

# Development of a Model for the Estimation of Indoor Volatile Organic Compounds Concentration Based on Experimental Sorption Parameters

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In order to model indoor air quality quantitatively, the adsorption and the desorption rate constants of toluene for four typical furnishing materials (namely, two polyacrylonitrile carpets, a cotton sofa and a polyester curtain) were obtained by analyzing the toluene evaporation experiments in a test chamber. The  $N_2$ -BET areas were also measured for these materials and they were found to be in the range of 0.5–1.5 m<sup>2</sup>/g. It was found that the constants, at 25 °C, ranged between 0.0009 and 0.0013 m·h<sup>-1</sup> for adsorption and between 0.03 and 0.06 h<sup>-1</sup> for desorption, depending on the type of the material. These constants were then incorporated in a model for predicting the toluene concentration as function of time and the theoretical results were compared with the measured data of a test house. The unsteady state mass-balance model which took into account adsorption and desorption rates and assumed negligible external and internal mass-transfer resistances were found to be satisfactory. Mixing factors were used in expressing deviations from complete mixing in the test chamber, and values around 0.7 were obtained experimentally by the SF<sub>6</sub> tracer gas technique. The same technique was also used to measure the hourly air exchange rates in the test house.

## Introduction

Any evaluation technique for indoor air quality (IAQ) requires a satisfactory understanding of three main factors: the sources of the indoor pollutants and their emission characteristics, the air exchange between the indoors and the outdoors, and finally the possible interaction of these pollutants with surfaces within the building. One important class of indoor pollutants consists of volatile organic compounds (VOCs) which have been identified, characterized, and categorized in several studies (1, 2). The objective of these studies was mainly to evaluate the potential sources of these compounds by using different strategies, which involved the combined use of sophisticated questionnaires and statistical techniques (3, 4). The indoor sources could be classified as random short-term on/off sources (e.g., cigarettes), long-term on/off sources (e.g., heaters), long-term steady-state sources (e.g., moth crystals), and sources with high initial emission rates that decay with time (e.g., floor wax or painted surfaces) (5). Sources of indoor airborne VOCs can also be categorized into those that are located outdoors,

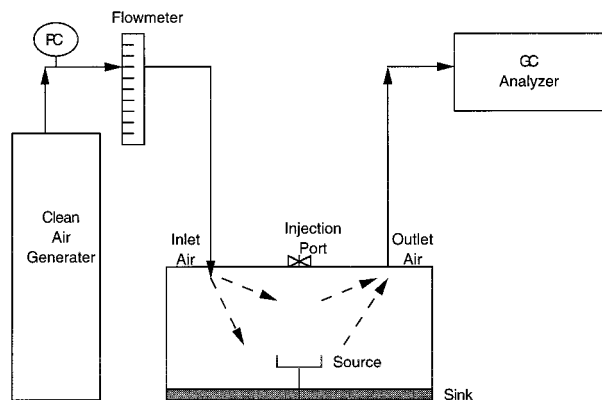


FIGURE 1. Schematic representation of the test chamber experimental apparatus.

such as infiltrated air and contaminated soil gas, and those within the building or structure envelope such as combustion sources, human activity sources, and surface emissions (6).

The second factor, which has a significant effect on indoor air pollutant concentrations, is the air exchange rate (7). In ventilation engineering a mixing factor (or ventilation effectiveness) of  $m$  is often used to express the deviation from perfect or complete mixing (8). For conventional ventilation, where supply diffusers are often located in ceilings adjacent to return or exhaust grills, bypass of air will be significant, leading to ventilation effectiveness which might therefore be significantly less than 1.0 (8). Here,  $(1 - m)$  represents the percent of air which bypasses the room without mixing.

The third factor that affects indoor air modeling is the interaction of the pollutant with surfaces. Adsorptive surfaces that remove pollutants from the indoor air act as sinks. Although the importance of sinks in determining indoor air quality has generally been recognized, there is only limited published work on the behavior of sinks or sink models (9–12). It is known that certain indoor surfaces can adsorb VOCs acting as sinks. The same surfaces can desorb the gas when the gas-phase concentration becomes too low to function as sources.

Previous studies by the authors (13, 14) emphasized the importance of surface sorption/desorption rates in modeling and assessment of indoor air quality. The aim of this work was therefore to obtain sorption/desorption characteristics of four typical furnishing materials (such as carpets) by controlled experimentation in a test chamber, which had mixing factors similar to that of a test house. These parameters were then incorporated in a model in order to predict the behavior of a test house VOC concentrations. In this study toluene in air was used as a typical VOC pollutant and all mixing factors were determined experimentally.

## Measurements and Analysis

**Test Chamber.** Figure 1 shows a schematic diagram of the test chamber. The chamber dimensions were proportional to those of the test house. It had dimensions of 50.3, 45.0, and 14.8 cm and was constructed from 6 mm thick glass sheets. The chamber had a surface area of 0.735 m<sup>2</sup>, a volume of 0.0335 m<sup>3</sup>, and a surface area-to-volume ratio of 21.9 m<sup>-1</sup>. The temperature was kept at 25 ± 0.5 °C.

The top sheet of the chamber had three openings, each 6 mm in diameter. The central opening was used to introduce toluene into a Petri dish, and the others were for inlet and outlet air flows. A zero air generator (Domnick Hunter ANG 600/2) supplied the clean air with virtually no VOC which

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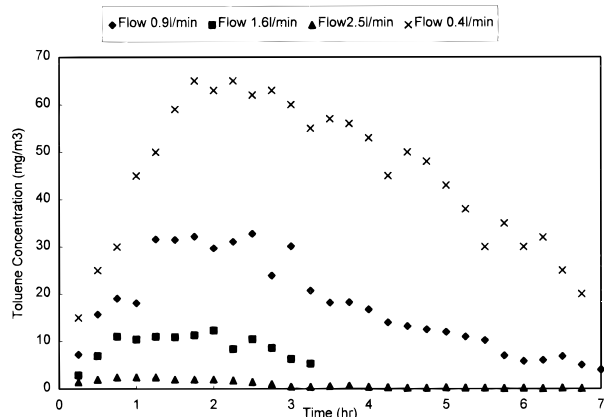


FIGURE 2. Results of chamber experiments at different airflow rates (chamber without any furnishing material).

was pumped through the chamber at a constant flow rate. Prior to each run, the chamber was purged with clean air for several hours. In order to determine the air mixing factor of the chamber, a known amount of  $\text{SF}_6$  (sulfur hexafluoride) tracer gas was injected into the chamber while the clean air was flowing. The gas samples were then collected at fixed time intervals, which were analyzed by using a gas chromatograph UNICAM 4600. The pertinent details of the tracer gas method can be found elsewhere (15, 16).

A set of experiments was first carried out where the test chamber contained no furnishing material. A known amount of toluene was introduced to the chamber in a 6.5 cm diameter Petri dish. Clean air was then pumped through the inlet air opening at a measured constant flow rate. Gas samples at constant time intervals were collected from the outlet air opening by an airtight gas syringe and were analyzed by a gas chromatograph (UNICAM 4600 with a 1.5 m long, 4 mm diameter porapak column equipped with thermal conductivity detector).

In one set of experiments the Petri dish was partially filled with toluene which acted as a source until it evaporated completely. The following decay in toluene concentration was also measured as a function of time (see Figure 2). For these experiments four different airflow rates in the range of (0.4–2.5 L/min) were employed. In the second set of experiments, the source was kept constant by the addition of toluene intermittently so that the level in the Petri dish

remained essentially the same. Here, experiments were carried out at two airflow rates only (see Figure 3).

In the next set of experiments the floor of the chamber was covered with one of the four different types of textile materials that were actually present in the living room of the test house. These were two polyacrylonitrile carpets of different thicknesses, one cotton sofa textile and one polyester curtain textile. The experimental procedure was exactly the same as before and during a run the toluene in the Petri dish evaporated completely so that a decay in the gas-phase concentration of toluene was also recorded (see Figure 4). The collected gas samples of the air/toluene mixture were immediately analyzed by gas chromatography.

**Test House.** The test house was a furnished, two-story villa-type building with a central air conditioning system. Experiments were carried out in the living room which were furnished mainly with the four materials studied here. Toluene was introduced in a 15 cm diameter dish that was placed in the center of the room and at about 100 cm above the floor. Indoor and outdoor pumped air sampling was performed simultaneously. The chemical analysis of toluene was carried out by the thermal desorption gas chromatography technique using a Perkin-Elmer automatic thermal desorber (ATD 400) coupled to a PE 8700 gas chromatograph. The pumped sampling of air was carried out by using Perkin-Elmer stainless steel air sampling tubes filled with Tenax-TA (60–80 mesh). The use of the Perkin-Elmer sequential tube sampler (STS25), specifically designed for pumped sampling and thermal desorption, was described elsewhere (14). The tubes were conditioned by heating in a flow of helium before each sampling, and the conditioned tubes tested to be free of any residual contaminants prior to sampling.

The air exchange rate of the test house was also determined experimentally by using the tracer gas  $\text{SF}_6$  and the pertinent details can be found elsewhere (16).

## Modeling

A simple, single-chamber unsteady-state mass-balance model, adopted from Tichenor et al. (17) and Ligocki et al. (18), was used as the framework for the development of the sorption model for both the chamber and the house. The model assumes that the resistance of external mass transfer for the transport of a VOC from bulk to the surface was negligible. Further, it was also assumed that the effects of diffusion in the pores of the furnishing material could be neglected. Thus,

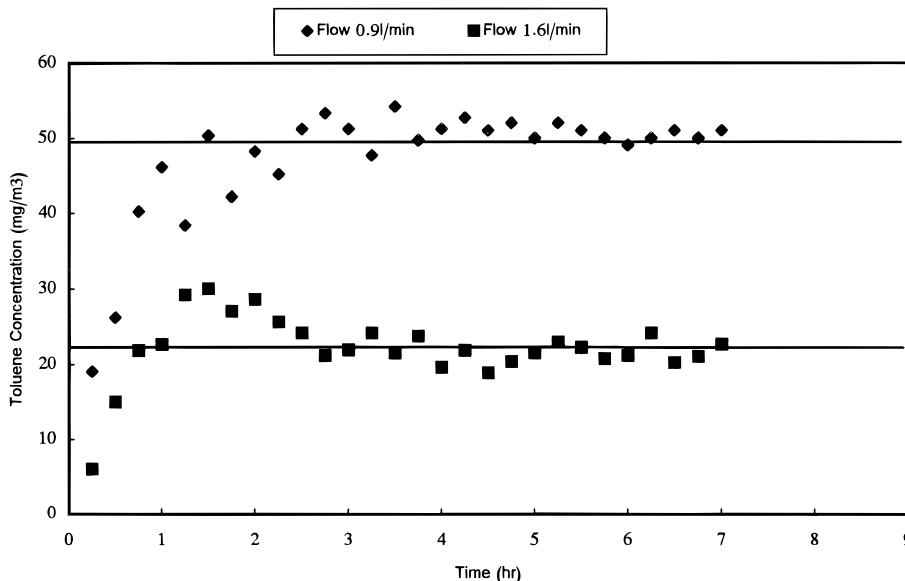


FIGURE 3. Results of chamber experiment at different airflow rates using a constant source (chamber without any furnishing material).

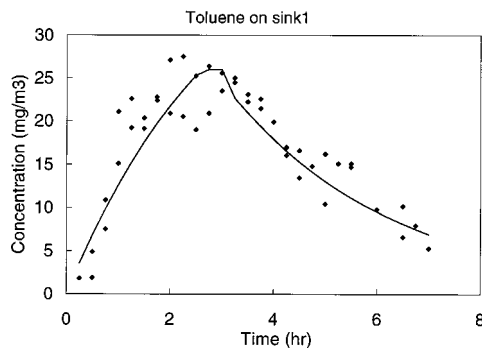


FIGURE 4. Comparison of measured and predicted toluene concentrations of the test chamber furnished with carpet A.

the only rate phenomena on the vicinity of the surface of the materials were assumed to be the adsorption and the desorption of the VOC. Then, the governing equations for indoor concentration ( $C_i$ ) and sorbed concentration ( $C_s$ ) are

$$\frac{dC_i(t)}{dt} = NC_o(t) - NC_i(t) - R_a(t) + R_d(t) + r_s(t)A_{\text{source}}/V \quad (1)$$

$$\frac{dC_s(t)}{dt} = k_a C_i(t) - k_d C_s(t) \quad (2)$$

$$R_a(t) = k_a (A_{\text{sink}}/V) C_i(t) \quad (3)$$

$$R_d(t) = k_d (A_{\text{sink}}/V) C_s(t) \quad (4)$$

where  $N$  is the effective air exchange rate which is equal to  $m(Q/V)$  (where  $Q$  is the volumetric flow rate and  $V$  is the indoor volume),  $C_o(t)$  is the outdoor concentration,  $A_{\text{sink}}$  and  $A_{\text{source}}$  are the surface areas of the sink and the source, respectively,  $k_a$  and  $k_d$  are the adsorption and the desorption rate constants, respectively, and  $r_s(t)$  is the source emission rate. The latter is either equal to  $k_c (C^* - C_i(t))$  or zero depending on whether there is any toluene left in the Petri dish or not. Here  $C^*$  is the vapor pressure of toluene divided by  $RT$  (which is then converted to  $\text{mg}/\text{m}^3$ ) and  $k_c$  is the gas-side mass-transfer coefficient.

The above model assumed also linear adsorption and desorption rates, which were found to be appropriate for the adsorption of ambient levels of volatile organic compounds (VOCs) on building materials (19). In addition, the adsorption isotherms of several VOCs on indoor surface materials (such as polyacrylonitrile carpet) were determined by thermal gravimetry in this laboratory and found to be in agreement with Langmuir theory leading to linear dependency at low concentrations of this study.

### Calculations during Evaporation

During the evaporation of toluene from the Petri dish, the source term was considered to be equal to  $k_c (C^* - C_i(t))$ . The gas-side mass-transfer coefficient  $k_c$  was calculated by using eq 1 from the results of the unfurnished chamber. Here, it was assumed that negligible adsorption/desorption took place by the glass walls of the chamber which appears to be reasonable for toluene (see Sollinger (11, 12)). Using such determined  $k_c$  values, eqs 1 and 2 were then solved simultaneously to determine  $k_a$  and  $k_d$  values for the four furnishing materials by making the regression analysis of respective measured concentrations versus time data. Finally, these rate constants, which were obtained from the chamber studies, were then used for  $k_a$  and  $k_d$  to predict the variation of test house concentrations with time.

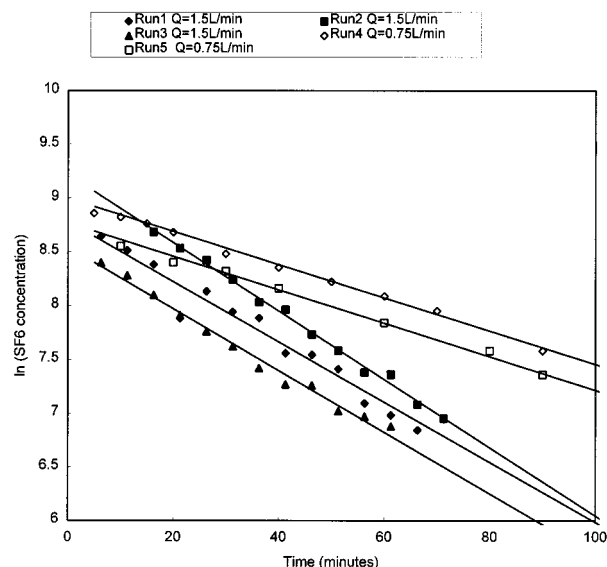


FIGURE 5. Tracer decay data for the test chamber to determine the mixing factor.

TABLE 1. Measured and Calculated Air Exchange Rate in the Chamber

air flow $Q$ (L/min)	measured $N$ ( $\text{h}^{-1}$ )	calculated $Q/V$ ( $\text{h}^{-1}$ )	mixing factor $m = N/(Q/V)$
1.5	2.056	2.687	0.765
1.5	1.904	2.687	0.709
1.5	1.720	2.687	0.640
0.75	0.925	1.343	0.689
0.75	0.941	1.343	0.701

Since the living room of the test house contained all four types of furnishings, eqs 3 and 4 were replaced by the following, respectively:

$$R_a(t) = [(\sum_{i=1}^4 k_{a,i} A_{\text{sink},i})/V] C_i(t) \quad (5)$$

$$R_d(t) = [(\sum_{i=1}^4 k_{d,i} A_{\text{sink},i})/V] C_s(t) \quad (6)$$

## Results and Discussion

**Test Chamber without Furnishing.** Figure 5 shows the results of tracer gas  $\text{SF}_6$  experiments. For a given air flow rate, experiments were repeated for different initial  $\text{SF}_6$  concentrations as shown in the figure. The slope of each line is equal to the measured air exchange rate ( $N$ ) (15, 16). Table 1 shows the measured and calculated air exchange rates for the five runs in addition to the calculated mixing factor. The mixing factor values ranged between 0.64 and 0.765 and had an average value of  $0.7 \pm 0.02$ . The estimated standard deviations of the measured air exchange rate were 0.097 and  $0.0084 \text{ h}^{-1}$  for the corresponding air flow rates of 1.5 and 0.75 L/min, respectively.

Figure 2 shows the experimental results of toluene evaporation at different flow rates. The concentration increases until the complete exhaustion of toluene in the Petri dish is achieved. Then the concentration decreases since there is no source of it, that is,  $r_s(t) = 0$ . As the air flow rate was decreased from 2.5 to 0.4 L/min, the concentration of toluene in air increased from 2.5 to  $65 \text{ mg}/\text{m}^3$ . The measured

TABLE 2. Estimated Values of Gas-Side Mass-Transfer Coefficient of Toluene in Air at Different Airflow Rates from Unsteady- and Steady-State Experiments

flow (L/min)	$k_c$ (m/hr)
Unsteady State	
0.4	0.00205
0.9	0.00243
1.6	0.00249
2.5	0.00275
Steady State	
0.9	0.00288
1.6	0.00289

TABLE 3. Adsorption and Desorption Rate Constants and the Specific Surface Area of Furnishing Materials

material	BET area (m <sup>2</sup> /g)	$A_{\text{sink}}$ (m <sup>2</sup> )	$k_a$ (m/h)	$k_d$ (h <sup>-1</sup> )
carpet A (Sink1)	1.4167	70.835	0.00106	0.0551
carpet B (sink2)	1.2431	53.080	0.00123	0.0415
sofa (sink3)	0.7572	22.716	0.00111	0.0382
curtain (sink4)	0.4796	10.072	0.00097	0.0359

toluene concentration versus time data were fitted to eq 1 with  $r_s(t) = k_c(C^* - C_i(t))$  and  $N = m(Q/V)$  using the measured mixing factors to yield  $k_c$ . Table 2 shows these results at four different flow rates and the gas-side mass-transfer coefficient increased gradually with the gas flow rate which was to be expected.

The decaying part of the curves in Figure 2 can now be used to obtain an independent estimation of the mixing factor since for this part  $r_s(t)$  is equal to zero and the only unknown in eq 1 is  $m$ . The results are in reasonable agreement with the SF<sub>6</sub> experiments; for instance, data at an air flow rate of 1.6 L/min yielded an  $m$  value of 0.63 which compares well with the values in Table 1.

Finally, Figure 3 shows experimental results in which the level of toluene in the Petri dish was kept constant. In this case noting that  $r_s(t) = k_c(C^* - C_i(t))$  and the derivative of  $C_i(t)$  was equal to zero, eq 1 can be solved algebraically to obtain  $k_c$  for the steady-state part of the experiments, that is, when the toluene concentration became essentially constant. Table 2 shows also these data which are in good agreement with the unsteady-state results.

**Test Chamber with Textile.** During these sets of experiments, the floor of the chamber, which had a surface area of 0.226 m<sup>2</sup>, was covered with one of the four furnishing materials. Since the N<sub>2</sub>-BET area is universally meaningful for sorption phenomenon, this was also measured for all four materials and the total BET areas are given in Table 3. Toluene evaporation experiments were carried out for all four materials at an air flow rate of 0.9 L/min. The concentration of toluene in air as a function of time was measured both during the evaporation and for the following decay period.

Figure 6 compares the bulk concentration of toluene in air versus time with and without one of the furnishing materials (that is, carpet A). Curve 1 represents experiments with no material, that is, with no effective sink during evaporation and with no additional source during the decay period. Curve 2 represents the results when the chamber floor was covered with carpet A. Comparison of these curves indicates clearly that during the evaporation period the material acts as a sink, resulting in a lowered toluene concentration. On the other hand, higher concentrations during decay shows clearly that the same material now desorbs previously adsorbed toluene and acts as a source. Similar results were also obtained by the other three materials, showing clearly that typical furnishing materials can adsorb

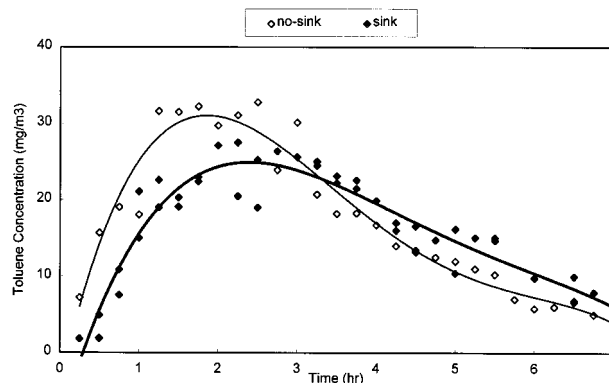


FIGURE 6. Comparison of measured toluene concentrations versus time for the chamber with and without furnishing material (i.e., carpet A) (curves represent fittings).

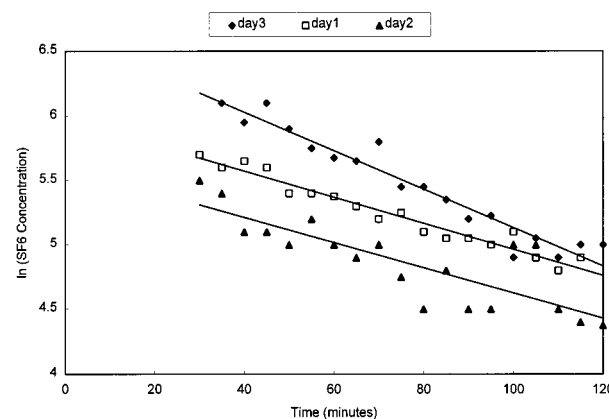


FIGURE 7. Tracer decay data for the test house to determine hourly air exchange rate.

a VOC which will then be desorbed if its concentration in the indoor air is lowered.

Equations 1 and 2 were then solved simultaneously taking  $r_s(t)$  as  $k_c(C^* - C_i(t))$  when there was toluene in the Petri dish and as zero during the decay. The surface area of the furnishing material was based on the N<sub>2</sub>-BET areas and they are given in Table 3. The regression based on the minimization of least-square error yielded the two rate constants, that is,  $k_a$  for adsorption and  $k_d$  for desorption. The values of  $k_a$  and  $k_d$  for all four materials are given in Table 3. Figure 4 compares the model prediction using the rate constants of Table 3 with measured toluene concentrations as a function time for carpet A. Similar results were also obtained for other three materials, indicating that the model, as proposed in eqs 1 and 2, appears to be an adequate representation of the actual case.

**Test House.** Figure 7 shows the tracer gas SF<sub>6</sub> experiments for determining the effective air exchange rate  $N$  in the test house which were carried out three times with differing initial concentrations. The linear regression of the data resulted in  $N$  values for this central air conditioned house as 0.61, 0.59, and 0.89 h<sup>-1</sup> and a mean value of them will be used for the latter part.

Prior to introducing a source for toluene evaporation experiments, the toluene levels of the test house was sampled for 5 h and five samples were collected using a sequential tube sampler. The toluene concentrations of these samples were analyzed by gas chromatography and ranged from 0.055 to 0.111 mg/m<sup>3</sup> and had an average value of 0.0754 mg/m<sup>3</sup> which was considered to be its initial value. After toluene was introduced in a 15 cm dish, its concentration increased rapidly and reached a maximum value after about 12 h where the total duration of the experiment was 60 h. Figure 8 shows



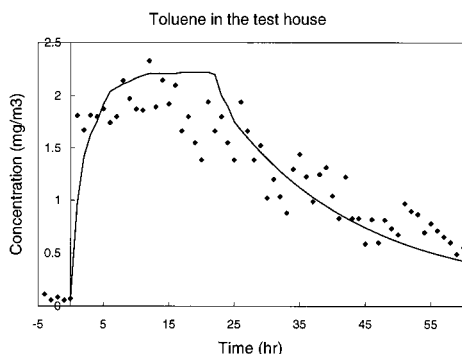


FIGURE 8. Comparison of measured and predicted toluene concentrations as a function of time for the test house.

the measured indoor concentration as a function of time. During the runs, the measured outdoor concentration ranged between 0.05 and 0.3 mg/m<sup>3</sup> and had an average value of  $0.1542 \pm 0.0686$  mg/m<sup>3</sup>. This average outdoor concentration was used while solving eqs 1 and 2.

Since the rate constants of the major furnishing materials were already determined in the test chamber, these values could be employed in solving eqs 1 and 2 by using also eqs 5 and 6. The surface areas of four materials were estimated from their weights by using also the BET values of Table 3. The estimated areas were 3996, 795, 1250, and 182 m<sup>2</sup> for carpet A, carpet B, cotton sofa, and the polyester curtain, respectively. For the source term, the value of  $r_s(t)$  was considered to be  $k_c(C^* - C_i(t))$ . The value of  $k_c$  was calculated from the steady-state data corresponding to a time span of 10–16 h (see Figure 8). Using this estimated value of  $k_c$ , eqs 1–4 can be solved simultaneously to predict the increase in the concentration of toluene. Figure 8 shows the result of this model. The prediction of the decaying part does not require a value of  $k_c$  since  $r_s(t)$  becomes zero after 18 h when all of the toluene has already evaporated. The predicted concentration profiles were also given in Figure 8 for this part. An acceptable fit indicates that the model proposed here together with the adsorption and desorption constants of typical furnishing materials obtained in the test chamber are probably appropriate.

In conclusion, this simple IAQ model proposed here could be considered as an adequate representation of the much more complex indoor environment. Apart from the  $k_c$  value in the source term, all other parameters, such as adsorption and desorption rate constants for toluene (which were obtained from the test chamber results), and the hourly air exchange rate for a certain house (which could be calculated from ventilation parameters) are universal. The remaining additional information required consists of furnishing material types and their surface areas for a certain case, which could be obtained from the furniture loading. The rate constants reported here are for toluene and for 25 °C only, and if such data were to be made available for other temperatures and for other typical indoor VOCs, the proposed model could then successfully be used to predict time dependency of them in an indoor environment.

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## Notation

$A_{\text{sink}}$	sink surface area, m <sup>2</sup>
$A_{\text{source}}$	source surface area, m <sup>2</sup>

$C_i$	indoor toluene concentration, mg/m <sup>3</sup>
$C_o$	outdoor toluene concentration, mg/m <sup>3</sup>
$C_s$	sorbed surface concentration of toluene, mg/m <sup>2</sup>
$C^*$	equilibrium concentration of toluene at the air–liquid interface, mg/m <sup>3</sup>
$k_a$	adsorption rate constant, m/h
$k_c$	gas-side mass-transfer coefficient, m/h
$k_d$	desorption rate constant, h <sup>-1</sup>
$k_s$	emission rate constant, h <sup>-1</sup>
$m$	mixing factor
$N$	total air exchange rate, h <sup>-1</sup>
$Q$	air flow rate, L/min
$R_a(t)$	adsorption rate, mg/m <sup>3</sup> ·h
$R_d(t)$	desorption rate, mg/m <sup>3</sup> ·h
$r_s(t)$	emission rate, mg/m <sup>2</sup> ·h
$t$	time, h
$V$	room volume, m <sup>3</sup>

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