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ON VALIDATION OF SOURCE AND SINK MODELS: PROBLEMS AND POSSIBLE SOLUTIONS

by

Zhishi Guo

Acurex Environmental Corporation
P. O. Box 13109
Research Triangle Park, NC 27709

ABSTRACT:

While model validation remains the weakest part of the entire process of indoor air quality (IAQ) model development, special problems have made the validation of indoor source and sink models even more difficult. Many source and sink models have been developed, but few have been properly validated. Major problems with current procedures include: elusive model parameters; confusion in parameter estimation methods; uncertainty in scale-up and misleading scaling factors; unspecified validity ranges; and weakness in quantitative comparisons between models and experimental observation.

To improve validation procedures, we have identified a number of potential areas including: proper definition of validation scope, proper use of statistical comparison methods, development of mass transfer indices to bridge the gap between test chambers and real buildings, and development of a cooperative effort to build a source and sink database to facilitate validation.

KEY WORDS: model validation, source, sink, indoor air quality

Introduction

Model validation is the process of evaluating the usefulness, accuracy and limitation of a model under various application conditions. While validation remains the weakest part in the entire process of IAQ model development, validating of source and sink models has its special difficulties. In fact, although many source and sink models have been developed, few have been properly validated.

General discussions on model validation can be found in the literature [1-4], but the special problems associated with source and sink models remain untouched. This paper identifies the major problems with current practice in validating source and sink models, and discusses some possible solutions. Most of the problems raised came from examining the author's own practice in model validation, and some came from reviewing other researchers' work. Although this paper is focused on validation, the author has found it difficult to completely separate model validation from model building. Some discussions here may be applicable to both steps.

Purposes of Validating Source and Sink Models

Why must we validate source and sink models? Before answering this question, we need to briefly discuss how they are developed and how they are used. It is generally agreed that, before any satisfactory verification scheme is adopted, it is necessary to determine the primary purpose or purposes to be served by the verification [4].

The development of source and sink models relies heavily on experimental observations and understanding of the mechanisms. From chamber data, one can often calculate the emission rate based on mass balance equations. For example, if we assume that the adsorption of pollutants on chamber walls is negligible, the mass balance for an area source in a chamber in an infinitesimal time period dt is:

Mass increased in chamber = Mass emitted - Mass exfiltrated

$$\text{or} \quad V \frac{dC}{dt} = S\,R(t) - Q\,C \qquad (1)$$

where V = chamber volume;

C = chamber concentration;

S = area of the source;

R(t) = emission rate at time t; and

Q = inlet/outlet air flow rate.

Let L = S/V be the loading factor, and N = Q/V be the air exchange rate, we then have:

$$R(t) = \frac{dC/dt + N\,C}{L} \qquad (2)$$

When the concentration data have reasonable time resolution, the term dC/dt can be well represented by $\Delta C/\Delta t$. The data represented by empty squares in Figure 1 is an example of using Equation 2 to calculate the emission rates from the chamber concentration data. The source tested was an indoor coating product. A total of 1.82

g of the product was applied to a 0.021 m^2 oak board, and tested for total organic emissions in a 53 L stainless steel chamber; the chamber was kept at 23° C and 45% relative humidity; and the air exchange rate was 0.514 h^{-1}.

Direct calculation of the adsorption and desorption rate is impossible because the two processes occur simultaneously, but it is possible to calculate the net mass transfer rate for the pollutant to or from the sink surface.

Based on experimental observations, a model can be developed in different ways. For the convenience of discussion, we can roughly divide all source and sink models into two categories: statistical models and fundamentally based models.

To find a statistical model suitable to the data in Figure 1, basic knowledge of analytic geometry will convince us that the emission pattern in Figure 1 can be approximated by a first order decay model[5]:

$$R(t) = R_0 \, e^{-kt} \qquad (3)$$

where R_0 = initial emission rate; and

k = first order decay constant.

The next step is to estimate the model parameters -- the constants (or coefficients) in a model's expression. In this case, we need to determine the values for R_0 and k that could give the best agreement between the model prediction and observation. The solid line in Figure 1 was obtained by log-linear regression imposed on the emission rate data.

In most cases, however, model parameters are estimated by fitting the model to concentration data. This is especially true for sink models. No matter what data are used, such model development depends heavily on statistical estimations.

To develop fundamentally based models, or mass transfer models, one needs to understand the physical-chemical phenomena involved such as evaporation, adsorption, molecular diffusion in the air-surface interface, and molecular diffusion within the source. The parameters of a mass transfer model often have well defined physical meanings such as vapor pressure, molecular weight, mole fraction, adsorption energy, diffusivity, and boundary layer thickness. Most parameters are obtained either directly from the literature or from well-established models. Therefore parameter estimation for mass transfer models does not rely heavily on curve-fitting. In some cases, however, one or two of the parameters may have to be estimated empirically. It should be pointed out that, among existing source and sink models, many are neither pure statistical models nor pure mass transfer models.

Like any other model, a source or sink model is at best a simplification and approximation of a real source or sink. Therefore, no one should expect the model to represent reality perfectly. There is a definite need, however, to know how good the agreement would be under certain conditions between the model and reality. In other words, we need some estimation of the model's predictive error.

A model may give satisfactory prediction in one case but fail in another. Then we must know the conditions under which the model

gives acceptable prediction, and those under which the model fails. We also need to know the sensitivity of the model to its parameters.

Source and sink models are seldom used alone. In most cases, they are part of an IAQ model. Since both source and sink models serve the IAQ model, any predictive errors they generate can be propagated during an IAQ simulation. We need to know how the inaccuracy of a source or sink model could affect the IAQ simulation output.

In summary, the purposes of validating source and sink models may include:

- estimating the model's predictive error by comparing the predicted values to observed ones;
- defining validity range and validity conditions;
- defining conditions of failure;
- defining the applicability of the model (good for a single product or for a type of product, or for several types of products);
- estimating the model's sensitivity to its parameters; and
- if possible, estimating the model's propagated error in IAQ simulation.

For those who want to use source and sink models, proper validation will provide them with a clear view of the conditions for reliable model application, and the uncertainty they may expect under certain conditions. Such information may prevent the user from misusing the models. For those who develop source and sink models, proper validation may enable them to learn how to improve

their models, and how to develop more advanced models.

Major Problems with Current Validation Procedures

Variable Model Parameters

Most statistical models have at least one variable parameter -- the parameter that is determined through statistical estimation and whose value changes as the environmental conditions change. Parameters R_0 and k in Equation 3 are such variable parameters. Unlike physical parameters (such as boiling point, vapor pressure, diffusivity, and air velocity), these parameters are sensitive to any change of the environment. The modelers often find it difficult to choose proper values under certain given conditions. The following example illustrates how those parameters may vary with test conditions.

A wood stain product was tested for its organic emissions in small environmental chambers [5]. The concentration data were fit by the first-order source model. Test conditions and estimated emission factors are summarized in Table 1. As one can see, both R_0 and k vary over a wide range. Unless correlations are found between these parameters and the environmental conditions (such as air exchange rate, loading factor, application rate, and degree of air turbulence), there is no way to tell what values to choose for R_0 and k under certain given conditions.

(N/L) - a Misleading Scaling Factor

The ratio of air exchange rate (N) over chamber loading factor (L) has been one of the most commonly used scaling factors in chamber experimental design and model validation. The concept

behind this factor is that, if we double the chamber loading and the air exchange rate simultaneously, the two effects will be canceled out, and the resulting chamber concentration should be the same [6]. This assumption is correct if, and only if, we are dealing with a constant source at steady state. The problem is that it has been used far beyond the above limitations. It is theoretically incorrect to apply such a scaling factor to either non-steady state situations and/or non-constant sources. To illustrate this problem, let's look at two theoretical concentration models: Equation 4 is the expression for a constant source and Equation 5 for the first-order decay source [5].

$$C(t) = \frac{L\,R}{N}\,(1 - e^{-Nt}) \qquad (4)$$

$$C(t) = \frac{L\,R_0}{N - k}\,(e^{-kt} - e^{-Nt}) \qquad (5)$$

where C(t) is the chamber concentration;

R = emission rate for constant source;

L = chamber loading;

N = air exchange rate; and

t = time.

Figures 2 and 3 were plotted based on Equations 4 and 5, respectively, assuming that chamber volume = 1 m^3 and $R = R_0 = 400$ $mg/m^2/h$. Figure 2 shows that, for the same constant source, the same N/L may not yield the same concentration curve if steady state is not approached; and Figure 5 shows that, for a given finite source, very different concentration curves are obtained with the

same N/L.

Confusion in Parameter Estimation

Estimating the parameters for a given model from a given set of data can be confusing, too, because there are many ways to fit a model to data. For example, to fit the first-order model to the indoor coating data described above, we have many ways of estimating R_0 and k. For illustration purposes, four different regression methods are discussed here: (A) Using nonlinear regression to fit Equation 5 to the concentration data without any data transformation; (B) Using nonlinear regression to fit Equation 5 to the concentration data with logarithmic transformation; (C) Using nonlinear regression to fit Equation 3 to directly calculated emission rate data (as shown in Figure 1); and (D) Using linear regression to fit Equation 3 to directly calculated emission rate with logarithmic transformation. The different results are given in Table 2. Parameters obtained from method C appeared the best in catching the peak (Figure 4) and also the worst in tracing the tail as illustrated in the semi-log plot (Figure 5). Just the opposite, curve D looked the worst in the high concentration region but gave the best prediction in the tail. The other two curves fell between the two extremes. The questions then become: Which estimation method should we choose? and which set of parameters should we report?

The Effect of Data Range

Many indoor sources last for a long period of time, and the effect of indoor sinks lasts even longer. Tracking such long term effects can be very costly and time-consuming. People usually

have to develop and validate their models by using chamber data within a limited time period. It is clear that, unless the validity range is defined, the statement of "validated" can be very confusing.

The following is an example of this problem. We tested the emission of ethylbenzene from a piece of 0.113 m^2 duct liner in a 53-L chamber (0.54 air change per hour, 23° C, and 45% relative humidity). The sample was taken from the air handling system in a test house, and was previously exposed to ethylbenzene polluted indoor air. Using the first 10 hours of chamber data, we found that the simple first order model (Equation 3) fit the data adequately (Figure 6). The model prediction failed after 10 hours, and the double exponential model $R(t) = R_1 \exp(-k_1 t) + R_2 \exp(-k_2 t)$ seemed more suitable to the wider data range (Figure 7). After 120 hours, however, it failed too. A second-order model, $R(t) = R_0/(1+ktR_0)$, fit the data almost perfectly within 400 hours (Figure 8). It is difficult to tell if or when the second order model fails beyond 400 hours.

Obviously, selection of data range plays an important role in both model building and validation. Without specifying the validity range of a model, the whole validation becomes meaningless. Information on conditions of model failure is especially important to IAQ modelers for IAQ simulation programs do not turn off a source or a sink automatically.

The Effect of Air Velocity and Turbulence

The degree of air turbulence above the source or sink surface can alter the rate of mass transfer in both directions. This means

that a model validated in one chamber may not work at all in another if the air turbulence conditions are significantly different.

To show how the degree of air turbulence affects the model parameters, we did a set of preliminary sink tests in a two-chamber experimental system [7]. A piece of wallboard was placed in the test chamber and insulted by a first-order decay ethylbenzene source. To alter the air circulation conditions, a small biscuit fan was placed inside the test chamber (fan speed could be adjusted by varying the voltage). Tests were conducted with the fan at 50 V, 70 V, and 110 V, respectively, and all other conditions were kept the same. Figure 9 shows one of the test results and the fitting of the dynamic Langmuir sink model [7,8] to the data. The estimated model parameters -- adsorption rate constant k_a and desorption rate constant k_d -- are given in Table 3. It seems that both adsorption and desorption were accelerated by the increased air circulation. During these tests, we did not measure the fan speed and air velocity in the chamber; therefore, the results presented below should be considered as a qualitative illustration. Since air movement and turbulence conditions in a building can be significantly different from that in a chamber, the applicability of chamber results to real buildings has been challenged. This so-called "scale-up" problem has been the most troublesome in validating source and sink models.

Oversimplified Illustration of the Goodness of Fit

Checking the agreement between model prediction and observation can be misleading, too. So far, most modelers,

including the author [9], have tried to show the validity of their models by presenting the model prediction and observation together in a diagram. Then the authors would claim that the presented model had been "validated" (or sometimes more cautiously, "preliminarily validated"). While there is nothing wrong with graphical comparison, something is missing here: What does "validated" or "preliminarily validated" mean? There have to be some criteria so that the model developer and, more importantly, the user can make an objective and quantitative judgement. Statistical methods for verification comparisons are available; unfortunately, many of us often neglect those useful tools.

Special Problems with Fundamentally Based Models

Mass transfer models are preferred to statistical models because the former emphasize the physical understanding of the real mechanisms and because their parameters are usually well defined. But these types of models have their own problems.

To model very complex reality with relatively simple models, the modelers have to exclude whatever they consider "unnecessary" details and focus on one or two mechanisms. Due to the omission of the remaining mechanisms, the resulting models are often unusable unless some fuzzing factors are introduced. These variables often make the mass transfer models less attractive because they have made the mass transfer models undistinguishable from empirical models.

Mass transfer models are often much more complicated than their corresponding statistical models. It is commonly true that a mass transfer model has better "validity" than a statistical

model, but is more difficult to use than the latter due to its complexity.

Recommendations

Three Levels of Validation

As just discussed, the primary purpose of validating source or sink models is to make sure they represent reality close enough under certain conditions so that they can be used in IAQ simulation without bringing in excessive propagation errors. Keeping this in mind, we can divide the validation process into three steps: (1) Checking the agreement between the model and a single set of observations; (2) Checking the agreement between the model and multiple sets of observations; and (3) Verification of scale-up.

After a model is formulated, it is usually compared to a single set of observations to determine if the model represents the real pattern in that particular case reasonably well. If there are variable parameters in the model, they can be estimated by this step. If the model concept is poor, the model may not "survive" this step at all.

Since many, if not all, source and sink models contain variable parameters, one set of parameters which give satisfactory prediction in one event may not work in others. By comparing the model with a few sets of data, one can either fine-tune the parameters or find correlations between the values of parameters and test conditions. The author believes that not all existing source and sink models can survive this validation step.

The last step -- scale-up verification -- requires data from

either real buildings or large test chambers. Without this step, the usefulness of a model cannot be justified.

Some Aspects in Validation Procedures

When comparing a model with observation, the modeler should clearly specify the conditions under which the observation was obtained. This will allow the modeler and the potential users to distinguish the conceptual errors of the model from those of the data. The list of conditions should include, but not be limited to, the following information:

— Chamber specification (type, material, volume, shape, temperature, humidity, pressure, etc.);

— Sample specification (material, size, sample preparation, and position in the chamber);

— Air exchange rate;

— Description of air movement in the chamber (qualitative description such as inlet/outlet pipeline design, with or without forced mixing; and quantitative description such as surface velocity, Reynold's number, or other fluid dynamic parameters);

— Sampling and analytical methods;

— Data quality; and

— Data range.

If some or all the model parameters are estimated using statistical means, a detailed description of the approach used should be given:

— The equation used in the regression (it may or may not be the source or sink model itself);

— Independent and dependent variables;

— Statistical method (linear regression, nonlinear regression, or other methods); and

— Data Transformation (no transformation, logarithmic transformation or, in more general terms, Box-Cox power transformation).

When comparing a model to multiple sets of observations, the chamber data should include:

— Observations under at least two air exchange rates; and

— Observations under at least two loading factors.

If a model is statistically based, the sensitivity of a model to its parameters, and the dependence of model parameters on environmental conditions (such as air exchange rate, loading factor, and degree of air turbulence) should be described.

When performing scale-up verification, data from large test chambers are preferred to those from buildings because the conditions in a building are difficult to control. The most important uncontrolled factors include: varying air exchange rate, multiple air zones, and strong adsorption on many different surface materials. If the model parameters established from previous validation steps need further adjustment in scale-up, such adjustment should be justified.

Finally, validity range and validity conditions should be specified.

Making Comparisons More Objectively

Graphic comparison of model performance is absolutely necessary, but using it alone isn't enough. Statistical tools

should be used to complement graphical comparison.

Quite a few statistical methods are available in making comparisons [10,11]. At least one statistical verification method should be used along with graphical comparisons. The modeler should make careful selection among those methods because different validation purposes require different statistical techniques. The danger of using any goodness-of-fit index in model verification is illustrated by Benarie [4]. Besides, some methods may not be suitable to our particular situation. We should emphasize the importance of physically understanding the model during validation. Some statistical comparison techniques do not help very much in this aspect.

Dealing with Scale-up Problems

Source and sink models based on basic mass transfer theories have received ever-increasing attention by the indoor air community in recent years [12-14]. Many of us, who have been frustrated by elusive statistical models, believe that fundamentally based models are the final solution to our problems, especially to the scale-up problem. Mass transfer models for emission and adsorption are not new: they can be found in many mass transfer monographs. The problem is that people can rarely find a proper model from the existing engineering literature that can be used by IAQ modelers on an "as is" basis because the processes being modeled are too complicated for those models.

To develop relatively simple mass transfer models, we need to select proper expressions for mass transfer coefficients (or mass transfer resistance). Criteria for selecting good expressions may

include:

— They should separate the properties of the environment from those of the source or sink;

— They should be simple enough to be used in source or sink models;

— They can be measured independent of source and sink models; and

— They can be correlated to more complicated boundary layer models.

Making a Cooperative Effort by Building a Source and Sink Database

Generating quality data to validate a model can be very costly and time-consuming. A modeler may have the talent to develop scientifically sound models but may not have the ability or resources to generate good quality data. This has left little choice to the modelers: they often have to accept whatever data they can get, regardless of the suitability of the data to serve their validation purposes.

All modelers would benefit if some organization (a professional society or university, for example) would assume the responsibility for collecting source and sink data from volunteer research organizations and build an indoor source and sink database. Such a cooperative effort could make a great difference in easing the shortage of quality data.

Ending Remarks

As stated earlier, the major purpose of this paper has been to raise issues regarding the validation of source and sink models. We have identified a number of potential problem areas in IAQ modeling. Due to the great difficulty in validating indoor source and sink models, we cannot expect all the problems to be solved overnight. Improvement can only be made gradually. Besides, as long as a new model is built on a sound scientific basis, the model can be published without complete validation. The model developer should be allowed to leave part of the validation work to other researchers. Validating a model is as important as creating one, and can be an original contribution to science.

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Table 1 -- Total Organic Emissions from Wood Stain

Chamber Type	166 L with a slow stirrer
Air Exchange Rate Range	0.35 to 4.6 [h^{-1}]
Application Rate Range	23 to 26 [g m^{-2}]
Chamber Loading Range	0.1 to 1.3 [m^{-1}]
Range of Estimated R_0	2.2 to 27 [g m^{-2} h^{-1}]
RSD (mean)	7%
RSD (range)	3% to 12%
Range of Estimated k	0.24 to 2.41 [h^{-1}]
RSD (mean)	11%
RSD (range)	4% to 16%

Table 2 -- Estimated Emission Factors with Four Regression Methods

	Method A	Method B	Method C	Method D
Model	Eq.5	Eq.5	Eq.3	Eq.3
Data Type[a]	Conc.	Conc.	Rate	Rate
Data Scale	Normal	Log	Normal	Log
Regression	Nonlinear	Nonlinear	Nonlinear	Linear
R_0 (g m^{-2} h^{-1})	10.1	6.68	14.0	4.96
RSD	4.2%	5.8%	4.3%	2.3%
k (h^{-1})	0.356	0.208	0.599	0.186
RSD	5.1%	2.5%	6.1%	2.0%

[a] Conc. = concentration data; rate = emission rate data.

Table 3 -- The Estimated Adsorption Rate Constant (k_a) and Desorption Rate Constant (k_d) at Three Fan Speeds

Fan Voltage (V)	50	70	110
k_a ± RSD (m h^{-1})	0.64±5.9%	0.91±6.0%	1.23±12%
k_d ± RSD (h^{-1})	1.48±6.4%	1.44±7.0%	2.54±6.7%

Sink material: 0.14 m^2 wallboard; pollutant: ethylbenzene; air exchange rate = 1.2 h^{-1}; temperature = 23°C; and relative humidity = 45%.

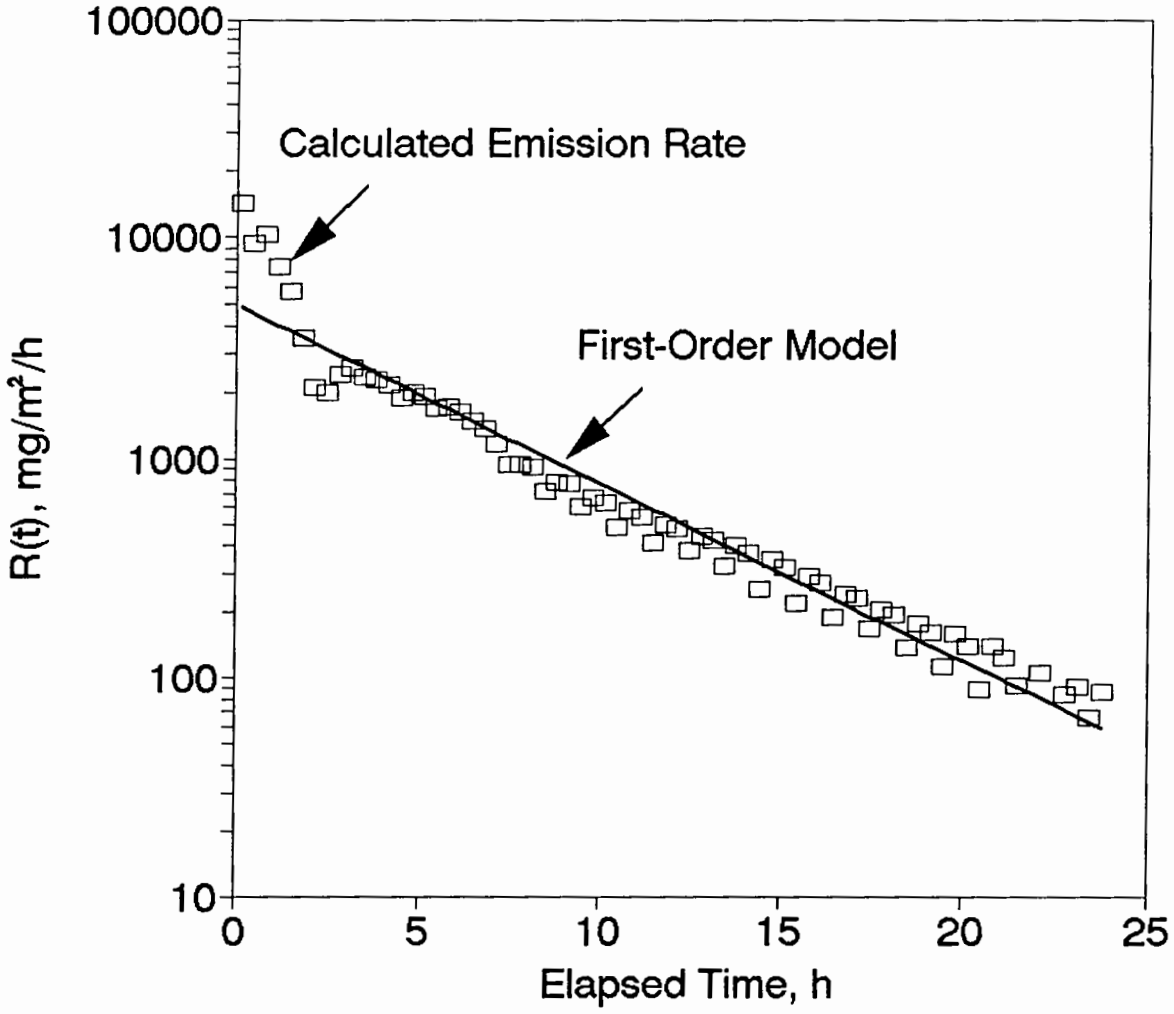

Figure 1. Calculated VOC emission rate for an indoor coating product based on chamber concentrations and first-order model prediction.

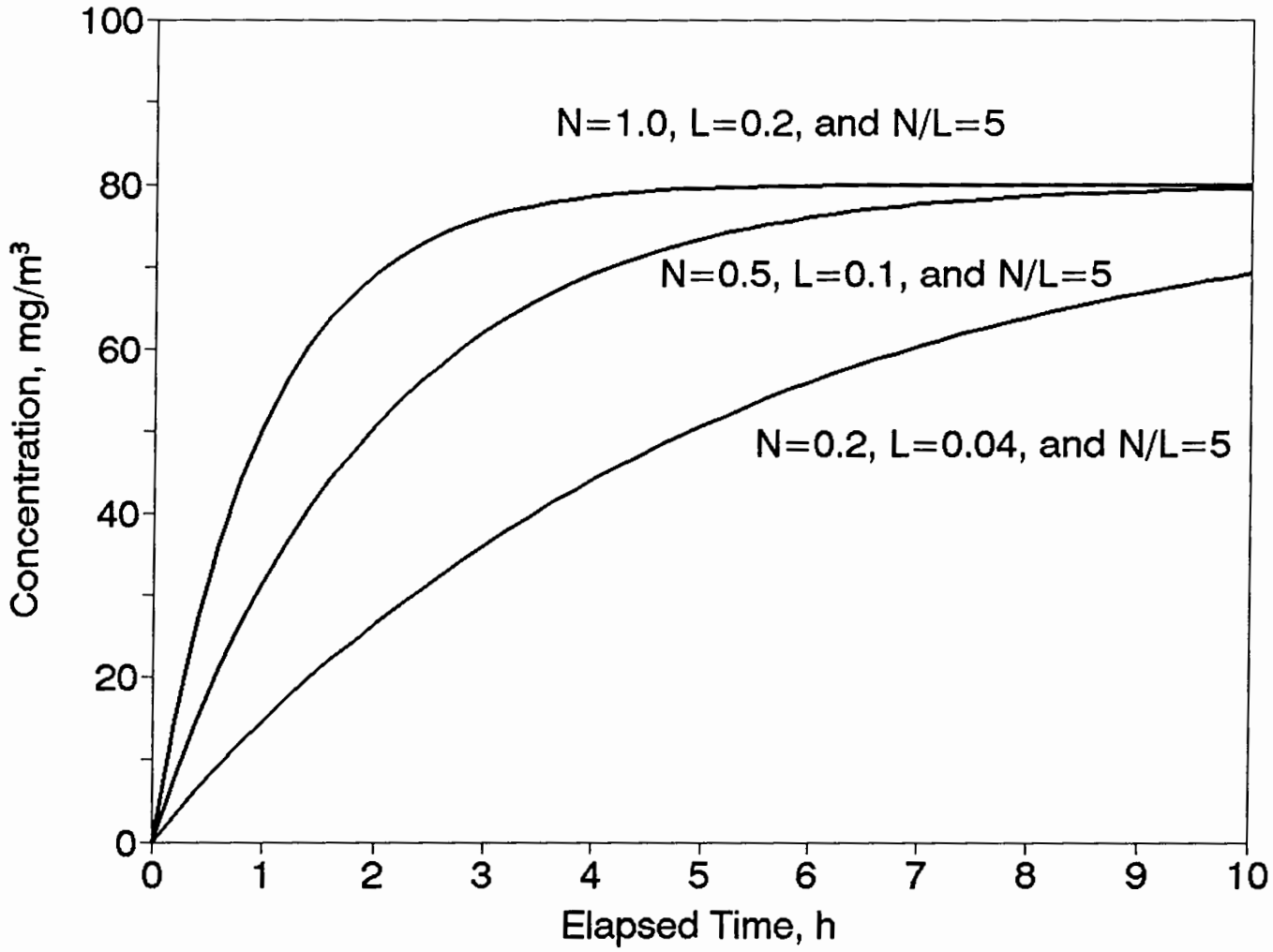

Figure 2. Improper use of (N/L) - constant source at non-steady state.

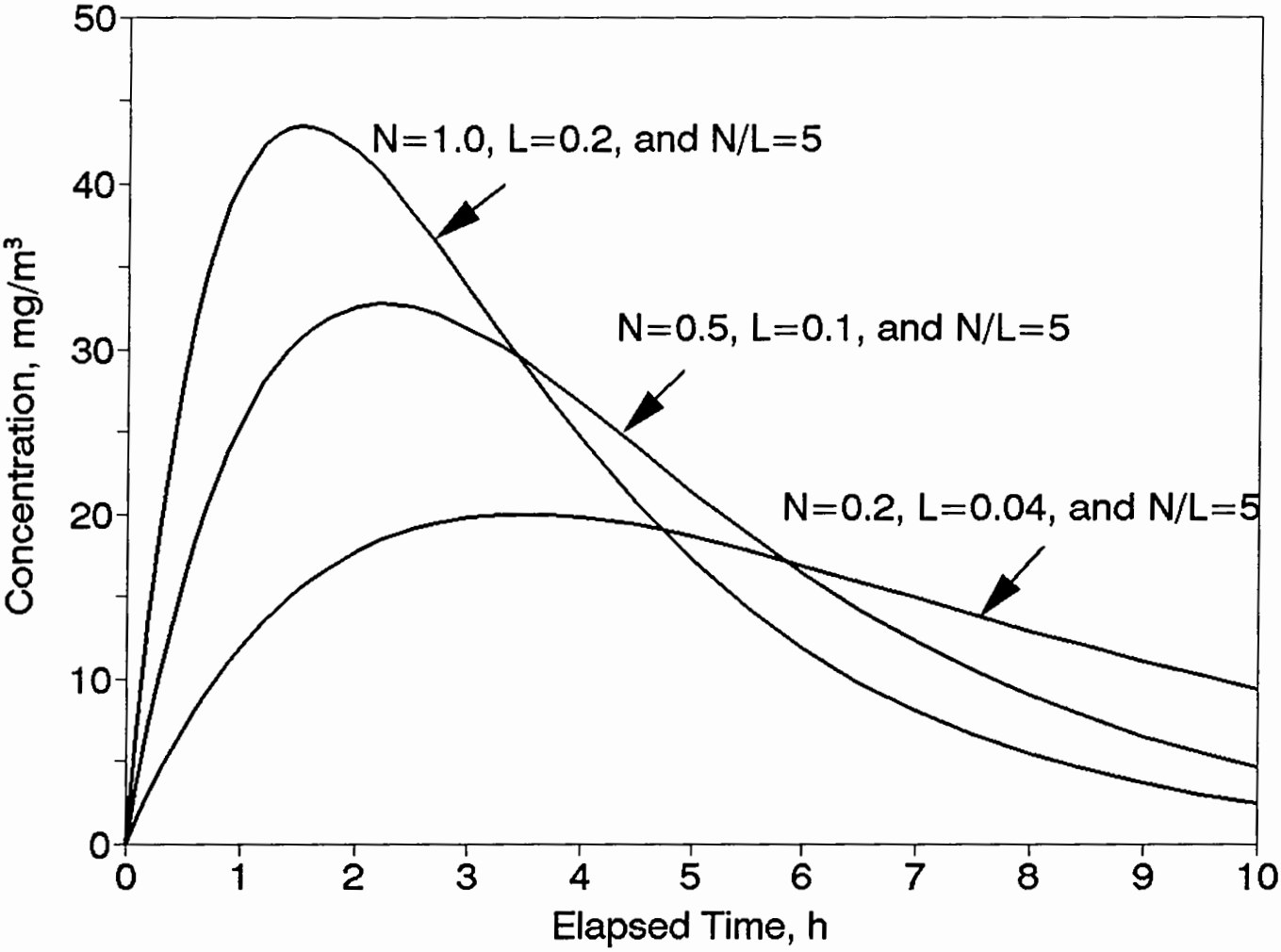

Figure 3. Improper use of (N/L) - finite source.

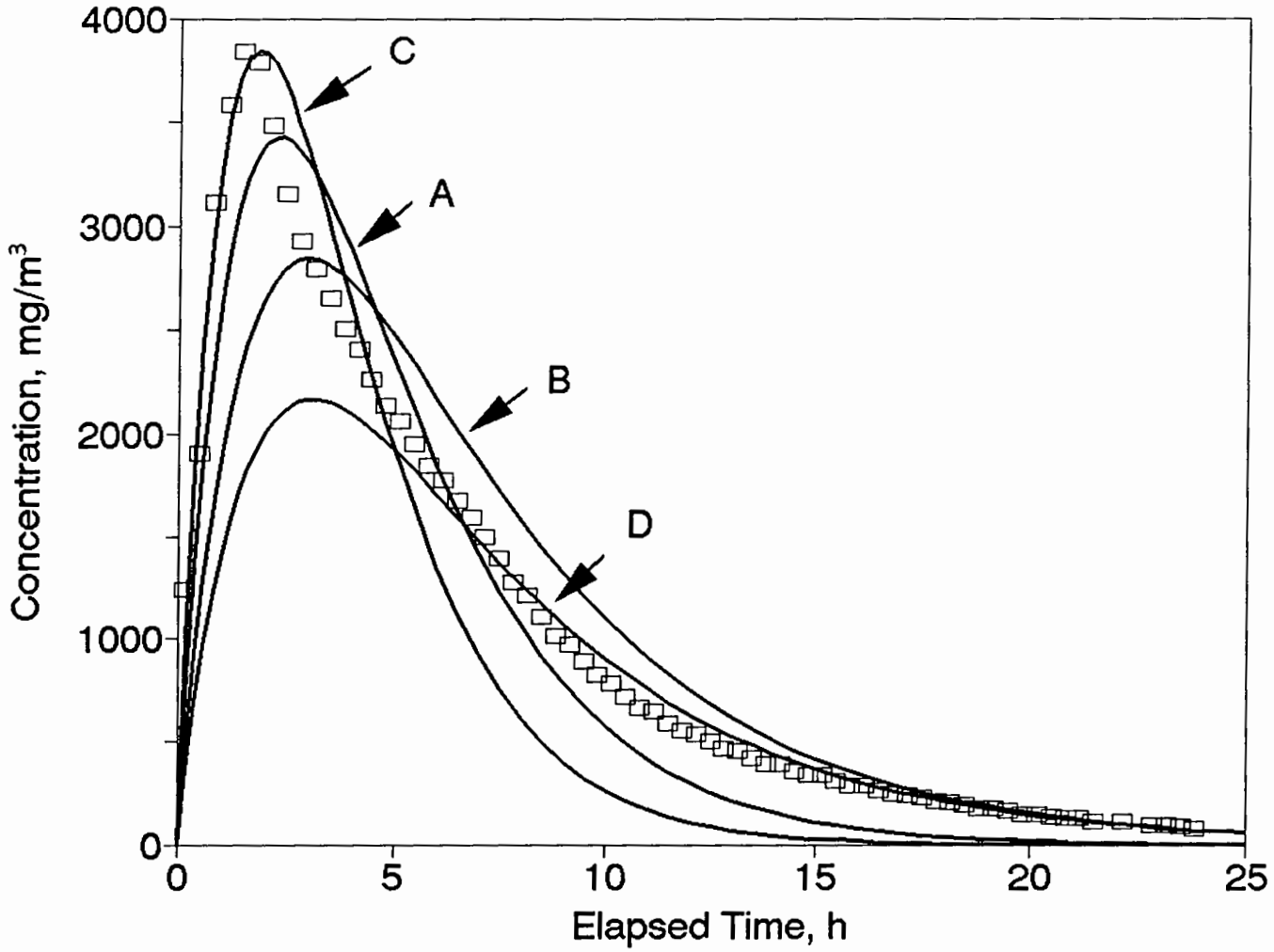

Figure 4. The effects of parameter estimation methods on model performance - normal scale.

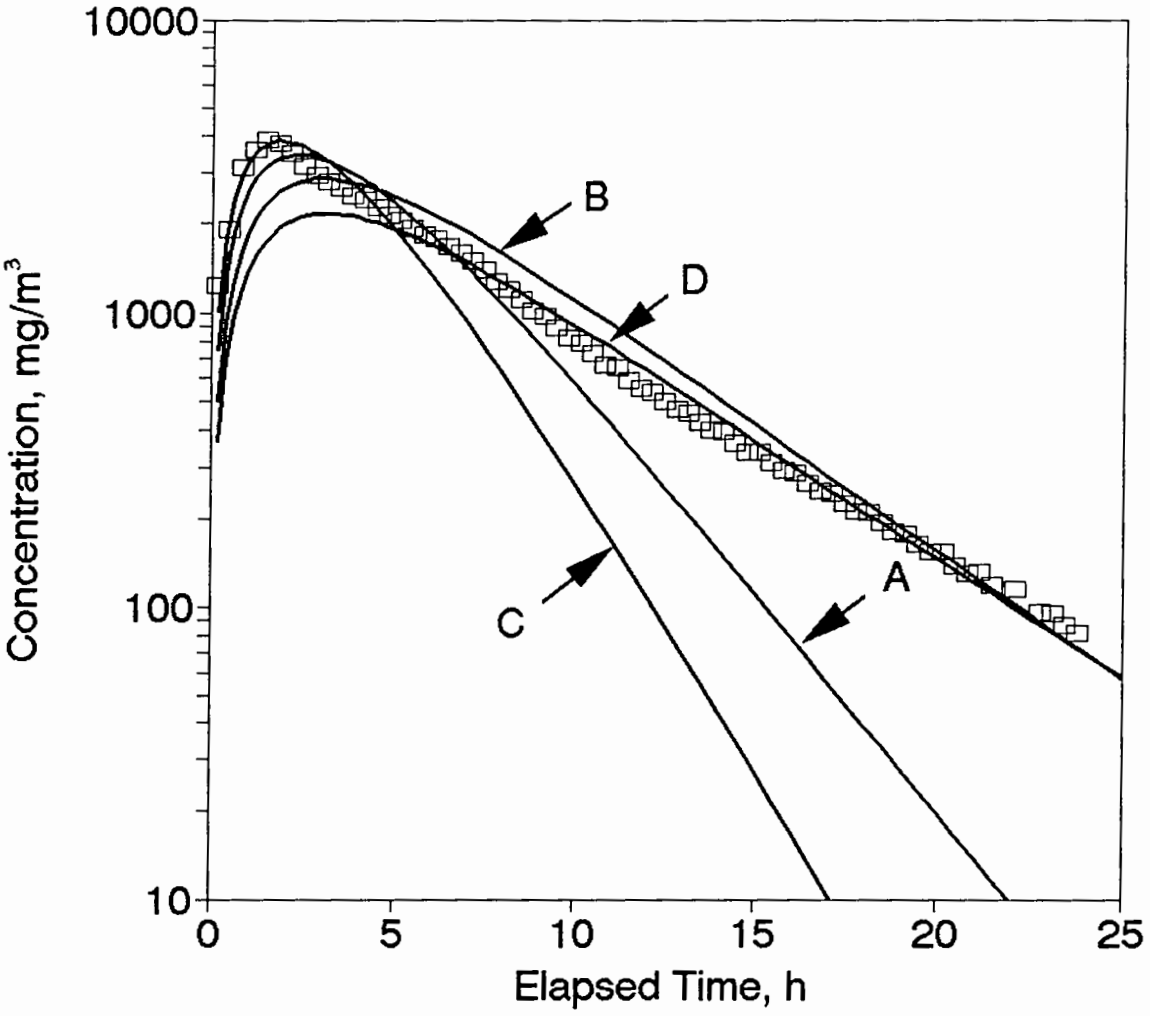

Figure 5. The effects of parameter estimation methods on model performance - log scale.

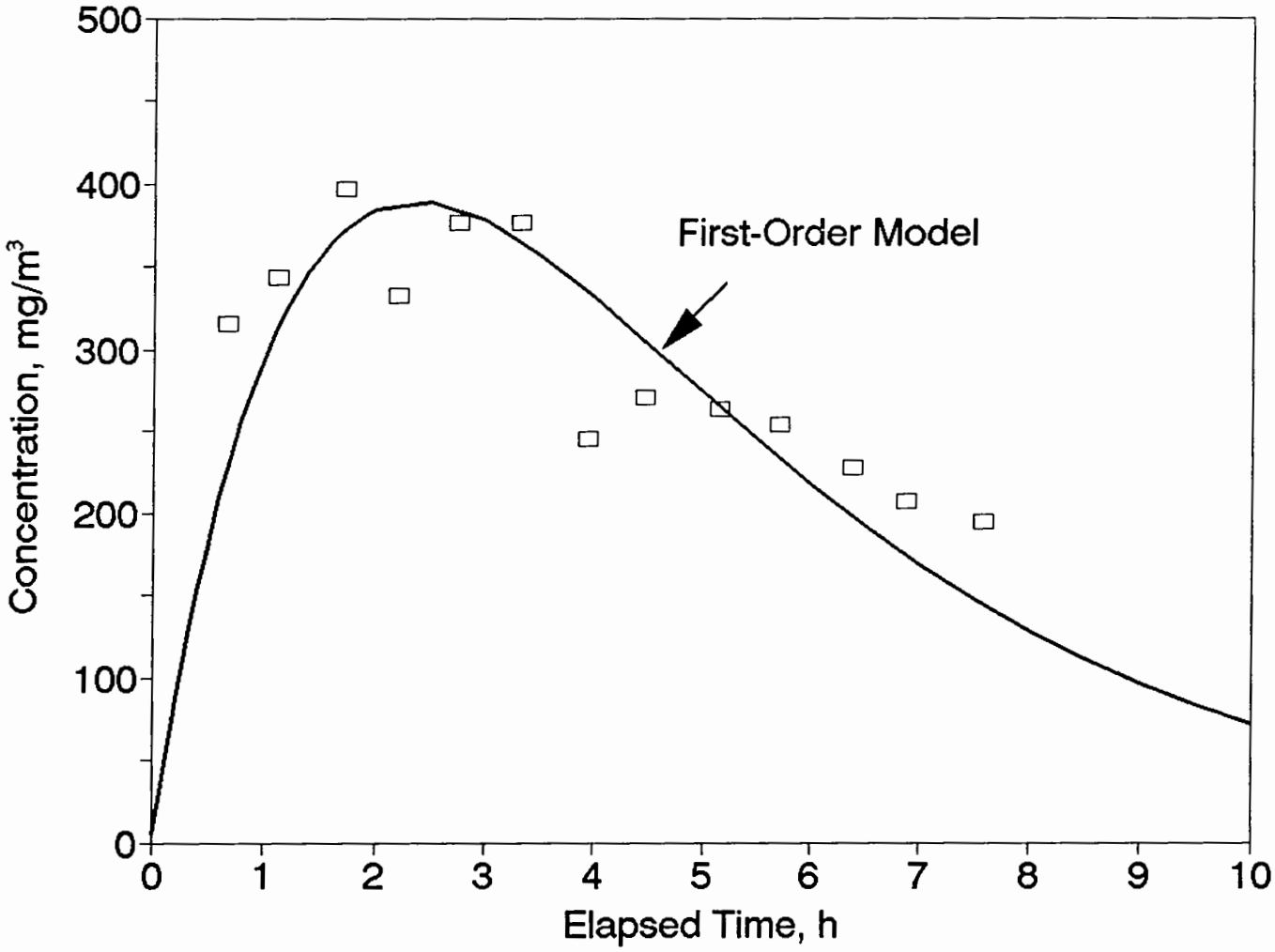

Figure 6. Modeling the re-emission of ethylbenzene from polluted duct liner - 10 hour data.

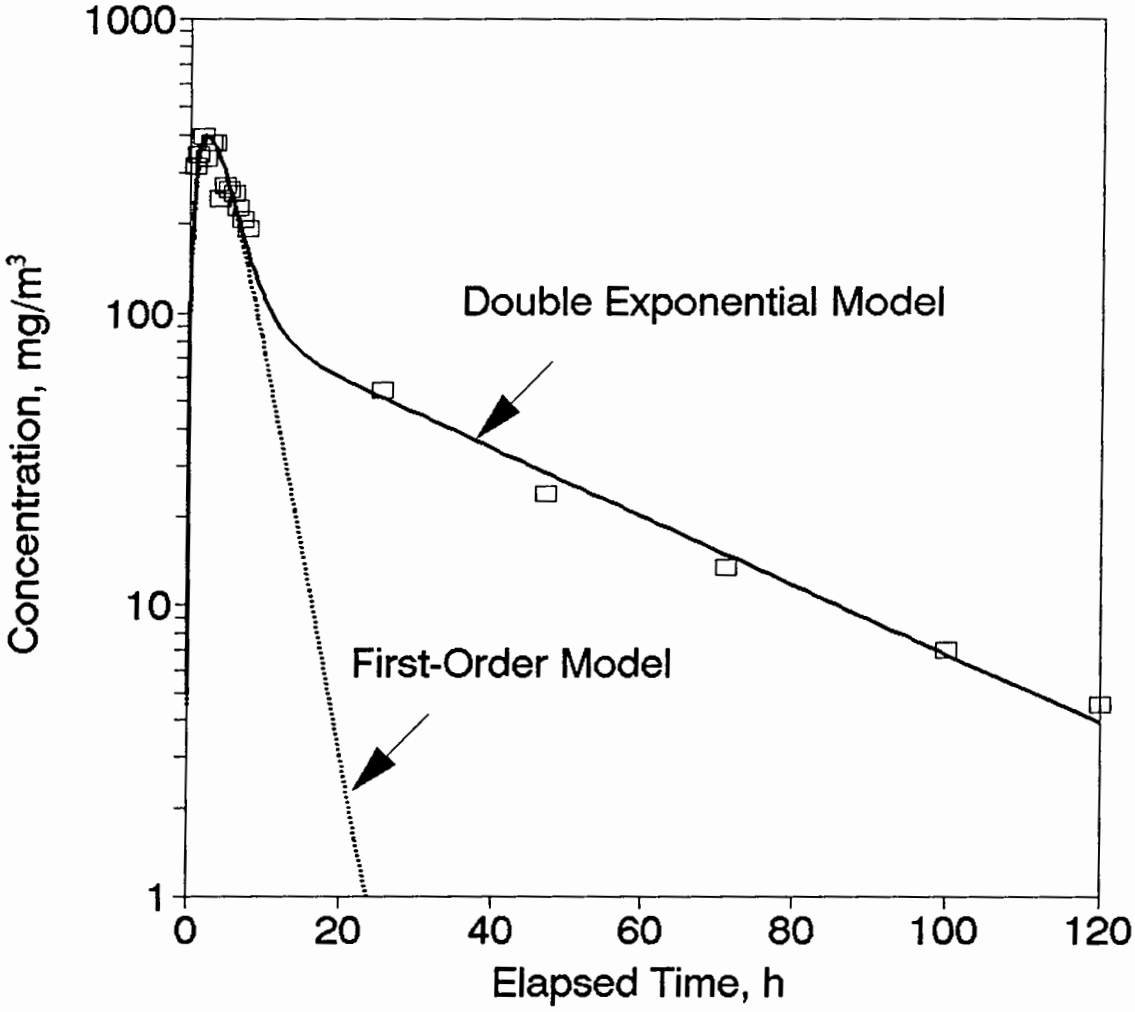

Figure 7. Modeling the re-emission of ethylbenzene from polluted duct liner - 120 hour data.

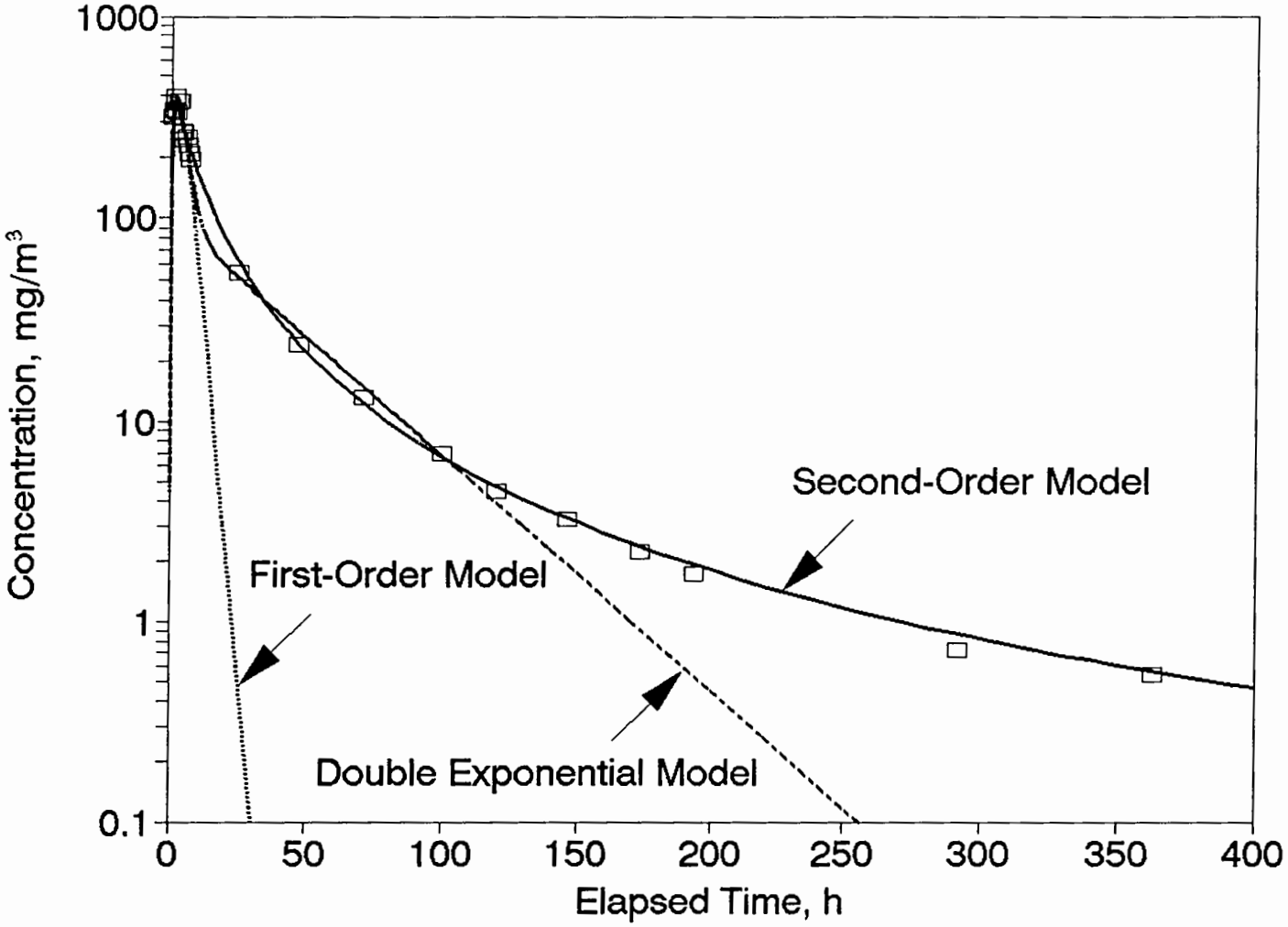

Figure 8. Modeling the re-emission of ethylbenzene from polluted duct liner - 400 hour data.

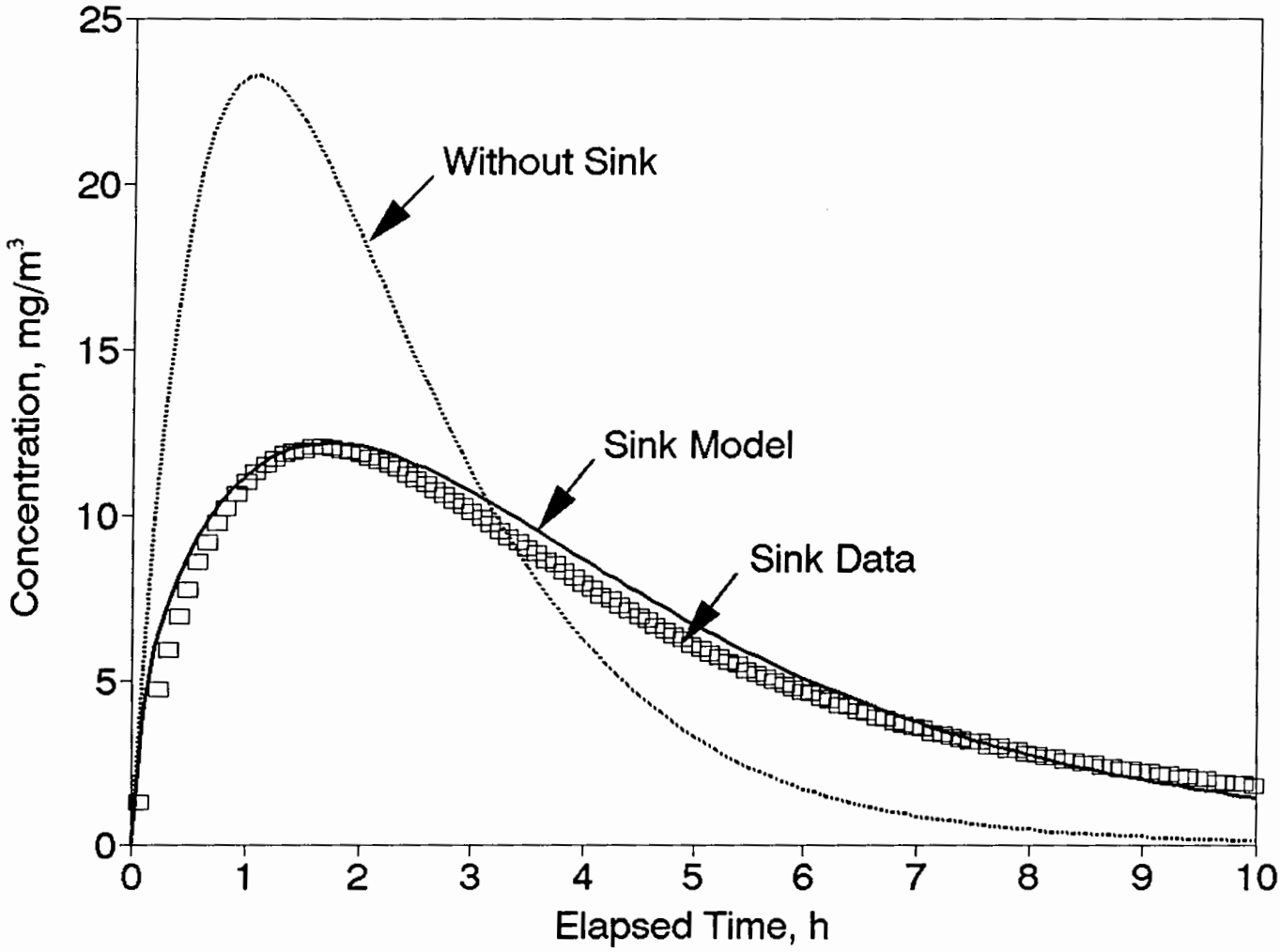

Figure 9. Dynamic sink test and model prediction.

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16. ABSTRACT The paper discusses solutions for problems relating to validating indoor air quality (IAQ) source and sink models. While model validation remains the weakest part of the entire process of IAQ model development, special problems have made the validation of indoor source and sink models even more difficult. Many source and sink models have been developed, but few have been properly validated. Major problems include: elusive model parameters, confusion in parameter estimation methods, uncertainty in scale-up and misleading scaling factors, unspecified validity ranges, and weakness in quantitative comparisons between models and experimental observation. Possible solutions include: proper definition of validation scope, proper use of statistical comparison methods, development of mass transfer indices to bridge the gap between test chambers and real buildings, and development of a cooperative effort to build a source and sink database to facilitate validation.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Pollution	Pollution Control	13B
Mathematical Models	Stationary Sources	12A
Proving	Indoor Air Quality	
Sources	Sinks	14G

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