

**Modeling the Reversible, Diffusive Sink Effect in Response
to Transient Contaminant Sources**

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Abstract – A physically based diffusion model is used to evaluate the sink effect of diffusion-controlled indoor materials and to predict the transient contaminant concentration in indoor air in response to several time-varying contaminant sources. For simplicity, it is assumed the predominant indoor material is a homogeneous slab, initially free of contaminant, and the air within the room is well mixed. The model enables transient volatile organic compound (VOC) concentrations to be predicted based on the material/air partition coefficient (K) and the material-phase diffusion coefficient (D) of the sink. Model predictions are made for three scenarios, each mimicking a realistic situation in a building. Styrene, phenol, and naphthalene are used as representative VOCs. A styrene butadiene rubber (SBR) backed carpet, vinyl flooring (VF), and a polyurethane foam (PUF) carpet cushion are considered as typical indoor sinks. In scenarios involving a sinusoidal VOC input and a double exponential decaying input, the model predicts the sink has a modest impact for SBR/styrene, but the effect increases for VF/phenol and PUF/naphthalene. In contrast, for an episodic chemical spill, SBR is predicted to reduce the peak styrene concentration considerably. A parametric study reveals for systems involving a large equilibrium constant (K), the kinetic constant (D) will govern the shape of the resulting gas-phase concentration profile. On the other hand, for systems with a relaxed mass transfer resistance, K will dominate the profile.

Key words – Indoor air, Diffusion, Emission, Sorption, Volatile organic compounds

PRACTICAL IMPLICATIONS

Many materials used to finish and furnish a building's interior potentially act as sinks for volatile organic compounds (VOCs) entering from outdoor and indoor sources (i.e., they act as sorbents to temporarily reduce indoor concentrations of these pollutants). When indoor concentrations decrease, sink materials release the pollutants back to air. The type of source and the chemical and physical characteristics of both pollutants and materials determine the resulting indoor

pollutant concentrations. Cumulative human exposures can be reduced, increased or unaffected depending upon occupancy patterns during these transient events. This modeling study provides mechanistic equations to predict the effect of reversible, diffusion-controlled, material sinks on indoor VOC concentrations using three realistic source scenarios and three common material/VOC combinations. Systems in which a VOC strongly partitions from air to a material or in which a VOC diffuses quickly in a material are shown to have the largest impacts on temporal concentration profiles.

NOTATION

A	area of flooring or carpet (m^2)
C	concentration of compound in material phase ($\mu\text{g m}^{-3}$)
D	diffusion coefficient for compound in material phase ($\text{m}^2 \text{s}^{-1}$)
D_c	characteristic diffusion coefficient ($\text{m}^2 \text{s}^{-1}$)
K	partition coefficient between material phase and gas phase (dimensionless)
L	thickness of flooring, carpet backing or cushion (m)
L_c	characteristic diffusion path length (m)
Q	volumetric air flow rate through chamber or room ($\text{m}^3 \text{s}^{-1}$)
r	mass emission rate from a source ($\mu\text{g m}^{-2} \text{s}^{-1}$)
t	time (s)
τ_c	characteristic diffusion time (s)
V	volume of air in chamber or room (m^3)
x	linear distance (m)
y	concentration of compound in gas phase ($\mu\text{g m}^{-3}$)

INTRODUCTION

A variety of building materials commonly found indoors can serve as sources of and sinks for indoor air pollutants. The resulting interactions between indoor materials and indoor pollutants may significantly alter the concentrations of contaminants in indoor air (Zhao et al., 1999a). Of the various mechanisms governing the source/sink behavior of indoor materials, material-phase

diffusion may be one of the most important (Clausen et al., 1993; Zhao et al., 1999b; Cox et al., 2001c).

A number of empirical or semi-empirical models have been proposed to describe diffusion-controlled emission processes (Matthews et al., 1987; Colombo et al., 1990; Clausen et al., 1993). However, estimates using such empirical models for conditions not covered by the experimental data are uncertain and provide little insight into the controlling mechanisms. Several physically based models have been proposed to describe the sorption/desorption of VOCs by various indoor sinks and sources. Some focused on surface effects (Silberstein et al., 1988; Dunn and Tichenor, 1988; Tichenor et al., 1991, Clausen et al., 1991; Chang and Guo, 1992), while others emphasized internal diffusion (Dunn, 1987; Dunn and Chen, 1992). The latter diffusion models assume an infinitely deep source, and cannot be applied to finite sources and sinks. Recently, a sink-diffusion model was introduced to characterize interactions between VOCs and material surfaces (Jørgensen et al., 2000). This model quantifies diffusion between a surface sink and an interior sink using an effective mass-transfer coefficient (combining the diffusion coefficient and the diffusion path length) and assuming a linear concentration gradient. As a result, it does not fully represent the physical characteristics of a diffusion-controlled sink and necessitates fitting of data to estimate model parameters (Jørgensen et al., 2000).

In a previous study, Little et al. (1994) developed a source model for predicting emissions from homogeneous, diffusion-controlled building materials. Recently, the model has been validated for emissions from vinyl flooring using values for the three parameters determined in a completely independent fashion – in other words, no fitting of parameters was required (Cox et al., 2000a,b; Cox et al., 2001a-c). For the source model, the key parameters are the initial concentration in the material phase (C_0), the material/air partition coefficient (K), and the material-phase diffusion coefficient (D). Little and Hodgson (1996) derived an extended version of the source model based on identical principles but focusing on the sink effect, and considering a variable influent VOC concentration. In this paper, the sink model is used to evaluate several cases where varying contaminant concentrations in the influent air to a room result in sorption and subsequent re-emission of compounds. For demonstration purposes, three contaminant source scenarios are considered, each of which simulates a realistic situation in a building. For

each scenario, a standard function of influent concentration versus time is derived or deduced based on existing knowledge or literature data. The predicted indoor concentration profile in response to each influent concentration function is discussed, and a parametric study is conducted with respect to model parameters, K and D.

DEVELOPMENT OF MODEL

The dynamic sink model is reviewed and the solution is repeated to correct a small typographical error in equation 10 of the original paper (Little and Hodgson, 1996). For modeling purposes, the system is represented by two interconnected chambers as shown in Figure 1. The first chamber represents the contaminant source. The concentration of the contaminant in this chamber is controlled by changes in source strength and is prescribed by a known function of time. The effluent from the first chamber enters the second chamber containing the homogeneous, diffusion-controlled indoor material. The concentration of the contaminant in the second chamber is controlled by the ventilation rate and the sorption/re-emission rate of the indoor material. Both chambers are assumed well mixed. This two-chamber scenario is mathematically equivalent to having both source and sink within the second chamber.

Transient diffusion through a homogeneous slab (the indoor material) is described by

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} \quad (1)$$

where $C(x, t)$ is the material-phase concentration, D is the diffusion coefficient, t is time, and x is distance from the base of the slab. The initial condition assumes the concentrations of contaminant in both the air and the slab are zero, or

$$y(t)|_{t=0} = C(x, t)|_{t=0} = 0 \quad \text{for } 0 \leq x \leq L \quad (2)$$

where y is the concentration of the contaminant in the chamber air. The first boundary condition assumes there is no flux through the base of the slab, or

$$\left. \frac{\partial C}{\partial x} \right|_{x=0} = 0 \quad (3)$$

The second boundary condition is imposed through a mass balance on the contaminant in the chamber air with four terms representing accumulation of the contaminant in the chamber air, contaminant entering the chamber, mass flux diffusing in or out of the slab, and contaminant leaving the chamber in the effluent air stream, or

$$\frac{\partial y}{\partial t} \cdot V = Q \cdot y_{in} - D \cdot A \left. \frac{\partial C}{\partial x} \right|_{x=L} - Q \cdot y \quad (4)$$

where $y_{in}(t)$ is the concentration function of the contaminant in the influent stream, V is the volume of air in the chamber, Q is the volumetric flow rate of air through the chamber, A is the area covered by the slab, and L is the thickness of the slab. Equilibrium is assumed between the contaminant concentration in the surface layer of the slab and the chamber air, or

$$K = \frac{C|_{x=L}}{y} \quad (5)$$

where K is the material/air partition coefficient. Combining equations (4) and (5) yields

$$\left(\frac{V}{A \cdot K} \right) \left. \frac{\partial C}{\partial t} \right|_{x=L} + D \left. \frac{\partial C}{\partial x} \right|_{x=L} + \left(\frac{Q}{A \cdot K} \right) C|_{x=L} - \left(\frac{Q}{A \cdot K} \right) K y_{in}(t) = 0 \quad (6)$$

The solution to equations (1)-(3) and (6) is (Little and Hodgson, 1996)

$$C(x, t) = \left(\frac{2 \cdot Q}{A \cdot K} \right) \cdot \sum_{n=1}^{\infty} \left\{ \frac{\exp(-Dq_n^2 t)(q_n^2) \cos(q_n x)}{\left[L(h - kq_n^2)^2 + q_n^2(L + k) + h \right] \cos(q_n L)} \cdot \int_0^t \exp(Dq_n^2 \tau) K y_{in}(\tau) d\tau \right\} \quad (7)$$

where

$$h = \frac{Q}{ADK} \quad (8)$$

and

$$k = \frac{V}{AK} \quad (9)$$

The q_n 's are the positive roots of

$$q_n \tan(q_n L) = h - kq_n^2 \quad (10)$$

and τ is a dummy variable.

Equation (7) gives the contaminant concentration in the slab as a function of time and distance from the base of the slab. The concentration of contaminant in the chamber at any time t can be obtained by first finding the concentration at the slab surface ($x = L$) and then applying the equilibrium condition defined by Equation (5).

Model parameters and additional assumptions

The area (A) of the air-sink interface is assumed to be 9 m^2 , and the volume (V) of the room or chamber is 20 m^3 . A typical ventilation rate of 1 air change per hour is used, with an air flow rate (Q) of $20 \text{ m}^3 \text{ h}^{-1}$, unless otherwise stated. Three material/VOC systems are examined. These are carpet with styrene-butadiene rubber backing and styrene (SBR/styrene) (Little, et al., 1994), vinyl flooring and phenol (VF/phenol) (Cox et al., 2001c), and polyurethane foam carpet cushion and naphthalene (PUF/naphthalene) (Zhao et al., 1999b). The thickness (L) of each material as well as the K and D values for the respective material/VOC systems are listed in Table 1. Several additional assumptions are implicit to the development of the preceding model. The parameters D and K are independent of concentration. Cox et al. (2001c) show this assumption holds for a wide range of VOCs in vinyl flooring. Schwoppe et al. (1989) found the approximation is good for material-phase concentrations below 1% by mass. The resistance to mass transfer in the boundary layer between the bulk chamber air and the material surface is assumed negligible. The characteristic diffusion time, τ_c , is given by

$$\tau_c = \frac{L_c^2}{2D_c} \quad (11)$$

where L_c is the diffusion path length and D_c is the diffusion coefficient of the VOC in the specific medium. For a 1-cm layer of stagnant air, and a gas-phase diffusion coefficient of $1 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$, a characteristic time of 5 s is obtained. Thus, the time taken to establish equilibrium at the material surface is fast relative to the time scale over which gas-phase chamber concentrations change. Finally, the air in the chamber must be well mixed within time scales that are short relative to the time for diffusion into the sink material. Characteristic diffusion times for each material/VOC combination are shown in Table 1, confirming this assumption.

Time-varying source functions

Indoor air contamination can be attributed to both outdoor and indoor sources. The nature of the source and the variability of ventilation conditions result in the dynamic behavior of VOCs indoors. Crump (1995) divided sources into continuous (long-term emission, constant source strength) and discontinuous categories (short-term emission, variable strength). From a modeling viewpoint, Sparks (1992) classified sources into long-term, steady state sources such as moth crystals; on/off sources such as HVAC systems; decaying or wet sources such as paints and waxes; and burst sources such as aerosol sprays.

The source concentration profile (the input function y_{in}) can be described using mathematical functions. For example, a first order exponential decay model was used to describe VOC emissions from a carpet, and a second order decay model was invoked to simulate TVOC emissions from a latex paint (Tichenor and Sparks, 1996). In this paper, three contaminant source scenarios are considered, each of which mimics a realistic situation in a building. Source functions are obtained by fitting appropriate source models to experimental data from the literature. Depending on the source scenarios, the concentration functions are defined either continuously or piecewise continuously.

RESULTS AND DISCUSSION

Scenario 1 – sinusoidal source

The first source scenario involves air entering the chamber at a constant ventilation rate, but with a sinusoidal concentration profile. Practical examples of this type of source include atmospheric contaminants from motor vehicle exhaust, and concentration patterns of toxic organic pollutants such as benzene in outdoor air in Los Angeles (Harley and Cass, 1994). In this case, the source function can be approximately described by a sinusoidal function, or

$$y_{in}(t) = 10 \cdot \sin\left(\frac{2\pi}{24}t\right) + 20 \quad (12)$$

where the parameters (y in ppb and t in hours) were inferred from the benzene concentration data reported for Los Angeles air (Harley and Cass, 1994). Substituting equation (12) into equation (7), the transient concentration profile of a contaminant in the slab, $C(x, t)$, is readily determined. Subsequently, the corresponding concentration profile in the chamber air, $y(t)$, is calculated from equation (5). Figure 2 shows y_{in} , the influent concentration, as well as the model predicted concentration profiles for SBR/styrene, VF/phenol, and PUF/naphthalene systems for a period of 48 hours. For the sinusoidal source scenario, the indoor material serves as both a sink and a source. During the rising stage of source concentration, the SBR, VF, or PUF functions as a sink (i.e., $y < y_{in}$), while on the down slope of the sinusoidal curve, the material acts as a source, (i.e., $y > y_{in}$). In all cases, significant concentration depression and a predominant sink effect is evident. Also, the period of the influent and effluent concentration functions remain identical although the effluent peaks are delayed to a small extent.

Scenario 2 – decaying source

The second scenario involves a constant ventilation rate in chamber 1, but with a slowly depleting source such as a carpet. Therefore, the gas-phase concentration entering chamber 2

decreases slowly with time. Such a decaying source may be approximated by a four-parameter double exponential function (Hodgson et al., 1993), or

$$y_{in}(t) = 790 \cdot e^{-0.15t} + 140 \cdot e^{-0.022t} \quad (13)$$

where y is in ppb and t is in hours. Again, the gas-phase contaminant concentration in response to equation (13) is predicted using equations (7) and (5). Figure 3 shows the input function and predicted concentrations as a function of time for the SBR/styrene, VF/phenol, and PUF/naphthalene systems. In accordance with the decaying input, the peak concentrations in the chamber air take place about four hours after the contaminant is introduced. Peak concentrations are suppressed, especially for the PUF/naphthalene system, which has a much higher D value. Again, the indoor material serves as a sink during the initial stage when the gas-phase concentration is higher than the solid phase concentration. Later, as the direction of the surface concentration gradient is reversed, the material acts as a contaminant source.

Scenario 3 – burst source

The third scenario considers a regular episodic introduction of a VOC in chamber 1. This situation approximates a chemical spill or the use of a consumer product such as an aerosol spray. The concentration in the first chamber has an instantaneous vertical rise followed by an exponential decrease, with decay determined by the ventilation rate.

Considering a frequency of two spills per day, one at 9 a.m. and the other at 1 p.m., and assuming the initial value for both spikes is 500 ppb, the following piecewise functions were constructed by mass balance to represent the influent concentration profiles:

$$y_{in}(t) = 0 \quad 0 \leq t < 9 \quad (14)$$

$$y_{in}(t) = \frac{500}{e^{-9 \cdot \frac{Q}{V}}} \cdot e^{-\frac{Q}{V}(t-24d)} \quad (9+24d) \leq t < (13+24d) \quad (15)$$

$$y_{in}(t) = \frac{500}{e^{-\frac{Q}{V}t}} \cdot \left(e^{-\frac{Q}{V}(t-4-24d)} + e^{-\frac{Q}{V}(t-24d)} \right) \quad (13+24d) \leq t < [9+24(d+1)] \quad (16)$$

where midnight is taken as the starting point, d is a date factor defined as $d = \text{integer}(t/24)$, i.e., on day 1 ($t < 24$ hour) $d=0$; day 2 ($24 < t < 48$ hour) $d=1$, and so on. Spills from the previous day are assumed not to affect the indoor air concentration on the subsequent day. However, the residual effect of the first spill on the second spill during the same day is considered, as shown in equation (16).

Using the SBR/styrene system as an example, Figure 4 provides the transient response. The presence of the SBR sink reduces the peak concentrations considerably even when the K and D values are relatively low. In contrast, the sink effect is much less significant for cases of more gradually varying sources as revealed in scenarios one and two.

Sensitivity of the model to K and D

As evidenced by the illustrative scenarios, the resulting indoor air contaminant concentration profile for different contaminant/sink systems can vary significantly. The model provides a convenient tool to examine the influence of K and D on indoor concentrations. For simplicity, a constant influent concentration (y_{in}) of 500 ppb is assumed. Accordingly, equation (7) simplifies (Little and Hodgson, 1996) to

$$C(x, t) = 2hKy_{in} \sum_{n=1}^{\infty} \left\{ \frac{[1 - \exp(-Dq_n^2 t)] \cos(q_n x)}{[L(h - kq_n^2)^2 + q_n^2(L + k) + h] \cos(q_n L)} \right\} \quad (17)$$

Equation (17) shows the concentration in the slab and, thus, in the chamber air, is linearly proportional to y_{in} . This confirms our intuition that source minimization is the most effective way to reduce the level of indoor air pollutants. On the other hand, impacts of K and D on $y(t)$ are less explicit in equation (17). A parametric study with respect to K and D reveals the relative

importance of each parameter and thus provides additional insight into the transient behavior of such material sinks.

Figures 5 through 8 display the calculated concentrations for various K and D values using equation (17). The material slab is assumed to be initially free of contaminant and an air exchange rate of 0.5 h^{-1} is assumed ($Q = 10 \text{ m}^3 \text{ h}^{-1}$). Although the predicted gas-phase concentrations will all ultimately reach the imposed influent concentration level of 500 ppb, the transient responses depend quite strongly on the relative values of K and D. Figure 5 shows $y(t)$ for D varying between 0.1×10^{-12} and $100 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$ with K fixed at 1,000 while Figure 6 shows $y(t)$ for D varying between 0.01×10^{-12} and $10 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$ with K fixed at 100,000. Figure 5 demonstrates for a relatively low K value of 1,000 (approximately the magnitude for many VOCs found indoors), a variation of D by three orders of magnitude hardly alters the shape of $y(t)$. However, if K equals 100,000, D becomes the dominant parameter governing the sink behavior, as shown in Figure 6. In a similar fashion, Figures 7 and 8 show $y(t)$ at fixed D values of 1×10^{-12} and $1 \times 10^{-14} \text{ m}^2 \text{ s}^{-1}$, respectively, while K varies from 100 to 100,000. Figure 7 shows for a relatively high D value ($\sim 10^{-12} \text{ m}^2 \text{ s}^{-1}$) the magnitude of K will dominate the influence of the sink. However, as shown in Figure 8, when the D value of a compound is so low ($\sim 10^{-14} \text{ m}^2 \text{ s}^{-1}$) as to become the limiting factor, a change of K will no longer impact the indoor air concentration profile to any significant extent.

In summary, the higher the values of D and K, the more significant the sink effect of an indoor material becomes. For systems involving a high material to air phase partition coefficient (K), the kinetic factor (D) will govern the shape of the resulting air concentration profile. The VF/phenol system is an example of this type. On the other hand, for systems with a relaxed material-phase mass transfer resistance, the equilibrium factor (K) will dominate the shape of $y(t)$. Systems involving permeable indoor materials such as PUF and many common VOCs found indoors may fall into this category.

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Tables

Table 1. Model parameters and characteristic diffusion time for three material/VOC systems.

Material	L (mm)	VOC	K	D (m ² s ⁻¹)	τ_c (day)
SBR	1	styrene	5,700	3.1×10^{-12}	2
VF	2	phenol	120,000	1.2×10^{-13}	200
PUF	10	naphthalene	6,400	2.3×10^{-10}	2

Figures

Figure 1. Schematic representation of model: Chamber 1 represents the time-dependent contaminant source, Chamber 2 symbolizes a room/chamber containing a sink material. Symbols are defined in the text or under notation.

Figure 2. Indoor air concentration profiles for SBR/styrene, VF/phenol, and PUF/naphthalene systems exposed to a sinusoidal source.

Figure 3. Indoor air concentration profiles for SBR/styrene, VF/phenol, and PUF/naphthalene systems exposed to a double exponential decaying source.

Figure 4. Indoor air concentration profile for SBR/styrene system exposed to an episodic source.

Figure 5. Plots of $y(t)$ for D varying between 0.1×10^{-12} and $100 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$ at a constant K of 1000 and y_{in} of 500 ppb.

Figure 6. Plots of $y(t)$ for D varying between 0.01×10^{-12} and $10 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$ at a constant K of 100,000 and y_{in} of 500 ppb.

Figure 7. Plots of $y(t)$ for K varying between 100 and 100,000 at a constant D of $1 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$ and y_{in} of 500 ppb.

Figure 8. Plots of $y(t)$ for K varying between 100 and 100,000 at a constant D of $1 \times 10^{-14} \text{ m}^2 \text{ s}^{-1}$ and y_{in} of 500 ppb.















