

Material Emission Rates: Literature Review, and the Impact of Indoor Air Temperature and Relative Humidity

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An extensive literature review of research on the impact of indoor air conditions; temperature, relative humidity and surface air velocity on materials emission rates is presented. This paper also presents the results of an experimental work to study the impact of room air temperature and relative humidity on materials emission rates. The results indicate that both the temperature and relative humidity have a significant effect on the emissions from paint and varnish. In the case of varnish, the results were consistent with earlier results. However, the paint results show inconsistent emission behaviour. Further, for both materials, the individual compounds did not necessarily follow the same trend established for the TVOC. © 1998 Elsevier Science Ltd. All rights reserved

INTRODUCTION

Research conducted over the last few years indicates that indoor air pollution in non-industrial buildings is affecting human health and productivity in a significant way [1–4]. It has been suggested that a significant percentage of respiratory illnesses and lung cancers may be caused by avoidable indoor air pollution. Furthermore, a number of cross-sectional surveys have detected that significant proportions of the population suffer from eye and respiratory discomfort, headaches, and feelings of lethargy among occupants of buildings with poor indoor air quality [1, 5].

Evidence from a variety of building investigations and systematic studies suggest that many of the materials used in buildings, either as structural materials or as furnishings, are the main source of pollution, due to their large surface area and their permanent exposure to indoor air in non-industrial buildings [6]. Another potential source of indoor pollution may be from the thermal and acoustical insulation materials commonly used to line the interior of air handling units, reheat coils, variable air volume units, mixing and diffuser boxes and ducts. These materials are the source of Volatile Organic Compounds (VOC). Over two hundred VOC have been identified in the indoor environment [1, 7]. These studies also speculated on or implicated the importance of indoor environment parameters; temperature, relative humidity and surface air velocity on the material emission rate. As an example, higher temperatures have been consistently associated with greater dissatisfaction with the indoor environment, and may adversely affect productivity [8].

The effect of temperature may be a result of direct effects, or increased material emissions.

This paper first presents a comprehensive review of earlier work on the impact of indoor air conditions on material emission rates and then will report the results of an experimental study on the impact of temperature and relative humidity on the emission rates from two commonly used materials in buildings.

REVIEW OF PREVIOUS WORK

Material emissions are the result of several mass transport processes. The interaction of these processes commonly occurs, but their effects on material emissions are somewhat complex. However, the emissions can be considered to stem from two main processes.

Diffusion within the material

Diffusion of a compound through a material can be the result of a concentration, pressure, temperature or density gradient. Diffusion is described by Fick's Second Law [9]:

$$\frac{\delta C_A}{\delta t} = D(\nabla^2 C_A) \quad (1)$$

where $\delta C_A / \delta t$ = rate of change in concentration of compound A ($\text{mg}/\text{m}^3 \cdot \text{h}$), D = diffusion coefficient (m^2/h), and ∇^2 = the Laplacian operator of C_A (x , y , and z directions).

Each compound has its own diffusion coefficient, dependant upon its molecular weight, molecular volume, temperature, and the characteristics of the material within which the diffusion is occurring [9]. For a given sample, such as paint, the overall diffusion factor is very difficult to determine.

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Surface emissions

The surface emissions occur between the material and the overlying air as a consequence of several mechanisms, including evaporation and convection. As long as a concentration gradient exists between the two phases, surface emissions will occur. This phenomenon can be expressed as:

$$E_A = k_A(C_s - C_a) \quad (2)$$

where E_A = surface emission rate of compound A ($\text{mg}/\text{m}^2\cdot\text{h}$), k_A = mass transfer coefficient (m/h), C_s = concentration of compound A at the surface of the material (mg/m^3), and C_a = concentration of compound A in the overlying air (mg/m^3).

In liquids, the evaporation of a compound is a dominating process, and therefore, the vapour pressures of the compound and overlying air can be shown to be proportional to their concentrations in Eqn (2). The mass transfer coefficient is dependant upon the environment in which these processes are taking place, including the surface velocity and turbulence, surface characteristics, and the properties of the overlying air.

Tichenor *et al.* report that it has been determined that diffusion is not a controlling process in water-based paints [10]. Rather, evaporation (surface emissions) controls the emissions of volatiles even after a long elapse of time. However, Clausen reports findings that established that after the sample has dried, the emissions may be controlled by either evaporation (surface emissions) or diffusion depending on the volatile organic compounds involved [11].

Adsorption/desorption processes also need to be considered. In a real room, materials may adsorb compounds emitted by other materials, releasing them at a later time. This complicates the investigation of material emissions in real rooms. However, in chamber tests, the only materials present are the test sample and stainless steel. As stainless steel is inert, its adsorption effects can be considered negligible.

From a literature review conducted to date, most chamber tests have been conducted under one set of environmental conditions [12]. Levin stresses the need for emission data on one hand, and the necessity of carefully investigating the test conditions that produced data on the other hand [13]. A summary of these efforts, with the environmental settings used is shown in Table 1.

Small chamber tests

Bremer reported the emission variations with change in temperature, and concluded that the emitted substances were temperature dependent [14]. However, the data presented does not reveal a systematic comparison of the effect of the different temperatures on the emissions from a PVC flooring. Rather, they show results for three different PVC (poly-vinyl chloride) floorings, each tested at a different temperature.

In the experiments conducted by Colombo *et al.*, several levels of relative humidity were used, depending on the material sample, for preliminary testing of the influence of relative humidity on the material adsorptivity [15, 16]. The authors concluded no apparent influence of relative humidity. Contrary to this, other researchers

have found that temperature and relative humidity do cause variations in material emissions.

Sollinger *et al.*, conducted experiments on ten types of textile floor coverings using two chambers: a 33 l glass chamber and a 1000 l stainless steel chamber [17, 18]. Both were equipped to vary temperature from room to 60°C in the glass chamber, and 70°C in the stainless steel chamber. Relative humidity and air change per hour could be varied within a wide range. The experiments were conducted under static and dynamic conditions. The purpose of the static experiments was to identify emitted compounds and to determine their dependence on temperature and relative humidity. In their study, it was found that temperature variations did not have a significant effect on the concentrations of volatile compounds, but showed a stronger effect on less volatile compounds. For example, the concentration of styrene did not change with increasing temperature but the less volatile compound, benzothiazole, was strongly influenced by temperature variations. The relative humidity was noted to have little effect on all compounds with the exception of aniline. The report does not record the ranges of temperature or relative humidity used. Tucker, (1988), notes the same varied effect of relative humidity and attributes that only products which contain highly polar compounds require testing for relative humidity effects.

The European Collaborative Action, called "Indoor Air Quality and its Impact on Man", published reports [19, 20] on an inter-laboratory comparison of small chamber measurements [21]. The importance of this experiment was based on the upcoming preparation of databases on material emissions, providing an even further reason to standardize small chamber tests. In this report, the effects of temperature, air exchange rate, and compound volatility on material emissions were measured. Most tests were carried out under controlled environmental conditions, namely 23°C, 45% relative humidity and 1.0 ACH. In the case of temperature, tests were carried out at two laboratories and results showed that with an increase in temperature from 23 to 60°C, the emission rate, both by concentration or weight loss for n-dodecane, increased. This was after the removal of the source; which, it was concluded, indicates the possibility of sink effects.

The U.S. Environmental Protection Agency (EPA) has performed tests to characterize the emissions from a variety of sources of indoor air contaminants and to identify the effects of various factors on the emission rates [22]. These factors included temperature, relative humidity, air changes, and chamber loading. The testing facility included two 166 l stainless steel environmental chambers. Environmental parameters were controlled within the following limits: temperature $\pm 0.1^\circ\text{C}$; relative humidity $\leq 2\%$; airflow $\leq 0.7\%$ of reading. All materials were first analyzed using headspace testing to identify the chemical compounds emitted. The results of the effects of air change rate and temperature on moth crystal emissions was reported. Two temperatures, 23 and 50°C; two relative humidities, 20 and 50%; and several ACH, from 0.25 to 2.0 ACH, were tested. The results show that for para-dichlorobenzene (main component in moth crystals) an increase in temperature

caused increased emissions; as did increased air change rates. The author states that the air change was especially of importance in its effect on wet materials. Emissions decreased sharply during the first hours of application, and was especially aided by a higher air exchange rate. The effects of relative humidity during these tests, were not reported [23–26].

Gebefugi and Korte studied the chemical accumulation on different textile materials, such as cotton, acrylic, and lambs wool, placed in a small chamber ($50 \times 50 \times 50$ cm) when the walls of the chamber were coated with such products as Pentachlorophenol (PCP), Lindane, Fumencyclohexane, and Dichlofluenid [27]. The textile samples were then analyzed by capillary gas chromatography with electron capture and flame ionisation detectors within 72 h of their exposure. The samples were tested at three different temperatures, 18, 23, and 28°C without ventilation. The results show that adsorption increased with a temperature increase. These reported results were for tests conducted using Fumencyclohexane. The group also noted that higher relative humidities caused higher adsorption by the textile samples, although no results were published.

Emission data related to formaldehyde have been extensively researched. The Housing and Urban Development Agency (HUD) in the U.S.A. has developed standards for formaldehyde use in pressed wood products. These standards were based on chamber tests and a mathematical model, which relates formaldehyde concentrations at 25°C , 50% relative humidity to those recorded in chamber tests at various temperatures and relative humidities. In this way, the predictions show increasing concentrations of formaldehyde with increasing temperature and relative humidity. Other models have been developed which relate formaldehyde concentrations to more factors, such as air change rate and product loading. One such model, developed by Matthews *et al.*; [28], predicts increasing concentration with increasing ACH and with decreasing product loading [29].

Levin reports on the results obtained while evaluating building products and materials during the design of a large office building situated in California [30]. The project involved reviewing materials/products to identify those most likely to emit contaminants; screening these materials using literature and manufacturer's data; testing selected material to determine chemical content, emission rate, or change in composition due to environmental exposure, and to make recommendations to the building owner and architect for material selection, modification, and handling to reduce indoor air contamination. When testing the materials, the bulk testing, environmental chamber and headspace air sampling methods were used. The effect of temperature variations on emissions using headspace and chamber testing was evaluated. However, the author reports that the number of temperature tests and the use of the results are limited. Two temperatures were used, 23 and 37°C . The chamber was made of sheet metal, 1.7 m^3 in volume with controlled airflow and temperature. Air change within the chamber was typically 12 h^{-1} . Humidity was not controlled during testing, though, it was recorded to be between 50–55% for most test runs. The author did not present the results for the temperature effect tests because insufficient data was obtained to

inspire enough confidence in the results obtained and in the testing facilities used.

Pickrell *et al.*, studied formaldehyde emissions from different products to evaluate the influence of chamber loading, relative humidity, temperature, and multiple products [31]. Samples of a particle-board, plywood, insulation material and carpet were tested. A 0.45 m^3 dynamic chamber was used. Temperature was varied to maintain at 25 or 35°C . The relative humidity was maintained at either 40% or 90%. Results show that the change in temperature or relative humidity did not have a significant effect on formaldehyde emissions for the particle-board or plywood. The authors explain that these results are contradictory to other researchers but explain that their product loading was higher than that normally used by other researchers.

NASA (National Aeronautics and Space Administration) in the U.S.A. has one of the most extensive databases, with over 5000 entries (in 1989), collected on material emissions [32, 33]. These products are strongly related to the spacecraft industry, including those used in construction and operation, as well as those that may be carried by personnel [34]. The testing method used by NASA involved a small chamber of, at least, 2 l in volume, commonly 4 l was used. The chamber was heated to a temperature of 49°C for a 72 h duration. The material was then analyzed; i.e. air samples within the chamber were taken and analyzed at room temperature [33, 34] and it was reported that the emission testing used by NASA was somewhat conservative for several reasons including the high temperature of 49°C used, the reduced pressure of 12 psia used, and that all material samples used were freshly prepared [33].

Baechler *et al.* compared some of the results recorded in the NASA database to other results obtained when similar products were tested by other researchers in small chambers or field tests [34]. The investigation concluded that the emission rates recorded in different studies were significantly different. The authors recognized some shortcomings involved in such a comparison. These included: differences in experimental facilities; NASA used a temperature of 49°C while others used room temperature; NASA did not consider relative humidity; comparisons with field measurements is made difficult because of the dynamics of the temperature and relative humidity in a true life building.

Zhang and Haghighat designed a velocity-controlled test chamber, to study the impact of surface air velocity on the emission rate [35]. The test chamber can generate a uniform flow field for a wide range of air velocities, turbulence fluctuations, and relative humidity and temperature variations. A series of tests from a constant source were conducted. Both the theoretically derived relationship and test results show that emission rates are a function of surface air velocity [36]. The relationship derived was further validated using the experimental results from literature [37].

Field tests

Shriever and Marutzky studied VOC emissions from varnished parquetry floors [38]. They compared measurements and results obtained from three sources, in two real-life rooms and in chamber testing. The authors did

Table 1. Summary of small chamber test conditions and procedures conducted to date

Reference	Temperature (°C)	Relative humidity (%)	Volume and material of chamber	Product loading (m ² /m ³)	Air change rate (ACH)	Conditions	
						Sampling procedures	Objective/purpose of experiment
[43]	23	50	52.7 l stainless steel	1.45	1.0	Emissions measured during first 24 h following application; paint applied with brush on one side of glass plate (828 cm ²) Alkyd paints: samples taken at 1, 4, 8, 24 h sample volumes of 0.1–10 ml Latex paints: extracted with methylene chloride; purified air used 96 h exposure time	Comparison of 7 methods for analyzing paint emissions, one of which was small chamber
[53]	23 ± 1	50 ± 5	0.05 m ³ stainless steel	0.41 ± 0.02	1.0 ± 0.05		Emissions from softback and hardback carpets
[54]	23	45	0.28 m ³ n/m ^a	1	0.5	VOCs sampled by drawing 1 l of air through Tenax cartridge; desorbed 20 min at 250 °C; GC program 0–280 °C, 4 °C/min Emissions tested over several weeks, during which time samples stay in climatic chamber	Analyse emission of formaldehyde, vinyl chloride, VOCs and plasticizers from different wall coating materials Tested PVC flooring
[14]	23, 30, 40	45	100 l cylindrical stainless steel	0.4	1.0		
[41]	23	45	1 m ³ stainless steel	0.17	1.0	Purified air; velocity = 0.3 m/s vertical; air samples taken at exhaust; drew 125–3000 ml of air through adsorption tubes Synthetic air used; 24 h sampling period; air flow = 100 ml/min; tested 4 weeks and 26 weeks after sample preparation Purified air used	Analyze wall paint emissions as a function of time and effect of material on which applied Tested VOC emissions from different floor coverings
[55]	23 ± 2	50 ± 5	FLEC ^b	n/m	171		
[56]	23	45	Stainless steel, glass, and teflon chambers 0.2 m ³ ; stainless steel	n/m	1.0		Tested VOC emissions from different floor coverings
[57]	23 ± 2	n/m		1	1.0	General BR1 (Building Research Institute) control method; 2 h sampling time	Tested VOC emissions from floor and wall coverings, insulations, paint, glue, furnishings
[58]	22	—	1.9 m ³ ; stainless steel	2.2	0.5	Ventilated with air that is filtered with carbon; 15 l of air samples absorbed on porous polystyrene filters using a flow rate of 1 l/min	To evaluate whether it is possible to re- establish concentrations of VOCs of a room in a small test chamber by transferring samples of predominating building materials from the room to the chamber VOC emissions from wall coverings, insulation materials, flooring
[59]	23	45	1 m ³ ; stainless steel	0.41	0.5	Paint allowed to dry for 2 weeks before measuring; samples conditioned in chamber for 2 days before testing; paint applied to gypsum board; sample volume 10 l; sampling time 30 min	

[19, 20]	23	45	two 450 l glass chambers	0.4 (particle board); 1.4 (gypsum board); 0.2 (plywood)	0.25	Three building assemblies were tested: particleboard with glued on carpet, gypsum board with wallpaper pasted on both sides, and a plywood sandwich with one layer stained and painted with a polyurethane lacquer	Compared the number of compounds emitted and their magnitude for the three building assemblies
[15, 16]	23 ± 0.5°C	< 10 to 70 ± 5%	2 glass chambers each 0.45 m ³ ; 1 stainless steel, 0.28 m ³	1.1 for dynamic tests; 1.8 for static testing	0 to 1	Sample of carpet, blown vinyl wall coating, and gypsum board	Tested the adsorptivity of common building materials to VOCs, such as n-dodecane, n-decane, 1-4-dichlorobenzene
[60]	n/m	50	FLEC	n/m	n/m however, flow rate specified as 100 ml/min	12 linoleum products; compared emissions after 24 h in FLEC and after one month's storage in a well ventilated room	Purpose was to identify emission characteristics of linoleum products; and to establish a link between emitted VOCs and odour perception
[61]	23 ± 0.5	45 ≤ 1 (3 products) 50 ≤ 1 (2 products)	n/m	n/m	0.507	Test equipment equipped with devices capable of controlling temperature, relative humidity, and air flow rate	Studied the VOC emissions from five household products by chamber testing and compared these results with those obtained from headspace analysis
[62]	23	50	55 l	various loadings depending on material	0.5	A dynamic chamber test procedure was followed; chamber under positive 0.05" water pressure; 37 material samples tested representing both conventional and recycled (Build Green) building materials; examples are carpet, carpet under-cushion, insulation, cabinetry, counter tops, and drywall	Compared the emissions from the conventional building materials with those of the Build Green materials
[63]	23 ± 1	50 ± 5	n/m	Various loadings depending on material	0.3 ± 0.0015	37 materials were tested for VOC emissions; 20 materials were tested for formaldehyde emissions	Developed an environmental chamber test procedure; used these to test materials to establish a data bank for selection of lower emitting materials, and to provide data for IAQ computer simulation
[22, 64]	37 ± 3 (source chamber) 24 ± 2 (exposure chamber) 23 ± 0.2	50	Four 10 gal (38 l) glass aquariums	n/m	n/m	Samples of in situ carpet were used: samples were heated to 35–70°C	EPA wished to replicate testing conditions achieved by Anderson Laboratories when exposure to carpet emissions killed laboratory mice
[65]	45 ± 3	45 ± 3	1 m ³ stainless steel	1	0.5 ± 0.1	Ventilated with clean filtered air; after each test temperature was raised to 110°C to eliminate traces of absorbed substances	To compare the emissions from 12 textile floor coverings
[51]	22	50	FLEC	n/m	507	Air velocity was 1 cm/sec; samples tested over 1 week duration	Compared emissions from 10 cleaning agents
[66]	21 ± 1	35 ± 5	1 m ³ stainless steel	1	1		To develop a quick evaluation technique for dynamic sorption effects of indoor composite materials
[40]	22	40 ± 6	4 aluminium boxes each 0.3*0.45*0.3 m	n/m	41.2	Different air velocities were used (0.05, 0.5, 1.0, 2.0 m/sec); samples of chipboard, carpet, rubber, and straw mat were used; used trained panel of 12 judges to evaluate air out of the box chambers	To evaluate influence of air velocity on perceived air quality

^a n/m: no mention of information in the report/paper.

^b FLEC stands for field and Laboratory Emission Cell; it is circular in shape with an internal diameter of 150 mm and made of stainless steel.

not control the temperature in the true-life rooms, but recorded them to range from 19 to 25°C in one room and 20 to 30°C in the other, and hold that this variation would have some effect on the VOC's reported, although this is not quantified in any way. The chamber tests were conducted in a 1 m³ chamber to evaluate formaldehyde emissions. The test was conducted at 23°C, 45% relative humidity, and 0.5 ACH.

Sensory tests

Etkin reports that material emissions vary inversely with relative humidity [39]. Thus as relative humidity is increased, emissions from materials, such as furnishings, carpeting and wall coverings are decreased. These were based on sensory test results.

Iwashita and Kimura studied the effect of surface air velocity on material emissions using sensory evaluation methods [40]. Four chambers were used, each corresponding to the four surface velocities (0.05, 0.5, 1.0 and 2.0 m s⁻¹) tested. These surface velocities were controlled by a fan installed within each chamber. Samples from four different materials; chipboard, carpet, rubber, and straw-mat, were tested. Temperature, relative humidity and air exchange rate within the chamber were kept constant at 22 °C, 40–60%, and 41.2 ACH, respectively. A trained panel of twelve judges evaluated the perceived air quality in each chamber. The results from this study show that the perceived air quality (and in turn, the material emission) increases with increasing surface air velocity. More specifically, increasing the surface velocity from 0.05–2.0 m s⁻¹ leads to an increase of close to 50% in perceived air quality. The results also showed that the rate at which the surface emissions increased varied between the four materials.

General findings

Gehrig *et al.* concluded that the characterization of emissions from building materials in real rooms is very difficult [41]. Many parameters influence the VOC concentrations, including furnishings, fluctuating temperatures, relative humidity, air changes, indoor activities, and sink effects.

While chamber testing was not used in the study conducted by Van der Wal *et al.* interesting results were discovered from the experiment on thermal insulation [42]. Results show that at elevated temperatures, 50°C, odours from wet insulation (insulation which may be exposed to environmental moisture) were significantly greater than when dry. Also the increased presence of aliphatic and aromatic aldehydes was noted under the same conditions. Quantities were almost one hundred times greater. The researchers concluded that the binder material of the mineral wool decomposed faster under wet and elevated temperatures.

Fortmann *et al.* compared seven different methods for analyzing the emissions from architectural coatings, where paint was predominantly used [43]. One of these methods was the small chamber. The research concluded that additional development in the area of evaluating methods of VOC emissions was required.

Plehn evaluated the solvent emissions from 25 low polluting paints and varnishes [44]. The objectives of his study were threefold: (1) to determine the solvent

composition of samples via headspace analyses; (2) to measure solvent emissions via chamber tests; and (3) to measure the emissions in a real-life situation. The chamber tests were conducted in a 17 m³ chamber. The walls of the chamber were covered by aluminum foil to avoid adsorption/desorption effects. The air change through the chamber was 0.8 ACH. The product loading was 100 g of paint for every 1 m² of aluminum foil used. The author found that the paints labelled as low-polluting had significantly lower emissions than the regular paints.

De Bortoli *et al.* report on a second inter-laboratory comparison, conducted by the European Collaborative Action, involving the comparison of the emissions from paint in small chamber tests conducted in 18 laboratories from 10 countries [45]. The paint was a waterborne styrene/acrylic paint, applied onto stainless steel plates with a nominal wet thickness of 200 µm. All laboratories met the following test conditions: chamber loading of 0.5 m²/m³, temperature of 23°C, relative humidity at 45%, 1 ACH, and an air speed of 10 cm s⁻¹. Chamber volumes varied between 35 cm³ to 1.47 m³. The results showed that there were considerable variations in the results from the different labs. It was concluded by the authors that these differences can be attributed for the most part to the paint sample thickness/uniformity and to the GC/MS analysis. For the latter, the laboratories were permitted to use their preferred methods.

Bjorseth and Malvik have investigated the number of components in water based paints which have been identified as causing allergic or irritating effects [46]. Three latex and one acrylate paint were tested. The authors used a 100 l stainless steel chamber equipped with small internal fans. The test conditions were 23 ± 1°C; 50 ± 5% rh; 0.5 ACH; and 1.2 m²/m³ product loading. The paints were applied to aluminum substrates. The results showed that the concentrations and, in some cases, the presence of the problem components decreased considerably in the more recent water-based paints as compared to water-based paints which have been on the market for a longer time.

Jorgensen *et al.* studied the influence of wall material on the emissions from paint [47]. The substrates tested were aluminum, gypsum board, and gypsum board with wall coverings. In addition, two different paints were tested, both of which were water-based. One was an acrylate paint, while the other was referred to as a "natural" paint. The paints were applied to the substrate and left at ambient temperature for 1 h. Following this the samples were placed inside a glass test chamber for 23 h. The chamber was conditioned at 23°C, 35% rh, 1.0 ACH, and 0.15 m s⁻¹ air velocity. The emissions were then tested using the Field and Laboratory Emission Cell (FLEC) [1]. After the initial 24 h conditioning period, the FLEC was placed onto the painted area and samples were taken. The sampling conditions were 23 ± 0.5°C, 48 ± 3% rh, and 507 ± 2 ACH. Results showed that the emissions from the aluminum substrate were initially higher than for the other tested materials, but with time (over 30 days) the emissions were lower. Results also showed that the "natural" paint emission profile was considerably different from conventional emission decays.

Myers *et al.* conducted an intensive study on the relationship between material emissions and the ventilation rate, as well as the product loading factor [48]. They found that the emission rate was proportional to ventilation rate and the loading factor.

Knudsen *et al.* identified that surface air movement has a non-negligible effect on material emissions [49]. Due to the limitation of the experimental facility, the authors could only qualitatively report that higher emissions from carpets and other dry materials occurred at higher velocities.

EXPERIMENTAL SET-UP AND METHODOLOGY

A small stainless-steel test chamber was built to evaluate the impact of indoor air temperature and relative humidity on the material emission rate. The chamber is 52.44 l in volume and made in accordance with the ASTM guideline D-5116-90 [50]. After some initial tests, the test chamber design was slightly modified, due to observation of a large variation in relative humidity during the placement of the sample in the chamber (removing the entire back plate). As a consequence, a slot (12.25" by 0.25") with cover (15.5" by 2.5") was machined at the base of the back face to allow for easy and quick insertion of the sample during testing. The air supply was provided by cylinder air of a pure medical breathing grade and the temperature control was achieved by placing this facility inside an environmentally controlled chamber: 2.29 m by 2.27 m \times 2.26 m. The desired level of relative humidity was achieved by passing one portion of dry air from the cylinder through a bubbler before introducing this to a second portion of purely dry air inside a mixing chamber.

Experiments were carried out under steady-state conditions. Pure fresh air was allowed to flow through the

chamber for a minimum of 12 h prior to each. The steady-state condition was also ensured by recording the air temperature and relative humidity during this period using a Protimeter Dew Point Meter (DP 989M). Figure 1 shows the schematic of the experimental set-up.

The tracer gas decay test was performed to test the air mixing in the test chamber. Air samples were collected at positions just above the surface of the sampling area and at the outlet port. The results indicated that the air in the chamber can be confidently considered to be mixed.

A paint and varnish were chosen in this study because of their extensive use in all buildings. Product loading is defined as the ratio of material surface area to the volume of the chamber it is occupying. It has been shown that the product loading effects the emission rate determined through small chamber tests. As such, the product loading should remain constant for all tests if it is to be eliminated as a variable in this study. Product loadings of 0.8 and 0.33 m²/m³ were chosen for paint and varnish, respectively.

A stainless steel substrate was chosen as it is an inert material. The stainless steel plates, however, could only be used for one application. Thus, it was decided to use a polyethylene wrap, containing no plasticizers, as a protective cover over the steel. The wrap itself was tested for emissions and yielded nothing. It is recognized that the substrate, hence the polyethylene wrap, has an effect on the emissions from the material samples. The extent of this effect is too complicated to predict or determine. For the purposes of the experiment, however, the interest lies in determining the general emission trends established by varying temperature and relative humidity. The impact of the substrate is common and relative to all tests performed. The paint or varnish was applied using a 2" polyester bristled brush.

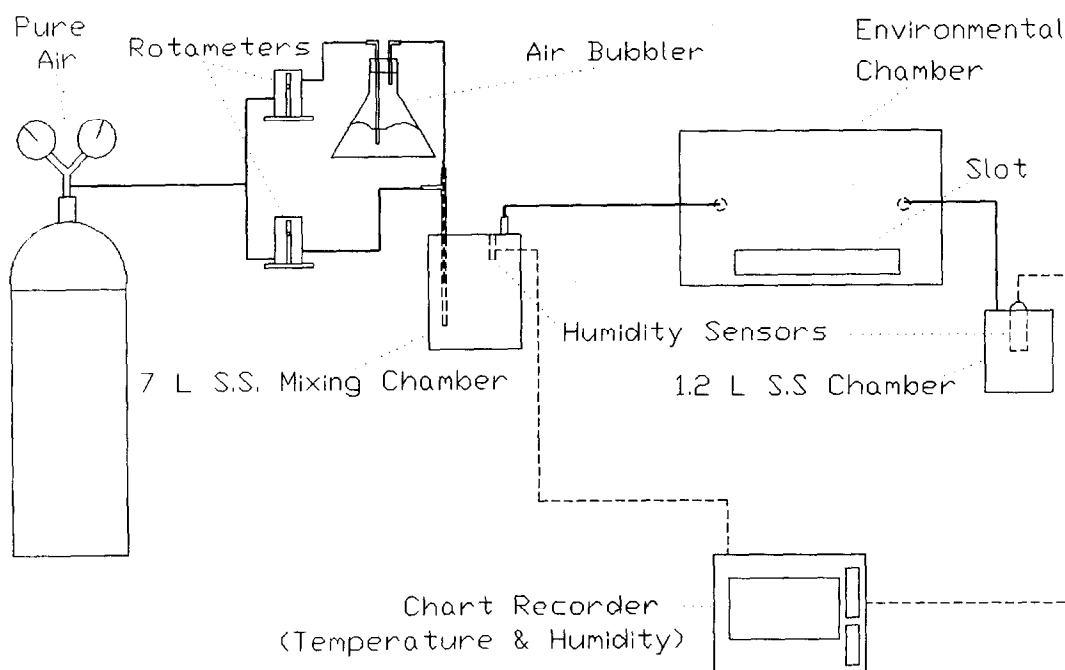


Fig. 1. Schematic drawing of the experimental test set-up.

Air samples from the chamber were taken at varying time intervals, depending on the environmental test conditions. At a minimum, a sample was taken at the following time intervals immediately following the insertion of the sample into the chamber: 15 minutes, 1, 2, 3, 4, 8, 12, 24, and every subsequent 24 h until the emissions curve reached steady state. The air samples were drawn from the exit port using a calibrated pump. The pump was calibrated to 100 ± 2 ml min⁻¹, and each sampling period lasted twenty min for the paint samples and ten min for the varnish samples, thereby drawing an air sample volume of 2 l and 1 l, respectively, from the chamber. The sampling tubes were multi-sorbent, consisting of glass beads/150 mg 20/35 mesh Tenax—TA/Ambersorb/Charcoal. The samples were thermally desorbed and analyzed using the Hewlett Packard 5890 Series II gas chromatograph. The chromatograph was equipped with a flame ionization detector. Prior to use, the chromatograph was calibrated using pure standards of known solution. They were from the alcohol, ketone, hydrocarbon and aldehyde families, as well as a premixed VOC standard.

The method used to determine Total Volatile Organic Compounds (TVOC) values present in the samples was a semi-quantitative method. Samples of the paint and varnish obtained from headspace analysis were sent to the National Fire Laboratory—National Research Council Canada for mass spectrum analysis. The reference response factor for toluene, identified as a component in each of the paint and varnish, was used to determine TVOC values.

DISCUSSION

Varnish

The varnish selected was a polyurethane plastic finish varnish for interior or exterior use. It has a drying time of within 8 h. All tests were conducted using a 2.0 gram

sample weight, a product loading of 0.33 m²/m³, and an air exchange rate of 1.2 ACH.

Temperature results. Figure 2 shows the TVOC emission rate profile for varnish at three air temperatures, 15, 25 and 35°C, and approximately 3% relative humidity. While the overall TVOC curve is generally shown to follow expected trends, it was decided to investigate the effect of temperature on individual compounds. These individual compounds were positively identified as components in the varnish tested. The chosen compounds include toluene, m, p, xylene, ethylbenzene, and 1,3,5-trimethylbenzene. The curves for these compounds (Fig. 3 depicts toluene), can be generalized in that their emission rates reach peak values very early (0–2 h) and are near depletion by the sixth hour. However, with the exception of 1,3,5-trimethylbenzene, the emission rates recorded for the 15°C test are higher than at 25°C, and in some instances, the 35°C, tests. This is not consistent with the TVOC curve. The curve for 1,3,5-trimethylbenzene is more akin to the TVOC behaviour. As its emission rate is somewhat higher than the other compounds, it may be a driving/influential factor in establishing the TVOC curve, in spite of the different patterns shown by the other compounds. In fact, then, it is truly impossible to generalize the behaviour of individual compounds based on the TVOC curve.

When comparing the maximum emission rate recorded for each compound to the limiting values summarized in Table 2, it can be seen that the measured values are well below these specifications. The values shown in Table 2 are meant for industrial environments, and are therefore, applicable to the workers applying these materials during the construction or renovation phases. It should be noted, however, that some renovation work can be conducted in a partially occupied building, in which case, measures should be taken to ensure that the higher levels of contaminants are not carried to occupied zones via the

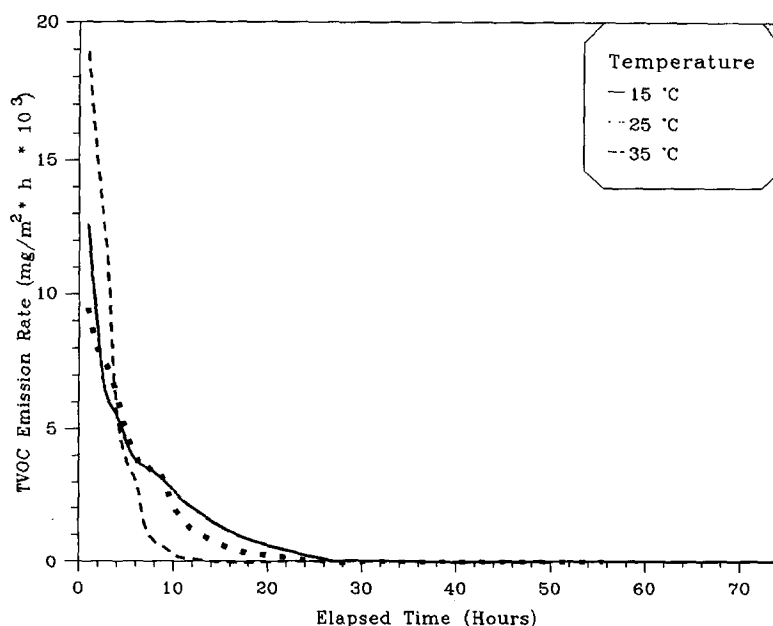


Fig. 2. TVOC emission rate vs time profiles for varnish at 1.2 ACH and temperature 15, 25, and 35°C.

Table 2. Comparative table of existing IAQ guidelines [20, 39]

VOC	ACGIH ^a TLV-TWA ^f (mg/m ³)	OSHA ^b PEL ^g (mg/m ³)	NIOSH ^c REL ^h (mg/m ³)	WHO ^d TLV ⁱ (mg/m ³)	MAK-Commission ^e MAK ^j (mg/m ³)
Benzene	32	3.1	0.32	30	16 (TRK) ^j
Toluene	377	375	375		380
Ethylbenzene				433	440
m, p-xylenes	434	435		435	440
Styrene				215	85
Heptane	1640	1600	350	1600	2000
Octane				1450	2350
Nonane				1050	
iso-propyl benzene					245

^a ACGIH: American conference of governmental Industrial Hygienists.

^b OSHA: Occupational Safety and Health Administration (U.S.).

^c NIOSH: National Institute of Occupational Safety and Health (U.S.).

^d WHO: World Health Organization.

^e MAK-Commission: Federal Republic of Germany.

^f TLV-TWA: average concentration to which workers may be repeatedly exposed over a normal 8-h day/40-h week without adverse health effects.

^g PEL: permissible exposure limit.

^h TLV: threshold limit value.

ⁱ MAK: maximum workplace concentration.

^j TRK: No MAK values are set for confirmed human carcinogens. Instead technical guidance concentrations are used.

HVAC system. In spite of this, it may be worth noting that though the concentrations were near depletion at test end, when the chamber door was opened, a strong odour was still detected.

Relative humidity results. Tests were conducted at three different inlet air relative humidity levels, $3 \pm 3\%$, $32 \pm 3\%$ and $62 \pm 3\%$, with air temperature at 25°C . The outlet relative humidity levels were also recorded and they were very near the inlet conditions for the duration of each test.

Figure 5 shows the TVOC emission rate profile for

three levels of humidity as a function of time. As shown in this figure, highest emission rates are recorded for the highest humidity level.

Unlike the TVOC, toluene, m,p-xylene and 1,3,5 trimethylbenzene show higher values for the 32% case than for the 62% case. Figure 6 depicts the curves for toluene. This again shows that trends established for the TVOC do not necessarily stand for individual compounds. The curve for ethylbenzene (Fig. 7) shows that the lowest concentration occurred for the 62% case. The values measured for the 3% and 32% cases are quite similar in order of magnitude.

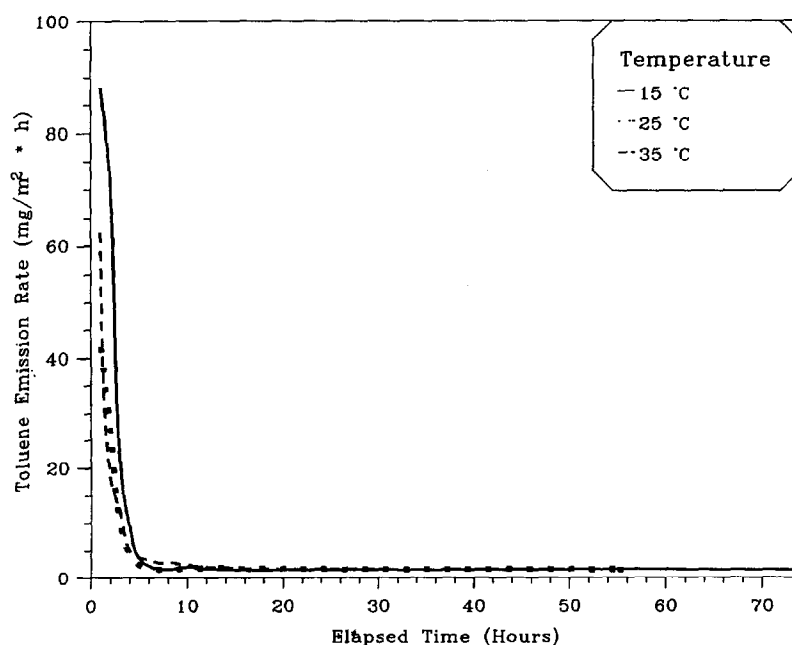


Fig. 3. Toluene emission rate vs time profiles for varnish at 1.2 ACH and temperature 15, 25, and 35°C .

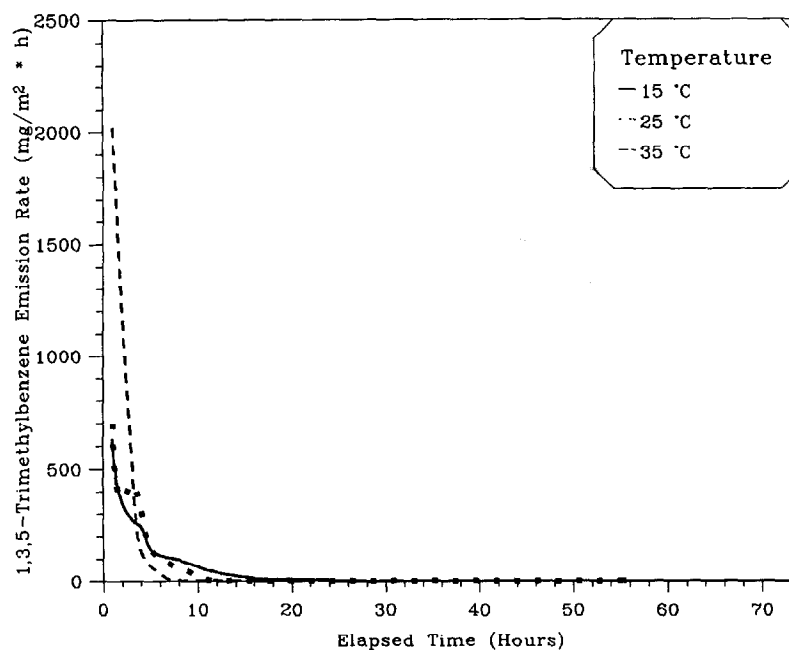


Fig. 4. 1,3,5-trimethylbenzene emission rate vs time profiles for varnish at 1.2 ACH and temperature 15, 25, and 35°C.

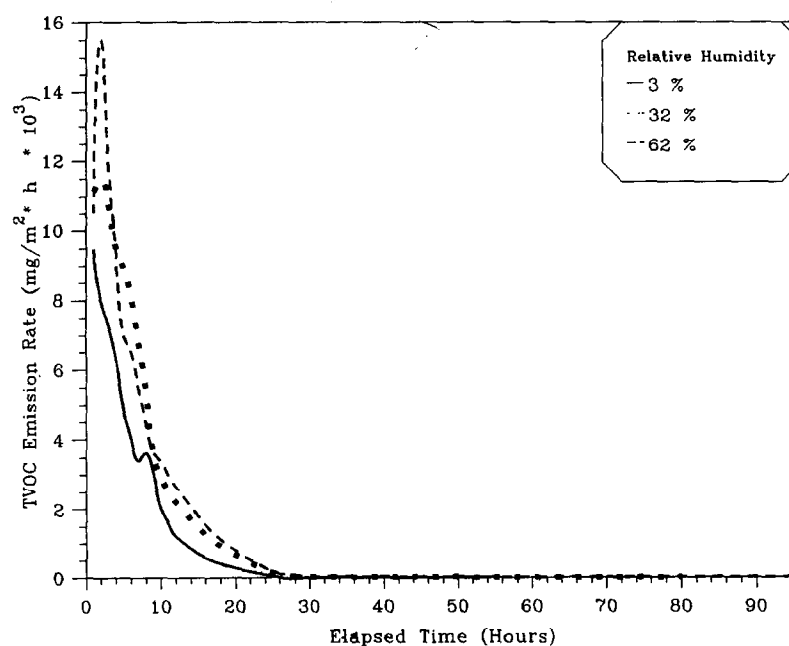


Fig. 5. TVOC emission rate vs time profiles for varnish at 1.2 ACH and relative humidity 3, 32, and 62%.

Paint

The paint used for this work was a water-based gloss acrylic. It is suitable for both interior and exterior applications. The manufacturer's label boasts that the paint is fast drying with low odour. However, health and safety advice on the label also suggests that when applying the paint, always ensure good ventilation. At 21°C, the paint should be dry to the touch within 1 h and ready for a second coat within 12 h. All tests were performed with a 10.2 g sample of paint, a product loading of 0.8 m²/m³, and an air exchange rate of 1.2 ACH.

Temperature results. Figure 8 illustrates the TVOC emission rate profile for the paint samples vs time for three air temperatures, 15, 25 and 35°C, and approximately 3% relative humidity. In general, it can be said that with an increase in temperature, TVOC emission rates increased. However, certain differences or idiosyncrasies can be noted. At the earlier stages of each test, fluctuations can be seen, the degree of which increases with temperature. At 35°C, these fluctuations last the longest, approximately 16 h, before the curve begins to decrease. The high levels of relative humidity recorded

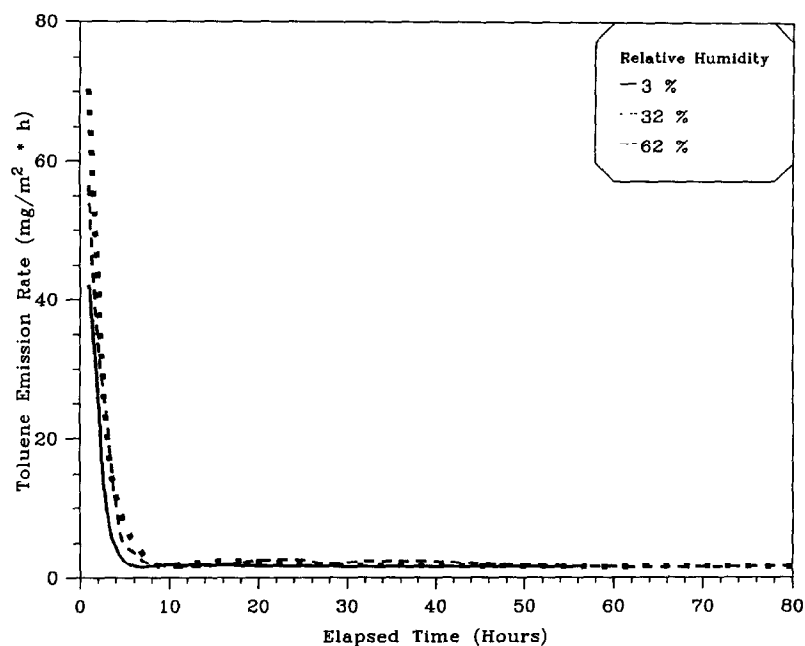


Fig. 6. Toluene emission rate vs time profiles for varnish at 1.2 ACH and relative humidity 3, 32, and 62%.

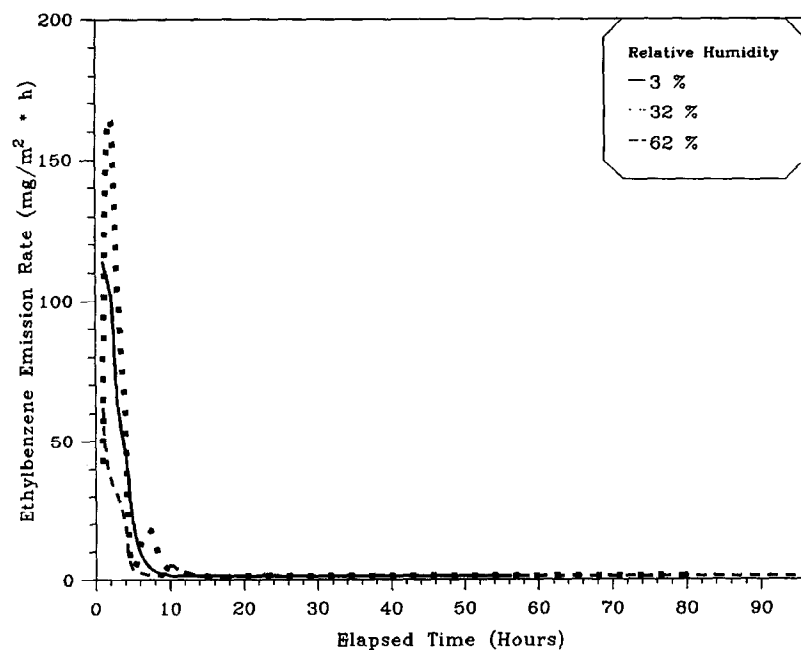


Fig. 7. Ethylbenzene emission rate vs time profiles for varnish at 1.2 ACH and relative humidity 3, 32, and 62%.

during this period must cause this strange phenomena. Though the air feeding into the system is very dry, the release of water vapour from the sample must impede the release of VOCs during this time. Vejrup and Wolkoff noted similar effects when they tested cleaning agents [51]. They attribute this to the water in the wet sample which prevents the more polar compounds from being released due to hydrogen bonding. As the recorded exit humidity drops to levels below 30% (12–24 h), the curve begins to behave exponentially as expected. The peak emission rates measured at each temperature vary sig-

nificantly. At 35, 25, and 15°C, the highest TVOC emission rates obtained were 115, 30, and 15 $\text{mg}/\text{m}^2\cdot\text{h}$ respectively. The time of peak concentrations occurs later with dropping temperature. The TVOC emission rates released at 35°C are significantly higher than those released during the 25 and 15°C tests. It was expected that as temperature increases, the vapour pressures of the VOCs will increase, thereby encouraging their release. This is indeed the case at 35°C. However, the jump from 15–25°C, while still showing an increase, is not nearly as significant as the jump from 25–35°C. The two profiles

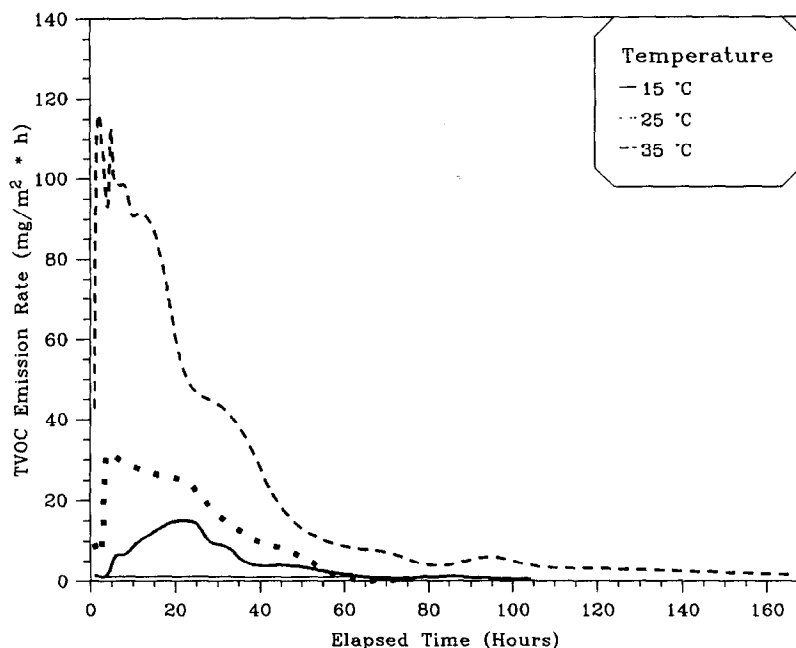


Fig. 8. TVOC emission rate vs time profiles for paint at 1.2 ACH and temperature 15, 25, and 35°C.

(15 and 25°C) follow each other rather closely. The 25°C curve has consistently higher values than the 15°C curve, but depletion is reached at approximately the same time (60 h).

As for the varnish, the emission rates from individual compounds present in the paint were also investigated. The compounds chosen were not necessarily the most abundant compounds found in the paint. It is recognized that glycols are more predominant in paints. However, four compounds which are more typically associated with indoor air quality were chosen. Also, where possible, the compounds chosen were also present in the varnish.

As can be seen from the profile for toluene (Fig. 9), the effects of temperature on the individual compounds are difficult to predict and generalize. The toluene curves for the early stages of the 25 and 35°C tests are erratic, but their patterns are somewhat similar. At 15°C, the toluene has comparatively very low emission rates, as well as fairly constant, when compared to the higher temperatures. It would seem that toluene, being a very volatile compound, is significantly affected by temperatures at levels as low as room temperature. The same argument can be said about the emission rates of m, p-xylene, shown in Fig. 10. Here, however, the emission rates are

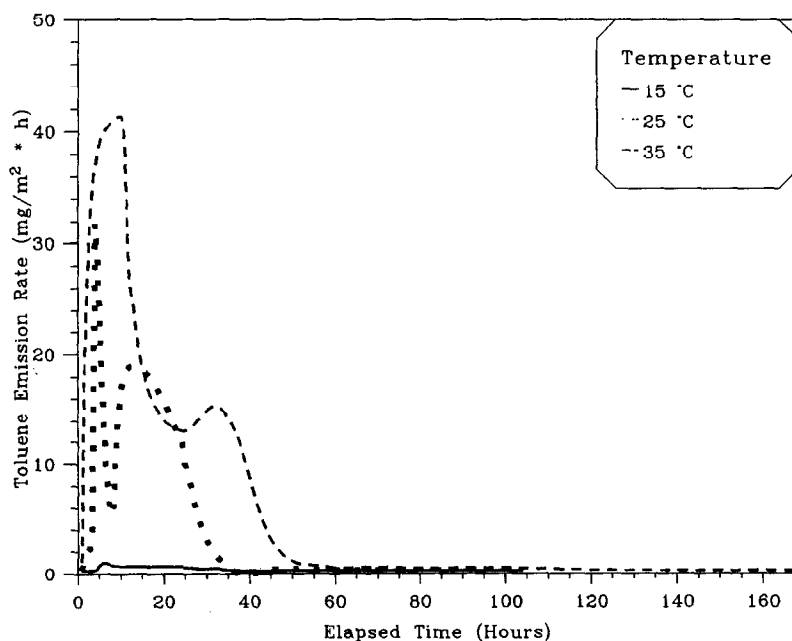


Fig. 9. Toluene emission rate vs time profiles for paint at 1.2 ACH and temperature 15, 25, and 35°C.

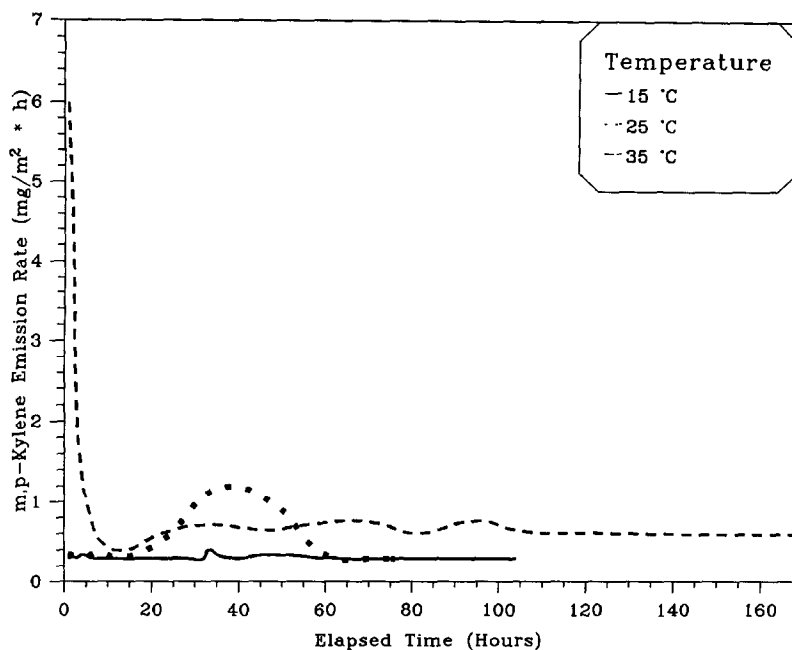


Fig. 10. m, p-xylene emission rate vs time profiles for paint at 1.2 ACH and temperatures 15, 25, and 35°C.

not as erratic. At 35°C, the peak is reached very early, and quickly (within 10 h) descends to values similar to the other temperatures. The 25°C curve shows a peak value of the compound fairly late in the test. At approximately the same time, the 35°C shows a slight increase in value before it becomes constant. This trend is not repeated for the 15°C curve, which behaves very similarly to the toluene curve, i.e., low and constant emission rates. Ethylbenzene and hexanal reach low peak values fairly early for all three temperatures, and go on to constant concentrations in a consistent pattern after approximately 12 h.

Relative humidity results. Figure 11 illustrates the TVOC emission rate profile for the paint samples vs time. As shown, the effect of relative humidity on the TVOC profile is erratic. The curves cannot be described as exponential decays. The highest emission rates occur when the humidity is 32%, but these decay faster than the emission rates recorded for the 62% test. Here, the emission rates fluctuate between 15 and 30 $\text{mg/m}^2\cdot\text{h}$ for almost 100 h before showing signs of decay. Up to 160 h the curve has reached steady state, but emission rates are not yet depleted. It is curious to see higher emission rates recorded for the 32% test rather than the 62% test. It

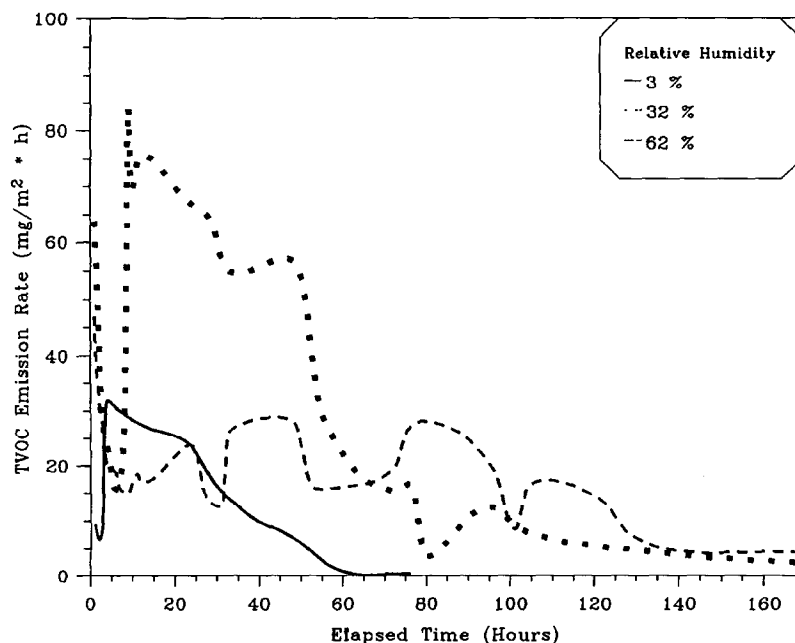


Fig. 11. TVOC emission rate vs time profiles for paint at 1.2 ACH and relative humidity 3, 32, and 62%.

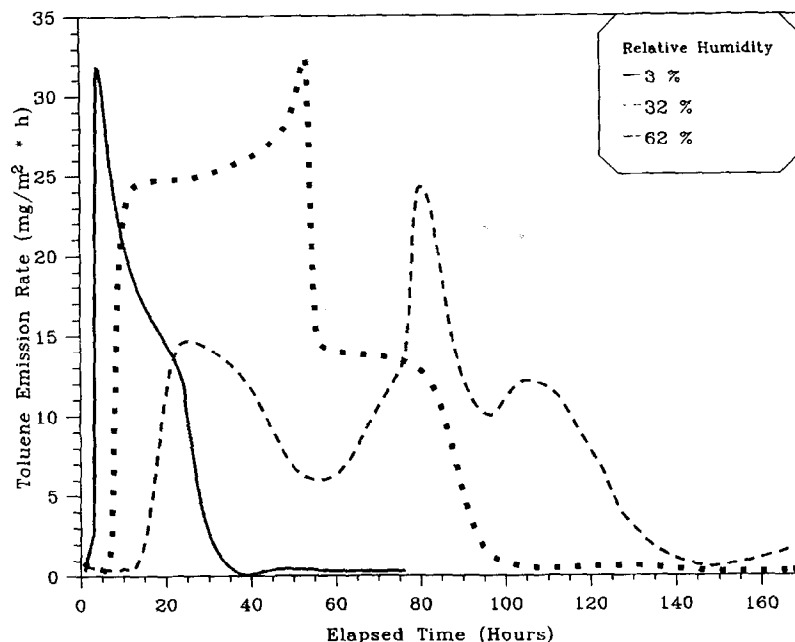


Fig. 12. Toluene emission rate vs time profiles for paint at 1.2 ACH and relative humidity 3, 32, and 62%.

would seem that the relationship between emission rate and relative humidity cannot be generalized as either directly or inversely proportional. At 32%, the humidity may be such that the vapour pressure of the overlying air encourages, rather than inhibits, the release of the VOCs, something which is not as evident at the other two levels tested. The emission rates recorded for the 3% test, not only yields lower emission rates, but reaches near depleted values just under 80 h.

The erratic behaviour of the TVOC curve is also demonstrated for the toluene and m, p-xylene curves (Figs 12 and 13). The effect of the 32 and 62% humidity levels on toluene are quite dramatic. The highest emission rates are noted for the 32% test. However, the 62% values are quite significant up to 160 h, where it appears to be on the rise again. The 32% values deplete after the 80 h mark. At 3%, initial peaks are as high as those at 32%, but diminish quite rapidly to near zero values after 24 h.

For m, p-xylene, the 62% humidity yields the higher emission rates which, though decreasing, are still significant after 160 h. At 32%, the emission rates are quite high for the first 12 h, then start to deplete. However, the depletion is slow with several minor peaks still occurring. At 3%, the highest emission rate is only recorded after 40 h.

Both ethylbenzene and hexanal do not show a significant difference between humidity levels tested. For the most part ethylbenzene emissions are quite low, and reach steady state within 24 h. The hexanal curve shows a minor peak for the 32% test early in the profile, while the 62% test shows several minor peaks.

The impact of the relative humidity on the different compounds considered is quite extraordinary. Toluene and m, p-xylene are greatly affected by the humidity but at different levels. As they also have very abundant emission rates, their reaction to the relative humidity must impact the TVOC emission rate profile.

It was noted that the behaviour of the TVOC emission rate profile and/or its compounds from the paint, were different from the varnish (i.e. Figs 5 and 11). The difference may have been caused by the high water content and the relatively low content of VOCs in the paint. The explanation may be due to the release of water vapour from the paint sample impeding the release of VOCs due to hydrogen bonding.

CONCLUSION

A comprehensive review of research on the impact of indoor air conditions; temperature, relative humidity; and surface air velocity on materials emission rates is presented.

The results of this study show that both the temperature and relative humidity have a significant effect on the emissions from paint and varnish. It was noted that as the temperature increased, the TVOC emission rates increased for both the paint and varnish. The results are consistent with the conclusions reached by other researchers [21, 25, 26]. However, in this study, the individual compounds did not necessarily follow the same trend established by the TVOC. Some compounds show secondary, less significant peaks later in their emission profiles; while other compounds show greater emission rates at the lower temperatures. These profiles contradict expected trends. From the profiles shown, the individual compounds present in greater concentrations follow the TVOC profile more closely. This indicates that the TVOC profile is influenced by the compounds found in greater abundance in the sample. This demonstrates the possibility that the TVOC value may not be a good indicator for indoor air quality. The emission profiles for these compounds should be determined independently of the TVOC values. In this way, immediately following the application or installation of a product, better judgement

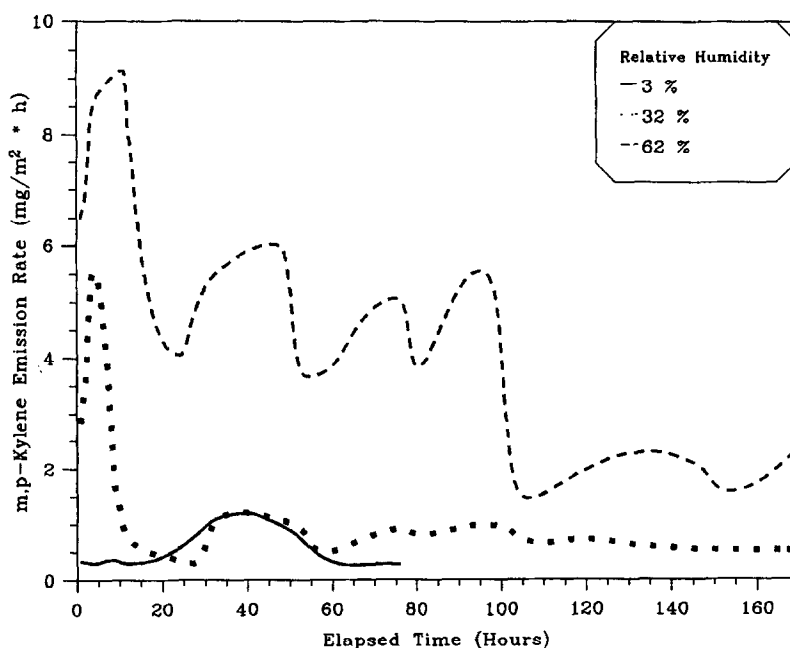


Fig. 13. m, p-xylene emission rate vs time profiles for paint at 1.2 ACH and relative humidity 3, 32, and 62%.

can be used when determining the time required before occupancy can take place.

The effect of the relative humidity on the emissions from the paint samples was very interesting. The fluctuations were quite significant, and no trend can be established. The emission rates measured for the 32% test approached the values reached during the 35°C test. While at the same temperature, the 3% and 62% tests showed lower emission rates. This demonstrates how sensitive the emissions are to the environmental conditions under which these materials are used.

The results for the varnish TVOC are more consistent

with expected trends. The effects of relative humidity could not be summarized into a general trend for the emissions from different materials. This is shown by the very different TVOC profiles established for paint and varnish. The two materials behave in very different ways. Tucker noted that highly polar compounds required testing for relative humidity effects [52]. This may be true of the water-based paint. However, the varnish, though following smoother trends, still showed varied effects. Its TVOC increased with increasing humidity, but some of its individual compounds showed higher emission rates for the 32% test.

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