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Modelling of Volatile Organic Compound (VOC) Emissions From Building Materials/Furnishings – A Literature Review

Internal report (Institute f
ANALYSE

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Internal Report No. IRC-IR-755

Date of issue: December 1997

Modelling of Volatile Organic Compound (VOC) Emissions
From Building Materials/Furnishings -- A Literature Review

IRC/NRC Internal Research Report by

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ABSTRACT

In this report, the emission characteristics of various building materials/furnishings are reviewed. Existing emission source models and their application for building materials/furnishings are described. It is concluded that various statistical models are available for analyzing and reporting emission test results of individual materials as functions of time. However, there is a need to develop mass transfer models that describe the emission process mechanistically. These mass transfer models would provide better predictions of the emission characteristics beyond the standard testing period and testing conditions. Furthermore, models for predicting the emission characteristics of material systems (assemblies) and methods for predicting the impact of ambient air conditions (temperature, humidity, air velocity and turbulence) on the material emission characteristics are especially needed.

1. INTRODUCTION

Building materials and furnishings have been recognized as one of the major sources of volatile organic compounds (VOCs) found indoors (Molhave 1978, 1982; Steel 1985; White et al.1988; Tichenor and Mason 1988). For example, Molhave (1982) tested 42 commonly used building materials and identified 52 different volatile organic compounds (VOCs). On average, each tested material emitted 22 different compounds. Berglund et al. (1992) summarized the effects of indoor air pollutants including VOCs on occupant's comfort and health. They reported that many VOCs are mucous membrane irritants and have been implicated as a cause of sick building syndrome. Molhave (1991) found that at levels below 0.2 mg/m^3 total volatile organic compounds (TVOC) appeared to have little or no effect on indoor air quality. TVOC would cause effect on indoor air quality when its concentration is above 3 mg/m^3. Understanding VOC emission characteristics of typical building materials will help to control the level of VOC concentrations in buildings and hence reduce potential health risks of building occupants.

The main purpose of modelling emission sources is to predict the emission rates of various VOCs as a function of time under typical indoor air conditions. This would allow us to evaluate the impact of various building materials on the VOC concentrations in buildings. Source models are

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also useful for analysing the emission data obtained from test chambers, for extrapolating the test results beyond the test period, and for developing simplified methods and procedures for emission testing.

The VOC emission rates of building materials and furnishings are affected by many factors such as product type, manufacturing process, product age (time), surrounding environmental conditions (temperature, humidity, air velocity and turbulence, and VOC concentration level), and the way they are packaged, transported, stored, and used in buildings. This report reviews and summarizes the emission characteristics of building materials and available mathematical models for describing them. It focuses on the emission characteristics of new or uncontaminated products under the environmental conditions that are typical for residential and office buildings.

2. VOC EMISSION CHARACTERISTICS OF BUILDING MATERIALS

2.1 Variations of Emission Rates with Time

Building materials/furnishings may be divided into the following four groups in terms of their emission characteristics: (1) "wet" individual coating materials; (2) "wet" individual installation materials (3) dry individual materials; and (4) material systems/assemblies.

Group 1: "Wet" Individual Coating Materials

Materials included in this group are wood stains, varnishes, paints, and wax. These materials are applied "wet" on certain substrate materials. Their emission characteristics are therefore dependent on both the coating material itself and the substrate used.

VOC emissions from the "wet" coating materials can generally be characterized by two periods (e.g., Tichenor 1987, Chang and Guo 1992a & 1992b, Wiles et al. 1996, and Zhang et al. 1996): (1) an initial period (drying period) in which the emission rate is high, but decreases quickly with time; (2) a later period (after the surface become dry or a thin film is formed) during which the emission rate decreases slowly with time. During the first period of the emission, the emission rate is primarily controlled by the interfacial evaporative mass transfer, while in the second period by the VOC diffusion within the materials (internal diffusion).

The duration of the initial period is relatively short. Depending on the coating material and substrate used, it can vary from a few hours to about 15 hours after the coating material is applied. The amount of VOCs emitted during each period varies significantly among different from product to product. Wilkes et al. (1996) measured the VOC emissions from latex paints and reported that the total VOC emitted during the first period accounted for up to 10% of the total VOC mass released, while the total VOC mass released was between 20-35% of the mass applied. On the other hand, for solvent-based materials (such as woodstain), the VOC mass

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emitted during the first period accounted for 40-50% of the total emittable VOC mass and the total emittable VOC mass was between 60-80% of total mass applied. Guo et al. (1996a) found that the VOCs emitted from a water-based latex paint only accounted for as low as 4% of the total mass, while the VOCs emitted from oil-based wood stains accounted for as high as 50% of the total mass. For the same type of materials, the emission rates also varied with the manufacturers (Tähtinen et al. 1996).

The emission characteristics of "wet" coating materials are strongly dependent on the substrate materials used. In general, a more porous substrate material (e.g., wood-based panel, gypsum board) would adsorbe more VOCs during the initial drying period than a less porous material (e.g., glass, metal sheet), resulting a smaller amount of VOCs emitted during the initial drying period and larger amount emitted after the drying period. The total emission period is also longer when a more porous substrate material is used. For example, Gehrit et al. (1993) measured VOC emissions from low solvent (1.5% of total mass) paint in a 1 m^3 chamber over a 4 day period. Both a glass plate and a paper-faced gypsum board were used as the substrate. Their data showed that most of the emissions occurred within the first 12 hours under both substrates. However, the paper-faced gypsum board substrate resulted in fewer emissions than the glass substrate, especially for the polar compounds measured. Guo et al. (1996) showed that paints applied on softwood had lower initial emission rates and longer emission period than paints applied on a hardwood.

Clausen et al. (1991) studied the long-term emission characteristics of waterborne paints. They found that the data collected within first three weeks of the test were sufficient to predict the emissions of VOCs for up to one year. Colombo et al. (1990) measured the steady-state emission rates of seven major VOCs (n-dodecane, hexane, n-undecane, pentanal, n-tridecane, n-decane, and propanal) and TVOC for a polyurethane lacquer on plywood in a well-stirred 450 L glass chamber, at 23 C, 45% RH and 0.25 ACH. The sample had been prepared and dried 2-4 months previously and sealed in aluminum foil. The test showed that the TVOC emission decreased to a "steady state" level (defined as ≤ 10% concentration change per day) in 65 days.

Group 2: "Wet" individual installation materials

This group includes materials such as adhesives, caulk and sealant, and varieties of joint, patching and texturing compounds. These materials may have similar emission profiles as that of "wet" individual coating materials, but the time period of the initial emission process for these materials are expected to be longer due to the relative "thicker layer" and slower drying process.

Group 3: Dry individual materials

This group includes a majority of building materials and furnishings such as carpets, gypsum wallboards, particleboard, oriented strand board (OSB), vinyl flooring materials, ceiling tiles,

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fabrics, etc.. VOC emissions from these materials are generally characterized by a nearly constant or very slowly decaying emission rate. For example, Tichenor (1996) reported that the initial emission rate for carpet ranged from 0.02 to 2 mg/m^{2}h, with an exponential decay constant of about 0.004 1/h. Similar order of magnitude in the initial emission rates and exponential decay constants were measured by Clausen et al. (1993) for vinyl flooring. These values are much smaller compared to that of "wet" materials. For example, Tichenor (1996) reported that the initial VOC emission rates of paints were in the range of 2000 to 50000 mg/m^{2}h, with an initial decay coefficient of about 0.5 1/h. VOC emissions from dry materials are mainly controlled by the VOC diffusion within the materials (i.e., the internal diffusion process).

Group 4: Material systems/assemblies:

In practice, different individual materials are usually used together to form material systems/assemblies such as:

carpet // adhesive // concrete,
paint // gypsum board // vapor barrier,
carpet // underpad // plywood (or OSB) subfloor // wood joists,
wax // vinyl sheet // adhesive // concrete,
polyurethane floor vanish // wood stain // hard wood // plywood (or OSB) // wood joists, etc..

In order to estimate the actual contributions of each individual material to the VOC concentrations in buildings, it is necessary to understand the emission characteristics of these material systems/assemblies as well as that of individual materials.

Very limited data are currently available about the emission characteristics of material systems. Tshudy and Little (1995) measured TVOC emissions from a "vinyl tile // adhesive // concrete" system as well as from the individual adhesive and vinyl tile. As expected, the adhesive, when tested alone, had the highest initial emission rate that decreased quickly with time. The emissions from the vinyl tile alone agreed well with the system initially. However, 48 hours after the testing, the TVOC emission from the system was larger than that from the vinyl tile alone, indicating that it took 48 hours for the adhesive to diffuse through the vinyl tile. Low et al (1996) showed that the initial emission rate of adhesive VOCs from a "carpet // adhesive // concrete" system was significantly smaller than that from the adhesive when tested alone. The carpet layer provided some resistance to the transport of the VOCs from the adhesive to the ambient air. It also increased the differences of emission profiles among individual VOCs since different VOCs diffused through the carpet layer at different speed. VOCs with a lower boiling point (i.e., more volatile) reached its maximum emission rate earlier than that with a higher boiling point.

2.2 Effect of Environmental Conditions on VOC Emission Rates

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Local environmental conditions can significantly affect the VOC emissions from building materials and systems. Major factors include temperatures, humidity, air motion (velocity and turbulence) and VOC concentrations in the ambient air. Ventilation rate in testing chambers (or space) and the loading ratio (defined as the ratio between the area of emitting surfaces divided by the air volume of the chamber/space containing the emitting materials) also affect the VOC emissions indirectly since they affect the VOC concentrations and airflow conditions in the chamber/space. These environmental factors can change the VOC emission rates at any given time, and therefore, result in different profiles of emission rates.

Temperature

An increase in the ambient air temperature would result in an increase in the emission rates of VOCs from either dry or "wet" materials. The significance of temperature effect on the emission rate profiles depends on the specific materials and the VOCs emitted. Wolkoff (1996) reported that an increase of temperature from 23 °C to 35 °C had resulted a 20% increase in the emission rate of VOCs from a nylon carpet with latex backing and a sealant. However, no significant impact was observed for VOC emissions from a PVC flooring material, a floor varnish on pretreated beechwood parquet and a waterborne wall paint on gypsum board, when these tests continued for more than a week.

Van der Wal et al. (1997) experimentally studied the effect of temperature on the VOC emissions from PVC flooring, carpet and paint. In general, an increase in temperature from 23 °C to 30 °C caused an increase in both the initial emission rate and the decay rate of the emission rate. For the carpet and PVC flooring tested, increasing temperature also increased the total emittable amount of VOCs. When the temperature was reduced to 23 °C from 30 °C, the emission rate was at the similar level as if the temperature was not increased. In other word, the high temperature period had no significant effect on the long-term emissions. However, increasing the temperature decreased the long-term emission rate of VOCs from the paint tested. The effect of temperature on the emission rates from building materials are expected to vary from material to material and VOC to VOC. More quantitative data on the relationships between the ambient air temperature and emission rate profiles are currently lacking.

Relative humidity

It is well known that relative humidity affects the emission of formaldehyde from pressed wood products (e.g., ASTM E1113-96). It is expected that relative humidity would also affect the emissions of VOCs that are water-soluble. However, experimental data in this regard are very limited. Wolkoff (1996) tested four building materials under 0% and 50% RH, respectively. He found that relative humidity had large effect on the emission of 2-ethylhexanol from a carpet during the first week of testing. The effect was less on the emission of 4-phenylcyclohexene

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from the same carpet. He reported that the humidity also had some effect on the VOC emissions from a sealant, a floor varnish and a wall paint, but had no impact on the VOC emissions from a PVC flooring material.

In measuring the emission rates of VOCs from latex paints, Roache et al. (1996) found that relative humidity had significant impact on the emission rate of ethylene glycol, but had little effect on the emission rate of Texanol from the latex paints.

<u>Air velocity and turbulence</u>

Air velocity and turbulence affect the evaporative mass transfer from solid or liquid to the air. By using a chamber in which air velocity and turbulence were precisely controlled, Zhang et al. (1996) showed that increasing the air velocity and turbulence led to an increase in the emission rates of VOCs from a wood stain, but the effect became smaller as the wood stain became drier. Guo et al. [b] (1996) also found that for p-dichlorobenzene, when air velocity over the substrate surface increased from <0.03 to 0.46 m/s, the initial emission rate increased by a factor of three. Wolkoff (1996) measured the emission rates of VOCs from a carpet, a PVC flooring material, a sealant, a varnish and a waterborne paint applied on a gypsum wall board, under four different velocity levels (1, 2, 5, and 9 cm/s nominal). They found that VOC emissions from the carpet, varnish and wall paint measured a few days after the beginning of the test (called primary emission rates) were not significantly affected by the air velocity. It is expected that emissions from both dry and dried "wet" materials be mainly controlled by internal diffusions. He also reported large variations in the emission data for carpet during the first 4 weeks, and the effect of air velocity could not be clearly identified.

The initial period of VOC emissions from "wet" materials is primarily a convective mass transfer process from the material surface to its ambient air. Sparks et al. (1996) developed a correlation to estimate the effect of air velocity on the convective mass transfer coefficient, and demonstrated its usefulness for predicting the emissions from evaporative controlled sources such as the initial period of "wet" coating materials. Zhang et al. (1996) also derived a method to estimate the convective mass transfer coefficient based on the airflow characteristics over the material surface.

In a recent study on the VOC emissions from a material system ("carpet // adhesive // concrete"), Low et al. (1996) found that increasing air velocity over the carpet surface from 0.05 to 0.25 m/s resulted in an increase in the emission rates only during the first 30 hours of the test. Little effect on the emission rates was observed afterwards.

<u>VOC concentrations in air, air change rate and loading ratio</u>

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For an emitting material, the VOC concentration at the material surface is higher than that in its surrounding air. A higher concentration in the air would lead to a lower concentration gradient between the material surface and the surrounding air, and hence reduce the convective mass transfer rate from the surface to the air. This will significantly reduce the emission rate if the emission process is controlled by the convective mass transfer over the material surface (e.g., during the initial emission period of "wet" materials). However, the effect is much less significant for dry materials whose emissions are primarily controlled by the internal diffusion.

VOC concentrations in a chamber/space are dependent on the air change rate (ventilation rate) and/or material loading ratio. Tichenor and Guo (1988) investigated the effects of air change rate (N - from 0.3 to 4.6 ACH) and loading factor (L - 0.06 to 1.3 m^2/m^3) on the emission rates of three wet materials: wood stain and polyurethane applied to hardwood substrates, and floor wax applied to aluminum plate, using a small chamber. They found that increasing ventilation rate increased emission rates while increasing loading ratio decreased emission rates, as expected. They also reported that wax emitted so fast in the first few minutes that the influence of N/L on the emission was negligible. A similar study on emission from caulking compound (Tichenor, 1989) showed that emission rates were higher initially, but lower in the later emission period when the air exchange rate was increased, thus indicating high air exchange rate caused the source to deplete faster.

Gunnarson et al. (1993) measured Texanol emission from aged acrylic latex paint. They found that increasing air change rate increased the emission rate at low air change rates, but had little effects on the emission rate at high air change rates. The emission appeared to be primarily limited by internal diffusion rather than the evaporation at the high air change rate conditions.

Andersen et al. (1996) studied the effect of concentration on VOC emission rates of five commonly used building materials: a PVC flooring, a waterborne acrylic varnish applied on a 12 mm thick beechwood parquet, a loomed polyamide carpet (nylon fibres on a latex foam backing), an acrylic sealant applied on a U-shaped aluminum channel, and a waterborne acrylic wall paint applied on a gypsum board. Different concentrations in the test chamber were achieved by setting the air change rate at three different levels. All test specimens were conditioned for 6 to 13 days at 1.0 ach air change rate before testing. Their results showed that the different concentration levels had no effect on the emission rates for the PVC-flooring and the varnish, moderate effect for the carpet and sealant, and strong effect for the paint.

It should be noted that increasing the air change rate could also result in significant changes in the airflow pattern, an increase in velocity and turbulence level over the emitting surfaces. This would result in an increase in the convective mass transfer coefficient. The results of the previous studies reviewed above should be viewed as the combined effect of both the concentration and the boundary layer flow over the material surface. Moreover, it is preferable to consider the concentration in the chamber/space as a dependant variable in modeling the effect of

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environmental conditions on VOC emissions. In this way, the fundamental convective mass transfer theory can be readily applied (see section 3.3.1).

3. MATHEMATICAL MODELS

3.1 Categorization of Emission Source Models

Emission source models for building materials may be divided into two categories:

1) Statistical models: These models are proposed/derived based on the observation and statistical analysis of emission data obtained from environmental chamber testing. The coefficients (or parameters) of these models are determined entirely by regression analysis of emission data obtained from chamber tests.

2) Mass transfer models: These models are derived partially or entirely by applying the mass transfer theory to the emission processes. The coefficients (or parameters) in these models have more distinct physical meanings than those of statistical models. The model coefficients may be determined by regression analysis of data from the dynamic emission testing and/or experiments that are independent of the environmental chamber tests.

Guo (1996) has summarized various indoor source models in his User's Guide for Z-30 Indoor Air Quality Simulator. In this report, the discussions are limited to models that are most relevant to building materials/furnishings.

Emission source models conventionally express the emission rate or emission factor as a function of time. In this report, the emission rate, R(t), is defined as the total amount of VOC or TVOC emitted per unit of time from the entire source.

The emission factor, E(t), is defined as follows:

1) For a surface source with a defined area, E(t), is the amount of VOC or TVOC emitted per unit time and per unit surface area of the source. Therefore,

$$R(t) = A\ E(t)$$

where,

A = area of the emitting surfaces, m^2;

2) For a non-surface source or a source whose surface area can not be easily measured or calculated (e.g., when an office workstation module is tested in a full-scale chamber), E(t), is

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the amount of emissions per unit time and per unit source (e.g., per workstation module). In this case,

$$R(t) = n\ E(t)$$

where,

      n   = number of units of the source.

3.2 Statistical Models

3.2.1 Constant Emission Factor Model

$$E(t) = constant$$

Chang and Krebs. (992) showed that this model described well the emission of p-dichlorobenzene from mothcakes. In practice, it may also be used to approximate the emission rates of building materials that have nearly constant emission rates for a certain period of time. For example, emission tests are often conducted by taking a single air sample after the test specimen is placed in a dynamic (i.e., ventilated) chamber for a fixed period of time (e.g., 24 hours or a week) for the purpose of comparing emission strength of different products (Levin and Hodgson 1996). These test data may be used for evaluating the exposure of occupants to VOCs in a worst case scenario, assuming that the emission rates of these products are approximately constant.

3.2.2 First-Order Decay Model

$$E(t) = E(0)\ exp\ (-k\ t)$$

where,

      $E(0)$ = initial emission factor, $mg/(m^2 h)$, or mg/h per unit source;
      k = first-order decay constant (or exponential decay constant), 1/h.

It is called first-order decay model because it can be derived from $d[E(t)]/dt = -k\ E(t)$ with the initial conditions: $E(t) = E(0)$, at $t = 0$. Both $E(0)$ and k are determined by the concentration data, $C(t)$, obtained from a dynamic chamber test. Several different calculation procedures are available for determining the model coefficients (ASTM 1990 and Zhang et al. 1997).

With this model, the total emittable mass per unit source (or area), M(0), may be calculated as

$$M(0) = E(0)/k$$

In a study of the effects of the film thickness on the VOC and SVOC emission rates from waterborne paints, Clausen et al. (1991) expressed the first-order decay model as

$$E(t) = M(0) \ k \exp(-k \ t)$$

They also further expressed k as k_{E1}/L and k_{D1}/L^2 for the evaporative and internal diffusion controlled period, respectively (where, L is the film thickness, k_{E1} and k_{D1} are decay coefficients per unit thickness for the evaporative and diffusion controlled period, respectively). Again, non-linear regression analysis was used to determine M(0) and k (and in turn k_{E1} and k_{D1}, respectively with the L measured independently). The model was as considered to fit the experimental data of 1,2-propanediol, 2-butoxyethanol, undecane, white spirit and TVOC reasonably well for the period from 24 hours to one year after the start of the test. However the model did not fit the data from the first 24 hours of testing. The model was also not valid for fitting the chamber data of Texanol and 2-(2-butoxyethoxy)-ethanol due to the presence of strong sink effects for this compounds.

A single first-order decay model is generally adequate for analyzing (representing) VOC emission data from chamber test only for a certain period of time. This time period is longer for a dry material or a "wet" material that is sufficiently dried than a "wet" material (e.g., from a week to one month for dry materials vs. 12 hours for the initial emission period of "wet" materials). The validity of applying the model to predict the emission rate beyond the testing period has to be justified by some longer-term emission testing data for the same type of materials considered.

3.2.3 Double Exponential Model

$$E(t) = E_{01} \exp(-k_1 \ t) + E_{02} \exp(-k_2 \ t)$$

where,

E_{01} = initial emission factor for the first (fast) decaying period, $mg/(m^2 \ h)$

E_{02} = initial emission factor for the first (fast) decaying period, $mg/(m^2 \ h)$

With this model, the total initial emission factor is $(E_{01} + E_{02})$, and the total emittable mass can be calculated by $(E_{01}/k_1 + E_{02}/k_2)$.

This model is adequate for representing chamber test data for both "wet" and dry individual materials. The coefficients of the model are determined by non-linear regression analysis, but several procedures have been used by different researchers:

<u>Direct non-linear regression</u>

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The coefficients are determined by fitting the emission rate data calculated from the measured VOC concentrations to the model directly. Colombo et al. (1991) used this method successfully to characterize the VOC emission profiles of five household products (two liquid floor detergents, one wax, a spray detergent for carpets and a spray for furniture).

Two phase approach

Chang and Guo (1992) used the double exponential model to describe the VOC emissions from a wood stain. To determine the model coefficients, they divided the whole emission-testing period into two phases:

(1) an initial phase with high and rapid decaying emission rates (first 10 hours), described by:

$$E_1(t) = E_{01} \exp(-k_1 t), \quad t \leq 10 \text{ h}$$

(2) a second phase with low and slowly decaying period (20 to about 150 hours), described by:

$$E_2(t) = E_{02} \exp(-k_2 t), \quad 0 < t \leq 150 \text{ h}$$

The total emission factor was then approximated by a simple addition:

$$E(t) = E_1(t) + E_2(t)$$

They found that the model fitted the measured results closely since $E_{01} \gg E_{02}$ and $k_1 \gg k_2$. The validity of this model to other materials has to be checked to ensured that the values of E_{01} and k_1 are significantly larger than E_{02} and k_2, respectively.

Two stage approach

Wilkes et al. (1996) used the following procedure to determine the coefficients of the double exponential model for the VOC emissions from interior latex paints:

(1) E_{02} and k_2 are first determined by fitting the equation, $E(t) = E_{02} \exp(-k_2 t)$ to the test data measured after $t > 48$ h.

(2) E_{01} and k_1 were then estimated by fitting the double exponential model to all measured data with E_{02} and k_2 evaluated from step (1).

The first period of emission testing data usually have greater experimental uncertainty than the second period since they are strongly dependent on the procedure of specimen preparation (e.g., the time used in applying a "wet" material onto the substrate, uniformity of the application,

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smoothness of the substrate surface, etc.). The two stage approach, have the advantage of minimizing the effect of experimental uncertainties on predicting the emission rates of the second emission period.

3.2.4 Second-Order Decay Model

$$E(t) = E(0) / [1 + k_2 E(0) t]$$

where,

k_2 = second-order decay constant, m^2/mg

It is called the second-order decay model because it is derived from $d[E(t)]/dt = k_2 E^2$ with the initial condition: $E(t) = E(0)$ at $t = 0$. It gives a very high initial emission rate followed by low and long-lasting emissions. Tichenor et al. (1991) showed that this model described well the emissions from wood finishing materials. Clausen et al. (1993) represented k_2 as k_{2l}/L to account for the effect of the thickness of a vinyl flooring (k_{2l} is the second-order decay constant for unit thickness, L is the thickness of the source), and showed a good fit between the model and the test data. However, the model coefficients were not reproducible based on the results of three repeat tests for the same material. The authors suggested that further validation of the model with better-characterized data were necessary.

3.2.5 The *nth*-Order Decay Model

$$E(t) = E(0) / [1 + (n-1) k_n E(0)^{(n-1)} t]^{1/(n-1)}, \quad n>1$$

where,

k_n = the *nth* order decay constant.

The *nth* order decay model is a generalization of the second-order decay model. The additional parameter, n, provides a slight improvement in fitting the emission data (Tichenor et al. 1991b).

3.3 Mass Transfer Models

From mass transfer point of view, VOC emissions from building materials/furnishings involve two processes: (1) interfacial mass transfer (i.e., the mass transfer from a material surface to its ambient air across the boundary layer airflow over the surface); (2) diffusion within the solid materials. Researchers have developed various mass transfer models that address either or both processes.

3.3.1 Interfacial mass transfer

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The interfacial mass transfer across the boundary layer may be described by (Kays and Crawford 1981):

$$r(t) = K[C_s(t)-C(t)]$$

where,

r	= rate of interfacial mass transfer, $mg/(m^2h)$;
C_s	= air-phase concentrations at the interface of the surface, mg/m^3;
C	= air-phase concentrations in the "free stream" over the surface, mg/m^3;
K	= mass transfer coefficient, m/h;
t	= time, h.

Note: r(t) is positive if the mass transfers from the surface to the ambient air (i.e., emission).

The above equation states that the interfacial mass transfer rate (i.e., the emission factor in this case) is proportional to the concentration differential between the emitting surface and the ambient air. This principle has been applied successfully to model the initial period of the VOC emissions from "wet" materials, which are mainly controlled by the evaporative process. Different assumptions about $C_s(t)$ has led to different interfacial mass transfer models as described below:

3.3.1.1 Constant vapor pressure model:

$$E(t) = K [Cv - C(t)]$$

where,

Cv = VOC concentration in air corresponding to the saturated vapor pressure at the temperature of testing, mg/m^3;

In this model, the concentration on the material surface is assumed to be constant and equal to the saturated vapor concentration in air. Chang and Krebs (1992) used this model to describe the emission of para-dichlorobenzene from a solid moth repellent. They assumed that Cv was equal to the measured headspace concentration, while K was determined by regression analysis on the VOC concentration data from dynamic chamber testing. The model is only applicable to evaporative sources with a constant vapor pressure.

3.3.1.2 VB (vapor pressure and boundary layer) model:

$$E(t) = K [Cv M(t)/Mo - C(t)]$$

where,

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$M(t)$ = amount of VOC mass in the source, mg/m^2;

Mo = total emittable VOC mass in the source, mg/m^2;

This model assumes that $C_s(t)$ is proportional to the VOC amount remained in the source (Guo and Tichener 1992). When the VB model is applied to a well mixed chamber/space that has a constant air change rate and zero concentration at time zero, this model results in the following equation for the concentrations in the chamber:

$$C(t) = [L \ Cv \ K/(r_1-r_2)] \ [\exp(r_1 \ t) - \exp(r_2 \ t)]$$

and

$$r_{1,2} = \{[-(N + L \ K + Cv \ K/Mo)] \pm [(N + L \ K + Cv \ K/Mo)^2 - 4 \ N \ Cv \ K/Mo]^{1/2}\}/2$$

where,

L = loading ratio, m^2/m^3;

N = air change rate, $1/h$;

The concentration of the most abundant VOC measured in the headspace analysis was used as Cv. Mo was the total amount of VOC emitted during the dynamic chamber testing. K was determined by fitting the model to the concentration data from the dynamic chamber testing (K was expressed as D_f/δ, D_f---average diffusivity based on the most abundant compound; δ--- apparent laminar boundary layer thickness). The model was found to predict well the initial period (up to first 20 hours) of TVOC emissions from wood stains, polyurethane varnish and wax.

3.3.1.3 VBX Model: VB Model for the emissions of individual VOCs

To quantify emission rate of individual VOCs from paints, Guo et al. (1996b) further developed the VB model to predict the emission rates of each individual compound:

$$E_i(t) = K_i \ (Cp_i - C_i)$$

and

$$Cp_i = Cv_i \ x_i$$
$$x_i = \Pi_i/\Pi, \text{ and } \Pi = \Sigma\Pi_i$$

where,

E_i = emission factor of VOC i, mg/m^2;

K_i = mass transfer coefficient for VOC i, m/h;

Cp_i = partial pressure for VOC i expressed in mass concentration unit, mg/m^3;

Cv_i = vapor pressure for pure VOC i expressed in mass concentration unit, mg/m^3;

x_i = molar fraction of VOC i remaining in the solvent, dimensionless;

C_i = vapor pressure for VOC expressed in concentration in the air, mg/m^3.

Π_i = molar amount for VOC i remaining in the source, $moles/m^2$, $\Pi_i=M_i/mw_i$;

Π = molar amount for TVOC remaining in the source, moles/m^2, Π=M/mw$_{TVOC}$;
M$_i$ = mass amount for VOC i remaining in the source, mg/m^2;
M = mass amount for TVOC remaining in the source, mg/m^2;
mw$_i$ = molecular weight of VOC i;
mw$_{TVOC}$= molecular weight of TVOC.

To simplify the implementation of the model, Guo et al. (1996) also proposed to use the original VB model instead of the summation for calculating the molar amount for TVOC (Π) and that the average molecular weight of TVOC was approximated by that of the most dominant compound in the mixture. The method was validated by experimental data from small chamber and test houses for "wet" materials including wood stain, polyurethane wood finish, and floor wax, and was found to be suitable for predicting the emissions from these materials during the initial period (up to first 20 hours of testing).

3.3.1.4 A semi-empirical model for evaporative sources

$$E(t) = K [C_s(0) \exp (-k_s t) - C(t)]$$

where,

C$_s$(0) = initial VOC concentration on the emitting surface, mg/m^2;
k$_s$ = first-order constant for the VOC concentration on the surface, 1/h.

This model assumes that C$_s$(t) decreases exponentially with time. When applied to a well-mixed chamber/space that has a constant air change rate, this model results in the following equation for the concentrations in the chamber:

$$C(t) = a [(\exp(-b\ t) - \exp(-c\ t)]$$

where,

a = L K C$_s$(0)/(L K + N - k$_s$)
b = k$_s$
c = L K + N

Zhang et al. (1996) used this model to describe the initial period (first 12 hours) of VOC emissions from wood stains applied on oak wood substrates. Non-linear regression analysis was used to determine a, b and c, and then solve the algebraic equations for K, C$_s$(0) and k$_s$. The value of C$_s$(0) was found to be significantly less than the saturation (equilibrium) concentration measured in the headspace analysis. This was attributed to the time delay (about 10 to 15 minutes) between applying the material onto the substrate and placing the specimen in the dynamic testing chamber.

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The above interfacial mass transfer models are only applicable to evaporative sources such as the initial period of emissions from "wet" building materials. In practice, the initial emission period is always followed by a second period during which the internal diffusion becomes the controlling process. A transition period is also expected between the two periods. Therefore, the surface concentration [$C_s(t)$], in general, needs to be solved in conjunction with equation for the internal diffusion process within materials. This is an area that needs further studies.

3.3.2 Internal Diffusion Mass Transfer Models

Considering that most interior building materials have a much larger surface area than their thickness, we limit our discussion to 1-D models:

For non-porous solid materials, Fick's law may be used:

$$\rho_s A_s \frac{\partial C}{\partial t} = \rho_s A_s D \frac{\partial^2 C}{\partial x^2} + \rho_s A_s r_s$$

where,

A_s = area of the surface, m^2;
C = concentration in the solid, mg-species/mg-solid;
D = diffusion coefficient in the solid, m^2/h;
r_s = local generation rate, mg-species/mg-solid;
ρ_s = density of the bulk solid, mg/m^3.

Assuming a zero local generation rate and a uniform initial VOC concentration within the material, Little et al. (1994) used the above model to describe the VOC emissions from carpets. The diffusion coefficients were estimated to be in the order of 10^{-14} to 10^{-11} m^2/s, and the model was found to fit reasonably well the emission data over a period of about 150 hours.

Christiansson et al. (1993) derived the following simplified model based on the above Fick's law:

$$E(t) = Co \ (D/\pi/t)^{0.5}, \ \text{when } M(t)/Mo \geq 50\%$$
$$E(t) = 2 \ Co \ D/z \ \exp(D \ \pi \ t/4 \ z^2), \ \text{when } M(t)/Mo < 50\%$$

where,

Co = initial pollutant concentration in the source, mg/m^3;
D = diffusivity of the pollutant in the source, m^2/h;
π = 3.14159;
$M(t)$ = amount of pollutant remaining in the source, mg/m^2;
Mo = initial amount of pollutant remaining in the source, mg/m^2;
z = thickness of the source.

Their results show that the above model described successfully the VOC emissions from PVC flooring materials tested.

For porous solid materials, diffusion within the solid can be attributed to three types: (1) diffusion in the air-phase, which can be characterized by an effective diffusion coefficient, D_e, in analogous to the Fick's law; (2) diffusion along the surfaces of the solid phase (surface diffusion), in which the diffusion coefficient is usually dependent on the concentration of the sorbed phase on the surfaces; (3) diffusion through the solid. The partial differential equation for describing the diffusion within porous materials is much more complicated. For example, for sheet-like porous solids, assuming a) the sorbed-phase concentration remains in equilibrium with the porous air-phase concentration, and b) the surface and solid-phase diffusions are negligible, we have (Axley 1995):

$$\rho A_s \varepsilon \frac{\partial C}{\partial t} + \rho_s A_s \frac{\partial C_s}{\partial t} = \rho A_s D_e \frac{\partial^2 C}{\partial x^2} + \rho_s A_s r_s$$

where,

D_e	= effective diffusion coefficient in the porous gas-phase, m^2/s;
ρ	= gas-phase density, g-air/m^3;
ρ_s	= density of the bulk porous solid, g/m^3;
C	= gas-phase concentration, g-species/g-air;
C_s	= sorbed-phase concentration, g-species/g-bulk solid.

The above equation simply states that the total accumulation of the compounds (gas-phase plus the sorbed-phase) in a control volume is equal to that transferred by porous gas-phase diffusion and that by the distributed generation.

While the mass transfer equations are readily available, their applications to building materials are hindered by the lack of data on the diffusion coefficients for various materials. Methods for experimentally determining the diffusion coefficients are therefore necessary.

3.3.3 Other Mass Transfer Related Models

3.3.3.1 A Model for Formaldehyde Emissions from Manufactured Wood Products

$$E(t) = [E(0) - K C]$$

where,

K	= a proportional coefficient.

This model assumes that the source has a maximum (constant) emission factor when the ambient concentration is zero and it decreases linearly with the increase of the concentration in air (Hoetjer and Koerts 1986, Matthews et al. 1987). This model is essentially equivalent to the constant vapor pressure model (see section 3.3.1.1) if one assumes that the surface concentration is constant and equal to E(0)/K.

3.3.3.2 A Model for Pollutant Emissions from Treated Wood Products

$$E(t) = E(0) [1 - C(t)/Cv] [M(t)/Mo]$$

where,

Cv	= saturated vapor concentration of the pollutant in air, mg/m^3;
M(t)	= amount of pollutant remaining in the source at time t, mg/m^2;
Mo	= total emittable amount of pollutant in the source, mg/m^2.

This model assumes that the emission factor at any given time is proportional to the amount of pollutant remaining in the source and the difference between the pollutant concentration in air and the saturation vapor concentration. Jayjock et al. (1995) showed that this model could describe well the emissions of biocides from isothiazolone-treated wood products.

4. SUMMARY AND CONCLUSIONS

The above review indicated that various statistical models are available for analyzing and reporting emission test results. As these models are derived from relatively short-term test data (mostly up to one-week period), their validity for predicting the emissions beyond the test period has, yet to be further validated with some long-term emission test data.

The trend in predicting the rates of VOC emissions from building materials/furnishings is to use mechanistic models that are based on fundamental mass transfer theory (i.e., the so called mass transfer models). For "wet" coating materials applied on a realistic substrate (such as wood stains on an oak wood, paints on a gypsum board, etc.), the existing interfacial mass transfer model for evaporative sources appears to be adequate only for the initial emission period which is dominated by evaporation (about first 12 hours). A more complete model that describes both the evaporative mass transfer and the internal diffusion (which becomes a controlling factor for the emission process after the initial emission period) is, therefore, needed for "wet" coating materials. For "wet" installation materials (such as adhesives, caulks and sealants), effective mass transfer models are yet to be developed. For dry materials (such as carpets, vinyl floorings, wood-based panels, gypsum wallboards, etc.), the 1-D internal diffusion mass transfer model has shown promising results, but comprehensive validations are necessary. The applications of mass transfer models are also hindered by the lack of data on the model parameters such as the in-material diffusion coefficients of VOCs for various building materials.

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Therefore, there is a need to develop mass transfer models that can better describe the emission process and thus for predicting the emission rates beyond the standard testing period and testing conditions. Models for material systems (assemblies) and data for validating such models are especially needed.

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