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Sink Effect Study for Common Building Materials
— A Literature Review & Research Plan

by Y. An, J.S. Zhang, and C.Y. Shaw

Internal Report No. (IRC-IR-681)

Date of issue: June 1997

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1. Introduction

Sink effect describes the sorption and desorption (re-emission) phenomena of indoor air pollutants on the interior surfaces of building materials and furnishings. These indoor air pollutants include many volatile organic compounds (VOCs) which have a wide range of physicochemical characteristics (e.g., vapor pressure, boiling point, and diffusion properties).

Studies have shown that the re-emissions of adsorbed organic vapors, the so called sink effect, can contribute to elevated concentrations of VOCs in the indoor environment (Korte and Gevefugi, 1987 and Nielsen, 1987). It has been noted that the sink effect of most building materials can last for a long period and even for the entire service life of a building (Nielsen, 1987). The building materials include floors (particularly carpets and rugs), walls, ceilings, HVAC systems, and furnishings.

Sink effect can have a significant impact on the material emission characteristics(Tichenor, 1992). Some experimental results suggested (Clausen, 1993 and De Bortoli, et al, 1995) that the emission characteristics of some materials could be more accurately described by a source-sink model (i.e., source emission process plus adsorption-desorption process) than a source model alone. Hence, a comprehensive description of sink effects is essential for developing models for predicting the overall indoor air quality level in buildings (Sparks, et al, 1993 and Neretnieks, et al, 1993).

This report reviews published work on sink effect in indoor environment, including models, experimental approaches and available data. The research plan for studying sink effect is also outlined.

2. Sink Models

Sink effects may involve two processes: the adsorption/desorption of VOCs on a material surface and the diffusion of VOCs from the surface into the material interior (i.e., in-material diffusion). Existing models for describing indoor sinks include: sorption models, boundary-layer models, diffusion models, hybrid models, and sorption-degradation model. Sorption and boundary-layer models ignore in-material diffusion and deals with the adsorption/desorption process only. Diffusion models regard the in-material diffusion as the controlling process in indoor sinks. Hybrid models consider both the sorption and the in-material diffusion processes. The sorption-degradation model considers the degradation of chemical compounds in the sink in addition to the sorption process.

2.1 Sorption Models

Sorption models describe the sink effect mainly as a surface phenomenon, namely an adsorption/desorption process, i.e., the transport of the adsorbate (VOC molecules) between two alternative phases: the bulk air phase and the adsorbed phase. The adsorption of any VOC species onto a material surface include: 1) the transport of the species from bulk air to an active site of the surface adsorbent; 2) the deposition of the species on the active site; and 3) possible post-deposition activities which include the transport of the desorbed species between adjacent active sites and the multilayer adsorption (i.e., adsorption on the adsorbed species). However, multilayer adsorption is normally not considered for studying indoor sink effects, since the VOC concentration in the indoor air is very low (< 1ppm).

There are two types of sorption models: equilibrium sorption model and two-sink model. The equilibrium sorption model assumes that the adsorption/desorption process takes place under the condition of thermodynamic equilibrium, in which the adsorption rate of an adsorbate is equal to its desorption rate under a constant environmental condition (humidity, temperature and pressure). The two-sink model assumes indoor sink has two types of adsorption processes: reversible adsorption and irreversible adsorption.

2.1.1 Equilibrium Sorption Model

Based on the *Langmuir isotherm* (Yang, 1987), the adsorption/desorption equilibrium of a gaseous compound on a materiel surface under the steady-state conditions can be described by the equation,

$$k_{aL}C_e(1 - Q/Q_S) = k_{dL}'Q/Q_S \qquad (1)$$

where k_{aL} is the adsorption rate constant; C_e is the equilibrium concentration of the adsorbate in the gas phase; k_{dL}' is a desorption constant; Q is the number of occupied sites per unit surface area; and Q_S is the total number of the available sites per unit surface area. Under the condition of low vapor concentrations (e.g., in the case of indoor VOCs), $Q/Q_S << 1$. Equation (1) can be simplified to a linear form:

$$k_{aL}C_e = k_{dL}'Q/Q_S \qquad (2)$$

Since k_{dL}' and Q_S are constants and Q is linearly proportional to the mass per unit area of the surface, M_e. It follows that (Tichenor, et al, 1991):

$$k_{aL}C_e = k_{dL} M_e \qquad (3)$$

where k_{dL} is the desorption rate constant. Let:

$$K_L = k_{aL}/k_{dL} \qquad (4)$$

Therefore:

$$M_e = K_L C_e \qquad (5)$$

where K_L is known as the equilibrium constant.

Equation (5) is called the linear sorption equilibrium model. Axley and Lorenzetti (1993) proposed a slightly different form of the linear sorption equilibrium model, which takes the form:

$$C_{se} = K_P C_e \qquad (6)$$

where C_{se} is the amount of the adsorbed species on the surface in terms of vapor concentration and K_P is called partition coefficient (i.e., the ratio between the vapor concentration in the adsorbed phase and in the bulk air). It is noted that the K_P is dimensionless and the K_L is in the unit of meter, e.g., if C_e is in mg/m^3, then M_e is in mg/m^2.

When a linear equilibrium sorption model is used, a mass balance has to be made on the control volume containing the adsorption surface. The following example (Tichenor, et al, 1991) illustrates the application of the linear equilibrium sorption model to a sink test using an environmental chamber.

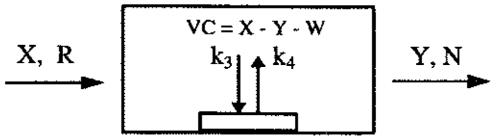

Figure 1. Schema of Mass Balance in an Environmental Chamber
(For Sorption Model)

As shown in Figure 1, a mixture of vapor compounds each with a known concentration is introduced at a constant rate into the chamber where the sink material is placed. The control volume is the chamber. The air in the chamber is well mixed. The concentrations of the vapor compounds are monitored at the chamber outlet. The concentration of each compound in the chamber increases at the beginning and eventually reaches a constant when an equilibrium is established.

In Figure 1, X is the vapor mass introduced into the chamber at a rate of R; Y is the mass exit from the chamber at an air change rate of N; W is the mass in the sink; k_3 and k_4 are the rate constants from and to the sink, respectively; C is the concentration of the compound of interest in chamber air; V is air volume of the chamber; and hence, VC is the mass of the compound in the chamber air. With these definitions, we have:

$$dX/dt = R \tag{7}$$

$$dY/dt = N(X - Y - W) = NVC \tag{8}$$

$$\begin{aligned} dW/dt &= k_3(X - Y - W) - k_4W \\ &= k_3VC - k_4MA \end{aligned} \tag{9}$$

where M is the mass deposited on sink surface, in mass/surface area of sink material; and A is the surface area of sink material. At equilibrium, dW/dt = 0. Thus,

$$k_3VC = k_4MA \tag{10}$$

If a Langmuir adsorption process is assumed, comparison of equations (3) and (10) yields: $k_{aL}A = k_3V$, or $k_3 = k_a(A/V)$ and $k_4 = k_d$. Based on the mass balance in the chamber, the concentration change rate of the interested compound in chamber air is derived (assuming a perfect air mixing in the chamber):

$$\begin{aligned} VdC/dt &= dX/dt - dY/dt - dW/dt \\ &= R - NVC - k_{aL}CA + k_{dL}MA \end{aligned} \tag{11}$$

At equilibrium, the initial conditions are t = 0, C(0) = C_e, and M(0) = M_e = $C_e(k_{aL}/k_{dL})$. The solution of equation (11) is:

$$C(t) = C_e\,[(N - r_2)e^{-r_1t} - (N - r_1)e^{-r_2t}\,] / (\,r_1 - r_2\,) \tag{12}$$

$$M(t) = k_{aL}C_e(\,r_1e^{-r_2t} - r_2e^{-r_1t}\,) / k_{dL}\,(\,r_1 - r_2\,) \tag{13}$$

where

$$r_{1,2} = (\,N + k_{aL} + k_d\,) \pm [(\,N + k_{aL} + k_d)^2 - 4Nk_d]^{1/2} / 2 \tag{14}$$

Values of k_{aL} and k_d are obtained by fitting equation (12) using a nonlinear regression curve fit routine.

The equilibrium sorption model is the most widely used model for the sink effect studies. It is relatively simple to use. Up to date, it has been used to estimate the sink effect of building materials in both chambers (Levsen and Sollinger, 1993 and Jorgensen, et al, 1993) and real buildings (Neretnieks, et al, 1993). The adsorption/desorption equilibrium coefficient (K_p or K_L) derived from this model can be related to the physicochemical properties of the adsorbate. However, the equilibrium sorption model is limited by the assumption of low vapor concentration and uniform adsorption sites. Two-sink model and boundary-layer model are, therefore, developed to solve these problems.

2.1.2 Two-Sink Model

Colombo, et al, (1993) suggested a two-sink model which classified the sink effects into two types: reversible sink and irreversible sink. The reversible sink indicates that the adsorption rate constant (k_{a-r}) and the desorption rate constants (k_{d-r}) are comparable. The adsorption/desorption coefficient (K_r) for reversible sink can be derived from k_{a-r} and k_{d-r} by equation 4. The irreversible sink comprises a large adsorption rate constant (k_{a-i}) and a very small desorption rate constant (close to zero). The mass (m) adsorbed by the material surface is a function of the vapor concentration (C_e) above the surface, K_r, and k_{a-i}, as the following:

$$m = f(K_r, k_{a-i}, C_e) \tag{15}$$

Another two-sink model was developed recently (De Bortoli, et al, 1996). It divided sink effects into two classes: fast sink and slow sink. The fast sink is characterized by a relatively large adsorption/desorption equilibrium coefficient (K_1) and the slow sink is characterized by a relatively small adsorption/desorption equilibrium coefficient (K_2). The mass adsorbed by the material surface is a function of the vapor concentration above the surface and the two adsorption/desorption equilibrium coefficients (K_1 and K_2):

$$m = f(K_1, K_2, C_e) \tag{16}$$

Two-sink model is an improved equilibrium sorption model, which describes sinks as a process involves two types of adsorption. Therefore it applies to materials with non-uniform adsorption sites.

2.2 Boundary-Layer Model

The boundary-layer model assumes (Axley and Lorenzetti, 1993) that a boundary layer exists (see Figure 2) immediately above the adsorption surface. The vapor concentration in this boundary layer varies from bulk air concentration to the near-surface concentration, i.e., a concentration gradient is established in the boundary layer. The net mass transport rate from the bulk air to the surface, dW/dt, may be described by:

$$dW/dt = k_b \rho_b A(C - C^*) \tag{17}$$

where k_b is the average boundary-layer mass transfer coefficient in m/s, ρ_b is the average air density of boundary-layer in mg/m^3, A is the projected surface area of the adsorber in m^2, C is the bulk air concentration, and C^* is the near-surface vapor concentration, both C and C^* in mg (of species)/mg (of air). Referring to Figure 1 and equation (11), the mass balance in the chamber will be:

$$\begin{aligned} VdC/dt &= dX/dt - dY/dt - dW/dt \\ &= R - NVC - k_b\rho_b A(C - C^*) \end{aligned} \qquad (18)$$

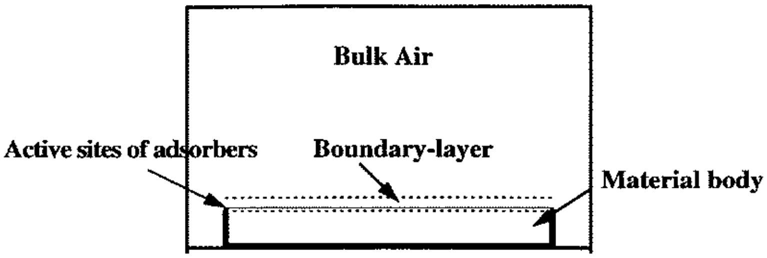

Figure 2. Diagram of Boundary-Layer

The boundary-layer model is developed on the basis of fundamental mass transfer theory. It is not limited by the conditions such as low vapor concentration and uniform adsorption sites. It can be used to describe sinks in which the diffusion through the boundary layer is the controlling process. It has also been shown that when the air mass flow rate above the surface is smaller than the boundary-layer mass transfer rate, the equilibrium sorption model is expected to be accurate, otherwise the boundary-layer model is expected to be accurate (Axley and Lorenzetti, 1993).

2.3 Diffusion models

The diffusion models (Little and Hodgson, 1996) suggest that the in-material diffusion is the dominant process once the sorption equilibrium is established at the surface. According to *Fick's law of diffusion* (Alkins, 1978), the equation for transient diffusion of species through a homogeneous polymer slab is:

$$\delta C_s/\delta t = D\,(\delta^2 C_s/\delta z^2) \qquad (19)$$

where $C_s(z,t)$ is the concentration of the adsorbed species in the polymer slab, t is time, D is the diffusion coefficient, which is assumed to be independent of concentration, and z is the distance from the material interior to the surface, as shown in Figure 3.

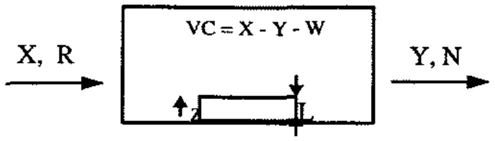

Figure 3. Schema of Mass Balance in an Environmental Chamber
(For Diffusion model)

In Figure 3, X is the vapor mass introduced into the chamber (volume V) at a rate of R, Y is the mass exit from the chamber at an air change rate of N, W is the mass in the sink, C is the VOC concentration in the chamber air, and L is the thickness of the polymer slab. Assuming that the adsorbate is uniformly distributed through the polymer at a concentration of C_s. The first boundary condition assumes that there is no flux out of the base of the polymer, then:

$$\delta C_s/\delta z_{(z=0)} = 0 \qquad (20)$$

A second flux boundary condition is imposed through a mass balance in the chamber air, as referring to Figure 3, Eq. (7), and Eq. (8), then:

$$\begin{aligned} VdC/dt &= dX/dt - dY/dt - DA\delta C_s/\delta z_{(z=L)} \\ &= R - NVC - DA\delta C_s/\delta z_{(z=L)} \qquad (21) \end{aligned}$$

where A is the surface area.

Diffusion model assumes that the sink effect is mainly controlled by the in-material mass transport and the surface sorption is a relatively fast process and negligible. Since the in-material diffusion of VOCs is solely governed by the physical properties of VOCs (e.g., molecular weight) and sink materials, the diffusion model is essentially chamber independent (Little and Hodgson, 1996).

2.4 Hybrid models

Hybrid models describe sink effect as both a sorption (or boundary-layer diffusion) and an in-material diffusion process. One example is the equilibrium sorption/diffusion model (Little and Hodegson, 1996). The VOC concentration on the surface can be obtained by combining Eq. (6) and Eq. (21):

$$VdC/dt = R - NVC - DA\delta C_s/\delta z_{(z=L)} - K_pC \qquad (22)$$

Another example (Axley and Lorenzetti, 1993) of hybrid models is the boundary-layer model (Eq. 18) and in-material diffusion model (Eq. 21):

$$VdC/dt = R - NVC - k_b\rho_b A(C - C^*) - DA\delta C_s/\delta z_{(z=L)} \qquad (23)$$

Hybrid models are recently developed to consider both surface effect and in-material diffusion. However, the hybrid models are complicated, which include several coefficients. The values of these coefficients depend on the type of adsorbate, type of sink materials, and environmental conditions (e.g., air velocity on the surface).

2.5 Sorption-Degradation Model

Jayjock and co-workers (1995) proposed the following sorption-degradation model:

$$dW/dt = k_3VC - k_4MA + k_{deg}MA \qquad (24)$$

where W is the mass in the sink; k_3 and k_4 are the rate constants from and to the sink, respectively; V is air volume of the chamber; C is the concentration of the compound of interest in chamber air; M is the mass deposited on sink surface, in mass/surface area of sink material; A is the surface area of sink material; and k_{deg} is the degradation rate constant of the compound in the sink. The sorption-degradation model was used to describe the sink effect of biocide on treated wood.

A systematic experimental evaluation of the existing models is needed in order to clarify when and which model can be adequately applied. Furthermore, the coefficients of existing sink models have been determined only for limited numbers of VOCs. Models for predicting the sink effects of different VOCs based on the physicochemical properties (i.e., boiling point, polarity, etc.) are yet to be developed.

3. Experimental Approaches

Sink effect is a complex phenomenon. It varies with the properties of sink materials such as surface structure and specific surface area, the properties of VOCs such as vapor pressure and boiling point, and the environmental conditions such as temperature, humidity, air change rate, and air flow condition above the material surface (Tichenor, et al, 1992). In addition, VOC concentrations in the indoor environment is generally low, i.e., < 1 ppm (Wolkoff, 1995), which creates additional challenge in quantifying the sink effects. Sink effect tests, therefore, have to be conducted under carefully controlled conditions.

Sink effect in the indoor environment is usually evaluated by determining the adsorption constant and desorption constant included in equilibrium sorption models, e.g., equation (11). The key data needed for determining these constants is the VOC concentration vs. time profile. VOC concentrations are commonly measured by gas chromatography (GC) with flame ionization detector (FID) or mass spectroscopic detector (MS). In general the measurement comprises three steps (Saunders, 1996): 1) collection of VOCs at a selected sampling location with sorption tubes, 2) thermal desorption of VOCs from sorption tubes by a desorber unit, and 3) quantitative analysis of VOCs by GC/FID or GC/MS. This practice is used for measurement of indoor VOCs in the range of 0.01 $\mu g/m^3$ to 1000 mg/m^3 approximately, which is equivalent to 0.05 ppb - 500 ppm depending on the molecular weight of the measured VOCs. Typical sampling volume is about 1-10 liters.

The VOC concentration level used in previous sink effect tests were usually in the range of 0.1 - 100 mg/m^3. Further tests may require the use of an analytical system with lower detection limit because the geometric mean concentration of individual indoor VOCs is below 0.1 mg/m^3 (Jackson, et al, 1987) and the change of VOC concentrations in the indoor environment caused by sink effect at any time point may be very small, e.g., less than 10 % of the individual VOC concentrations. Thus, the appropriate concentration range for sink effect studies is approximately 0.1 to 10 $\mu g/m^3$. To conduct the measurement accurately, a detection limit of less than 0.01 $\mu g/m^3$ is required.

3.1 Chamber Test

A commonly used sink test protocol includes a dynamic chamber test, followed by a regression analysis of the data using a suitable sink model. The conditions of the test chamber are usually set at 23^0C, atmospheric pressure, 45 - 50% relative humidity (RH), and 0.5 - 1 air change per hour (ACH). These parameters should be well controlled, e.g., temperature: $\pm 0.5^0$C; RH: ±5%; and ACH: ±2% (Borrazzo and Davidson, 1990). The test material is placed in the chamber, as shown in Figure 4, and exposed to a mixture of known concentrations of VOCs. The test chamber is made of either stainless steel (Levsen and Sollinger, 1993) or glass (Seifert and Schmahl, 1987 and Jorgensen, et al, 1993). An empty glass chamber or stainless steel chamber itself may have slight sink effect (Colombo, et al, 1993), which has to be taken into account in analyzing the experiment data.

A typical test procedure developed by Tichenor, et al (1991), is described below. The test material is placed in the chamber. After it is conditioned with clean air for a certain period of time (e.g. 24 hours), a portion of the supply air is replaced by the test organic compound which is supplied by a commercially available permeation tube inside a constant temperature oven. The test compound is injected into the chamber continuously for 48 hours. At the end of the 48 hours, the permeation tube is disconnected. The amount of the compound introduced into the chamber can be calculated by its permeation rate (provided by the manufacturer) at the temperature and the air flow rate of the test chamber.

During the 48 hours, the VOC (or VOCs) inside the test chamber is measured by collecting the gas sample with sorption tubes. VOC samples should be collected as frequently as possible immediately after the injection of the organic vapor. Once the concentration reaches an apparent equilibrium value, the sampling frequency may be decreased. At the end of the 48 hours, when the permeation tube is disconnected, the sampling frequency has to be increased again in order to accurately measure the decay of concentration in the chamber. The VOCs collected by the adsorption tubes are quantitatively analyzed by GC/FID or GC/MS.

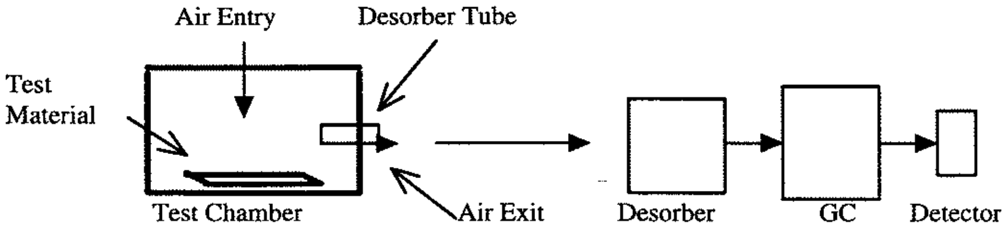

Figure 4. Diagram of Dynamic Chamber Test and GC/FID Analysis

Some researchers have successfully tried to introduce the organic vapor by directly placing the VOC diffusion tube(s) in the test chamber (Levsen and Sollinger, 1993). The VOC concentration in the chamber was monitored using the same method as above. The sink effect test was conducted with and without the sink material sample placed in the chamber. The sink effect was determined by comparing the results. This method has the advantage of simplifying the experimental procedure.

An accurate determination of the model coefficients is also dependent on the experimental design and computation method. De Bortoli, et al, (1995) reported that the same equilibrium sorption model may result in different k_a and k_d when using different initial values in the computation. In addition, the calculated k_a and k_d can vary from case to case for the same sink material, suggesting a possible influence of test conditions. In order to obtain good estimation results, Guo (1997) suggested that in performing the chamber test, two test periods should be included: an adsorption dominated period and a desorption dominated period. Thus, the experimental data provided for estimating k_a and k_d represent a broad range of adsorption/desorption processes which may minimize the estimation error when experimental data are limited.

3.2 Packed Columns

In addition to the dynamic chamber test the packed column test has also been applied to sink effect studies (Borrazzo and Davidson, 1990). Samples of sink materials (e.g. fibers) were packed into a column. The organic vapor was introduced into the column with carrier gas. An electron-capture detector (ECD) which was connected to the end of the column was used to record the retention time of the interested organic compound.

In gas-solid chromatography, the retention time of a compound can be used to evaluate the partition coefficient describing the equilibrium distribution of the compound between the vapor and sorbed phases (Kiselev and Yashin, 1969). A linear partition isotherm is usually applicable when adsorption at very low vapor concentrations. In this case, the retention time t_R (min) of the compound of interest can be expressed as:

$$t_R = t_m (1 + K M_s / V_m) \tag{25}$$

where t_m (min) is the retention time of an unretained component, K is the partition coefficient which equals to the sorbed phase concentration in g (adsorbate)/g(adsorbent) divided by the gas phase concentration in g (adsorbate)/m^3 (air) at equilibrium, M_s is the mass of adsorbent fiber in the column (g), and V_m is the volume of the column occupied by the gas phase (m^3). Note that the units for K imply that gas phase concentrations are expressed on a per volume basis, while sorbed phase concentrations are expressed as mass sorbed per mass of adsorbent.

The method is good for evaluating K values for various VOCs. Since K values are characteristics of the adsorption-desorption equilibrium, they can be used to evaluate the sink effects in the indoor environment if the linear equilibrium adsorption model (e.g., Eq. 6) applies. However, the method is limited to certain materials such as carpet or upholstery fibers.

3.3 Field Tests

The sink effect of building materials can also be evaluated in field test sites using small test chambers. A simple test procedure used by Kjaer, et al (1996) was described as follows: Sink materials such as carpet or paint applied on a steel plate were placed in small chambers. After well ventilated for a few days, several of the test chambers (called field-chamber hereafter) were moved to a test site. The remaining test chambers (called lab-chamber hereafter) containing the same sink material were left in the laboratory to produce the reference data for comparison. The field test site was a newly renovated office and was contaminated by a variety of VOCs. The environmental conditions (temperature, humidity, air exchange rate, etc.) of the office were well controlled and the field-chambers were ventilated with the office air, and were left in the office for 20 days. The VOC concentrations in the field-chambers and office air were monitored during this period. After 20 days, the field-chambers were moved back to the laboratory with both its air intake and exhausted sealed. After conditioned in the laboratory for 3 more days, the air samples in the field-chambers were analyzed and compared with those from the lab-chambers. The results indicated that most VOCs presented in the office air were detected in the air samples of field-chambers, indicating the adsorption/desorption of the VOCs in the office air on material samples tested in the chambers. The sink effect was estimated by comparing the results obtained from the lab- and the field-chambers.

An alternative method to investigate sink effect in field test site is to compare the measured material emissions with those measured under no sink conditions (e.g., those measured in a stainless steel chamber). By this method the sink effect in a test house was revealed (Tichenor, et al, 1991). The experiment was conducted by applying a coated parquet in the test house. The concentration of the total vapor organics was monitored over time and recorded. The concentrations after 48 hours were much higher than the values predicted by the emission model which assumes no sink effect. The results suggested the presence of the sink effect in the test house. The equilibrium sorption model satisfactorily fitted the experimental data.

4. Data for indoor sinks

To date, over 20 organic compounds have been tested for sink effects on different building materials. These materials include carpet (Kephalopoùlos, et al, 1996), ceiling tile (Tichenor, et al, 1991), wallboard (Colombo, et al, 1993), latex caulk (Dunn and Tichenor, 1988) and blown vinyl (Kephalopoulos, et al, 1996). The tested VOCs include alkanes, alkenes, aromatics, chlorine substituted alkenes, alcohols, ketones, and esters. Their boiling points (BP) range from 86.7^{0}C (chloroethylene) to 270^{0}C (pentadecane). The results indicated that most of these materials exhibited slight sink effects. In addition, adsorption/desorption process of formaldehyde which is a very volatile compound (BP = -19^{0}C) on unpainted gypsum wallboard has also been reported (Matthews, et al, 1987).

The evaluation of the sink effect is benefited by the knowledge of the properties of the VOCs such as boiling point, function group, and structure, and the properties of sink materials such as the surface type (textile surface or synthetic polymer surface) and the existence of polar compounds in the material surface. Moreover, careful design and control of the experimental conditions is especially important (Tichenor, 1996). It is generally assumed that only physisorption is involved in indoor sink effects. Physisorption results from the *van der Waals interactions* which include: a) dipole/dipole interactions between polar molecules, b) dipole/induced-dipole interactions between polar and non-polar molecules, and c) induced-dipole/induced-dipole interactions between non-polar molecules. The strength of the interactions decrease from type a to type c. The strong interactions between the vapor compounds would lead them to condense on solid surface easily. The polarity of the compounds used to describe the interaction strength between molecules is a key character during adsorption-desorption processes. Therefore, the molecular polarities are usually taken into account for characterizing sink effects in the indoor environment. However, the polarities of VOCs are not quantitatively available. They are commonly represented by the vapor pressure (VP) or the boiling point (BP) of the compounds. The compound with polar groups has higher BP or lower VP than those with no polar groups.

The dependence of sink effect on the VP or BP of a vapor compound has been frequently observed. Levsen and Sollinger (1993) investigated the sink effects of alkanes on carpet. The result showed that the adsorption-desorption equilibrium coefficients increased with the BP, but no simple correlation was found. Chang and Guo (1994) studied sink effect related to wood stain and floor wax in a test chamber. The adsorption-desorption equilibrium coefficients of four alkanes were estimated from experimental data by using an equilibrium sorption model. It was found that the adsorption-desorption equilibrium coefficients of the compounds were correlated to the corresponding VP with an approximately logarithmic relationship.

Tichenor, et al (1990 and 1991) studied the adsorption-desorption processes of tetrachloroethylene (BP = 121^0C) and ethylbenzene (BP = 136^0C) on different building materials. He found that for sink materials such as wallboard and ceiling tile the adsorption rate constants (k_a) of ethylbenzene were higher than that of tetrachloroethylene, while the desorption rate constants (k_d) of both compounds were the same. The adsorption-desorption equilibrium coefficients of ethylbenzene derived from those rate constants were higher than that of tetrachloroethylene. The result also showed that the adsorption-desorption equilibrium coefficients of the two compounds on carpet were almost identical. These results indicated that the correlation between the adsorption behavior of each of these two compounds and VP (or BP) is affected by the type of the adsorber material. Borrazzo, et al (1993) investigated four types of sink systems: 1) polar compound (ethanol) adsorbed on hydrophilic surface (cotton which contains hydrophilic group -OH); 2) non-polar compound (trichloroethylene) adsorbed on the same material surface as above; 3) ethanol adsorbed on hydrophobic surface (polyesters); and 4) trichloroethylene adsorbed on polyesters. They reported that the adsorption/desorption equilibrium coefficients obtained from sink systems 1 and 4 were higher than those obtained from sink systems 2 and 3. The high equilibrium coefficients for 1 and 4 was due to the high affinity between the adsorbed compound and the surface. For instance, in system 1 both the adsorbed compound (ethanol) and the surface contain hydroxyl groups. The interactions between the hydroxyl containing groups are very strong according to *van der Waals* theory (Alkins, 1978). These results indicated that the *van der Waals forces* control the indoor sink processes. Borrazzo, et al, also estimated the heat of adsorption for tetrachloroethylene deposited on the sink materials. Some of the values (e.g., -20.6 kcal/mol) were out of the range of physisorption. The average heat of adsorption for physisorption various from -15 kcal/mol to -10 kcal/mol (Hudson, 1992).

It is noted that the accuracy of estimating the adsorption coefficients depends strongly on the experimental data which are affected by the quality of test chamber and the sensitivity of the analytical instrument. De Bortoli, et al, (1996) recently reported that the poor repeatability of the experimental results with n-decane was due to the chamber's sink effect. Better results were produced after purging the chamber with clean air for one month. The adsorbed mass was accounted for the deposition into the "deep" sinks on the stainless steel surface.

The available adsorption and desorption rate constants (k_{aL} and k_d or k_3 and k_4, referring to section 2.1.1) are listed in Appendix 1. Note that k_3, k_4 and k_d are in h^{-1}, while k_{aL} is in mh^{-1}. The results indicated that a certain disparity exists among those adsorption coefficients reported by different laboratories. This disparity may be resulted from different test conditions. Colombo, et al (1993) indicated that a small variation of humidity had no significant effect on the adsorption/desorption process. While Dunn and Tichenor (1988) reported that the chamber's air change rate had a significant effect on the adsorption and desorption rate constants. The deduced adsorption-desorption equilibrium coefficients appeared to increase with the air change rate. Guo (1993) reported that both adsorption and desorption rate constants increased with the air change rate. Tichenor, et al (1991) examined the temperature effect on the adsorption-desorption coefficients. They reported that both the adsorption and desorption rate constants tested at 35^0C were significantly higher than those at 23^0C. Moreover, the desorption rate constant increased much more than the adsorption rate constant, which, in turn, caused a decrease in the adsorption/desorption equilibrium coefficient, as the temperature raised from 23^0C to 35^0C.

The different adsorption/desorption coefficients for similar materials obtained in different studies may be due to the different models used to evaluate them. As mentioned in section 2.1, the equilibrium adsorption model may not be suitable for describing some complex indoor sinks, since the surface of the building materials are normally uneven in terms of energetic distribution. Some adsorber sites on the surface may be more active than others on the same surface towards adsorption of VOCs. De Bortoli and his co-workers (1996) applied the two-sink model to study the sink effect of carpet in a small chamber. The adsorption and desorption rate constants estimated from the experimental data were 1.548 and 0.730 for the first sink compartment and 0.442 and 0.0164 for the second sink compartment. The results indicated the existence of the two sink compartments, with the first one representing a surface sink and the second one representing a deep sink. When the two-sink model was applied to test the sink effect of an empty chamber, the adsorption and the desorption rate constants in the first sink compartment were 930 and 39374, respectively. The corresponding rate constants for the second sink compartment were 0.178 and 0.0032, respectively. The result suggested that the first sink compartment had a dominant effect. In fact, for this particular case the single compartment model, i.e. the equilibrium adsorption model would be equally applicable.

Diffusion models have also been applied to investigate the indoor sinks (Dunn and Chen, 1993, Little and Hodgson, 1996, and Axley and Lorenzetti, 1993). Dunn and Chen (1993) applied both the sorption equilibrium model and diffusion model to estimate the sink effect of ethylbenzene and tetrachloroethylene adsorbed on pillows or carpet. The result showed that the sink effect of ethylbenzene on the pillow was better described by the equilibrium adsorption model, while the sink effect of tetrachloroethylene on the pillow was better described by the diffusion model. Moreover, the sink effect of ethylbenzene on carpet was best described by a hybrid model (Eqs. 14 and 16) which comprises the adsorption-desorption equilibrium and VOC diffusion into the material. It was further pointed out that when vapor compounds with higher boiling point, e.g. 136^0C for ethylbenzene, adsorbed on a sink surface, the equilibrium sorption model can be successfully applied, whereas when vapor compounds with lower boiling point, e.g. 87^0C for tetrachloroethylene, the diffusion model is preferred to be used. The authors suggested that the selection of equilibrium sorption model or the diffusion model depends on the ratio of mass flow rate in chamber air vs. the mass transport rate to/from the surface. When the mass flow rate in chamber air is lower than the mass transport rate to/from the surface, the equilibrium adsorption model is adequate. Otherwise the diffusion model is more suitable.

5. Research plan

The above literature review describes the up-to-date progress in the investigation of sink effects in the indoor environment. It shows that: 1) Several sink models, e.g., sorption models and diffusion model, have been developed and applied to examine the sink effect of building materials in test chambers; 2) Dynamic chamber technique is widely used in sink effect studies; and 3) The adsorption-desorption profiles of selected VOCs on some individual materials have been obtained by different research groups. The review also indicates:

- There is a certain variation among the sink test results. The variation may be due to the difference in test procedures and regression methods used in analyzing the data;
- The available sink effect data are insufficient for use in indoor air quality modeling;
- Differences between individual VOCs and a mixture of VOCs in terms of sink effect on building materials have rarely been studied.

Further studies are needed to identify/develop sink models for various VOCs and building materials, and to experimentally determine the model constants (e.g., adsorption and desorption constants, diffusion coefficients, etc.).

5.1 Objective

The objective of the sink effect study within the scope of the *IRC/NRC Consortium for Material Emissions and Indoor Air Quality* (CMEIAQ) project is to develop a method for predicting sink characteristics of typical building materials for typical indoor VOCs. The method will be recommended for use in indoor air quality (IAQ) prediction models.

5.2 Methodology

The dynamic chamber technique described in section 3.1 will be adopted for obtaining experimental data. Both sorption and diffusion models will be examined for their validity in describing the sink effects.

5.3 Experimental design

Experiments will be conducted under the following chamber conditions: temperature of 23^0C, humidity of 50%, supply air flow rate of 1 ACH, and air velocity of <0.05 m/s. Chamber sink effect and the system detection limit will be evaluated first. The VOC concentrations during the tests will be well above such detection limit. In order to low VOC concentrations (down to 0.01 $\mu g/m^3$), GC/MS will be used for VOC detection. Conditions for specific experiments (e.g., test of environmental effects) are described below:

5.3.1 Chemicals and materials

The selected building materials are carpets, ceiling tile, painted gypsum wall boards, and hard wood floor surfaces covered with polyurethane varnish. The above materials represent: 1) different macrostructural surfaces, such as the porous surface of carpet and the flat surface of gypsum wall board; 2) different microstructural surfaces, i.e., the flat surfaces which contain different chemical functions, e.g., painted gypsum wall board vs. hard wood floor surfaces covered with polyurethane varnish. Selection of materials with different properties is important in terms of modeling purpose, because the applicability of a sorption model or a diffusion model may basically depend on the structure of the material surface, as well as the VOCs of interest.

Individual VOCs and VOC mixture to be tested are octane, decane, dodecane, ethylbenzene, octanal, and p-dichlorobenzene. All these chemicals have been frequently detected in the indoor air environment and have been used as "sink markers". As shown in the Appendix 1, these chemicals were usually used to investigate the sink effect of building materials. The selection of these VOCs is intended to determine the relationship between physico-chemical properties of a compound and the sink effect. Tests will be conducted for the VOCs with similar structures but different boiling points. They are octane, decane and dodecane with boiling points of 126^{0}C, 174^{0}C and 216^{0}C, respectively. The purpose is to determine the correlation between boiling points and partition coefficients. Then experiments will be conducted for ethylbenzene (an aromatic with boiling point of 136^{0}C), octanal (an aldehyde with boiling point of 171^{0}C) and p-dichlorobenzene (a chlorine substituted aromatic with boiling point of 173^{0}C) to determine the effect of chemical functions on the adsorption/desorption processes.

5.3.2 Test Procedures

Experiments will first be conducted in a small chamber to determine the sink characteristics of individual material samples. An aged material sample will be placed in a small chamber and preconditioned with clean air at 1 ACH for 24 hours. A permeation tube containing the individual sink compound or a mixture of VOCs will be used to provide a constant concentration(s) at the chamber inlet air until the concentration(s) in the chamber reach the equilibrium condition. In the case of the compound with high boiling point, it may be necessary to heat the permeation tube to avoid any condensation at the outlet of the tube. The chamber is continuously ventilated during the testing period. In addition, SF_6 will also be injected to simulate the "no-sink" condition. Air samples will be collected by Tenax TA tube at the exhaust of the chamber. The sampling rate is about 250ml/min and the sampling volume is 1-5L. The VOCs will be analyzed by the a GC/MS equipped with a thermal desorber. The concentration(s) will be compared with the SF_6 concentration profile to determine the sink effect. Both existing sorption and diffusion models will be compared and used in analyzing the data. The following experiments will be conducted in the small chamber in series:

- Test of sink effect of carpet in a small chamber: A mixture of VOCs will be tested at the individual equilibrium concentration of approximately 0.5 - 1 ppm, which will allow the VOC concentrations above the detection limit of the system (~1ppb) during the whole test period. The same experimental conditions will be used for testing the selected individual VOCs. The results will be compared to the previous results to investigate the difference between the individual VOC and a VOC mixture containing the same VOCs. The experimental data will be used to evaluate the coefficients of a proper existing model. A relationship between these coefficients and the characteristics of VOCs (e.g., boiling point, vapor pressure, etc.) is expected to be established.
- Test of sink effects of other materials in small chamber: The same VOCs will be tested with different material samples under the same experimental conditions in

order to get adsorption rate constants, desorption rate constants and diffusion coefficients for different type of materials.

- Test of sink effect of materials in full-scale chamber: The selected materials will be tested in a full-scale chamber in order to investigate the interaction between different building materials as sinks and to validate the prediction method developed from the small chamber tests.

Acknowledgment

The authors are grateful for the insightful comments made by Dr. Zhishi Guo of Acurex Environmental Corporation and Mark Mason of US EPA's Indoor Environment Management Branch at Research Triangle Park, NC.

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Appendix 1. Literature Data of Sink Test

Materials	VOCs	B.P.	Ads.Const. (h^{-1})	Des.Const. (h^{-1})	Dif.Coef. (m^2/h)	Modeling fit	Ref.
Blown vinyl	1,2,4-Trimethylbenzene	168	1.68/0.21	0.60		Two-sink model	1
Blown vinyl	1,4-Dichlorobenzene	173	3.26/0.64	0.49		Two-sink model	1
Blown vinyl	2-Butoxyethanol	171	2.78/0.54	0.87		Two-sink model	1
Blown vinyl	2-Ethylhexanol	184	2.68/0.23	0.38		Two-sink model	1
Blown vinyl	♋-Pinene	155	0.36/0.24	0.41		Two-sink model	1
Blown vinyl	n-Decane	174	1.13/0.17	1.01		Two-sink model	1
Blown vinyl	n-Dodecane	216	3.29/0.20	0.40		Two-sink model	1
Blown vinyl	Tetrachloroethylene	121	2.04/0.17	7.13		Two-sink model	1
Carpet	1,2,4-Trimethylbenzene	168	1.34/0.32	0.21		Two-sink model	1
Carpet	1,4-Dichlorobenzene	173	2.46/0.71	0.20		Two-sink model	1
Carpet	2-Butoxyethanol	171	1.14/0.28	0.28		Two-sink model	1
Carpet	2-Ethylhexanol	184	0.85/0.26	0.11		Two-sink model	1
Carpet	α-Pinene	155	0.74/0.23	0.68		Two-sink model	1
Carpet	n-Decane	174	1.14/0.28	0.28		Two-sink model	1
Carpet	n-Dodecane	216	2.05/0.85	0.13		Two-sink model	1
Carpet	Tetrachloroethylene	121	1.04/0.17	0.91		Two-sink model	1
Empty chamber	1,2,4-Trimethylbenzene	168	0.98/0.17	2.56		Two-sink model	1
Empty chamber	1,4-Dichlorobenzene	173	6060/0.52	8130		Two-sink model	1
Empty chamber	2-Butoxyethanol	171	2200/0.22	9610		Two-sink model	1
Empty chamber	2-Ethylhexanol	184	1.25/0.24	1.47		Two-sink model	1
Empty chamber	♋-Pinene	155	56.4/0.18	196		Two-sink model	1
Empty chamber	n-Decane	174	91.2/0.17	332		Two-sink model	1
Empty chamber	n-Dodecane	216	0.53/0.21	0.59		Two-sink model	1
Empty chamber	Tetrachloroethylene	121	332/0.15	1410		Two-sink model	1
Gypsum board	1,2,4-Trimethylbenzene	168	1.33/0.14	0.54		Two-sink model	1
Gypsum board	1,4-Dichlorobenzene	173	1.64/0.23	0.31		Two-sink model	1
Gypsum board	2-Butoxyethanol	171	1.24/0.08	0.40		Two-sink model	1
Gypsum board	2-Ethylhexanol	184	1.37/0.76	0.11		Two-sink model	1
Gypsum board	♋-Pinene	155	1.19/0.13	0.98		Two-sink model	1
Gypsum board	n-Decane	174	1.22/1.28	0.13		Two-sink model	1
Gypsum board	n-Dodecane	216	2.00/0.09	0.16		Two-sink model	1
Gypsum board	Tetrachloroethylene	121	0.21/0.06	0.04		Two-sink model	1
Carpet	n-Decane	174	1.55/0.44	0.73/0.016		Two-sink model	2
Glass chamber	n-Decane	174	930/0.18	39374/0.003		Two-sink model	2
Carpet	Ethylbenzene	136	0.095	0.048	0.0031	Hybrid model	3
Pillow	Ethylbenzene	136	0.034	0.017	0.0098	Hybrid model	3
Latex caulk	2-Butanone	80	49/0.27	4238		Two-sink model(ACH=0.36)	4
Latex caulk	2-Butanone	80	9.3/0.39	41		Two-sink model(ACH=1.85)	4
Latex caulk	Butyl propionate	145	794/0.24	1896		Two-sink model(ACH=0.36)	4
Latex caulk	Butyl propionate	145	14/0.014	6.7		Two-sink model(ACH=1.85)	4
Latex caulk	Octanol	196	168/0.32	221		Two-sink model(ACH=0.36)	4
Latex caulk	Octanol	196	2.1/0.003	0.088		Two-sink model(ACH=1.85)	4

Appendix 1. Literature Data of Sink Test (Cont'd)

Materials	VOCs	B.P.	Ads.Const (h^{-1})	Des.Const. (h^{-1})	Dif.Coef. (m^2/h)	Modeling fit	Ref.
Nylon carpet	α-Pinene	155	0.43	0.19		Eq.sorption model	5
Nylon carpet	Toluene	111	0.18	0.34		Eq.sorption model	5
Wool carpet	α-Pinene	155	0.45	0.19		Eq.sorption model	5
Wool carpet	Toluene	111	0.26	0.43		Eq.sorption model	5
Carpet	Tetrachloroethylene	121	0.14	0.14		Eq.sorption model	6
Carpet	Ethylbenzene	136	0.07	0.08		Eq.sorption model	6
Ceiling tile	Ethylbenzene	136	0.25	0.59		Eq.sorption model	6
Ceiling tile	Tetrachloroethylene	121	4.2	0.61		Eq.sorption model	6
Pillow	Ethylbenzene	136	0.004	0.016		Eq.sorption model	6
Pillow	Tetrachloroethylene	121	0.02	0.10		Eq.sorption model	6
Wallboard	Ethylbenzene	136	0.45	1.5		Eq.sorption model	6
Wallboard	Tetrachloroethylene	121	0.21	0.14		Eq.sorption model	6
Test house	Nonane	151	0.15	0.09		Eq.sorption model	7
Test house	Decane	174	0.19	0.08		Eq.sorption model	7
Test house	Undecane	196	0.15	0.02		Eq.sorption model	7
Empty chamber	Decane	174	0.36/0.03	0.75		Two-sink model(RH=70%)	8
Empty chamber	Decane	174	0.83/-0.04	1.06		Two-sink model(RH=35%)	8
Empty chamber	2-Butoxyethanol	171	0.11	0.09		Two-sink model(RH=35%)	8
Carpet	Decane	174	0.63/0.14	0.14		Two-sink model(RH=70%)	8
Carpet	Decane	174	1.22/0.12	0.31		Two-sink model(RH=35%)	8
Carpet	2-Butoxyethanol	171	1.42/0.21	0.35		Two-sink model(RH=35%)	8
Carpet	2-Butoxyethanol	171	1.23/0.22	0.33		Two-sink model(RH=75%)	8

Legend:

B.P.	VOC boiling point;
Ads.Const.	Adsorption rate constants, the unit of which is m/h^{-1} for Eq.sorption model and is h^{-1} for the rest; Values separated by "/" indicate the two adsorption rate constants in the two sinks;
Des.Const.	Desorption rate constants; Values separated by "/" indicate the two desorption rate constants in the two sinks;
Eq.adsorption model	Equilibrium adsorption models.

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