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Publisher's version / Version de l'éditeur:

Proceedings of the Joint NSC-NRC Workshop on Construction Technologies: 26 April 2004, Taipei, Taiwan, pp. 173-180, 2004-04-01

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NRCC-47056

A version of this document is published in / Une version de ce document se trouve dans :
Proceedings of the Joint NSC-NRC Workshop on Construction Technologies, Taipei, Taiwan, April 26-27, 2004, pp. 173-180

INVESTIGATION OF BUILDING MATERIALS AS VOC SOURCES IN INDOOR AIR

Doyun WON[1] and C.Y. SHAW[2]

ABSTRACT

Source control using low emission building materials followed by ventilation has been considered as one of the most effective strategies for controlling volatile organic compounds (VOCs) indoors. To apply this strategy, it is necessary to have a decision-making tool for assessing the impact of material emissions on indoor air quality under various ventilation conditions. Therefore, a multi-year, client-supported project on material emissions and indoor air quality modelling was launched from 1996 to 2000 to develop such a tool, known as Material Database and Indoor Air Quality Simulation Program (MEDB-IAQ). On the project sponsors' suggestion, a follow-up project (Phase II) was conducted between 2000 and 2004 to improve the MEDB-IAQ. In this project, efforts were made to characterize VOC emissions for a total of 60 building materials and 90 chemicals. The emission data were incorporated into the MEDB-IAQ. Additional tasks were also conducted, including emission model development based on fundamental mass transfer theory and the investigation of environmental factors on material emissions. This paper provides a brief summary of the main tasks undertaken in both phases of the project.

***Keywords**: building materials, volatile organic compounds, emissions, chamber test.*

INTRODUCTION

There is a growing awareness of the importance of maintaining good indoor air quality, and with it a growing need to understand what constitutes, and contributes to, poor indoor air quality. This need spurred a major research initiative at the National Research Council Canada, with the overall goal of developing guidelines for indoor material selection and ventilation strategies to meet specific indoor air quality requirements. This objective will be achieved through a series of projects, including the first and second phase of the Consortium of Material Emission and Indoor Air Quality (CMEIAQ) project.

The major driving force behind the first and second phase of the material emissions project was the creation and maintenance of a material emission database and single-zone indoor air quality (IAQ) simulation program (called MEDB-IAQ). The other main tasks in the project, while important in their own right, were largely necessary because they provided the required input for the simulation program and database. This paper provides brief descriptions of the tasks conducted in Phase I and Phase II of the project.

CHAMBER TESTS

One of the main tasks of the project was to produce emission data for commonly used building materials in Canada for MEDB-IAQ. To obtain the emission characteristics of building materials over time, dynamic chamber tests were conducted. A set of concentration versus time data was obtained through the sampling of the exhaust air from the chamber on multi-sorbent tubes and DNPH cartridges and the analysis of them with GC/MS and HPLC. Two different sampling media were necessary due to the broad range of VOCs emitted from materials.

The materials tested under the project include 60 materials from 21 different types of materials: 6 wet material types (wood stain, polyurethane, wax, paint, adhesive, and caulk and sealant) and 15 dry material types (carpet, under pad, particleboard, plywood, oriented strand board, gypsum board, solid wood, ceiling tile, vinyl flooring, counter top, laminate flooring, linoleum, medium density fiberboard, pre-finished hardwood flooring, and vinyl-faced wallboard panel). Among the total 60 materials, 4 material systems were included: carpet/adhesive/concrete, laminate flooring/foam underlay, laminate flooring/foam underlay/OSB, and linoleum

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tile/adhesive/plywood.

More detailed information on the chamber tests can be found in the ASTM standards, two of which are the direct outcomes of this project: D6330-98 for testing wood-based panels in small-scale chambers (ASTM, 1998) and D6670-01 for full-scale chamber tests (ASTM, 2001). The expertise developed through this project also contributed to the revision of ASTM standard D5116-97, which is the standard guide for small-scale chamber tests for indoor materials and products (ASTM, 1997). The development of test methods and protocols were intended to promote the compatibility and comparability of the test results obtained by different laboratories. Even though substantial efforts were made to develop the database in this project, more data are required to make this database useful, and combining the emission data produced by different labs is the only practical way to meet this need.

TARGET VOC LIST

From a chamber test, one can notice that a building material can emit hundreds of chemicals. Therefore, identifying and quantifying all emitted chemicals is impractical. Certain criteria are needed for selecting specific chemicals for identification and quantification when conducting a material emission test. The most common approach is to focus on chemicals in greater abundance. The advantage of the approach is that the data analysis is relatively easy. The disadvantages are that: 1) the most abundant chemicals are not necessarily the most worrisome compounds, 2) quantifying the chemical against the calibration standard of the same chemical may not be possible if the chemical is not included in calibration mixtures, and 3) sometimes the true identity of a peak is obscured when the ion characteristics of the chemical are not properly identifiable. Another approach is to look for relevant chemicals to support a specific purpose (i.e., potential health concerns). The advantages of this approach are that finding chemicals with specific interests can be assured and quantifying a chemical can be done against the calibration standard of the chemical in question. The disadvantage is that chemicals for other important applications may be missed.

For this project, both approaches were combined. Chromatographs obtained from the GC/MS operation were analyzed for 5 to 6 most abundant VOCs and 90 Target VOCs for 55 materials. For the five materials analyzed with GC/FID, only abundant VOCs were monitored. The Target VOC list was established under the guidance of a Health and End-User Advisory Committee and was selected from 11 published lists by national and international agencies such as the World Health Organization, Health Canada, and the US Environmental Protection Agency (Table 1) and IRC material emissions data. The target VOC list for material emissions was intended to include chemicals that were: 1) known or suspected to have health or irritation concerns (health criterion); 2) known to be emitted from the building materials (building material criterion); 3) often found in indoor air (indoor air criterion), and 4) suitable for sorbent sampling and analysis with GC/MS or carbonyl analysis with HPLC (analysis criterion).

Table 1. Referenced Existing Lists for IRC Target VOCs

#	List name	Purpose of the List	Reference
1	EC VOC List	Compounds to be included in TVOC analysis in Indoor Air Quality Investigations	EC (1997)
2	Japanese List	Japanese Indoor Air VOCs Standards Mix for chemical analysis	Sigma-Aldrich (2004)
3	PSL1, PSL2	Priority Substances List under CEPA (Canadian Environment Protection Act)	Health Canada (2003a & b)
4	DSL	Domestic Substances List for the Pilot Program	Health Canada (2003c)
5	Proposition 65	Chemicals Known to the State of California to Cause Cancer or Reproductive Toxicity	OEHHA (2004)
6	2001 CERCLA	CERCLA (The Comprehensive Environmental Response, Compensation, and Liability Act) Priority List of Hazardous Substances	ATSDR (2003)
7	HAPs	Hazardous Air Pollutants	US EPA (2003a)
8	Air Pollutants	Guidelines for air pollutants with non-carcinogenic and carcinogenic health endpoints	WHO (1999)
9	C-REL	Chronic reference exposure level	OEHHA (2003)
10	VCCEP	Voluntary children's chemical evaluation program	US EPA (2003b)
11	Method 2549	List of Common VOCs for Screening	NIOSH (2003)

The resulting IRC Target VOCs are summarized in Table 2. The chemicals are grouped into 10 chemically different groups, including aldehyes, ketones, alcohols/glycols/glycol ethers, esters, halocarbons, aliphatic hydrocarbons, aromatic hydrocarbons, cycloalkanes, terpenes, and others. All Target VOCs are included in at least one of the 11 published lists with 7 exceptions, which are from IRC emission data. Exceptions are 1,2-propanediol, 2-methyl-2-propanol, isopropyltoluene, ethyl cyclohexane, propyl cyclohexane, decahydronaphthalene, and gamma terpiene, which were observed to be emitted in a large amount from building materials in Phase I of the project. Of the 90 VOCs, 47 have human health implications and 43 have large emission rates. The outcomes of the emission testing in this project are expected to contribute to the source characterization of chemicals with health hazard potentials indoors.

The IRC Target VOC list is intended to provide a manageable number of chemicals for the building material emissions study. For more general application to indoor air quality, the list needs to be expanded. For example, most halocarbons that are widely detected indoors are excluded from the current list based on the judgment that building materials are not the major sources of these compounds.

Table 2. IRC Target VOCs (90 Compounds)

Aldehydes	Acetaldehyde Acrolein Benzaldehyde Butanal Decanal Formaldehyde Furfural Heptanal Hexanal Nonanal Octanal Pentanal	**Aliphatic Hyddrocarbons**	2-Methylpentane 3-Methylpentane Decane Dodecane Heptane Hexadecane Hexane Nonane Octane Pentadecane Tetradecane Tridecane Undecane	**Cyclo-Alkanes**	Cyclohexane Butylcyclohexane Ethylcyclohexane Propylcyclohexane Decahydronaphthalene
				Esters	1 - Methylethyl acetate 2 - ethoxyethyl acetate Butyl acetate Ethyl acetate 2,2,4-Trimethyl-1,3-pentanediol diisobutyrate
				Halo-Carbons	1, 2-dichlorobenzene 1, 4-dichlorobenzene Methylene chloride Trichloroethylene
Alcohols, Glycols, GlycoEthers	1,2 Ethanediol 1,2 Propanediol 1-Butanol Propanol, 1-methoxy-2- 1-Propanol Ethanol, 2-butoxy- Ethanol, 2-butoxyethoxy- Ethanol, 2-ethoxy- Hexanol, 2-ethyl-1- Ethanol, 2-methoxy- Propanol, 2-methyl-2- 2-Propanol Ethanol Phenol	**Aromatic Hydrocarbons**	2-Ethyltoluene 3-Ethyltoluene 4-Ethyltoluene 4-Phenylcyclohexene Benzene 1,2,3-Trimethylbenzene 1,2,4-Trimethylbenzene 1,2,4,5-Tetramethylbenzene 1,2-Dimethylbenzene 1,3,5-Trimethylbenzene 1,3-Dimethylbenzene 1,4-Dimethylbenzene Isopropylbenzene Propylbenzene Ethylbenzene Naphtalene Isopropyltoluene Styrene Toluene	**Ketones**	Methyl ethyl ketone Acetone Acetophenone Cyclohexanone Methyl isobutyl ketone
				Terpenes	.alpha.-Pinene .alpha.-Terpinene .beta.-Pinene .gamma.-Terpinene 3-Carene Camphene Limonene
				Other	2-Pentylfuran 1-Methyl-2-pyrrolidinone Acetic acid Hexanoic acid n-Butyl ether Pentanoic Acid

MODELS FOR EMISSIONS

For the simulation of VOC concentrations in a realistic setting, models that can describe the emission characteristics over time need to be obtained. In general, the emission models can be categorized into empirical models and mass-transfer models. Empirical models are selected based on the statistical analysis of emission data from chamber testing without any consideration of physical phenomena. Since the coefficients of empirical models are determined by fitting the models to the material emission data using regression technique, there can be multiple solutions when more than one coefficient needs to be obtained from a set of data. Thus, the coefficients have little or no physical meaning. On the other hand, mass transfer models are based on fundamental mass transfer principles such as diffusion and evaporation. The model coefficients of mass-transfer

models can be determined by chamber tests as well as experiments that are independent of chamber tests. The coefficients have more distinct physical meaning and therefore, the results based on one condition can be applied to other conditions, including the scale-up of experiments.

Empirical and Semi-Empirical Models

Simplicity was the most important factor for selecting emission models for MEDB-IAQ since the software is mainly aimed for designers and engineers. Therefore, simple empirical and semi-empirical models are used in MEDB-IAQ. A power-law decay model (Equation 3) was used for dry materials. To determine the coefficients, data after 24 hours were used for the curve fitting. For wet materials, three different models were used to represent a particular curing stage of a wet building material from wet, to partially wet and dry: a vapor pressure and boundary layer model (Equation 1) for $0<t<t_1$, an exponential decay model (Equation 2) for $t_1<t<t_2$, and a power-law decay model (Equation 3) for $t>t_2$.

$$E = K_m\left[\left(C_v \frac{M(t)}{M_{o1}} - C(t)\right)\right] \qquad (0 < t < t_1) \qquad \textbf{(1)}$$

$$E = E(t_1)\, e^{-k(t-t_1)} \qquad (t_1 < t < t_2) \qquad \textbf{(2)}$$

$$E = a\, t^{-b} \qquad (t > t_2) \qquad \textbf{(3)}$$

where E = the emission factor (mg/m^2/h); a and b = empirical constants, t = the elapsed time (h), K_m = the mass transfer coefficient (m/h), C_v = the initial vapor pressure at the surface (mg/m^3), $C(t)$ = the concentration in the air phase (mg/m^3); M_{01} = the initial mass at the surface available for evaporation (mg/m^2), $M(t)$ = the concentration in the material phase (mg/m^2), $E(t_1)$ = the emission factor (mg/m^2/h) at $t = t_1$, k = the emission decay constant (h^{-1}), t_1 = the time at which the transition period began (h), t_2 = the time at which the diffusion controlled period began (h). The model coefficients for several abundant VOCs emitted from 48 building materials are reported in Won et al. (2003). Similar results will be produced for the combination of 90 Target VOCs and 55 materials.

One problem associated with emission testing and modelling is that because the models are based on short-term tests (e.g., 72 h), they tend to over- or under-predict the long-term emissions. To provide some answers to the question, long-term emission tests were conducted for a year with two OSB samples. The data will be used to verify the prediction capability of the empirical models developed based on short-term emission data

Mass-Transfer Models

Mass transfer models based on fundamental mass transfer principles have been recommended as a better alternative to empirical models in the research community. Mass-transfer models involve solving fundamental equations governing the emission processes from building materials to the air. Two such models have been developed for dry and wet materials under this project.

For dry materials, as the boundary layer effect is negligible, it was assumed that VOC concentrations are uniform within the air, including the boundary layer. For simplicity, it was assumed that the emission processes for dry materials were governed by the diffusion of a VOC through the source material and that instant equilibrium existed between the air phase and the material phase. The determination of the key model parameters including the diffusion coefficient in the material (D), the partition coefficient (K_{ma}) between the air phase and the material phase, and initial concentration (C_o) is described as follows:

- An experimental method was developed under this project to measure the diffusion (D) and partition coefficients (K_{ma}) for various VOCs diffused through common building materials.
- A headspace analysis was conducted to determine the equilibrium concentrations of VOCs at the air-side of the material-air interface.
- The results and the corresponding partition coefficients were used to estimate the initial VOC mass in the material.

In addition, correlations for the relationship between D and molecular weight (MW) and between K_{ma} and vapor pressure (VP) were also determined. These correlations will provide a tool to predict D and K_{ma} based on

chemical properties. Details can be found in Bodalal et al. (2000 and 2001).

For wet materials, the mass-transfer model developed accounts for surface evaporation as well as diffusion through the material film. A companion experimental method was also developed to determine both the evaporation (α) and diffusion coefficients. In addition, six aliphatic and six aromatic hydrocarbons from solvent-based paint were analyzed to determine the correlations between D and MW and between α and VP, which can be applied to a similar model to determine D and α for other compounds. Example correlation equations can be found in Won and Shaw (2003).

The mass-transfer models are useful for researchers to understand the emission processes, but are still too complicated for general applications. Therefore, these models are not included in the current version of MEDB-IAQ. More work, including model validation, is required before the mass-transfer models are incorporated in the MEDB-IAQ software.

EFFECTS OF ENVIRONMENTAL FACTORS

Emission testing is typically conducted under standardized conditions (i.e., at 23 ^{o}C and 50% RH). While the standardized testing condition is desirable to produce comparable and compatible emission data, it is also necessary to understand the effects of various environmental conditions on material emissions. The knowledge can lead to a more accurate assessment of the effect of building materials as a main indoor VOC source on the indoor air quality in buildings where a wide range of environmental conditions can exist. The information can also be used to provide better recommendations to minimize occupant exposure to VOCs indoors. For instance, increasing ventilation rate for several days is often recommended to speed up VOC emissions, particularly before the occupancy of a newly built or renovated building. However, it is still questionable whether the short-term increase of ventilation rate has any significant effects on reducing long-term occupant exposure to VOCs. The investigation on the effects of environmental factors on VOC emissions from building materials will provide information that will better answer those questions.

The effects of environmental factors including air velocity, temperature, relative humidity and initial concentration on the VOC emission from solvent-based paint were investigated. To report the results in a quantitative format, air temperature, air velocity, relative humidity and initial concentrations were correlated with the model coefficients of the aforementioned mass-transfer model for wet materials. The knowledge can lead to a more accurate assessment of the effect of building materials as a main indoor VOC source on the indoor air quality in buildings where a wide range of environmental conditions can exist. The model coefficients were obtained from dynamic chamber tests with a paint sample contained in a Petri dish. It was observed that the diffusion coefficient is a function of air temperature (T), initial concentration (C_o), and molecular weight (MW), while the evaporation coefficient is a function of air velocity (v) and vapor pressure (VP) of chemicals. On the other hand, relative humidity had no effects on emissions of alkanes and alky benzenes, which are non-polar compounds

Equations 4 was suggested to express the correlation of α with v and VP:

$$\alpha = \frac{a_1 VP}{a_2 + VP}\left(a_3 + a_4\ EXP(a_5\ v)\right) \tag{4}$$

Where α = evaporation coefficient (mm/h), VP = vapour pressure (mmHg), v = air velocity (m/s), and a_1, a_2 , a_3, a_4 a_5 = correlation constants.

Fitting Equation 4 to all the measured data from the experiments for studying the effect of air velocity led to the correlation constants summarized in Table 3. The excellent R^2 values show that the evaporation coefficient is a strong function of air velocity and vapor pressure in the form of Equation 4.

Table 3. Correlation Constants for α as a Function of *v* and VP.

		Alkanes	Alkyl benzenes
Correlation constants	a_1	0.23	0.18
	a_2	18.17	56.89
	a_3	5.26	43.96
	a_4	2.60	0.20
	a_5	12.48	36.19
R^2		0.9835	0.9984
# of data		30	30

For the correlation of D with T, C_o, and MW, Equation 5 was used:

$$D = (b_1 + b_2\ MW)(b_3 + b_4\ T)(b_5 + b_6\ C_o) \tag{5}$$

Where D = diffusion coefficient ($\mu m^2/h$), MW = molecular weight, T = air temperature (^oC), C_o = initial concentration of a chemical in a paint sample (g/L), b_1, b_2, b_3, b_4, b_5 and b_6= correlation constants.

The results of curve-fitting of Equation 5 to all the data from experiments on varied temperature and initial concentration levels are given Table 4. The R^2 values are reasonably good, which again demonstrates the validity of the relationship given in Equation 5.

Table 4. Correlation Constants for *D* as a Function of *MW*, *T*, and C_o.

		Alkanes	Alkyl benzenes
Correlation constants	b_1	6.91E+04	2.20E+02
	b_2	-4.00E+02	-1.54E+00
	b_3	-1.75E+01	-2.06E+03
	b_4	1.34E+00	1.58E+02
	b_5	1.38E-01	4.48E-01
	b_6	3.50E-03	6.80E-03
R^2		0.9342	0.9252
# of data		47	35

Note that the correlations shown in Equations 4 and 5 are only for alkanes and alkyl benzenes. The results may be different for other groups of chemicals, particularly polar compounds.

MEDB-IAQ SIMULATION SOFTWARE

The ultimate goal of the tasks mentioned so far is to create and improve the MEDB-IAQ simulation program. The outcomes of emission testing and empirical modeling were packaged into the material emission database. Although the mass-transfer models and the effects of environmental factors are not included in the program, the outcomes of these two tasks will be used to improve the program in the future.

The computer simulation program MEDB-IAQ was developed to serve several purposes. The user can view material emissions information (e.g., emission factors and coefficients for Equations 1, 2, and 3) on the display in tabular or graphical form, print reports, and export data to other programs for further processing. It also allows the user to browse the database and to formulate queries to search the database for specific information. For example, one can search for materials that emit a chemical of interest through the query mode.

Ultimately, this tool will assist building designers in making informed choices of low emission building materials. MEDB-IAQ has the capability to predict the concentrations of individual VOCs that occur in a room, based on the emission data for selected building materials in the database. The single zone model assumes that perfect mixing of the contaminants with indoor air exists and that air-borne VOC compounds do not interact with each other. Input data include loading ratio and emission source areas, emitted contaminants and their emission rate – time profiles, room or house dimensions, and air change rate. The database provides two of the key input data: emitted VOCs and their emission rate – time profiles for selected building materials.

Work is ongoing to link MEDB-IAQ to CONTAMW developed by NIST for the multi-zone simulation (Howard-Reed et al., 2003). CONTAMW is a multi-zone indoor air quality and ventilation analysis computer program designed to predict airflows, chemical concentrations, and personal exposure (NIST, 2004).

SUMMARY AND FUTURE PLANS

Two multi-year research projects (Phase I and Phase II) on material emissions and indoor air quality were initiated to determine the emissions from building materials and their effects on indoor air quality. The most important output of the project is a material emission database and a single-zone indoor air quality (MEDB-IAQ) simulation software for use with the database. The database includes 60 commonly used building materials in Canada. For each building material, the database lists the emission characteristics of 90 Target VOCs. The Target VOCs consist of chemicals known or suspected to have health or irritation concerns and/or known to be emitted from the building materials. MEDB-IAQ can be used as a decision making tool for designers and building engineers for selecting low emission building materials to minimize indoor VOC sources and/or for exploring trade-offs between selection of building materials and ventilation strategies to achieve a pre-determined indoor air quality level in their buildings.

Other important outputs include two mass-transfer-based models to describe emission characteristics: one for dry and the other for wet building materials. These models are useful in understanding the material emission processes and help to analyze emission test data. In addition, a method based on fundamental mass transfer principles has been developed to mathematically describe the effects of environmental factors such as air temperature, air velocity and relative humidity on material emissions. The results would be useful in predicting long-term indoor air quality levels in buildings where large variation in air temperature, air velocity, and relative humidity can exist.

The following tasks are recommended as future research:

- More data for the database: In spite of the efforts of this project, the number of materials investigated was small compared to the numerous building materials on the market. With the help of standardized testing protocols such as ASTM guidelines, collaborations among international laboratories are recommended to create a high quality and diverse material emission database.
- Mass-transfer models: More efforts are needed to further understand the fundamental mass-transfer mechanisms from building materials. Although the development of the mass-transfer models and the methodology to determine model coefficients was accomplished in this project, validation of these models and coefficients is required. In particular, the paint sample used to develop the mass-transfer model for wet materials was contained in a Petri dish rather than applied on a substrate. Thus, validation experiments with more realistic substrates are recommended. The correlations between model coefficients and chemical properties also need to be expanded to more diverse chemical groups in addition to alkanes and alkyl benzenes tested in this project.
- Incorporation of environmental effects: In the present MEDB-IAQ, the simulation is only for a standardized condition. The findings from the environmental factor study need to be included in the software. This task is only possible after the incorporation of mass-transfer models to MEDB-IAQ.

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