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The effect of wet film thickness on VOC emissions from a finishing varnish

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Abstract

Finishing varnishes, a typical type of oil-based varnishes, are widely used to shine metal, wood trim and cabinet surfaces in Hong Kong. The influence of wet film thickness on volatile organic compound (VOC) emissions from a finishing varnish was studied in an environmental test chamber. The varnish was applied on an aluminium foil with three different wet film thickness (35.2, 69.9 and 107.3 μm). The experimental conditions were 25.0 °C, 50.0% relative humidity (RH) with an air exchange rate of 0.5 h^{-1} . The concentrations of the major VOCs were monitored for the first 10 h. The air samples were collected by canisters and analysed by gas chromatography/mass selective detector (GC/MSD). Six major VOCs including toluene, chlorobenzene, ethylbenzene, *m,p*-xylene, *o*-xylene and 1,3,5-trimethylbenzene were identified and quantified. Marked differences were observed for three different film thicknesses. VOC concentrations increased rapidly during the first few hours and then decreased as the emission rates declined. The thicker the wet film, the higher the VOC emissions. A model expression included an exponentially decreasing emission rate of varnish film. The concentration and time data measured in the chamber were used to determine the parameters of empirical emission rate model. The present work confirmed that the film thickness of varnish influenced markedly the concentrations and emissions of VOCs.

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1. Introduction

A number of investigations on the VOC emissions from indoor sources have shown that varnishes contribute to indoor air pollution. According to their chemical constituents, varnishes can be

classified into four types: bituminous varnishes, oil-based varnishes (film primarily by chemical reaction), spar varnishes and spirit varnishes (film primarily by solvent evaporation) (ASTM, 1995). Each type contains specific chemicals that aid in the application of the wax or oil to the coating surface. Varnishes can also be classified according to their polarity: oil-based and water-based. Finishing varnishes, a typical type of oil-based varnishes,

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are widely used to shine metal, wood trim and cabinet surfaces in Hong Kong. They are coatings designed not only with excellent drying properties, but also with high gloss and resistance to water and dirt.

Studies have been conducted to identify the chemical constituents in varnish products in the United States and European countries (Howard et al., 1998; Silva et al., 1999). Saturated aliphatic, cyclic hydrocarbons, aromatic hydrocarbons and oxygenated hydrocarbons have been identified both in oil-based varnishes and spirit varnishes. McCrillis et al. (1999) identified that the major VOCs emitted from oil-based varnishes were isobutanol, ethylbenzene, *m,p*-xylene, *o*-xylene and formaldehyde.

Studies on VOC emissions from wet materials like paints, varnishes or wood stains were usually conducted in inert substrates. Stainless steel, aluminium and glass plates were substrates normally recommended for testing due to their properties of non-adsorption and non-porosity. Several parameters, including temperature, relative humidity (RH), substrate, air exchange rate and air velocity, have been shown to affect the VOC emissions from the building materials in many literatures (Chang et al., 1997; Gunnarsen et al., 1993; Van der Wal et al., 1997; Wolkoff, 1998). Although some studies were conducted on the characteristics of VOC emissions from varnishes, most were focused on the substrate effect on VOC emissions (Howard et al., 1998). The thickness of film source is always an overlooked variable in emission testing of materials. Although the influence of film thickness in paint research was investigated for many years (Sullivan, 1975), only Clausen (1993) investigated the thickness effect on VOC emissions from waterborne paints. No study investigated the influence of film thickness on VOC emissions from varnishes.

Models are often adopted to predict VOC emissions from indoor sources. There are several types of empirical models used to describe such emission based on dynamic chamber testing (ECA-IAQ, 1992). The concentration and time data measured in the chamber are used to determine the parameters of empirical emission rate models. In particular, mass transfer models based on fundamental

theory for indoor air pollution sources provide a good fit between modelled and measured VOC concentrations. However, it is impossible to use these models because of the difficulty in estimating or measuring the large number of parameters required by these models (Tichenor et al., 1993; Dunn and Tichenor, 1988; Sollinger et al., 1993; Little et al., 1994). The first-order decay model (Tichenor et al., 1988; Mølhave et al., 1995; Zhang et al., 1997; ASTM, 1998, 1999) is the most commonly used model because of its simplicity and adequacy to represent the emission test data. It is suitable for predicting the short-term concentrations and emission rates from wet coatings.

In the last decade, many studies focused on the emission characterisation of volatile organic compounds (VOCs) from building materials (Nielsen et al., 1994; USEPA, 1994; Wolkoff, 1999). Zhu et al. (1999) reported large data sets of VOC emissions from building materials under dynamic chamber test conditions. The purpose of the emission characterisation was to find out the potential indoor pollutant concentrations, the emission rates and changes in emissions over time (Godish, 2001). The research was provoked by the wish to reduce the exposure of humans to toxic substances from building materials and, in general, to improve the indoor air quality (IAQ) by the use of environmentally sustainable materials.

The goal of this work presented here is to investigate the influence of the wet film thickness on the emissions of VOCs from a finishing varnish. Varnish was selected in the present study because of the potential exposure of painters and building occupants to high VOC concentrations following application. The emission data obtained could be used to estimate the population exposure to the VOCs emitted and to reduce the exposure through selection of appropriate indoor materials. A small dynamic environmental chamber with controlled climatic conditions was used to investigate the time dependence of VOCs emissions from the finishing varnish. A varnish was selected as the most representative product available on the Hong Kong markets. VOC emissions from the finishing varnish with regard to three different film thicknesses were compared and evaluated. A first-order decay model was used to obtain emission para-

meters and to predict the VOC concentrations in the chamber. The correlation between chamber measurements and emission modelling results was statistically evaluated.

2. Experimental

2.1. Materials

After investigation of available information on market sales, a typical finishing varnish was purchased at local retail outlets for the investigation of the VOC emissions. The varnish was a commercially available and typical product that would be purchased by homeowners for application to metal or woodwork in residences. The varnish was thinned in accordance with manufacturer's instructions. Aluminium foils with an area of 200×250 mm were painted with the three different quantities of varnish. The average weights applied were 1.51 ± 0.20 g ($35.2 \mu\text{m}$), 3.02 ± 0.20 g ($69.9 \mu\text{m}$) and 4.60 ± 0.20 g ($107.3 \mu\text{m}$). The application was conducted by a synthetic short-haired brush in order to obtain a surface similar to a painted metal surface. Simple gravimetric method by weighing immediately before and after application was adopted to calculate the average film thickness. Sample weight loss was determined by weight difference to the nearest 0.1 mg. The painting method has been demonstrated to provide reproducible film applications on the substrates. Aluminium foil was selected as a substrate because it may eliminate the possibility of any confounding substrate effect on the material surface. After preparation, the specimen was placed immediately on the floor in the centre of the chamber, and the experiment started when the chamber door was closed (time=0). Before application the aluminium foil was cleaned by acetone, followed by tap water and distilled water. It was then placed inside the chamber 12 h before the experiment started.

2.2. Sampling and analysis

An electropolished stainless steel environmental chamber (Lee et al., 2001; Kwok et al., 2001) of a volume of 2.38 m^3 was used. The material loading was $0.021 \text{ m}^2/\text{m}^3$. The experimental con-

ditions for all emission measurements in the chamber were representative of indoor environments and as follows:

- Temperature: 25.0 ± 1.0 °C;
- Relative humidity: $50.0 \pm 5.0\%$;
- Air exchange rate: $0.5 \pm 0.02 \text{ h}^{-1}$;
- Air velocity: $10.0 \pm 5.0 \text{ cms}^{-1}$ at 1 cm above the surface of the tested materials.

The chamber was purged by compressed air, which was passed through a clean air system with particle filters and activated charcoal to supply VOC-free air. Four mixing fans were installed at the bottom corners of the chamber to ensure adequate mixing of air. The temperature and RH were continuously monitored by portable Q-Trak monitor (Model 8550, TSI, MN, USA). The air velocity was measured by portable VelociCalc Plus monitor with hot wire anemometers (Model 8388, TSI, MN, USA). The air exchange rate was measured by flooding with carbon monoxide (CO) with concentration to 100 ppm inside the dynamic chamber. When the CO concentration was achieved, the CO supply was stopped. The concentration was then measured over time and correlated with the airflow rate. The CO concentration was measured with Gas Filter Correlation CO Ambient Analyser (Model 48, Thermo Environmental Instrument Inc., MA, USA). The analyser was based upon comparison of the detailed structure of the infrared absorption spectrum of the measured gas to that of other gases also present in the sample being analysed. The airflow rate was then monitored and checked by a flow calibrator (Gilian, FL, US). An internal mixing test using CO was also performed to assure the reproducibility of the results (ECA-IAQ, 1992). After each test, the chamber was cleaned by scrubbing the inner surfaces with acetone, tap water and distilled water. The chamber was then conditioned by purging pure air and heated to 25 °C and 50% RH for at least 48 h prior to application. The chamber background level was measured before each experiment. Any materials that will be used during the experiment, such as substrate and monitoring equipment, were included to account for the background concentration.

A sampling port with Teflon tubing was inserted into the centre of the chamber at 0.6 m above floor. Air samples in the chamber were collected through the sampling port to a SUMMA canister at a rate of 4.0–6.0 l/min. The flow rate through the canister was measured with rotameter. The rotameter was calibrated in the laboratory against a soap bubble flow meter. All samples were analysed within 48 h. The samples were analysed using a cryogenic preconcentrator and gas chromatography/mass selective detector (GC/MSD) (Model 6890/5973, Hewlett Packard, CA, US) by US-EPA TO-14A method to identify and quantify individual volatile organic compounds (USEPA, 1999).

2.3. Quality assurance and quality control

Identification and quantification of VOCs by GC/MSD were based on retention time and peak areas of the corresponding calibration standards, respectively. It was performed by matching spectra using the National Institute of Standards and Technology (NIST) mass spectra library and with external standard gas based on retention times. The instrument was calibrated using 40 standard concentrations covering the concentrations of interest for indoor work. Calibration curves for all measured compounds were made daily and had linear correlation coefficients (R^2) greater than 0.95. The experiment was performed in triplicate

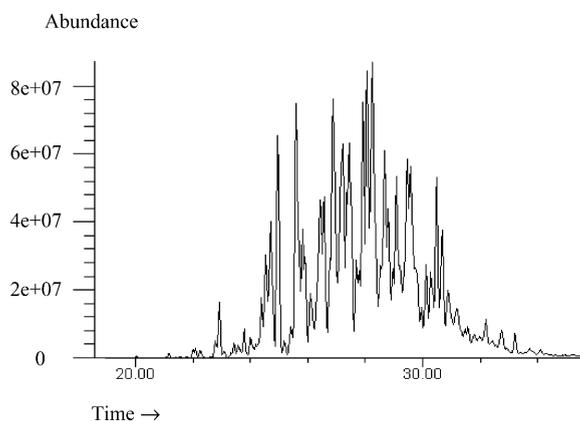


Fig. 1. Gas chromatograph of a finishing varnish.

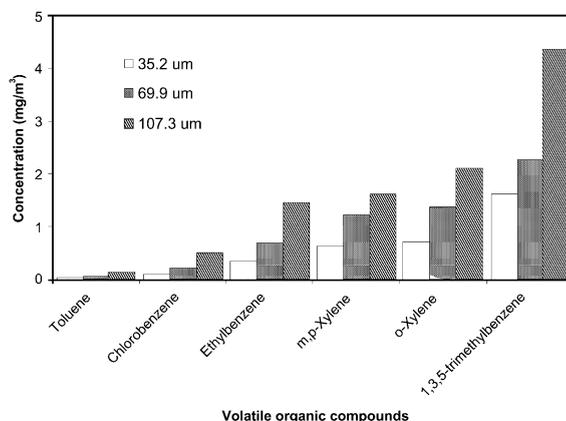


Fig. 2. The peak VOC concentrations from varnish under different film thickness.

for each varnish sample with same thickness. Duplicate analysis was performed for 10% of air sample randomly selected for precision checking. The precision of triplicate analysis was within 10%.

2.4. Modelling of VOC emissions

In the present study, VOC concentrations from the varnish emissions were fitted using a first-order decay model, instead of a double-exponential model previously used (Guo and Murray, 2000a; Guo et al., 2000b). It provided comparatively reliable estimates of emission parameters when VOC emissions from indoor materials were mainly controlled by evaporation.

The emission rates of VOCs from the varnish were calculated and analysed using a first-order decay model (Tichenor and Guo, 1991):

$$E(t) = E_0 e^{-kt} \quad (1)$$

where $E(t)$ = emission rate ($\text{mg}/\text{m}^2 \cdot \text{h}$), E_0 = initial emission rate ($\text{mg}/\text{m}^2 \cdot \text{h}$), k = first-order decay constant (h^{-1}). The mass balance for the chamber over a small time increment dt is:

mass in chamber = mass emitted – mass leaving chamber

Table 1
Vapour pressure of the major VOCs (1 atm and 25 °C) (Perry et al., 1984)

Compounds	Vapour pressure (mmHg)
Toluene	21.8
Chlorobenzene	38.9
Ethylbenzene	42.2
<i>m,p</i> -xylene	43.7
<i>o</i> -xylene	44.7
1,3,5-Trimethylbenzene	64.8

This can be expressed as:

$$VdC = AE(t)dt - QCdt \tag{2}$$

where V =chamber volume (m^3), A =sample area (m^2), Q =flow rate through chamber (m^3/h), C =chamber concentration (mg/m^3).

The chamber mass balance Eq. (2) with the source term defined by Eq. (1) was integrated and the initial concentration was assumed zero to give the equation:

$$C = LE_0(e^{-kt} - e^{-Nt}) / (N - k) \tag{3}$$

where L =material loadings (m^2/m^3), $N = Q/V$ =air exchange rate (h^{-1}).

The first order decay model used a non-linear least-square best-fit routine to analyse the chamber emission data (Statistica program, 1993). The two emission parameters E_0 and k were then obtained. The correlation coefficient (R^2) and the sum of

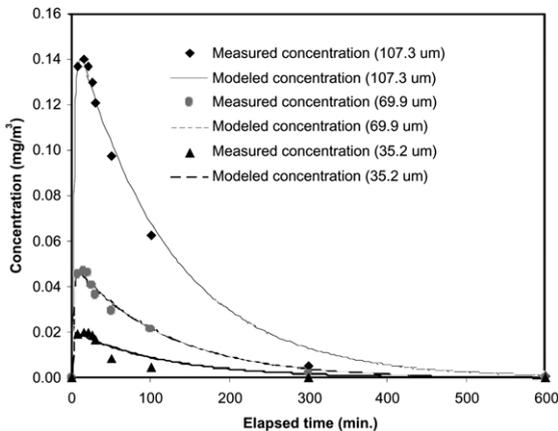


Fig. 3. The toluene concentrations from varnish under different film thickness with time after application.

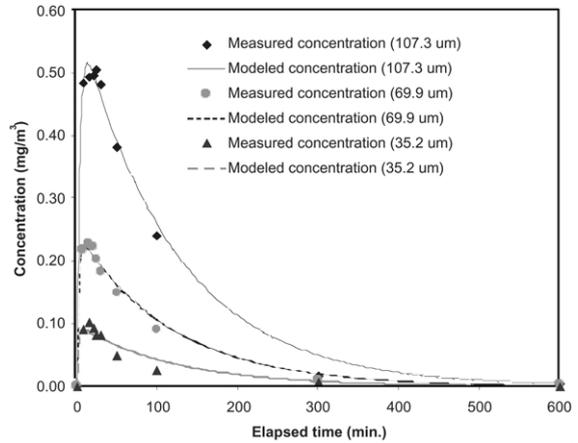


Fig. 4. The chlorobenzene concentrations from varnish under different film thickness with time after application.

squared error (SSE) between measured and predicted values were assessed automatically. The program used an exponential peering procedure to calculate the best estimates of the emission parameters (Serber et al., 1989). It was clear that models should be applied only to physical situations similar to those used to derive the model. With known emission parameters from indoor sources, the indoor air concentrations under specific materials loading (L) and air exchange rate (N) can be simulated.

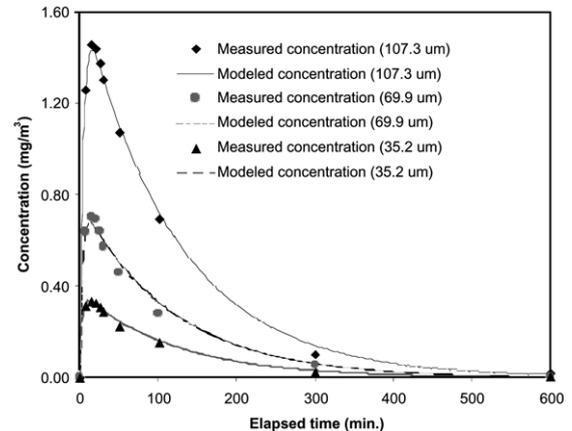


Fig. 5. The ethylbenzene concentrations from varnish under different film thickness with time after application.

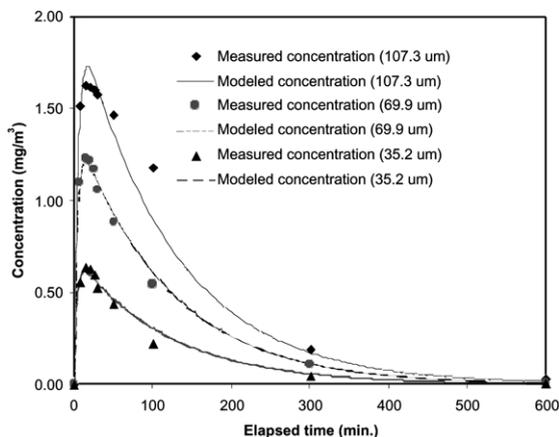


Fig. 6. The *m,p*-xylene concentrations from varnish under different film thickness with time after application.

3. Results and discussion

3.1. Thickness effect of varnish

The major VOCs emitted from the varnish were toluene, chlorobenzene, ethylbenzene, *m,p*-xylene, *o*-xylene and 1,3,5-trimethylbenzene. Fig. 1 shows that the major VOCs in the varnish emissions appeared at the end of the spectrum, which indicated that the varnish mainly emitted high molecular-weight VOCs commonly found in petroleum-related products.

The most abundant VOC was 1,3,5-trimethylbenzene, followed by *o*-xylene and *m,p*-xylene. The average concentrations of 1,3,5-trimethylbenzene, *o*-xylene and *m,p*-xylene were 4.39, 2.14 and 1.63 mg/m³, respectively, when the varnish with thickness 107.3 μm was applied. The peak concentrations of the VOCs under three different thicknesses are shown in Fig. 2. The linear relationships between the film thickness and the peak VOC concentrations were statistically significant (the linear correlation coefficients R^2 were between 0.92 and 1.00). The abundance of the major VOCs emitted was shown to be closely related to the vapour pressure. Boubel et al. (1994) found that vapour pressure can affect the rate of evaporation of organic compounds into the atmosphere. As shown in Table 1, the vapour pressure of 1,3,5-trimethylbenzene was the highest and that

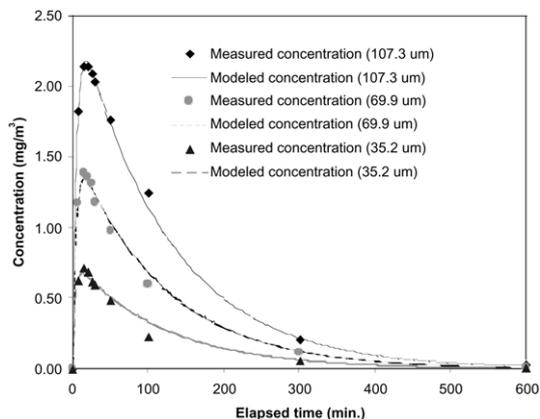


Fig. 7. The *o*-xylene concentrations from varnish under different film thickness with time after application.

of toluene was the lowest among six VOCs. The higher the vapour pressure of a compound, the higher the rate of evaporation and thus the higher the concentration found in the emission.

Figs. 3–8 show the temporal change of VOC concentrations measured in the chamber and derived from the model from the varnish emissions under three different thicknesses. The VOC concentrations increased rapidly to a maximum value within 30 min. After that, a systematic decrease of the concentration towards an equilibrium or undetectable was observed. The average film thickness of sample was 35.2, 69.9 and 107.3 μm. The

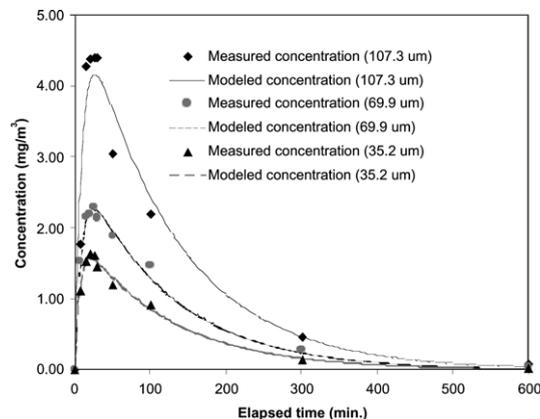


Fig. 8. The 1,3,5-trimethylbenzene concentrations from varnish under different film thickness with time after application.

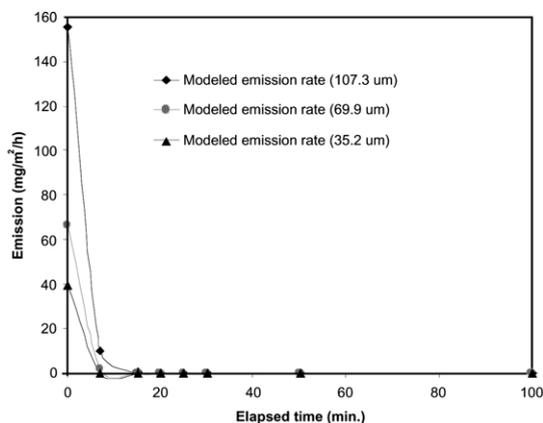


Fig. 9. The toluene emission rates from varnish under film thickness with time after application.

thicker the wet film, the higher the VOC concentrations. After 5 h of exposure, the VOC concentrations were reduced by 89.5% (for 1,3,5-trimethylbenzene) and 97.4% (for toluene) relative to the peak concentrations. After a period of 10 h, the majority of the VOCs were completely removed. The emission rate profiles of the major VOCs are shown in Figs. 9–14. The VOC emissions started at the maximum values and then decreased rapidly within 30 min (except 1,3,5-trimethylbenzene where its emissions decreased within 50 min).

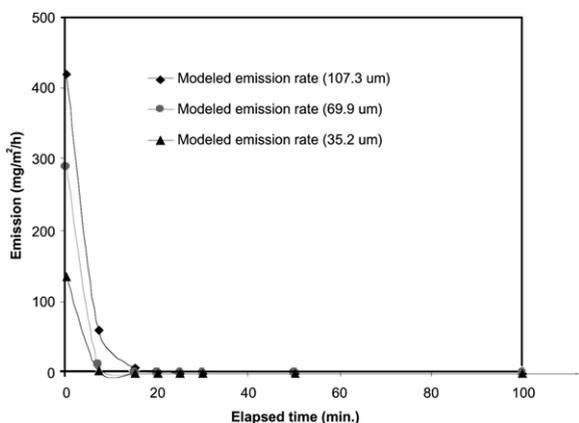


Fig. 10. The chlorobenzene emission rates from varnish under different film thickness with time after application.

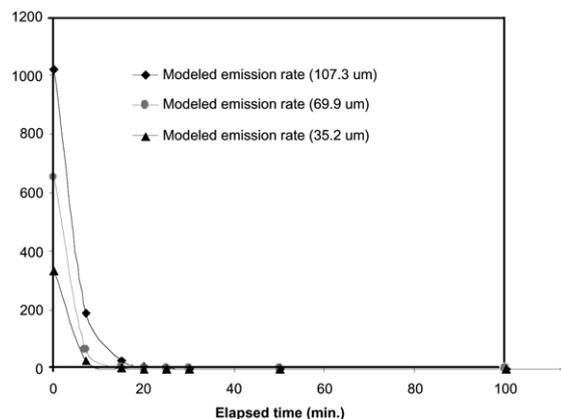


Fig. 11. The ethylbenzene emission rates from varnish under different film thickness with time after application.

3.2. Emission parameters of the model

The model-derived emission parameters for the varnish emissions under three different film thickness are reported in Table 2. The emission parameters can be obtained by using a non-linear regression curve fit implemented on a computer and fitting the measured concentration vs. time data to Eq. (3). The estimation used Quasi-Newton method to determine initial emission rate E_0 (mg/m²·h), first-order decay constant k (h⁻¹) and source strength E_0/k (mg/m²).

The correlation coefficient (R^2) for the varnish

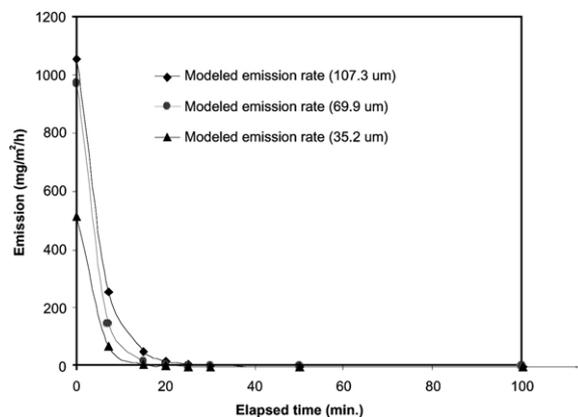


Fig. 12. The m,p-xylene emission rates from varnish under different film thickness with time after application.

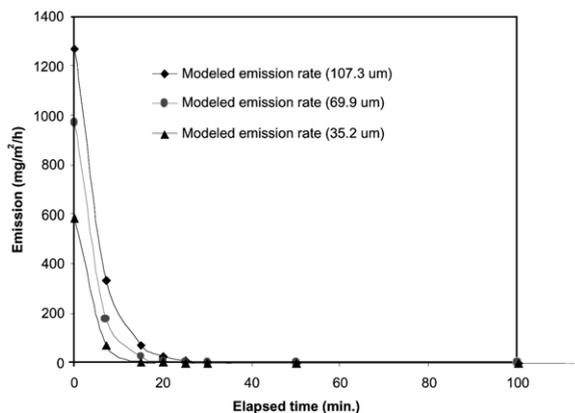


Fig. 13. The *o*-xylene emission rates from varnish under different film thickness with time after application.

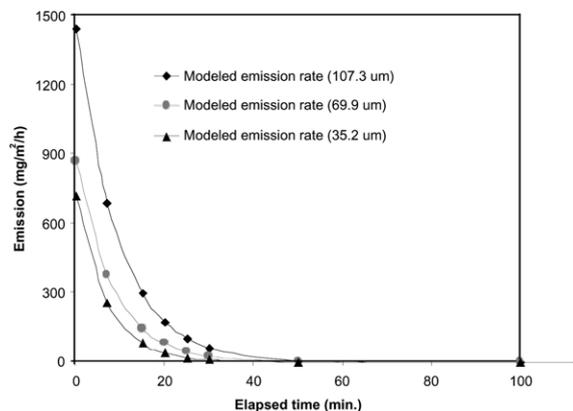


Fig. 14. The 1,3,5-trimethylbenzene emission rates from varnish under different film thickness with time after application.

emissions ranged from 0.918 to 0.998. The sum of squared error (SSE) between the measured and predicted VOC concentrations ranged from 4.61×10^{-5} to 1.61. Toluene had the smallest SSE while 1,3,5-trimethylbenzene had the largest SSE.

A value of zero for SSE indicated a perfect fit between the measured and predicted values.

The modelled parameters for the major VOCs are shown in Table 2. According to the model, E_0/k represented the total quantity of pollutants

Table 2

Model-derived VOC emission parameters for a varnish under three different wet film thickness

Thickness	E_0 (mg/m ² /h)	k (h ⁻¹)	E_0/k (mg/m ²)	R^2	SSE
107.3 μm					
Toluene	156.1	23.1	6.7	0.996	1.47E-04
Chlorobenzene	420.4	16.6	25.4	0.992	3.76E-03
Ethylbenzene	1024.2	14.4	71.4	0.998	6.41E-03
<i>m,p</i> -xylene	1055.2	12.1	87.4	0.974	1.19E-01
<i>o</i> -xylene	1270.5	11.5	110.8	0.998	1.57E-02
1,3,5-Trimethylbenzene	1444.8	6.3	228.3	0.949	1.61
69.9 μm					
Toluene	66.7	30.6	2.2	0.987	4.61E-05
Chlorobenzene	289.5	27.4	10.6	0.988	9.84E-04
Ethylbenzene	650.9	19.8	32.8	0.989	8.78E-03
<i>m,p</i> -xylene	970.1	16.3	59.6	0.995	1.12E-02
<i>o</i> -xylene	970.4	14.6	66.5	0.994	1.79E-02
1,3,5-Trimethylbenzene	870.9	7.2	121.6	0.994	4.75E-02
5.2 μm					
Toluene	39.9	43.9	0.9	0.918	6.08E-05
Chlorobenzene	135.3	31.8	4.3	0.935	9.74E-04
Ethylbenzene	335.6	20.8	16.2	0.995	9.08E-04
<i>m,p</i> -xylene	514.0	17.1	30.1	0.982	1.17E-02
<i>o</i> -xylene	585.2	17.7	33.0	0.989	8.25E-03
1,3,5-Trimethylbenzene	718.8	8.8	82.1	0.993	2.76E-02

emitted. The E_0 values of the VOCs ranged from 39.9 to 1444.8 mg/m²/h and the E_0/k values ranged from 0.9 to 228.3 mg/m². It indicated that increase in film thickness increased the E_0 and E_0/k values. The thicker the wet film was, the more the quantity of VOCs emitted. As shown in Table 2, increased film thickness decreased the k values. The results were similar to those found by Clausen (1993), in which the effect of an increased film thickness should decrease the emission decay constant. It implied that the thicker the wet film was, the longer the VOC emissions lasted.

4. Conclusion

The influence of wet film thickness on the volatile organic compound (VOC) emissions from a finishing varnish was studied using an environmental test chamber. The most abundant VOCs was 1,3,5-trimethylbenzene, followed by *o*-xylene and *m,p*-xylene. This research demonstrated that varnishes can be a significant source of VOCs in indoor environments, but that the exposures to these compounds were predicted to be of relatively short duration. The data showed that the major VOCs in varnish emissions were emitted during the first 10 h after application. The results also indicated that the film thickness of the varnish was closely associated with emissions of VOCs. The thicker the wet film, the higher the VOC concentrations and emissions. The change of VOC concentrations showed a good fit with results from a first-order decay model. The first order decay model provided reliable estimates of the initial emission rate and emission decay constant.

Decrease in film thickness reduced the initial amount of VOCs emitted and increased the emission decay constant. The results implied that high quality varnishes (i.e. low VOC emissions varnishes) should be used indoors. Only one or two thin layers of the varnish needs to be applied. VOC emissions from varnishes therefore can be effectively reduced.

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