a diameter of approximately 45 mm before a test. The edges and rear surface of the specimen were

wrapped in aluminum foil to reduce edge effects. Then, the specimen was directly fit into a micro-chamber with the emitting surface raised using a spacer until the surface reached the collar projecting down from a chamber lid, which enabled the surface to form one wall of a reduced-volume chamber. In the case of liquid building materials, the material was applied on a glass disc (44 mm diameter) before a test with a dedicated clean brush. The glass disc was placed into a micro-chamber with a spacer to ensure a similar chamber volume for each specimen.

For the bulk emission testing, the specimen of solid consumer products was cut to fit a micro-chamber. A small Petri dish-type glass container (44 mm diameter, 10 mm depth) was used to avoid direct contact with the chamber. In the case of liquid consumer products most of which were not suitable for the glass disc application, the liquid was poured into a small Petri dish-type glass container mentioned previously. A spacer was also used when the Petri dish was put into a micro-chamber.

The chamber was operated at an ambient temperature of 23°C and relative humidity of 0 % with an air supply at $5 \times 10^{-5} \text{ m}^3 \text{ min}^{-1}$ which was determined to achieve a comparable vapour concentration between the micro-chamber and the 0.05 m^3 emission chamber by using an identical specific air flow rate (Schrupp et al., 2007). The specific air flow rate was calculated using the air change rate and the loading factor as shown in Equation (1).

$$q = n/L \tag{1}$$

where q is the specific air flow rate ($\text{m}^3 \text{ m}^{-2} \text{ h}^{-1}$), n is the air change rate (h^{-1}) and L is the loading factor ($\text{m}^2 \text{ m}^{-3}$). In a typical setting, the specific air flow rate for the 0.05 m^3 emission chamber was estimated to $2.5 \text{ m}^3 \text{ m}^{-2} \text{ h}^{-1}$ by a loading factor of $0.4 \text{ m}^2 \text{ m}^{-3}$ and air change rate of 1 h^{-1} . To replicate the parameters, an air flow rate of $5 \times 10^{-5} \text{ m}^3 \text{ min}^{-1}$ was used for the micro-chamber based on a loading factor of $400 \text{ m}^2 \text{ m}^{-3}$ (surface area of $1.25 \times 10^{-3} \text{ m}^2$, air volume of $3.14 \times 10^{-6} \text{ m}^3$) and an air change rate of 1000 h^{-1} .

The equilibration time for the micro-chamber test was decided by performing a pre-test. The equilibration time represents the time between the initiation of air flow after placing the test specimen into a micro-chamber (time zero) and the beginning of sampling, which allows the chamber concentration to stabilize (ASTM, 2011). For the pre-test, eight sequential samples were collected during the 120-minute testing period. Sampling was initiated 15 minutes after starting the test and lasted 10 minutes. Figure 1 shows the plot of chamber concentrations of naphthalene emitted from vinyl flooring for 120 minutes with 15-minute intervals. The results demonstrated that the rate of concentration changes was fast initially and then stabilized after about 30 minutes, which indicates that 30-minute conditioning is required for a micro-chamber to reach sufficient equilibrium conditions. The selected equilibration time was found to satisfy the range of equilibration time suggested in the ASTM standard (ASTM, 2011). Based on the pre-test results, three air samples were taken at 30, 60, 120 minutes after time zero in actual tests. The samples were collected using glass tubes $178 \times 6 \times 4 \text{ mm}$ (L \times O.D. \times I.D.) filled with adsorbent Tenax TA. The sampling volume was $5 \times 10^{-4} \text{ m}^3$ with a sampling rate of $5 \times 10^{-5} \text{ m}^3 \text{ min}^{-1}$. Table 2 compares the chamber dimensions and flow conditions for the micro-chamber used in this study with acceptable ranges

suggested in the ASTM standard (ASTM, 2011).

[Figure 1 near here]

[Table 2 near here]

2.4 Chemical analysis

The analysis of naphthalene collected on an adsorbent tube was performed using a Gerstel thermal desorption system (TDS-3) with an autosampler (TDS-A) connected to a Varian 4000 Gas Chromatograph - Ion Trap Mass Spectrometer (GC-MS) system equipped with a SPB-624 capillary column (30 m \times 0.25 mm I.D. \times 1.4 μ m thickness). The desorbed analytes were injected using a programmable temperature vaporizer called as Cooled Injection System (CIS). The Gerstel system was operated in the TDS splitless/CIS split mode and the MS was in the Selected Ion Scan (SIS) mode (target ion 128 for naphthalene). Temperature profiles for the thermal desorption and GC-MS are as follows.

- Thermal desorption: initial temperature 30°C, final temperature 320°C
- CIS condition: initial temperature -90°C, final temperature 350°C
- GC condition: initial temperature 30°C, ramp 10°C min⁻¹, final temperature 210°C

Naphthalene was identified by comparing its mass spectrum with the NIST Mass Spectral Library (NIST08, V1.0.2.2), and quantified by a 5-point calibration of its own standard. The detection limit was 0.51 ng for the CIS split ratio of 1:30 (solid specimens), and 0.98 ng for the split ratio of 1:200 (liquid specimens).

2.5 *Quality assurance*

Before each test, the chamber assemblies were cleaned with detergent and rinsed with distilled water. Then, the chamber temperature was elevated to 80°C for at least 12 hours to minimize background concentrations. To check the cleanliness, background samples were taken in chambers prior to each test. Sampling tubes were conditioned before each use at 320°C for 15 minutes with a helium flow using the Tube Conditioner TC 2 (Gerstel Inc.). During the sampling process, flow rate checks were conducted before and after each sampling using Digital Flow Meter 650 (Humonics Inc.). This was to confirm that a constant flow rate was maintained through each micro-chamber.

To understand the reproducibility of chamber tests, two identical vinyl flooring specimens were tested in two chambers simultaneously. The measured concentrations of 25 chemical compounds showed good agreement between two chambers. The average difference was 34 % and 8 % for samples taken at 60 and 120 minutes, respectively.

2.6 *Data analysis*

The naphthalene emission factor ($\mu\text{g m}^{-2} \text{ h}^{-1}$ or $\mu\text{g g}^{-1} \text{ h}^{-1}$) was calculated as the product of the naphthalene concentration ($\mu\text{g m}^{-3}$) and flow rate ($5 \times 10^{-5} \text{ m}^3 \text{ min}^{-1}$) divided by the amount of the specimen (m^2 or g). The mean and standard deviation values of the three time-series emission factors at 30, 60 and 120 minutes were calculated for each material, and the mean value was used as a representative emission factor of naphthalene. The emission rate ($\mu\text{g h}^{-1}$), which is defined as the mass of naphthalene emitted from a material per unit time, was calculated by the product of

emission factor ($\mu\text{g m}^{-2} \text{ h}^{-1}$ or $\mu\text{g g}^{-1} \text{ h}^{-1}$) and surface area (m^2) for the surface emission testing or mass of specimen (g) for the bulk testing.

Assuming a steady-state condition, the indoor concentration of naphthalene from multiple sources was estimated using Equation (2).

$$C = \sum_{i=1}^N C_i = \frac{1}{Vn} \sum_{i=1}^N A_i EF_i \quad (2)$$

where C_i is the naphthalene concentration contributed by the i^{th} material ($\mu\text{g m}^{-3}$), A_i is the amount of the i^{th} material (m^2 or g), EF_i is the emission factor of the i^{th} material ($\mu\text{g m}^{-2} \text{ h}^{-1}$ or $\mu\text{g g}^{-1} \text{ h}^{-1}$), N is the total number of materials, V is the house volume (m^3) and n is the air change rate (h^{-1}).

Scenarios were developed for the required information such as house volume, air change rate and amount of materials used (Table 3). The values for the house volume and air change rate were adopted from the field study conducted in 115 homes in Quebec City, Canada (Won et al., 2011). The values were similar to the typical house volume of 500 m^3 and the air change rate of 0.3 h^{-1} , suggested by CAN/ULC (2003) for the indoor air quality prediction. The amount of building materials and some furnishings was based on the material loading factor given by Hodgson et al. (2005), who reported the typical surface areas in 9 residences in San Francisco. The amount of caulking and wax applied on surfaces was estimated using the representative product coverage (Construction Resources, 1999; US DOE, 2011). The floor was assumed to have a mixture of carpet, vinyl flooring and area mat. The amount of consumer products and furnishings other than wooden furniture were estimated based on the amount of

products obtained from the distributors through the Internet search.

[Table 3 near here]

Four scenarios, shown in Table 8, were used to predict the indoor air concentration based on Equation (2). The scenarios were developed to model the use of products in a house, assuming different products have different emission factors to reflect the variability of the emission measurements (with minimum and maximum emission factors). An important note is that mothballs were not tested according to label's directions (e.g., being placed in an air tight container). They were chosen as a reference level considering that they would represent maximum naphthalene emissions as they are essentially composed of naphthalene. The scenarios with mothballs were to analyze the worst cases in terms of naphthalene emissions and concentrations.

3. Results and discussion

3.1 Naphthalene emission factors of moth preventatives and deodorizers

Table 4 summarizes the mean and standard deviation (S.D.) values of time-series naphthalene emission factors of moth preventatives and deodorizers. These sources have been considered as major contributors to indoor naphthalene. The two moth preventatives were ball-type products and the three deodorizers were block-type products designed for closet or toilet bowl. The results showed that mothballs emitted high levels of naphthalene. The emission factors of mothballs ranged from 841 to 1015

$\mu\text{g g}^{-1} \text{h}^{-1}$. This is not surprising since the mothballs contained crystalline naphthalene. A study reported that the naphthalene emission factor from a mothball was $192 \mu\text{g g}^{-1} \text{h}^{-1}$ (Jo et al., 2008), which is smaller than those reported in this study. The difference is likely due to the fact that our results were obtained within 2 hours of emission periods in a micro-chamber and the results from Jo et al. (2008) were obtained over 24 hours in a 0.05 m^3 chamber. Also, the mothball specimens used in this study consisted of fragments of a mothball which was broken into small pieces in order to fit into the micro-chamber. This may have allowed active volatilization from the increased surface area of a fragmented specimen in this study.

[Table 4 near here]

On the other hand, the deodorant blocks emitted lower levels of naphthalene compared to the mothballs. The emission factors of deodorant blocks ranged from 0.52 to $17.9 \mu\text{g g}^{-1} \text{h}^{-1}$. Although elevated naphthalene emissions were not observed from the deodorant blocks, small quantities of naphthalene were still detected. This is an interesting finding, because the deodorant products contained pure 1,4-dichlorobenzene according to the MSDS and this chemical has been used as a substitute for naphthalene in deodorizers to avoid the hazard associated with naphthalene. One possible explanation is that there may be cross-contamination during manufacturing or storage. To confirm whether naphthalene is the ingredient of 1,4-dichlorobenzene deodorant blocks, further experiments should be performed.

3.2 Naphthalene emission factors of building materials, furnishings and other con

sumer products

Tables 5 to 7 show the mean and standard deviation values of time-series naphthalene emission factors of building materials, furnishings and other consumer products. The emission factors were calculated as a mass per unit area for the solid materials tested in the cell mode and a mass per mass for the liquid and bulk materials tested in the bulk mode. Figures 2 and 3 show the average value of emission factors by category which is expressed as a bar, and the emission factor of each individual material indicated as a symbol.

3.2.1 Surface-emitting solid household materials

The top two material subcategories ranked among the surface-emitting materials were vinyl furniture, including vinyl chair cushion covers, and wooden furniture most of which were engineered wood panels with finishes. It should be noted that the emissions from the covers used in chair cushions were much higher than those from the engineered wood panels with finishes, showing averaged emission factors of 447.3 and 32.0 $\mu\text{g m}^{-2} \text{h}^{-1}$, respectively. The emission factors of naphthalene from the wooden furniture reflected the variations stemming from the differences in the types of engineered wood and finishes. The emission factors of flooring also varied according to the type of flooring. The category of area mat, including door mats, showed higher emission factors than those of carpet, under-pad and vinyl flooring. Within the subcategories of carpet and mat, the emission factors of the mat with polypropylene top surface and vinyl backing (CRP3) and that with the PVC coated polyester filament (CRP4) were higher than those from the carpet made of synthetic fiber (CRP2). Foam

mat and foam toys were the least emitting materials among the surface-emitting solid materials.

[Table 5 near here]

[Figure 2 near here]

In the literature, there are few data sets of naphthalene emissions from household materials. A study performed by Won et al. (2005) analyzed naphthalene as one of many target compounds. The study examined emissions of 90 target VOCs from 69 building materials and listed the emission factors of naphthalene from solid building materials including wood panels, flooring and installation materials. The study reported emission factors for naphthalene ranging from 0.001 to 310 $\mu\text{g m}^{-2} \text{h}^{-1}$. Carpet and caulking showed high emission factors, while vinyl flooring and adhesive showed low emission factors, which are similar to the results of the present study. In comparison to the emission factors of wooden furniture in this study (1.33 – 73.79 $\mu\text{g m}^{-2} \text{h}^{-1}$), wood panels in the study by Won et al. (2005) exhibited lower emission factors ranging from 0.02 to 0.2 $\mu\text{g m}^{-2} \text{h}^{-1}$. The discrepancy is likely to be attributed to the difference in the test method (micro-chamber in this study and 0.05 m^3 chamber in Won et al. (2005)) and/or the difference in test materials reflecting variations in wood types and finishes.

3.2.2 Liquid and bulk household materials

The mean value of emission factors for liquid building materials and consumer products showed a maximum of 87.99 $\mu\text{g g}^{-1} \text{h}^{-1}$. The highest ranked category was coatings, followed by caulking, primer, paint and adhesive which also showed relatively

high emission factors. Of note; some of coating materials showed higher naphthalene emission factors than deodorant blocks, which confirms the possible impact of wood finishes discussed above. Among the liquid building materials, naphthalene was prominently detected from a caulking (CAK8), a stain (STA2), and varnish specimens (VAR2, VAR3). CAK8 consisted of 10-30 % of light aliphatic solvent naphtha (CAS number 64742-89-8). Also, STA2 contained 6-9 % of heavy naphthenic petroleum oil (CAS number 64742-52-5) and highly refined naphthenic oil (CAS number 64742-53-6). These ingredients are assumed to be the main sources of naphthalene. In the case of two polyurethane based varnishes (VAR2 and VAR3), which showed higher concentrations than the other varnishes, it is hypothesized that mineral spirits (CAS number 64742-88-7) could be the cause of elevated naphthalene emissions since it was the main ingredient of the two varnishes. However, a low naphthalene emission was detected in a wax (WAX1) despite the fact that it contained mineral spirits as a main ingredient. Primers (PRM1 and PRM2) also emitted significant amounts of naphthalene. These primers contained 10-30 % and 5-10 % of hydro-treated light petroleum distillates (CAS number 64742-49-0), respectively.

[Table 6 near here]

[Table 7 near here]

[Figure 3 near here]

In addition to the investigation of ingredients of liquid building materials, the correlation between the measured naphthalene emission factors and the VOC contents in the liquid building materials provided by manufacturers was analyzed in Figure 4.

Generally, it appears that materials with low VOC contents have low naphthalene emission factors. The opposite, however, was not always true as several materials with a high VOC content did not show a high emission factor of naphthalene. This supports that the VOC content information given by manufacturers may not be directly applicable to gauging emissions of individual compounds with accuracy.

[Figure 4 near here]

Among liquid and bulk household materials, consumer products showed lower emission factors than liquid building materials. Candles, incense, air fresheners and cleaning products were the four lowest ranked categories. In this study, candles, incense and some of air fresheners were tested for emissions in the non-combustion or non-heating mode. These products are designed to be operated using a flame or other heat source to heat and vaporize a fragrance formulation, and several studies have pointed out that combustion is the single largest naphthalene emission source (ATSDR, 2005; Price and Jayjock, 2008). Plug-in air fresheners and combustion sources such as candles and incense may have different emission characteristics when in actual operation.

3.3 Source strength

By using the emission factor and the amount of materials typically used in a home, the respective emission rates were estimated as shown in Figure 5. The emission rate allows for determination of the overall source strength, which depends on both the quantity of a source present and the rate emitted into the indoor air per specific area or

mass. The minimum and maximum emission factors for each source category were adopted from Tables 4-7, which resulted in a range of emission rates expressed in a bar. The longer the bar, the larger the difference in emission factors between the minimum and maximum values.

[Figure 5 near here]

The results show that mothballs are one of the major sources with high source strengths, even though the small usage amount of 4.2 g was assumed. It was found that the most dominant source was vinyl furniture followed by painted walls and ceilings, when it was assumed that each material emitted naphthalene at its maximum emission factor within the relevant product category. This is an interesting observation considering the major research focus has been other sources such as mothballs (Jo et al., 2008), tobacco smoking (Singer et al., 2002; Charles et al., 2007) and other combustion sources (ATSDR, 2005; Price and Jayjock, 2008; Lu et al., 2009). The results with the maximum emission factors indicate a possibility that building materials and furnishing could be important naphthalene emission sources. Wax on the furniture and caulking around windows, which also belong to the categories of building materials showed relatively high source strengths in general. On the other hand, consumer products, except for the mothballs and deodorizer blocks, showed lower source strengths than building materials and furnishings. This is due to small emission factors and small usage amounts. Moreover, the use of consumer products generally occurs episodically, unlike building materials and furnishings. Therefore, it is suggested that the impact of most consumer products on indoor naphthalene would be limited.

Figure 6 illustrates the extent to which building materials and furnishings contribute to naphthalene emissions indoors compared to mothballs and deodorant blocks. When the minimum emission factors were assumed, the contribution of furniture and building materials was found to be 1/20th and 7/50th of mothball/deodorant blocks, respectively. On the other hand, when the maximum emission factors were assumed, the contribution of furniture and building materials was greater, that is, 4.1 and 1.7 times that of mothball/deodorant blocks, respectively. The results may vary with the assumptions used in the product usage amount. However, considering the large surface areas of building materials and furnishings and their continuous emission patterns after installation, the results strongly indicate that building materials and furnishings where naphthalene was unintentionally included could be the most dominant naphthalene sources.

[Figure 6 near here]

3.4 Predicted indoor concentrations

The indoor air concentrations were predicted using Equation (2) for the scenarios identified in Table 8. The naphthalene concentration in a home in Scenario 1 with items showing maximum emission factors was 187.5 $\mu\text{g m}^{-3}$. This is comparable to the high peak concentration of 144 $\mu\text{g m}^{-3}$ measured in the study involving 75 residences in Ottawa (Zhu et al., 2005). This indicates that the assumptions for Scenario 1 are reasonable for extreme cases. The naphthalene concentration under Scenario 2 was 4.9

$\mu\text{g m}^{-3}$, which is in the same range of the mean value (0.27 to $4.1 \mu\text{g m}^{-3}$) in non-smoking homes reported in a review study by Jia and Batterman (2010). The concentration also satisfies the WHO guideline of $10 \mu\text{g m}^{-3}$ for naphthalene (WHO, 2010). Again, this shows that the assumptions for Scenario 2 are realistic. For Scenario 2, the emission factors of the liquid-based materials were mostly from water-based products as opposed to oil-based ones for Scenario 1. Due to the presence of one mothball, the naphthalene concentration was elevated from 187.5 to $219.7 \mu\text{g m}^{-3}$ in Scenario 3. In Scenario 4, the presence of one moth ball also increased the naphthalene concentration from 4.9 to $30.5 \mu\text{g m}^{-3}$.

The results show that it is plausible to reduce the contribution of building materials and furnishings to naphthalene concentrations in indoor air by selecting materials with low naphthalene emission factors.

[Table 8 near here]

3.5 Study strengths and limitations

This study demonstrates that the emission information from a fast screening method can be useful in comparing various products and identifying potentially dominant sources. Ranking indoor sources can be achieved based on the generated emission information along with the product usage information. However, it should be noted that this method is mainly for screening sources as the test conditions do not reflect the reality completely. For example, liquid-based building materials were applied on a glass

disc. This may have led to maximum emission factors for those materials compared to the normal application scenarios involving absorptive substrates such as wood and gypsum board. Also, the relative humidity (RH) was set at 0 % as opposed to the typical setting of 50 % for emission chamber testing. There have been conflicting reports with respect to the RH effect on material emissions, including no general trend (Haghighat and De Bellis, 1998), no effects (Sollinger et al., 1994) and increased emissions with increasing RHs (Fang et al., 1999). The effect of RH on naphthalene emissions needs to be evaluated in future work.

Another issue can be raised on the renewability and longevity of the sources. All of the building material and furniture items used in this study were purchased as new. This would represent a period of high level emission within their lifetime. The emission factors in this study may be more suitable to represent sources like mothballs and other consumer products that are capable of being regularly renewed. On the other hand, the emission factors are likely to be overestimated for more permanent objects such as building materials and furniture that are expected to decay in the course of their lifetime. Additionally, the high standard deviation values of emission factors in Tables 4 to 7 indicate that the chamber conditions were not at equilibrium for some specimens. It appears that the emission rates decreased between 30 and 120 minutes. The observation was more prevalent with liquid building materials and consumer products. More research is recommended to develop a screening method that can capture the equilibrium emissions for liquid products.

It has yet to be proven whether this type of screening method can be expanded to

more rigorous modeling for the fate of naphthalene from sources to exposures. More systematic comparison studies, involving conventional emissions tests under more realistic conditions and validation studies in the full or field-scale settings, should be considered.

4. Conclusions

The health risks of naphthalene have been recognized as a public concern. This study identified household materials that may contribute to the naphthalene concentrations in indoor air in residential homes. Ninety-nine household materials including building materials, furnishings and consumer products were tested using a fast screening method with a micro-chamber system. The results suggest that some building materials and furnishings, particularly vinyl furniture and painted walls and ceilings, where naphthalene could have been either added as part of a liquid formulation or used as a chemical intermediate in the manufacture of solid materials and product components may be the dominant naphthalene sources. This study shows that selecting materials with lower naphthalene emission factors could be one of many ways to reduce the indoor naphthalene concentration.

Acknowledgements

The authors would like to thank Mr. Gang Nong and Ms. Judy Kwan for their assistance with setting up the test system and operating the GC-MS system.

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Figure captions

Figure 1. Chamber concentrations of vinyl flooring during 120 min for the investigation of appropriate conditioning time

Figure 2. Averaged emission factors of naphthalene according to the categories of surface emitting solid materials expressed in bars and emission factor of each material indicated as a symbol

Figure 3. Averaged emission factors of naphthalene according to the categories of liquid and bulk materials and expressed in bars and emission factor of each material indicated as a symbol

Figure 4. Correlation between measured naphthalene emission factors and VOC contents provided by manufacturers for the liquid building materials

Figure 5. Range of naphthalene emission rates for sources used in a home with the application of maximum and minimum emission factors of respective materials

Figure 6. Comparison of naphthalene emission rates between mothball/deodorant blocks and other sources according to the maximum and minimum emission factors

Figure 1.

Chamber concentrations of vinyl flooring during 120 min for the investigation of appropriate conditioning time

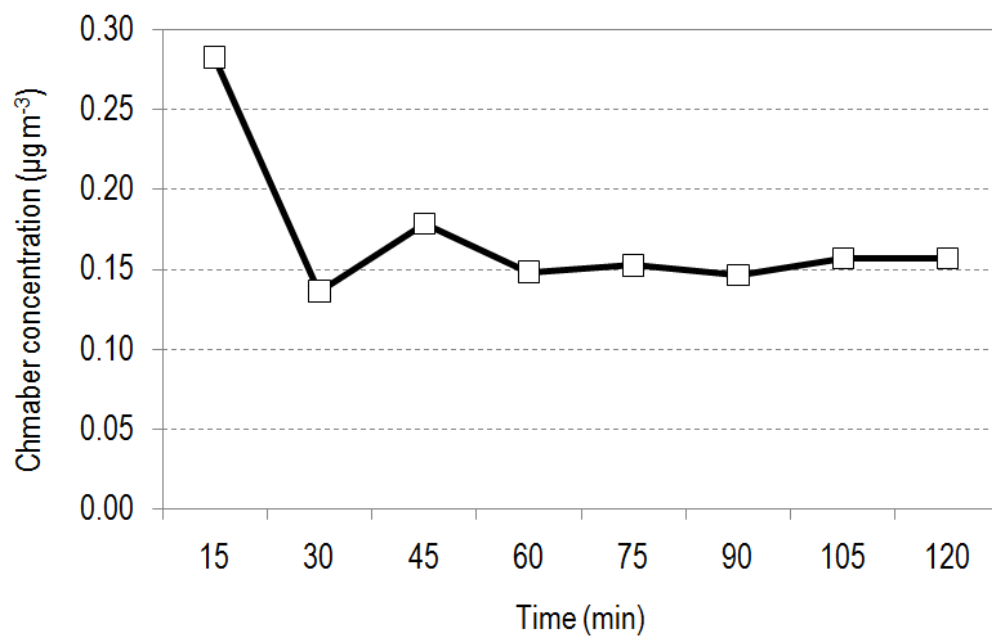


Figure 2.

Averaged emission factors of naphthalene according to the categories of surface emitting solid materials expressed in bars and emission factor of each material indicated as a symbol

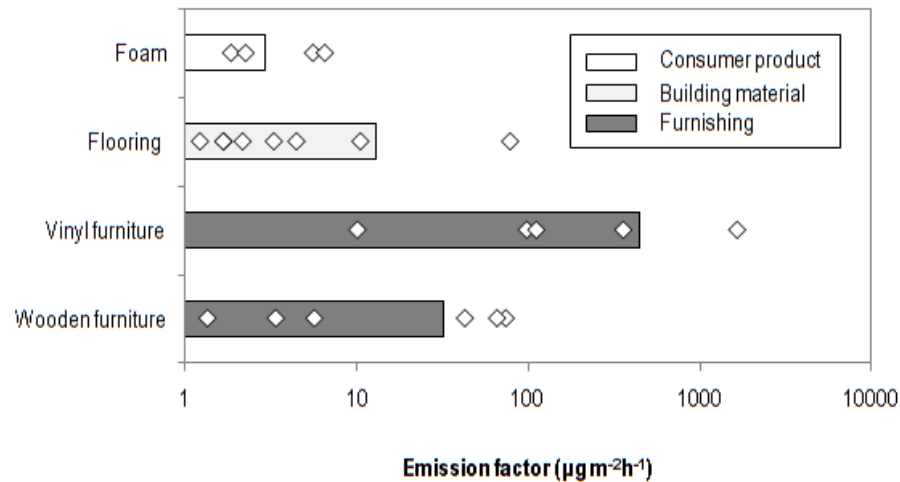


Figure 3.

Averaged emission factors of naphthalene according to the categories of liquid and bulk materials and expressed in bars and emission factor of each material indicated as a symbol

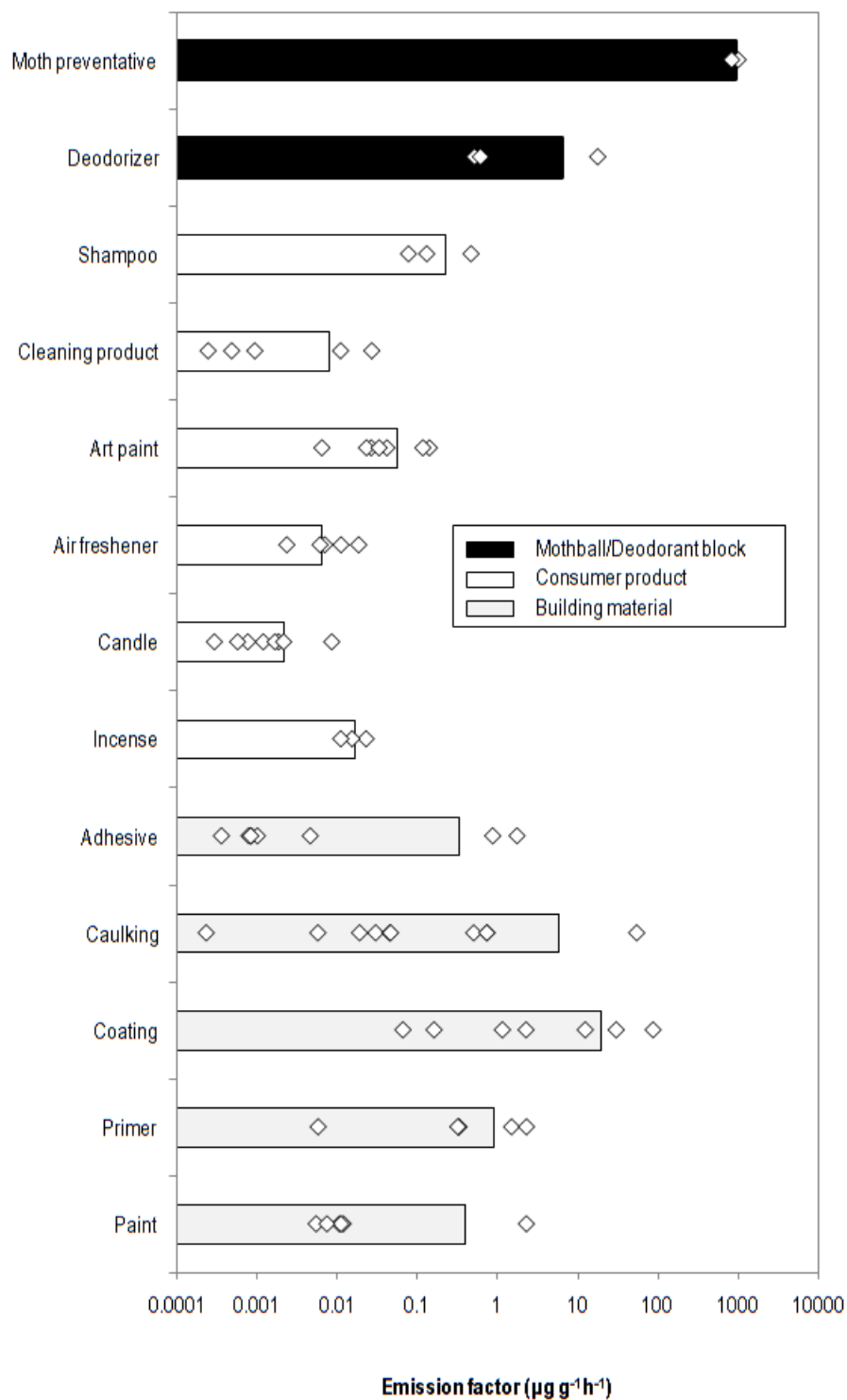


Figure 4.

Correlation between measured naphthalene emission factors and VOC contents provided by manufacturers for the liquid building materials

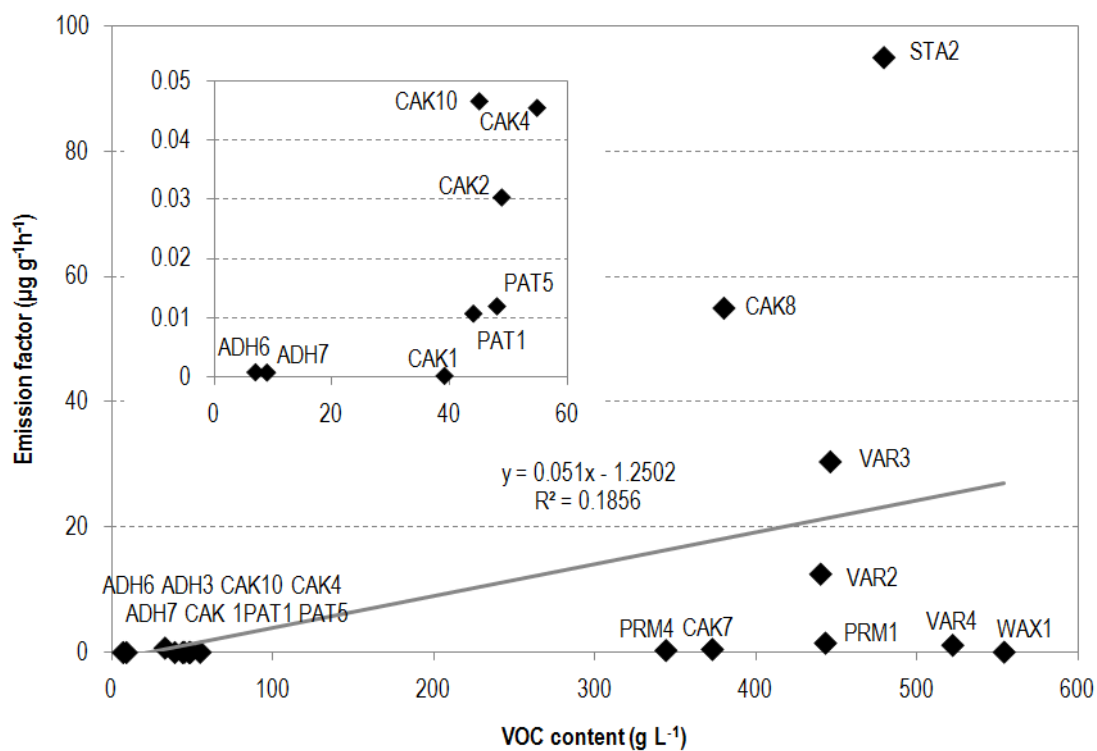


Figure 5.

Range of naphthalene emission rates for sources used in a home with the application of maximum and minimum emission factors of respective materials

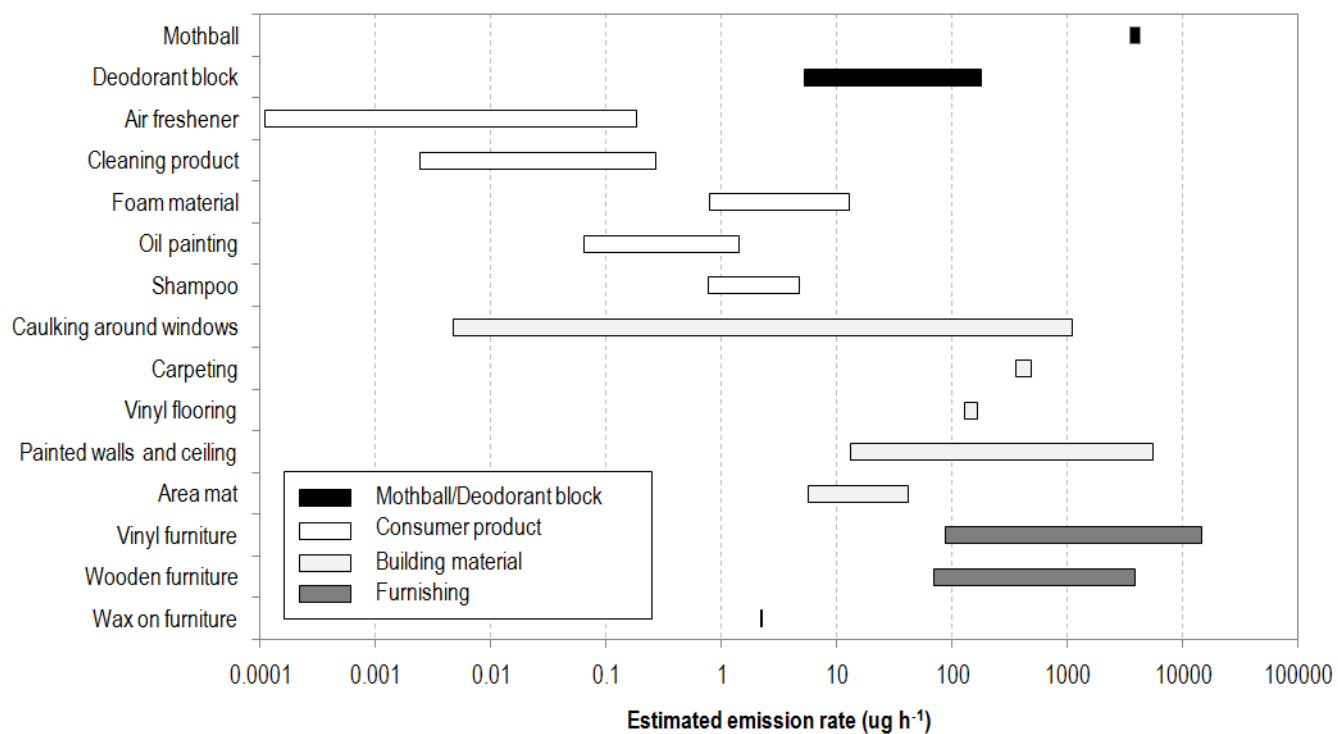


Figure 6.

Comparison of naphthalene emission rates between mothball/deodorant blocks and other sources according to the maximum and minimum emission factors

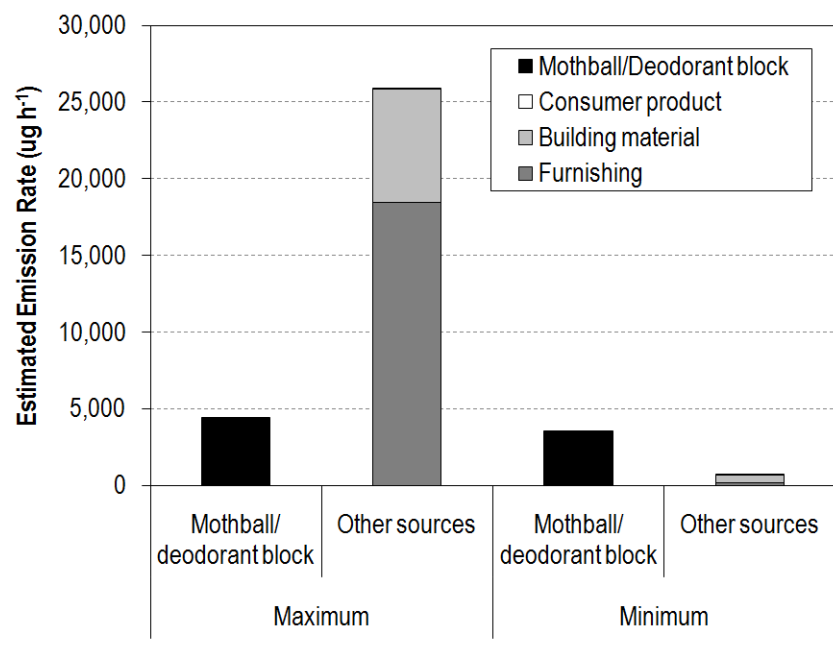


Table captions

Table 1. Number of household materials tested in this study

Table 2. Dimensions and flow rate conditions for micro-chamber used in this study (μ -CTE) and range of typical conditions suggested in the ASTM practice standard

Table 3. House volume, air change rate and amount of materials used for the scenario analysis

Table 4. Naphthalene emission factors of moth preventatives and deodorizers

Table 5. Naphthalene emission factors of surface-emitting solid household materials

Table 6. Naphthalene emission factors of liquid materials

Table 7. Naphthalene emission factors of bulk household materials

Table 8. Naphthalene indoor concentrations under different scenarios for indoor air quality modeling using the emission factors from micro-chamber experiments

Table 1.

Number of household materials tested in this study

	Category	Number of materials
Building material	Flooring	8
	Paint	6
	Primer	5
	Coating	7
	Caulking	10
	Adhesive	8
Furnishing	Wooden furniture	6
	Vinyl furniture	5
Consumer product	Foam	6
	Incense	3
	Candle	8
	Air freshener	7
	Deodorant block	3
	Moth preventative	2
	Art paint	7
	Cleaning product	5
	Shampoo	3

Table 2.

Dimensions and flow rate conditions for micro-chamber used in this study (μ -CTE) and range of typical conditions suggested in the ASTM practice standard

Parameter	Cell mode		Bulk mode	
	μ -CTE	ASTM	μ -CTE	ASTM
Diameter (mm)	40	56	45	64
Depth (mm)	2.5	3	10	36
Volume (m^3)	3.14×10^{-6}	7.39×10^{-6}	1.59×10^{-5}	1.16×10^{-4}
Exposed surface area (m^2)	1.25×10^{-3}	2.46×10^{-3}	N/A ^a	3.22×10^{-3}
Equilibration time (min)	30	20 - 40	30	20 - 40
Sampling time (min)	10	10 – 15	10	10 – 15
Inlet air flow rate ($\text{cm}^3 \text{min}^{-1}$)	50	50 (30 – 500)	50	50 (30 – 500)

^a N/A: not available

Table 3.

House volume, air change rate and amount of materials used for the scenario analysis

House volume			418 m ³	Won et al. (2011)
Air change rate			0.33 h ⁻¹	Won et al. (2011)
Amount of material	Building material	Caulking around windows ^c	405 g	Material surface area estimated using the material loading ratio given in ^a Hodgson et al. (2005)
		Carpeting ^a	110.62 m ²	
		Vinyl flooring ^a	77.45 m ²	
		Painted walls and ceiling ^{a, b}	48461 g	
		Area mat	0.54 m ²	
	Furniture	Vinyl furniture	8.76 m ²	Material coverage given in ^b Construction Resources (1999) and ^c US DOE (2011)
		Wooden furniture ^a	52.85 m ²	
		Wax on furniture ^{a, b}	661 g	
	Consumer product	Mothball	4.2 g	
		Deodorant block	10 g	
		Air freshener	10 g	
		Cleaning product	10 g	
		Foam material	2 m ²	
		Oil painting	10 g	
		Shampoo	10 g	

Table 4.

Naphthalene emission factors of moth preventatives and deodorizers

Category	Subcategory	Code	Emission factor ($\mu\text{g g}^{-1} \text{h}^{-1}$)		
			Mean \pm S.D.		
Moth preventative (2 ^a)	Mothball	MB1	1015.18	\pm	109.80
		MB2	841.14	\pm	17.82
Deodorizer (3)	Deodorant block	DEO1	0.52	\pm	0.67
		DEO2	17.93	\pm	7.83
		DEO3	0.62	\pm	0.48

^a The number of specimens tested within the category

Table 5.

Naphthalene emission factors of surface-emitting solid household materials

Category	Subcategory	Code	Emission factor ($\mu\text{g m}^{-2} \text{h}^{-1}$)	
			Mean \pm S.D.	
Flooring (8 ^a)	Vinyl flooring	VF1	1.66	\pm 0.27
		VF2	2.14	\pm 0.27
	Carpet	CRP1	4.41	\pm 0.15
		CRP2	3.25	\pm 0.23
	Area mat	CRP3	10.43	\pm 0.79
		CRP4	78.17	\pm 2.74
	Under pad	UP1	1.65	\pm 1.78
		UP2	1.21	\pm 1.06
Wooden furniture (6)	Wooden furniture panel	FUR1	73.79	\pm 34.69
		FUR2	65.60	\pm 109.55
		FUR3	5.62	\pm 8.20
		FUR4	42.55	\pm 72.12
		FUR5	3.34	\pm 2.79
		FUR6	1.33	\pm 1.37
Vinyl furniture (5)	Vinyl chair cushion cover	FUR7	10.02	\pm 4.56
		FUR8	97.84	\pm 42.69
		FUR9	1659.08	\pm 797.86
		FUR10	111.27	\pm 87.71
		FUR11	358.46	\pm 222.08
Foam (6)	Foam mat	FOM1	5.51	\pm 1.85
		FOM2	0.99	\pm 0.65
	Foam toy	FOM3	6.46	\pm 3.57
		FOM4	1.83	\pm 1.73
		FOM5	2.23	\pm 2.59
		FOM6	0.40	\pm 0.40

^a The number of specimens tested within the category

Table 6.

Naphthalene emission factors of liquid materials

Category	Subcategory	Code	Emission factor (μg g ⁻¹ h ⁻¹)		
			Mean ± S.D.		
Paint (6 ^a)	Oil-based paint	PAT1	0.01	±	0.00
		PAT4	2.31	±	0.96
		PAT5	0.01	±	0.01
	Water-based paint	PAT2	0.01	±	0.00
		PAT3	0.01	±	0.00
		PAT6	0.01	±	0.00
Primer (5)	Oil-based primer	PRM1	1.51	±	0.81
		PRM2	2.32	±	1.10
		PRM4	0.33	±	0.16
		PRM5	0.32	±	0.35
	Water-based primer	PRM3	0.01	±	0.00
Coating (7)	Wax	WAX1	0.07	±	0.09
	Wood stain	STA1	2.29	±	0.86
		STA2	95.01	±	23.68
	Varnish	VAR1	0.16	±	0.01
		VAR2	12.52	±	5.30
		VAR3	30.46	±	11.09
		VAR4	1.16	±	0.18
	Caulking (10)	Window/Door caulking	CAK1	0.00	±
CAK2			0.03	±	0.05
CAK5			0.02	±	0.01
Kitchen/Bath caulking		CAK4	0.05	±	0.08
		CAK6	0.01	±	0.01
Roof caulking		CAK3	0.75	±	0.33
Multi-purpose caulking		CAK7	0.51	±	0.65
		CAK8	55.00	±	3.53
		CAK9	0.74	±	0.38
		CAK10	0.05	±	0.07
Adhesive (8)	Flooring adhesive	ADH1	0.00	±	0.00
		ADH2	0.00	±	0.00
		ADH6	0.00	±	0.00
		ADH8	0.88	±	0.23
	Tile adhesive	ADH3	0.71	±	1.59
		ADH4	0.00	±	0.00
	Multi-purpose adhesive	ADH5	0.00	±	0.00
		ADH7	0.00	±	0.00

^a The number of specimens tested within the category

Table 7.

Naphthalene emission factors of bulk household materials

Category	Subcategory	Code	Emission factor ($\mu\text{g g}^{-1} \text{h}^{-1}$)		
			Mean \pm S.D.		
Incense (3 ^a)	Incense stick	INS1	0.02	\pm	0.00
		INS2	0.02	\pm	0.00
		INS3	0.01	\pm	0.01
Candle (8)	Scented candle	CND1	0.00	\pm	0.00
		CND2	0.00	\pm	0.00
		CND3	0.00	\pm	0.00
		CND4	0.00	\pm	0.00
		CND6	0.00	\pm	0.00
		CND7	0.01	\pm	0.00
	Unscented candle	CND5	0.00	\pm	0.00
		CND8	0.00	\pm	0.00
Air freshener (7)	Lamp oil	AF1	0.01	\pm	0.01
	Scented oil plug-in	AF2	0.01	\pm	0.00
		AF6	0.00	\pm	0.00
	Scented gel	AF7	0.00	\pm	0.00
		AF3	0.02	\pm	0.01
	Liquid fragrance diffuser	AF4	0.00	\pm	0.00
	Scented reed diffuser	AF5	0.01	\pm	0.01
Art paint (7)	Art oil paint	AOP1	0.04	\pm	0.02
		AOP2	0.01	\pm	0.00
		AOP3	0.14	\pm	0.07
	Oil paint thinner	OPT1	0.03	\pm	0.03
		OPT2	0.12	\pm	0.09
	Oil paint varnish	OPV1	0.02	\pm	0.01
		OPV2	0.03	\pm	0.01
		CL2	0.01	\pm	0.02
Cleaning product (5)	Degreaser	CL3	0.00	\pm	0.00
		CL4	0.03	\pm	0.02
		CL1	0.00	\pm	0.00
	Multi-purpose cleaner	CL5	0.00	\pm	0.00
		CL5	0.00	\pm	0.00
Shampoo (3)	Coal tar shampoo	SHP1	0.08	\pm	0.06
		SHP2	0.13	\pm	0.15
		SHP3	0.47	\pm	0.17

^a The number of specimens tested within the category

Table 8.

Naphthalene indoor concentrations under different scenarios for indoor air quality modeling using the emission factors from micro-chamber experiments

		Scenario 1 ^a	Scenario 2 ^b	Scenario 3 ^c	Scenario 4 ^d
Emission factor of building material ($\mu\text{g m}^{-2} \text{h}^{-1}$)	Caulking around windows	2.75	0.00	2.75	0.00
	Carpeting	4.41	3.25	4.41	3.25
	Vinyl flooring	2.14	1.66	2.14	1.66
	Painted walls and ceiling	0.12	0.00	0.12	0.00
	Area mat	78.17	10.43	78.17	10.43
Emission factor of furniture ($\mu\text{g m}^{-2} \text{h}^{-1}$)	Vinyl furniture	1659.08	10.02	1659.08	10.02
	Wooden furniture	73.79	1.33	73.79	1.33
	Wax on furniture	0.00	0.00	0.00	0.00
Emission factor of consumer product ($\mu\text{g g}^{-1} \text{h}^{-1}$)	Mothball	-	-	1015.18	841.14
	Deodorant block	-	-	17.93	0.52
	Air freshener	0.02	0.00	0.02	0.00
	Cleaning product	0.03	0.00	0.03	0.00
	Foam material	6.46	0.40	6.46	0.40
	Oil painting	0.14	0.01	0.14	0.01
	Shampoo	0.47	0.08	0.47	0.08
Indoor concentration ^e ($\mu\text{g m}^{-3}$)		187.5	4.9	219.7	30.5

^a Maximum emission factors of materials without mothball and deodorant block

^b Minimum emission factors of materials without mothball and deodorant block

^c Maximum emission factors of materials with mothball and deodorant block

^d Minimum emission factors of materials with mothball and deodorant block

^e See Table 3 for the house volume, air change rate and amount of materials used for the scenario analysis