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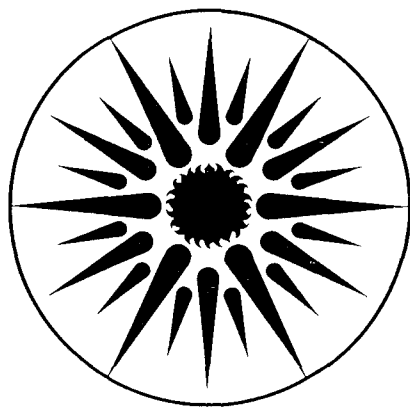
ENERGY & ENVIRONMENT DIVISION

Submitted to Atmospheric Environment, Part A

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February 1993



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**MODELING EMISSIONS
OF VOLATILE ORGANIC COMPOUNDS
FROM NEW CARPETS**

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February 1993

This work was supported by the Director, Office of Energy Research, Office of Health and Environmental Research, Human Health and Assessments Division of the U. S. Department of Energy under contract DE-AC03-76SF00098 through Lawrence Berkeley Laboratory.

Abstract

A simple model is proposed to account for observed emissions of volatile organic compounds (VOCs) from new carpets. The model assumes that the VOCs originate predominantly in a uniform slab of polymer backing material. Parameters for the model (the initial concentration of a VOC in the polymer, a diffusion coefficient and an equilibrium polymer/air partition coefficient) are obtained from experimental data produced by a previous chamber study. The diffusion coefficients generally decrease as the molecular weight of the VOCs increase, while the polymer/air partition coefficients generally increase as the vapor pressure of the compounds decrease. In addition, for two of the study carpets that have a styrene-butadiene rubber (SBR) backing, the diffusion and partition coefficients are similar to independently reported values for SBR. The results suggest that predictions of VOCs emissions from new carpets may be possible based solely on a knowledge of the physical properties of the relevant compounds and the carpet backing material. However, a more rigorous validation of the model is desirable.

Key Words: Diffusion model, diffusion coefficient, polymer backing, styrene-butadiene rubber, environmental chamber.

Introduction

In a previous study, Hodgson et al. (1992, In press) measured the emissions of selected volatile organic compounds (VOCs) released by new carpets that are typical of those used in residences, schools and offices. Carpet samples were each placed in a 20-m³, continually-mixed, controlled, environmental chamber at a fixed air exchange rate. The emission rates of VOCs from a carpet sample were determined by monitoring, for a week, the changes in the VOC concentrations in the chamber.

An examination of the experimental data revealed several interesting features. The concentrations of most VOCs in the chamber air quickly reached a peak, and then decayed rapidly over the first 12 hours. The initial decay rates of all compounds could be well

approximated with exponential curves. After this period, the decay rates slowed, with emissions of most compounds continuing for the entire week. The dominant VOCs emitted by the carpets were, for the most part, constituents of the polymer backing materials. An experiment with one carpet, in which the fibers and backing were separated and tested individually for emissions, confirmed that the backing was by far the dominant source. Finally, a rough check of the characteristic time for a VOC to diffuse through a thin polymer layer appeared to confirm that the polymer backings were acting as slow diffusive sources of the VOCs released into the chamber air.

Others have used empirical models to describe emissions processes. For example, Colombo et al. (1990) fit double exponential transient mass-balance equations to the concentration versus time curves for the emissions of VOCs from wood products and gypsum board. Four parameters were estimated for each case by non-linear least squares regression of a data set, and reasonably accurate fits were obtained. However, this approach lacks a physical basis and provides no insight into the mechanisms controlling desorption. As a consequence, estimates for regions beyond the data range are uncertain.

Several physically-based models have been developed to describe the sorption/desorption of VOCs by various indoor sinks/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), including an example where the boundary layer resistance between the bulk air and the source/sink was taken into account (Axley, 1991), while others considered internal diffusion (Dunn, 1987; Clausen et al., 1992; Dunn and Chen, 1992). Dunn (1987) emphasized the value of such models in de-coupling the source/sink behavior from the experimental apparatus, usually an environmental chamber, so that the results can be applied more widely.

The applicability of the existing physically-based models is briefly considered in the light of the experimental observations. Models that focus on surface effects appear inappropriate because many of the dominant compounds originate in the polymeric

materials and seem to be subject to diffusion controlled release. The diffusion models of Dunn (Dunn, 1987; Dunn and Chen, 1992) assume an infinitely deep source, and can not be used for finite sources that are significantly depleted. Clausen et al. (1992) considered a finite source, but invoked concentration dependent diffusion, which may not be necessary at the relatively low concentrations found in the backing in this study. They also neglected equilibrium partitioning between the bulk air and the surface of the source as the VOCs in their study had relatively high vapor pressures. As discussed later, the slow VOC emission rates and the well-mixed conditions in the chamber allowed concerns about boundary layer resistance between the carpet surface and the bulk chamber air to be ignored in this study.

In this paper, a simple physically-based diffusion model, which assumes that all of the carpet emissions come from a thin layer of polymer backing material, is proposed. The basic model parameters (the initial concentration of VOC in the polymer, a diffusion coefficient and an equilibrium partition coefficient) are obtained from the experimental data and, where possible, are compared to expected values. In addition, the relationship between the model parameters and the physical properties of the VOCs is examined, and the influence of the parameters on the emissions profiles is briefly discussed.

Experimental data

The initial study (Hodgson et al, 1992a, 1992b) measured the emissions of selected VOCs from samples of four new carpets. The original sample numbers have been maintained to facilitate comparisons with the initial study. Carpet 1 is typical of residential carpets with Nylon fibers and a secondary backing consisting of a coarse polypropylene mesh bonded to the primary backing with styrene-butadiene rubber (SBR) latex adhesive. Carpet 2 has nylon fibers and a flexible polyurethane foam (PUF) secondary backing. This carpet was excluded from the re-analysis because it was assumed that a simple diffusion model would not accurately describe the emissions of VOCs from the relatively porous PUF. Carpet 3, which is manufactured in the form of tiles, had Nylon fibers and a hard polyvinyl chloride (PVC) secondary backing. Carpet 4 has mixed polypropylene and

Nylon fibers with the same backing as Carpet 1. All carpet samples were collected directly from the finish lines at the manufacturers' mills and were packaged in two layers of heat-sealed Tedlar bags to preserve their chemical integrity. Two samples of Carpet 1 (1a and 1b) were collected simultaneously. The emissions measurements were made two to five weeks after sample collection.

Each carpet sample was initially screened for emissions of VOCs using two procedures. First, aliquots of headspace gas was withdrawn directly from the sample storage bags and analyzed. This analysis was conducted after the bags had been stored unopened in the laboratory for about one week. Second, samples were screened for emissions using small-volume (4-L) chambers. Compounds for quantitative analysis during the environmental chamber experiments were selected based on the results of the screening measurements. Five to seven of the most abundant VOCs were selected for each experiment. Those VOCs chosen for re-analysis are listed in Table 1 along with abbreviations and certain physical properties.

A 20-m³ environmental chamber, constructed of low-emitting materials and lined with stainless steel, was used for the emissions experiments. The ranges in the operating parameters of the chamber for all experiments were: 0.98-1.00 h⁻¹ for ventilation rate; 22.8-23.5°C for temperature; 46.5 to 50.2 percent for relative humidity; and 6.5-9 cm s⁻¹ for air velocity near the floor. The ventilation rate and temperature had relative standard deviations of one percent for the week-long experiments. On the day that an experiment began, the chamber was entered, and the carpet sample was installed to cover the floor of the chamber. The loading ratio was 0.44 m² m⁻³. The installation took about 15 minutes. The initial time for the experiment was established by the closing of the chamber door. Air samples for VOCs and low-molecular weight aldehydes were periodically collected over the next seven days. During the first day, the samples were collected starting at elapsed times of 1, 3, 6, 12, and 24 h. Subsequent samples were collected at 24 h intervals.

Development of model

Theory. The model assumes that all of the contaminants emitted from the carpet diffuse out of a single uniform layer of polymer backing material. This ignores contributions from the carpet fibers and other potential sources within the carpet. Furthermore, the boundary layer resistance between the carpet surface and the well-mixed bulk air in the chamber is assumed to be small in comparison to the resistance to diffusion within the polymer layer. This assumption is based on calculations using expected diffusivities of the selected VOCs in air and in the backing material. A schematic representation of the idealized carpet in the chamber is shown in Figure 1.

The governing equation describing transient diffusion through a polymer is

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

where $C(x,t)$ is the concentration of the contaminant in the polymer slab; t is time; and x is the linear distance. The diffusion coefficient, D , determines the rate of diffusion of a VOC through the layer and is assumed to be independent of concentration. The initial condition assumes that the compound of interest is uniformly distributed throughout the polymer layer, or

$$C = C_0 \text{ for } 0 \leq x \leq L, \quad (2)$$

where L is the thickness of the polymer layer, and C_0 is the initial concentration. Since the carpet is resting on the stainless steel floor of the chamber, the first boundary condition assumes that there is no flux out of the base of the polymer slab, or

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

A second flux boundary condition is imposed through a mass balance on the VOC in the chamber air. The three terms represent the accumulation of the VOC in the chamber air, the mass flux diffusing out of the polymer slab, and the VOC leaving the chamber in the outflowing air stream, or

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

where y is the concentration of VOC in the chamber air, Q is the volumetric flowrate of clean air through the chamber, V is the volume of air in the chamber, and A is the area covered by the carpet. Equilibrium is assumed to exist between the contaminant concentrations in the surface layer of the polymer and the chamber air, or

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

where K_v is a linear partition coefficient. Combining equations 4 and 5 yields the appropriate boundary condition

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

The solution to equations 1-3 and 6 is obtained from an analogous heat transfer solution (Carslaw and Jaeger, 1959), which is first transformed into mass transfer terms and then adjusted to account for equilibrium partitioning, yielding

$$C(x, t) = 2C_o \sum_{n=1}^{\infty} \left\{ \frac{\exp(-Dq_n^2 t)(h - kq_n^2) \cos(q_n x)}{[L(h - kq_n^2)^2 + q_n^2(L + k) + h] \cos(q_n L)} \right\}, \quad (7)$$

where

$$h = \frac{(Q/A)}{(D \cdot K_v)}, \quad (8)$$

$$k = \frac{(V/A)}{K_v}, \quad (9)$$

and q_n are the positive roots of

$$q \tan(qL) = h - kq^2. \quad (10)$$

Equation 7 gives the contaminant concentration in the polymer slab as a function of distance from the base of the slab, and also of time. The concentration of contaminant in the chamber air at any time, t , is obtained by first finding the concentration at the surface of the polymer slab and then substituting this value into equation 5, the equilibrium relationship. Care must be exercised when evaluating equation 7 as the number of terms required for an accurate solution depends strongly on the selected parameters. The series converges

especially slowly for early times and at low values of D . However, at no time in this study were more than 200 terms necessary.

Estimation of parameters. Equations 7 and 5 give the concentration of a VOC in the chamber air as a function of time and various other parameters, most of which may be obtained from experimental measurement. Thus, Q , V , and A are all known, while L was obtained by direct measurement of the polymer slab thickness. This leaves the initial polymer concentration, C_0 , the partition coefficient, K_v , and the diffusion coefficient, D . These last three parameters were obtained in an iterative fashion (described below) using the chamber concentration data, $y(t)$, the mass per unit area emitted from the carpet over the duration of the week-long experiment, M_{emit} , and the concentration of the VOC present in the air of the storage bags prior to opening, y_{bag} (available for carpets 1 and 4 only).

An initial estimate of the total mass per unit area, M , of the VOC in the polymer slab at the start of an experiment was obtained by setting $M = M_{emit}$, recognizing that this would be a poor estimate if some of the VOC remained in the slab at the end of the experiment. C_0 was estimated by dividing M by the thickness of the polymer slab, L . For carpets 1 and 4, an initial estimate of K_v was obtained by dividing C_0 by y_{bag} , as the carpets had all been stored in the bags for a period of at least two weeks prior to opening and it was assumed that the polymer and air concentrations were at equilibrium. Next, equations 7 and 5 were used to find D , and in the case of carpet 3, also K_v , using an interval weighted (Dunn and Chen, 1992) relative least-squares (Saez and Rittmann, 1992) iterative fitting procedure, which assumed a constant variance in experimental error. Equation 7 was then used to calculate the concentration distribution within the polymer slab at the end of the experiment, and hence to find the fraction of the VOC, f , emitted from the slab. This value of f was used to provide an improved estimate of the initial mass in the slab, or $M = M_{emit}/f$. The entire procedure was repeated until estimates of C_0 , D and K_v converged (i.e., successive iterations produced insignificant changes in the estimates).

Discussion of results

The experimental data and the best-fit model curves for carpets 1, 3 and 4 are shown in Figures 2-5, while the fitted parameter values and the relative least-squares residuals (Σ^2) for the compounds are given in Tables 2-4. For carpets 1 and 4, C_0 and K_v were based on experimental measurements, and the iterative parameter estimation procedure had only one degree of freedom in parameter D, which was obtained by fitting the model to the transient chamber concentration data. For carpet 3, however, no data were available for the air concentrations in the storage bag. Therefore K_v could not be calculated independently, and the fitting procedure had two degrees of freedom in parameters D and K_v .

The proposed model appears to provide a reasonable fit to most of the experimental data over the one-week time period. Although the fit to the PCH data of carpets 1a and 1b does not appear to be as good as for the other compounds, an examination of the residuals in Table 2 shows that the relative degree of fit is similar. Generally, Σ^2 for the carpet 3 compounds is about an order of magnitude lower than for carpets 1 and 4; reflecting the additional degree of freedom in the fitting procedure.

The parameters for STY, C2B and PCH from the experiments with carpets 1a and 1b show reasonable reproducibility with the exception perhaps of D for C2B, which varies by about a factor of two between the duplicate experiments. The values for D and K_v for carpets 1 and 4 are also quite similar despite the fact that for STY, C_0 varies by about an order of magnitude between the two carpets. These results appear to further support the hypothesis that the backing materials govern the emissions, because the backings for carpets 1 and 4 are similar, while the fibers are different.

Diffusion coefficients for VOCs in polymer materials depend on the molecular weight of the compound and also on the type of polymer (Schwope et al., 1989). Those authors correlated diffusion coefficients of a wide range of VOCs in six polymers as a function of molecular weight of diffusant at 25°C, revealing that D can vary by orders of

magnitude as polymer type or molecular weight change. For carpets 1 and 4, the range of observed values of D is 10×10^{-12} to $0.5 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$ for VOCs with molecular weights varying between 104 and 158. As expected, the largest molecule, PCH, has the smallest diffusion coefficient. Park et al. (1989) measured diffusion coefficients for a range of VOCs in SBR and found values of D varying between 2.3×10^{-12} and $1.0 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$ for molecular weights varying from 72 to 166. These diffusion coefficients are in reasonable agreement with those obtained from the carpet data, lending credibility to the assumed physical basis of the model.

Carpet 3 has a flexible plasticized polyvinyl chloride backing with polyvinyl acetate as a co-polymer. No independently measured diffusion coefficients are available for this material. However, the observed D values of between 6×10^{-12} and $0.06 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$ are close to those that would be expected for high density polyethylene (Schwope et al., 1989). In addition, the two low molecular weight compounds have the largest diffusion coefficients, although it is not certain that these two compounds originate in the backing. The low diffusion coefficients found for ISO, PRO and ETH in Carpet 3 result in a large fraction (about 85%) of the amount initially present remaining within the polymer backing at the end of the one week period. Emissions of these compounds could be expected to continue for several weeks.

Schwope et al. (1989) suggested that the concentration dependence of diffusion coefficients of a VOC in polymers may be neglected when the concentrations of the VOC in the polymer is less than 1%. An examination of the observed values of C_0 for all three carpets show concentrations of well below 0.1%, assuming a polymer density of 1 g cm^{-3} . Therefore, the assumption made in deriving equation 7 that D is independent of C seems justified. A similar argument applies to the use of the partition coefficient, K_v , which may also depend on concentration. At the relatively low contaminant concentrations present in the carpet backing material, the use of a linear partition coefficient appears reasonable.

Ideally, the observed values for K_v should also be compared to expected values. Park et al. (1989) measured K_v for various VOCs in SBR at varying polymer concentrations. For the lowest concentration case, the values of K_v ranged from about 200 for n-hexane to 28,000 for 1,2-dichlorobenzene. The range of K_v values found in carpets 1 and 4, which should be similar to SBR, varies from 1,400 to 170,000, which appears plausible. In principle, K_v should increase as the vapor pressure of the compound decreases, and as the solubility of the compound in the polymer increases. For carpets 1 and 4, PCH has the lowest vapor pressure, and the observed values of K_v are indeed the largest. This trend is also observed for values for K_v for carpet 3, which follow an increasing trend as the vapor pressure decreases. The value of $K_v = 1$ for ACE appears somewhat inconsistent, although this might be due to the relative insensitivity of the model to K_v at low values of K_v , as is shown below.

While the proposed model appears to be consistent with the observed emissions profiles for most of the VOCs released from the carpets, the iterative fitting procedure did not converge for the VA data of carpet 3 (data not plotted). The model under-predicts the early data points and over-predicts the later portion of the observed chamber data. No clear reasons can be found for this unexpected behavior.

Behavior of model

The influence of the model parameters on the resulting contaminant concentration in air, $y(t)$, is briefly examined. Equation 7 shows that the concentration in the chamber air depends linearly on C_0 , the initial concentration in the polymer, and that the shape of $y(t)$ will scale proportionally. This suggests that the most effective way to reduce emissions from new carpets is to reduce C_0 , the initial concentration of the VOCs in the polymer backing material.

Figures 6 and 7 show the effect of a variation in D and K_v , respectively, for a constant C_0 of 10^4 mg m^{-3} . Figure 6 shows plots of $y(t)$ for values of D varying between 0.1×10^{-12} and $10 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$ at a constant K_v of 1000. Figure 7 gives $y(t)$ for K_v

varying between 100 and 100,000 at a constant D of $1 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$. Increasing the diffusion coefficient results in a higher emission rate at early times and more rapid depletion of the contaminant in the polymer slab. Increasing the partition coefficient decreases the emission rate at early times and results in a much slower depletion rate of polymer contaminant. However, the influence of a change in K_v is virtually insignificant below a value of about 1000.

Conclusions

The simple physically-based diffusion model provides a reasonable fit to the experimental chamber data for most compounds. The model assumes that emissions of VOCs from new carpets originate predominantly in a uniform slab of polymer backing material. The diffusion and partition coefficients obtained by fitting the model to the observed concentration data behave in fashions that are consistent with the physical properties of the compounds. In addition, for the two carpets that have an SBR backing, the fitted parameters are similar to those determined independently in SBR. The power of the proposed model lies in its ability to predict source strengths of VOCs emitted by carpets into indoor air using only a few physically meaningful parameters. This can provide a useful guide for establishing carpet specifications, and for improving carpet design and manufacturing procedures to reduce emissions. However, further work is necessary to more rigorously validate the model by independently measuring the model parameters and testing the simplifying assumptions.

Notation

A	area of chamber floor and of carpet (m^2)
BPt	boiling point ($^{\circ}\text{C}$)
C	concentration of compound in polymer (mg m^{-3})
C_0	initial concentration of compound in polymer (mg m^{-3})
D	diffusion coefficient for compound in polymer ($\text{m}^2 \text{s}^{-1}$)
f	fraction of VOC emitted from polymer layer (dimensionless)
K_v	partition coefficient between polymer and air (dimensionless)
L	thickness of carpet polymer layer (m)
M	total mass per unit area of VOC initially in polymer layer (mg m^{-2})
M_{emit}	mass per unit area of VOC emitted from polymer layer (mg m^{-2})
MWt	molecular weight (amu)
P_{vap}	vapor pressure (mm Hg)
Q	volumetric air flow rate through chamber ($\text{m}^3 \text{s}^{-1}$)
t	time (s)
V	volume of air in chamber (m^3)
x	linear distance (m)
y	concentration of compound in chamber air (mg m^{-3})
y_{bag}	concentration of compound in storage bag (mg m^{-3})
Σ^2	relative least-squares residual (dimensionless)

Acknowledgments

We thank Sajid Hakim for his participation in the early phases of this analysis and Bill Fisk, Tsair-Fuh Lin, and Peter Persoff for their review comments. This work was supported by the Director, Office of Energy Research, Office of Health and Environmental Research, Human Health and Assessments Division, the U.S. Department of Energy under contract No. DE-AC03-76SF00098.

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Table 1. Physical properties of compounds evaluated (vapor pressure at 23°C).

Name (abbreviation)	Formula	MWt	BPt (°C)	P _{vap} (mm Hg)
Styrene (STY)	C ₈ H ₈	104	145	5
Ethylbenzene and xylenes (C2B) [†]	C ₈ H ₁₀	106	136	6
4-Ethenylcyclohexene (VCH)	C ₈ H ₁₄	110	128	
4-Phenylcyclohexene (PCH)	C ₁₂ H ₁₄	158		
Formaldehyde (FOR)	CH ₂ O	30		760
Acetaldehyde (ACE)	C ₂ H ₄ O	44		740
1,2-Propanediol (PRO)	C ₃ H ₈ O ₂	76	187	0.2
Vinyl acetate (VA)	C ₄ H ₆ O ₂	86	72	83
2,2,4-Trimethylpentane (ISO)	C ₈ H ₁₈	114	98	39
2-Ethyl-1-hexanol (ETH)	C ₇ H ₁₆ O	130	184	0.05

[†]Average values for ethylbenzene and ortho-, meta-, and para-xylenes.

Table 2. Parameters for carpets 1a and 1b, L = 1.25 mm.

Parameter	Compound (carpet)						
	STY (a)	STY (b)	VCH [†] (b)	C2B (a)	C2B (b)	PCH (a)	PCH (b)
M _{emit} (mg m ⁻²)	2.20	3.41	0.80	0.40	0.64	12.5	9.80
f	0.98	0.97	0.95	0.95	0.92	0.49	0.47
C _o (mg m ⁻³)	1,800	2,800	670	340	560	20,000	17,000
K _v	4,200	6,500	1,400	1,500	2,400	81,000	67,000
D (m ² s ⁻¹) × 10 ¹²	4.1	3.6	5.2	10.2	4.3	0.59	0.50
Σ ² × 10 ³	9.1	4.6	8.3	10.6	1.9	10.3	7.2

[†]Not measured for carpet 1a.

Table 3. Parameters for carpet 3, L = 2.0 mm.

Parameter	Compound				
	FOR	ACE	ISO	PRO.	ETH
M_{emit} (mg m ⁻²)	6.61	2.52	7.55	72.0	7.20
f	0.74	0.96	0.14	0.14	0.13
C_o (mg m ⁻³)	4,500	1,300	25,900	255,000	26,500
K_v	11,000	1	59,000	180,000	450,000
D (m ² s ⁻¹) × 10 ¹²	3.2	6.4	0.060	0.065	0.088
$\Sigma^2 \times 10^3$	0.40	2.3	0.92	1.1	0.31

Table 4. Parameters for carpet 4, L = 1.0 mm.

Parameter	Compound			
	STY	VCH	C2B	PCH
M_{emit} (mg m ⁻²)	25.9	2.62	1.19	11.2
f	0.99	0.96	0.91	0.67
C_o (mg m ⁻³)	26,000	2,700	1,300	16,700
K_v	5,700	1,700	5,300	170,000
D (m ² s ⁻¹) × 10 ¹²	3.1	2.1	1.5	1.2
$\Sigma^2 \times 10^3$	14.1	5.3	7.9	3.3

Figure captions

Figure 1. Idealized schematic representation of a carpet in a chamber. Symbols are defined in text and glossary.

Figure 2. Experimental chamber concentrations with time and best-fit model curves for carpet 1a. The fitting procedure had one degree of freedom in parameter D. See Table 1 for chemical abbreviations.

Figure 3. Experimental chamber concentrations with time and best-fit model curves for carpet 1b. The fitting procedure had one degree of freedom in parameter D. See Table 1 for chemical abbreviations.

Figure 4. Experimental chamber concentrations with time and best-fit model curves for carpet 3. The fitting procedure had two degrees of freedom in parameters D and K_v . See Table 1 for chemical abbreviations.

Figure 5. Experimental chamber concentrations with time and best-fit model curves for carpet 4. The fitting procedure had one degree of freedom in parameter D. See Table 1 for chemical abbreviations.

Figure 6. Plots of $y(t)$ for D varying between 0.1×10^{-12} and $10 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$ at a C_0 of $10,000 \text{ mg m}^{-3}$ and a constant K_v of 1000.

Figure 7. Plots of $y(t)$ for K_v varying between 100 and 100,000 at a C_0 of $10,000 \text{ mg m}^{-3}$ and a constant D of $1 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$.

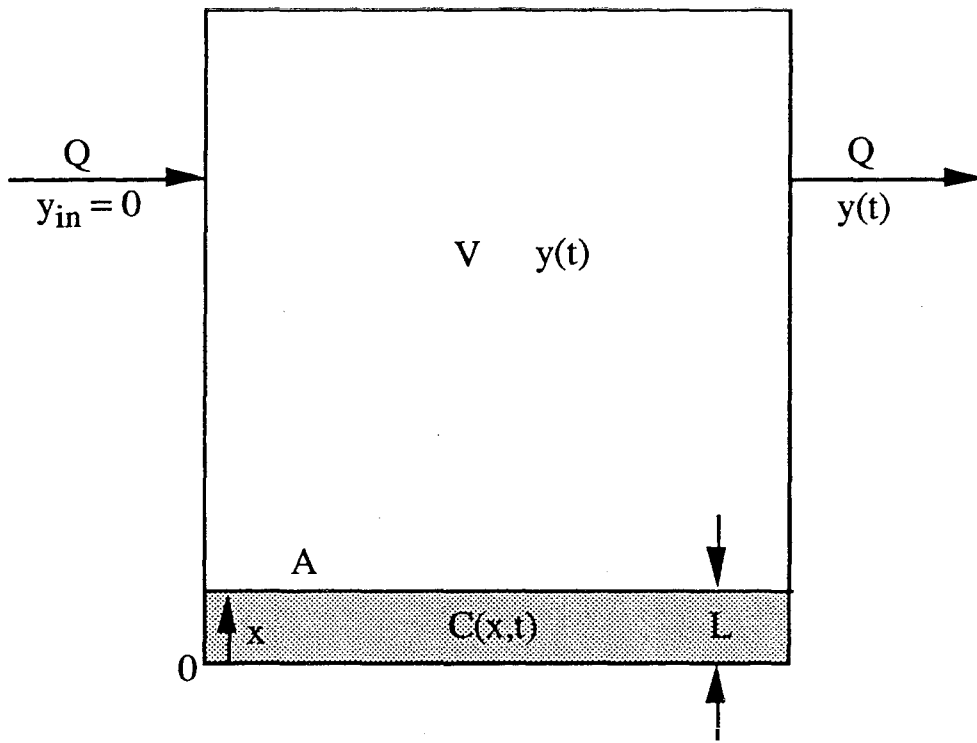
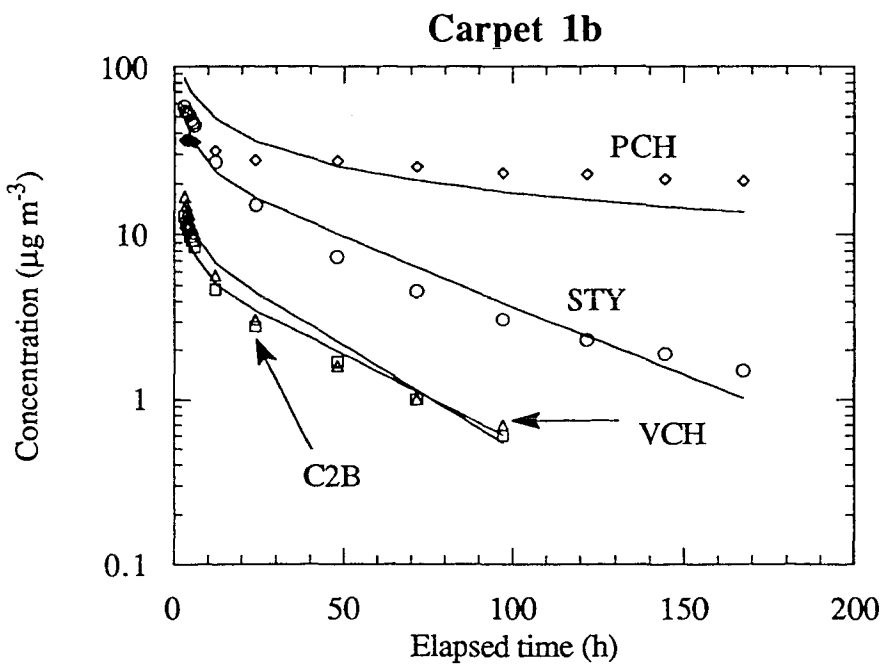
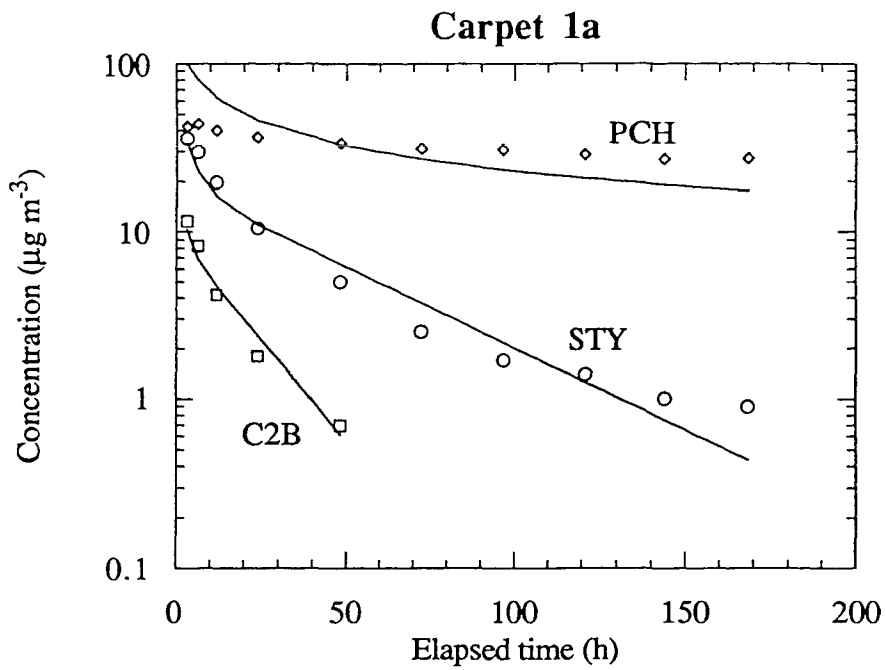
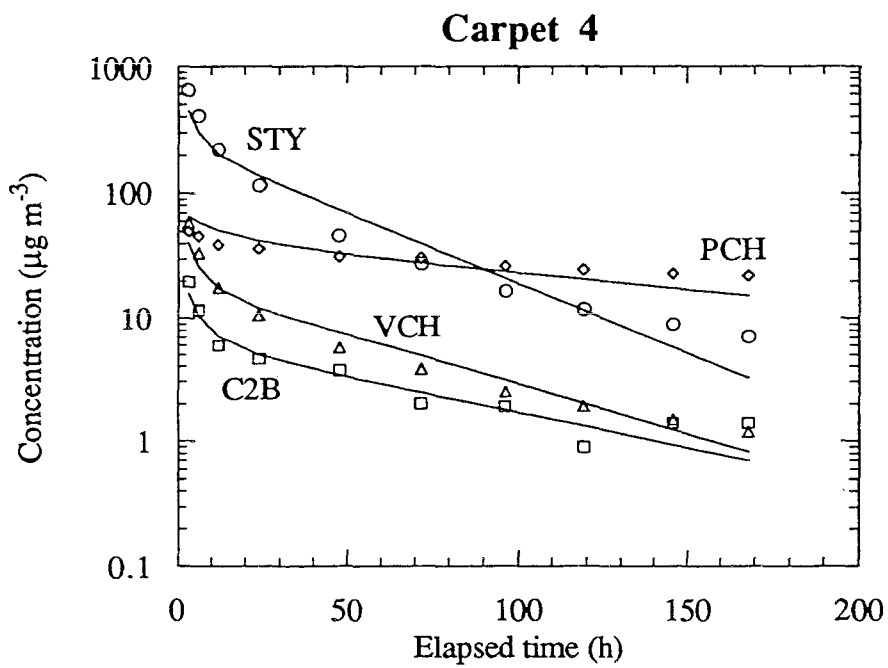
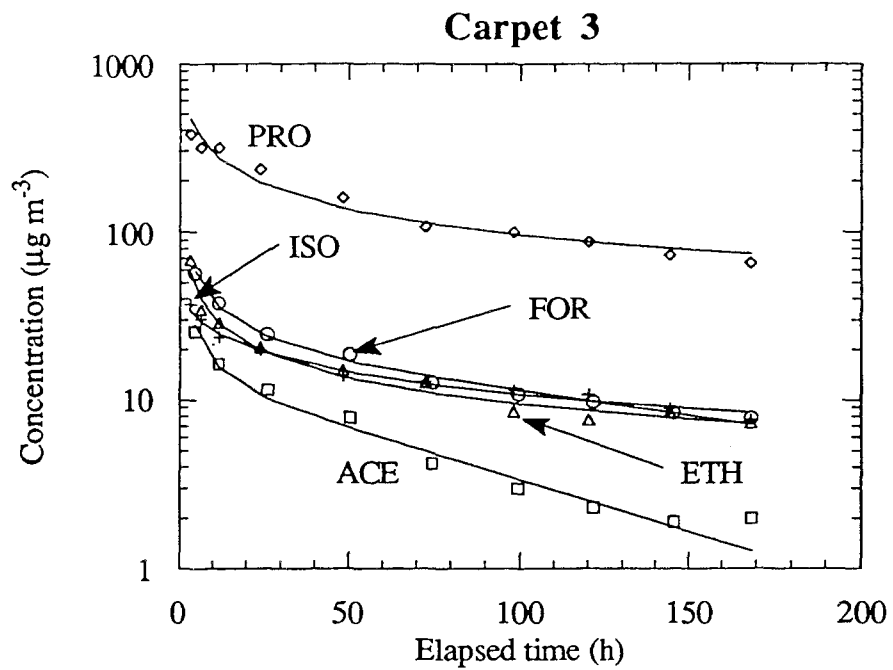


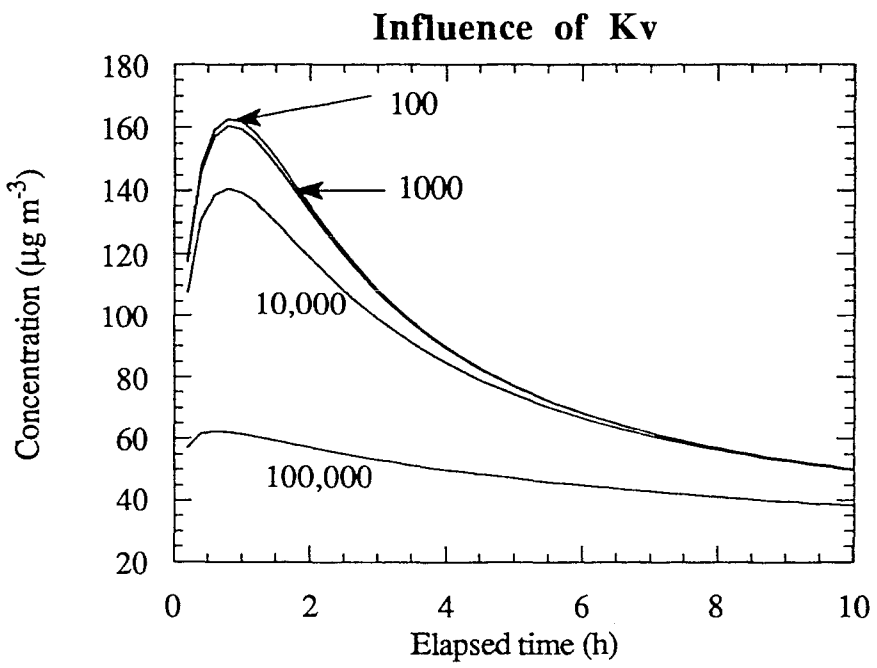
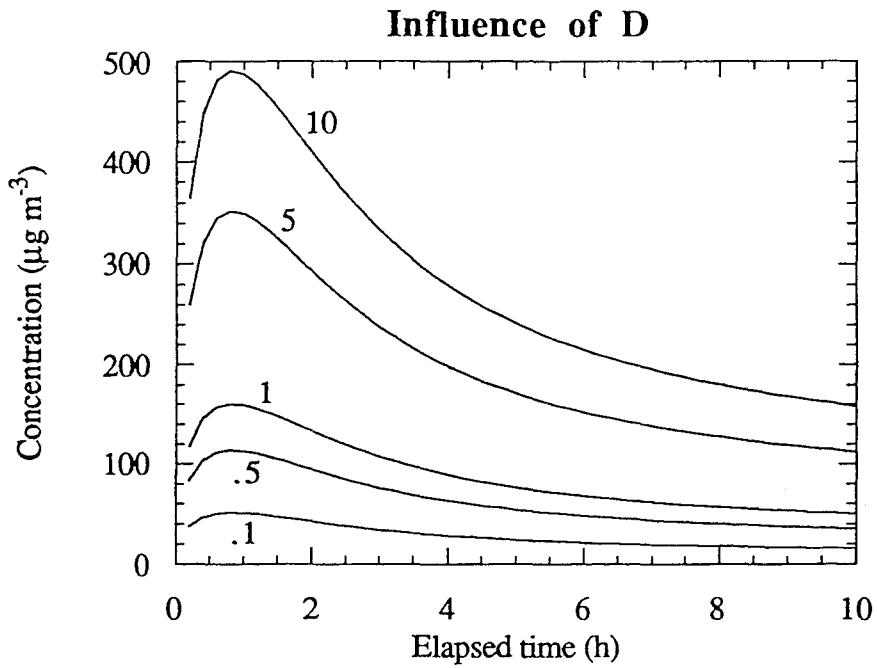
Figure 1.



Figures 2 and 3.



Figures 4 and 5.



Figures 6 and 7.