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Nonlinear Least-Squares Minimization Applied to Tracer Gas Decay for Determining Airflow Rates in a Two-Zone Building

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Abstract We developed a method based on tracer gas decay measurements to quantify the airflow rates, including the interzonal airflows, in a two-zone building: different tracer gases were simultaneously pulse-injected into each of the two zones and the evolution of the gas concentrations in each zone was measured; theoretical concentration profiles obtained by solving dynamic material-balance equations for two coupled, well-mixed zones were fit to the experimental data using nonlinear least-squares minimization; and estimates of the airflow rates were iteratively refined until a best fit was achieved between the model and the data. We conducted experiments validating the method in two full-sized rooms of a test house. Airflows were controlled using blowers, and mixing was ensured by the use of fans. Airflow rates inferred by the tracer gas technique agreed with imposed airflow rates within an average absolute error of 8%. Results are also reported for two experiments conducted in the same structure under uncontrolled conditions. Goodness-of-fit tests revealed no statistically significant differences between measured tracer gas concentrations and theoretical concentration profiles constructed using the least-squares parameter estimates.

Key words Air-exchange; Minimization; Modeling; Tracer gas; Ventilation; Interzonal airflow rates

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Introduction

A single, well-mixed reactor is the most common representation of indoor environments for analysis and modeling of air pollutant concentrations. This representation, in which pollutant concentrations are assumed to be uniform throughout, yields simple equations that describe the evolution of pollutant concentrations. Indoor environments, however, are not always well mixed. Several approaches have been suggested for predicting indoor concentrations when mixing is incomplete, including ventilation efficiency and age-of-air concepts (Skäret and Mathisen, 1982; Breum, 1993), and the application of computational fluid dynamics (Awbi, 1991; Chen et al., 1992). In another common approach, the indoor environment is represented as two or more idealized reactors, each independently well mixed (Rodgers, 1980; Özkaynak et al., 1982; Nazaroff and Cass, 1986; Ryan et al., 1988). Pollutant concentrations in each reactor are coupled by airflows between them.

Practical application of multizone models to indoor environments requires quantitative information on airflows between zones. Airflow rates are difficult to measure directly, however, and are usually estimated indirectly using tracer gases. Tracer gas techniques have been used extensively to measure the air infiltration of buildings considered as single zones (Hunt, 1980; Lagus and Persily, 1985; Sherman, 1990a). Tracer gas techniques have also been extended to complex buildings that behave as multizone systems, providing information on the airflows between rooms (Sinden, 1978; Lagus and Persily, 1985; Sherman, 1989). Multizone techniques have not received the same level of investigation and use, however, as single-zone techniques (Sherman, 1990b; AIVC, 1991).

There are three common classes of tracer gas techniques: decay, constant injection, and constant concentration. The decay method is a transient technique in which a tracer gas is released as a pulse into a zone, allowed to mix within the zone to establish an initial uniform concentration, and monitored as the concentration evolves. The constant concentration method is a steady-state technique in which the tracer gas injection rate is continuously adjusted to maintain constant

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concentrations within the zone. In the constant injection technique, tracer gas is injected at a constant rate and the time-varying concentration is measured. For detailed discussions of these techniques, see Lagus and Persily (1985), Sherman (1990b), AIVC (1991), or ASHRAE (1993).

Each of the above techniques can be applied to multizone structures. In addition, multizone measurements can be made using single or multiple tracer gases. Single-tracer techniques used in a multizone structure require sequential injections: tracer gas is released into one zone and the concentration is measured in all zones; this procedure is repeated for each of the other zones, one at a time (Afonso et al., 1986). Multiple tracer gas experiments are conducted by introducing many different tracer gases simultaneously, one into each zone of the building, and the concentration evolution of each tracer is measured in each zone. Multiple-tracer methods usually require more instrumentation than single-tracer methods, since more than one type of gas is injected and analyzed, but require less experimental time since only one measurement period is needed. Single-tracer methods are typically more sensitive to temporal variations in the airflow rates (Sherman, 1990b; AIVC, 1991).

Four approaches have been described for deriving airflow rates from multiple tracer gas decay data: the eigenvalue, differential, integral, and system identification approaches. In eigenvalue analysis, the system of material-balance equations is solved analytically, then linear regression is used over discrete sections of the experimental concentration profiles to estimate model parameters (Sinden, 1978; Hernandez and Ring, 1982; Irwin and Edwards, 1990; Heidt et al., 1991). The differential approach is implemented by directly estimating quantities in the differential material balance with experimentally measured tracer gas concentrations (Irwin and Edwards, 1990; Enai et al., 1993). In the integral approach, the experimental data are numerically integrated over a specified time interval. Airflow rates are inferred from this integral value and measured concentration values at the start and end points of that time interval (Axley and Persily, 1988; Heidt et al., 1991; Irwin and Edwards, 1990; Enai et al., 1993). In the system identification approach, the material-balance equations are regarded as state equations and the unknown airflow rates are coefficients in the state equation, and statistical estimation methods are used to obtain the airflow rates (Hedin, 1990; Okuyama, 1990; O'Neill and Crawford, 1990; O'Neill and Crawford, 1991; AIVC, 1991).

We have been investigating the effectiveness of engineering techniques for controlling environmental tobacco smoke (ETS) exposure in multizoned buildings using a combination of modeling and experiments (Miller-Leiden et al., 1993; Miller-Leiden and Nazaroff, 1994; Miller-Leiden and Nazaroff, 1996; Miller, 1996). Briefly, we conducted a suite of experiments in two full-sized interconnected rooms. After smoking a cigarette in one room, particle concentrations were measured over time in each room. A multiple tracer gas decay method was used to characterize the airflow rates in the two rooms during each experiment. The decay method was chosen over alternatives because it imitated the release and decay of ETS particles due to smoking a cigarette. It also did not require equipment other than a gas chromatograph and it did not require the control or measurement of gas injection rates. Two tracer gases were used rather than one because the interzonal airflows were buoyancy-driven in some experiments and may have varied significantly over the course of the experiment.

Initial application of the eigenvalue, differential, and integral approaches to extract airflow rates from our experimental tracer gas data did not yield realistic airflow rates. Other investigators have reported similar problems: negative airflow rates were obtained, noise in the data prevented accurate determination of airflows, and the approach sometimes required trial and error-type analyses (Hernandez and Ring, 1982; Prior and Littler, 1986; Afonso et al., 1986; Heidt et al., 1991; Enai et al., 1993). These problems stem from a number of factors, including measurement error, incomplete mixing of tracer gas, and too few measurements during the early time period.

On the basis of this experience, we decided to use the system identification approach, expecting that this method would be effective over a wide range of experimental conditions, even with data of varying quality. Although system identification techniques have existed for several years, their application to field data has not been widely reported. Hedin (1990) developed a method using quadratic programming to interpret data from single tracer gas injected into multiple zones, and illustrated its use with numerical simulations. O'Neill and Crawford (1990) presented a recursive least-squares algorithm for use with a single tracer gas in multiple zones and reported simulation results. Other researchers applied the least-squares approach to interpret measurements: Honma (1975), O'Neill and Crawford (1991), and Okuyama (1990), for a single tracer gas in multiple zones. The use of a multiple tracer gas system for determining airflows between multiple zones was demonstrated in Prior et al. (1985) and Prior and Littler (1986); their calculation of airflow rates involved matrix analysis, based on system identification theory.

In this paper, we describe the system identification technique of nonlinear least-squares minimization that we developed for analysis of two-tracer, two-zone decay experiments. This method has several advantages when compared to published methods for interpreting tracer gas data; in particular, it is not as sensitive to measurement error and is constrained against generating negative airflow rates. In addition to describing the method, we summarize the experiments we used to validate the accuracy of this method, and illustrate its application in experiments in which unknown airflow rates were to be determined.

Model of a Two-Zone Building

Figure 1 illustrates the essential features of a two-zone model. Each zone is considered well-mixed and communicates by airflow with the other zone and with the outside air. The airflow rates are assumed to be time invariant, and the indoor air pressure is assumed to be constant in each zone. Furthermore, isothermal conditions are assumed to prevail so that volumetric flows of air are balanced in each zone. The fractional error associated with applying this formulation in the case of non-isothermal conditions is approximately $\Delta T/T$, where ΔT is the temperature difference between two zones and T is the indoor air temperature, in K. For the present application, $\Delta T \leq 10K$, yielding a model formulation error of $\leq 3\%$. If a larger ΔT is expected, material-balance equations for non-isothermal conditions should be used (Roulet and Compagnon, 1989).

Discrete quantities of tracer gas are separately and simultaneously introduced into zone 1 (tracer S) and zone 2 (tracer R) at time t=0. Each tracer is assumed to be instantaneously and uniformly mixed within that zone upon injection and to remain well mixed for all subsequent times. The tracer gases are assumed to be conservative; that is, they do not react chemically within the space and are removed only by airflow out of the zones. It is also assumed that there are no tracer gases in the indoor air prior to injection, nor in the outdoor air. For time t>0, the tracer gas concentrations as a function of time can be described by four differential equations based on the principle of material conservation. Denoting the volumetric airflow rate from zone i to zone j by F_{ij} ($m^3\ h^{-1}$), where zone 0 is outdoors, the material-balance equations take the following form:

$$\frac{dC_{S1}(t)}{dt} = \frac{1}{V_1}[F_{21}C_{S2}(t)-(F_{12}+F_{10})C_{S1}(t)] \qquad (1)$$

$$\frac{dC_{S2}(t)}{dt} = \frac{1}{V_2}[F_{12}C_{S1}(t)-(F_{21}+F_{20})C_{S2}(t)] \qquad (2)$$

$$\frac{dC_{R1}(t)}{dt} = \frac{1}{V_1}[F_{21}C_{R2}(t)-(F_{12}+F_{10})C_{R1}(t)] \qquad (3)$$

$$\frac{dC_{R2}(t)}{dt} = \frac{1}{V_2}[F_{12}C_{R1}(t)-(F_{21}+F_{20})C_{R2}(t)] \qquad (4)$$

These equations are solved subject to the following initial conditions:

$$\text{at } t=0,\ C_{R1}(t)=0,\ C_{R2}(t)=C_{R0} \qquad (5)$$

$$\text{at } t=0,\ C_{S1}(t)=C_{S0},\ C_{S2}(t)=0 \qquad (6)$$

The symbols have the following definitions:

$C_{S1}(t)$ $C_{S2}(t)$ = mole fraction (ppb_v) of tracer gas S in zones 1, 2 at time t (h)

$C_{R1}(t)$, $C_{R2}(t)$ = mole fraction (ppb_v) of tracer gas R in zones 1, 2 at time t (h)

C_{S0}, C_{R0} = initial mole fraction (ppb_v) of tracer gas S in zone 1, tracer gas R in zone 2

V_1, V_2 = volume of zones 1, 2 (m^3)

Two material-balance equations can also be written on airflow rates:

$$F_{21}+F_{01}=F_{12}+F_{10} \qquad (7)$$

$$F_{12}+F_{02}=F_{21}+F_{20} \qquad (8)$$

The system of linear first-order ordinary differential equations (1) through (4) can be solved analytically using eigenvalues (Sinden, 1978; Luenberger, 1979). Let $a = \frac{F_{12}+F_{10}}{V_1}$, $b = \frac{F_{21}}{V_1}$, $c = \frac{F_{12}}{V_2}$, and $d = \frac{F_{21}+F_{20}}{V_2}$.

The system eigenvalues are

$$\lambda_1 = \frac{-(a+d)+\sqrt{d^2-2ad+4bc+a^2}}{2} \text{ and}$$

$$\lambda_2 = \frac{-(a+d)-\sqrt{d^2-2ad+4bc+a^2}}{2}.$$

Given initial conditions (5) and (6), model equations describe the evolution of tracer gas mole fractions as follows:

$$C_{S1}(t) = C_{S0}\left(\frac{1}{\lambda_2-\lambda_1}\right)[(a+\lambda_2)e^{\lambda_1 t}-(a+\lambda_1)e^{\lambda_2 t}] \qquad (9)$$

$$C_{S2}(t) = C_{S0}\left(\frac{c}{\lambda_2-\lambda_1}\right)[(e^{\lambda_2 t}-e^{\lambda_1 t}] \qquad (10)$$

$$C_{R1}(t) = C_{R0}\left(\frac{b}{\lambda_1-\lambda_2}\right)[(e^{\lambda_1 t}-e^{\lambda_2 t}] \qquad (11)$$

$$C_{R2}(t) = C_{R0}\left(\frac{1}{\lambda_1-\lambda_2}\right)[(d+\lambda_1)e^{\lambda_2 t}-(d+\lambda_2)e^{\lambda_1 t}] \qquad (12)$$

Nonlinear Least-Squares Minimization

Overview

The least-squares minimization method operates as follows: given a set of experimental tracer gas data, the mathematical model described by equations (9)–(12) is fit to the data, subject to the constraints that equations (7) and (8) must be satisfied and that all flow rates must be non-negative. More formally, a mathematical model $C(t) = C(t;\vec{a})$ is fit to a set of experimental data points (t_i, C_i), $i = 1,...,N$, to determine M unknown parameters, $\vec{a} = a_1,...,a_M$, in the model. The parameters are systematically and iteratively adjusted so that the output of the model is the best match, in some sense, to the observed data. The least-squares minimization technique uses the chi-square statistic, X^2, to find the set of parameters that are considered to be best-fit values (Everitt, 1987). X^2 measures the deviation of model predictions from the observed data:

$$X^2(\vec{a}) = \sum_{i=1}^{N} \frac{[C_i - C(t_i;\vec{a})}{\sigma_i^2} \tag{13}$$

In equation (13), C_i is the tracer gas concentration measurement at time t_i, σ_i is the standard deviation of the measurement (describes the uncertainty in the measurement), and $C(t_i;\vec{a})$ is the model prediction at time t_i.

Because the modeled tracer gas concentrations depend nonlinearly on the parameters we wish to estimate, a closed-form solution of the best parameter values does not exist. Instead, an iterative minimization procedure is used. Given a set of parameters $\vec{a}^i = a_1^i,...,a_M^i$, successive estimations are made according to the following equation:

$$\vec{a}^{i+1} = \vec{a}^i + \vec{\delta}^{i+1} \tag{14}$$

where $\vec{\delta}^{i+1}$ is the correction vector and $\vec{a}^i$ is the vector of M parameters determined during the ith iteration. The minimization procedure ensures that each iteration generates a new set of parameter values producing a X^2 statistic that is at least as good as the previous value:

$$X^2(\vec{a}^{i+1}) \leq X^2(\vec{a}^i) \tag{15}$$

We used the Levenberg-Marquardt (LM) method for finding a suitable correction vector, one of the more robust estimation methods (Davis, 1993). For a general discussion of these methods, see Everitt (1987), Bates and Watts (1988), Press et al. (1990), or Bevington and Robinson (1992).

Implementation

Our application of nonlinear least-squares minimization to the two-zone, two-tracer gas decay problem required the simultaneous fit of four mathematical equations to four sets of measurement data. Our final objective was to estimate six airflow rates: airflow from each room into the other room, airflow from each room to the outdoors, and airflow from the outdoors into each room (Figure 1). Only four of the six airflow rates were estimated from the tracer gas data: the two interzonal flows and the two flows to the outdoors. The flow rates from outdoors into the two zones were determined using equations (7) and (8). During each iteration, the airflow rate estimates were checked for non-negativity. If one or more of the flows was predicted to be less than zero, the correction vector was decreased by a factor of 2 and the airflow rates were recalculated according to equation (14). This procedure was repeated until all six flows were non-negative before proceeding to the next iteration.

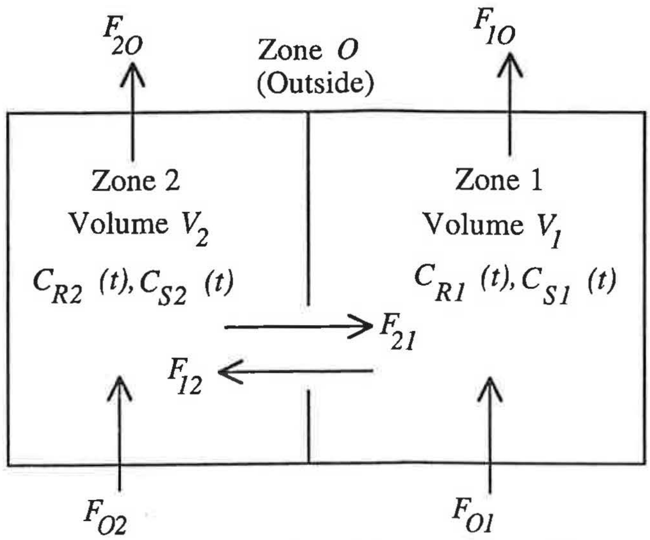

Fig. 1 Schematic representation of the two-zone model. Tracer gases S and R are pulse-injected into zones 1 and 2 respectively

It was determined during preliminary analysis that the minimization was sensitive to the model parameters C_{R0} and C_{S0}. Although these parameters can be independently determined from the quantity of tracer injected and the zone volumes, there is some uncertainty in these values due to losses during injection, inaccuracy in measuring the exact amount injected, inaccuracy in measuring zone volumes, or incomplete mixing of tracer into the zone volume. Consequently, these two parameters were estimated in addition to the airflow rates. Note that it is common, even in single-zone tracer techniques, not to specify the initial tracer gas concentration *a priori* (ASHRAE, 1993). Similarly, other investigators have estimated effective zone volumes (volume in which the tracer is completely mixed) in addition to the airflows (O'Neill and Crawford, 1990).

The X^2 parameter measures the "fit" between the data and the model. To ensure that a best fit was simultaneously achieved for our four sets of tracer gas data, four values of X^2 were calculated, one for each set of data, and summed to obtain an overall X^2. During our experiments, the measured concentrations of tracer gas R were roughly four times larger than the concentrations of tracer gas S. If we computed the overall X^2 as a simple sum of the four X^2 values, it would be more heavily weighted by the fit between the data and the model for tracer gas R than for tracer gas S. To avoid this bias, the data were first normalized to the respective peak measured concentrations.

Initially, our X^2 calculations assumed that all measurements had the same standard deviation. This assumption is appropriate when the true uncertainty associated with the measurements is not known in advance. It is a good assumption when all of the measurement errors are limited to statistical errors; the main implication is that all data values are equally reliable (Bates and Watts, 1988). Eventually, we determined that this assumption was not appropriate in some cases. We reformulated the X^2 calculations in an attempt to better represent the data variation, as discussed in the following section of this paper.

Convergence of the minimization technique was based on two criteria: the relative size of terms in the correction vector as compared with respective previous parameter values, and the relative change in X^2 on successive iterations. After each iteration, X^2 was evaluated to determine if it had decreased. If it did decrease, we checked both the size of the decrease and the size of the correction vector for convergence. More formally, if $\left|\frac{X^2(\vec{a}^i)-X^2(\vec{a}^{i+1})}{X^2(\vec{a}^{i+1})}\right|$ and $\left|\frac{a_j^i-a_j^{i+1}}{a_j^{i+1}}\right|$, for all $j = 1,\ldots,M$, were less than or equal to 10^{-6}, then convergence was assumed to have occurred.

A computer program to implement the minimization technique was written in Fortran-77, using computer routines from Press et al. (1990) to implement the LM method. Double precision was used for all calculations, as recommended by Bates and Watts (1988).

Assessing the Goodness-of-Fit

Once the parameters of the model were estimated, we used several methods to evaluate the agreement between the measured tracer gas concentrations and the predictions of the fitted model. As a qualitative evaluation tool, we examined plots of residuals over time. To quantitatively assess the model, we tested for goodness-of-fit using the Kolmogorov-Smirnov statistic.

Residuals

A residual analysis can assess the degree to which the underlying assumptions of the least-squares minimization are met. In this context, a residual is the difference at a given time between the model prediction and the corresponding measurement. The method of least-squares fitting assumes that the residuals are independent and normally distributed with constant standard deviation. In Equation (13), the assumption of constant standard deviation has been relaxed to obtain what is essentially a weighted least-squares, where σ_i is the weighting factor for data point i. Residuals from a good fit have a uniform, random spread about zero; any systematic pattern indicates problems of non-constant standard deviation (Bates and Watts, 1988), that the underlying model is incorrect, or that the measurements errors are neither normally distributed nor independent (McCuen, 1985; Press et al., 1990). If the "strict" assumption of normally distributed residuals is violated, the least-squares minimization can still be useful for estimating parameters; however, the X^2 can not be interpreted as the actual squared standard errors of the parameter estimates (McCuen, 1985; Press et al., 1990).

Analysis of our validation experiments showed that the residuals were small and spread randomly about zero; thus, we concluded that the assumption of constant data standard deviation was appropriate. However, analysis of the uncontrolled experiments, conducted during our ETS research, revealed residuals that were very large during the first hour and biased either positive or negative. After the first hour, the residuals decreased to smaller values and the bias was reduced. This behavior suggests that there are other sources of variation in the data from the uncontrolled experiments besides statistical measurement error and that the real variance in the data is time-dependent. A potential source of variation is imperfect mixing of tracer gas within each zone.

Because the residual plots indicated non-constant standard deviation, the minimization method was modified to better represent the variation in the data. Following the approach suggested by Bates and Watts (1988), the standard deviation of the data was modeled using a power function: a function of the form αt^{β} was fit to the absolute value of the residuals, where α and β are constants, determined by the fit. The minimization procedure was then repeated, using the square of this power function in equation (13). Using a time-dependent expression for the weighting factor in the X^2 equation gives less weight to the data points measured in the beginning of the experiment and more weight to the data points measured later in the experiment.

Kolmogorov-Smirnov Test

We used goodness-of-fit statistical testing to assess whether the measured data agreed with the underlying model. The Kolmogorov-Smirnov (KS) test is a non-parametric test for determining whether two independent samples could reasonably be supposed to come from the same continuous distribution (McCuen, 1985; Sprent, 1989). The test statistic, D_{KS}, is the maximum absolute deviation between the theoretical and observed cumulative relative frequencies. For our application, we compared the empirical distribution functions from the data points and the model predictions. If the functions did not depart significantly from each other, then we concluded that there was a good fit between the model and the data.

The X^2 goodness-of-fit test is a well-known, frequently used approach for assessing model fit, and it would appear to be an obvious choice for this application since X^2 is an output of the least-squares minimization. However, we believe that it is not a suitable test for this method, for two reasons: (1) residual analysis revealed that the data standard deviation was neither normally distributed nor independent, thus violating the assumptions of the X^2 test; and (2) the X^2 goodness-of-fit test is most appropriately used to compare discrete functions, not continuous functions from which our data derived.

Model Parameter Uncertainties

We estimated the uncertainties in the predicted airflow rates for the uncontrolled experiments using a Monte Carlo method. Starting with the predicted parameters, many sets of "artificial" data were generated using model equations (9)–(12) (Press et al., 1990). These data were corrupted by adding a random error term based on the power function model of the standard deviation; the least-squares minimization was then repeated for each artificial data set. We generated and analyzed more than 200 data sets to derive a probability distribution for each predicted parameter.

Validation Experiments

Method

To validate the least-squares minimization method, we conducted tracer gas decay experiments under controlled conditions at the Indoor Air Quality Research House, located at the University of California's Richmond Field Station. The test facility is a two-story, wood building, within which three rooms have been retrofitted to have small air leakage (Offermann et al., 1985). The validation experiments were carried out in two of these rooms, denoted zone 1 (36 m^3) and zone 2 (31 m^3) (Figure 2). The two rooms are connected by a doorway. An exhaust hood is located in the corner of zone 1. Each room has four small instrument-cooling fans mounted at the center of each wall that can be remotely operated to promote mixing.

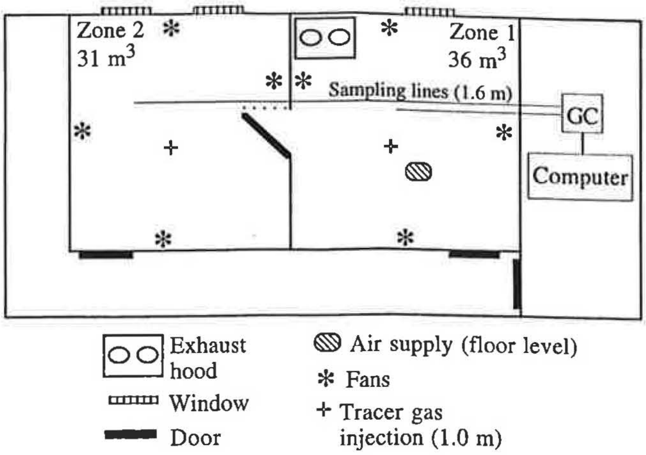

Fig. 2 Configuration of the test rooms used to conduct the two-zone, two-tracer gas decay experiments (plan view). GC=gas chromatograph. Height above the floor is given in parentheses

For the validation experiments, both rooms were tightly sealed (including the connecting door), and all six of the airflows in the two rooms were mechanically controlled. We installed six blowers to provide the airflows: supply and exhaust blowers penetrated the ceiling of each room, and interchamber blowers penetrated the wall connecting the two rooms. The two exhaust blowers discharged air to the outdoors to ensure that tracer gas was not re-entrained into the test rooms. During the experiments, we continuously monitored the airflow through the supply and exhaust blowers for zone 1 with two orifice plate systems.

Power to the blowers was provided by variable transformers. We used an orifice plate method to determine the appropriate settings on the transformers to achieve desired airflow rates. A calibration system was configured with each blower in turn discharging into a large, airtight plenum from which air was extracted using a separate fan. The fan discharged to a 20-m straight pipe which contained an orifice plate. Airflow generated by the fan was adjusted to the desired target value, as measured by the pressure drop across the orifice plate. Then the blower airflow rate was adjusted to yield atmospheric pressure in the plenum; at this point, the fan and the blower airflows were assumed to be equal.

After setting the blower transformers by this method, the blower airflow rates were accurately measured using a tracer gas technique (Drescher et al., 1995). Compressed air containing 109 ppm$_v$ of SF_6 was

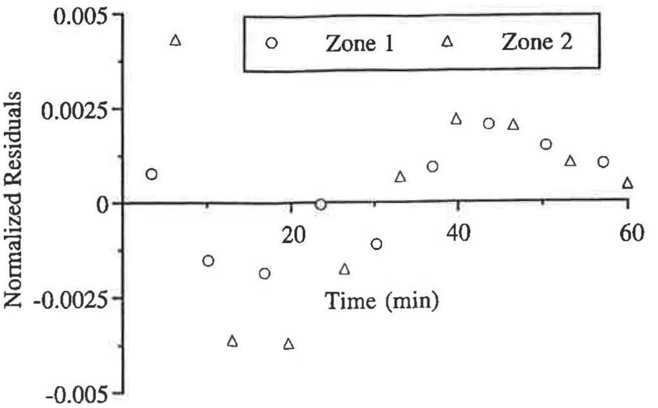

Fig. 3 R13B1 concentration residuals for the validation experiment, scenario A (run 1). The residuals are calculated as the difference between measurements and model predictions from the minimization with constant data standard deviation. The magnitude of the residuals indicates the size of the deviation relative to the peak normalized concentration: a residual of 0.005 indicates a 0.5% deviation

injected through a mass-flow controller (Matheson, Model 8274) into the inlet of the blower. The steady-state concentration of SF_6 was measured at the outlet of the blower with a gas chromatograph (GC) equipped with an electron capture detector (ECD) (Lagus Applied Technology). The airflow rate was determined by applying a material balance on SF_6.

We conducted duplicate experiments for each of the following airflow scenarios: in scenario A, flow rates were relatively high from outdoors into zones 1 and 2, from zone 2 to zone 1, and from zone 1 to outdoors; in scenario B, the flow rates were relatively high between zone 1 and outdoors and between the zones. In each experiment, wall fans plus a 0.3-m diameter oscillating fan were operated continuously in each room to promote mixing. Prior to the start of each experiment, the blowers were off. A small volume of tracer gas was released in the center of each zone by injection through norprene tubing: 37 cm^3 of 17.6% SF_6 (in helium) into zone 1 and 15 cm^3 of R13B1 ($CBrF_3$) into zone 2. The tracer gases were mixed for seven minutes, then the blowers were turned on. Tracer gas concentrations were monitored continuously for one hour using a GC with an ECD (Hewlett Packard, Model 5890). Air was sampled at 7-minute intervals through norprene tubing from a position at the center of the room, 1.6 m above the floor.

Results

We applied the least-squares minimization method to the validation data using measurements taken after the blowers were turned on. In the minimization procedure, we assumed that the data standard deviation was constant. The residuals were small and had no systematic bias, as shown in Figure 3, thus modifications in the minimization procedure were not required.

Figures 4 and 5 compare model predictions with measurements using both the measured blower airflow rates and the airflow rates estimated from least-squares minimization. It can be seen that a very good fit was obtained. Results of the KS goodness-of-fit tests, presented in Table 1, show that there are no statistically significant differences (significance level: 0.05) between observed and modeled values.

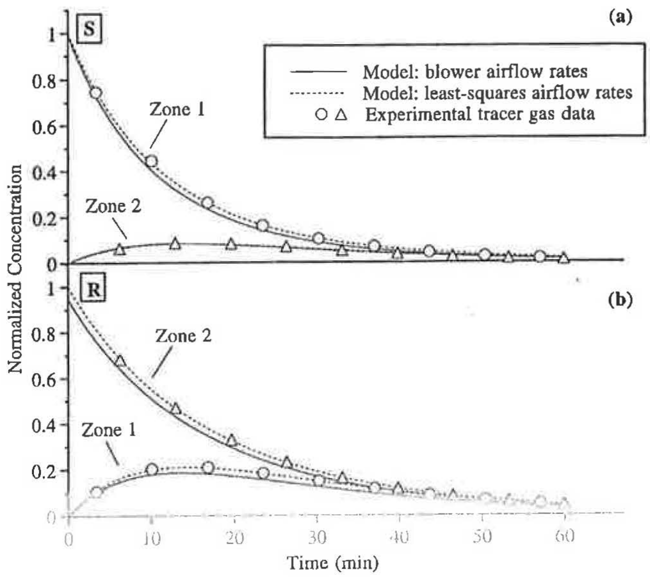

Fig. 4 Comparison between model predictions and tracer gas measurements for the validation experiment, scenario A: (a) SF_6 concentrations (denoted by S) and (b) R13B1 concentrations (denoted by R). Model predictions are separately plotted for the least-squares and measured blower airflow rates

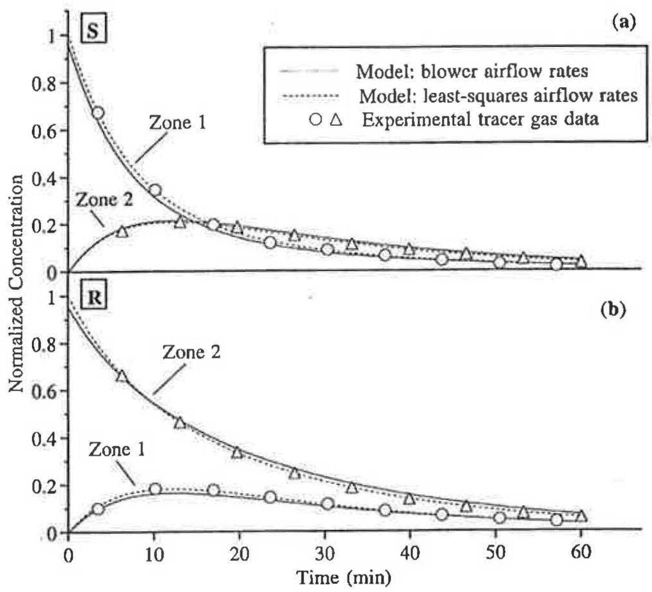

Fig. 5 Same as Figure 4, for the validation experiment, scenario B

Table 1 Results of goodness-of-fit analysis for all experiments

Experiment	Zone	Tracer	D_{KS}[a]
Validation, Scenario A[b]	1	*S*	0.11
υ^{c}=29		*R*	0.11
	2	*S*	0.22
		R	0.11
Validation, Scenario B[b]	1	*S*	0.10
υ=29		*R*	0.10
	2	*S*	0.10
		R	0.10
INFILT	1	*S*	0.14
υ=101		*R*	0.21
	2	*S*	0.24
		R	0.14
VENT	1	*S*	0.07
υ=101		*R*	0.11
	2	*S*	0.07
		R	0.07

[a] At 0.05 significance level, D_{crit}=0.41 for scenario A and B, and D_{crit}=0.24 for INFILT and VENT; if $D_{KS}<D_{crit}$, agreement between measured and modeled data is good
[b] Analyses for run 1 data only
[c] Degrees of freedom, $\upsilon=N-M-1$, where N=total number of data points used in minimization, M=number of estimated parameters

Table 2 Results of least-squares minimization analysis for the validation experiments

Parameters	Scenario A			Scenario B		
	Meas-ured[a]	Least-squares[b] run 1	Least-squares[b] run 2	Meas-ured[a]	Least-squares[b] run 1	Least-squares[b] run 2
F_{21} (m^3 h^{-1})	89	90	90	89	94	96
F_{12} (m^3 h^{-1})	33	30	32	100	92	94
F_{10} (m^3 h^{-1})	167	156	154	167	161	162
F_{20} (m^3 h^{-1})	32	30	25	32	34	39
F_{01} (m^3 h^{-1})	101	97	96	165	159	160
F_{02} (m^3 h^{-1})	91	89	83	31	36	41
C_{S_0} (ppb$_v$)	166±3	167	169	167±2	172	175
C_{R_0} (ppb$_v$)	444±3	463	475	434±1	457	455

[a] Airflow rates as supplied by blowers; initial tracer gas concentrations are mean±standard deviation of GC measurements in the test chamber prior to turning on the blowers for runs 1 and 2
[b] Two tracer gas experiments were run consecutively; both sets of data were separately analyzed using the least-squares minimization method

Table 2 compares the imposed blower airflow rates with the rates inferred using nonlinear least-squares minimization. The agreement is very good: the mean absolute relative error for individual flow rates is 8%. The four cases in which errors are greater than 10% correspond to low airflow rates; on a volume flow rate basis, the errors for low airflow rates are similar to the overall absolute average error of 5 m^3 h^{-1}. Table 2 also compares the amount of tracer gas injected during the experiments, as measured by the GC after injecting tracer and allowing it to mix for 7 minutes, with the prediction using least-squares minimization. Again, the agreement is good, with average and maximum deviations of 4% and 7%.

Under ideal conditions, the blower airflow rates will satisfy equations (7) and (8). In these experiments, the material balances based on the measured airflow rates are not satisfied: the errors are 3–5% for scenario A and 5–8% for scenario B. There are two main sources of error that could account for uncertainty in the blower airflow rates. First, in setting the rates we found that slight changes in the curvature of the ducting attached to a blower outlet could result in a flow rate change of up to 8%. In addition, we observed that while F_{01} was steady during the experiments (continuous monitoring during the experiments showed fluctuations of <1%), F_{10} fluctuated by 7%. The higher fluctuations are thought to be due, at least in part, to exhausting the air from this blower into a time-varying outdoor wind. Similar behavior is expected for blower flow F_{20} since it was also exhausted to the outdoors. Due to these factors, we expect the blower flow rates were accurate to within about 8%.

Experimental Application

Methods

As part of our larger study on ETS-particle exposures, two-zone, two-tracer gas decay experiments were conducted at the Indoor Air Quality Research House. The experimental configuration was similar to the controlled experiments described above, except that the connecting door between zones 1 and 2 was not sealed, mixing fans were not operated after an initial tracer

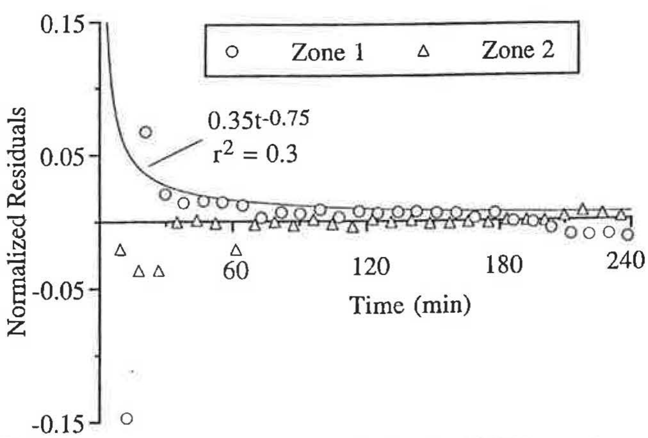

Fig. 6 R13B1 concentration residuals for the VENT experiment. The residuals are calculated as the difference between measurements and model predictions from the minimization with constant data standard deviation. The power function fit to the absolute value of residuals for both zones is represented by the solid line. The magnitude of the residuals indicates the size of the deviation relative to the peak normalized concentration: a residual of 0.15 indicates a 15% deviation

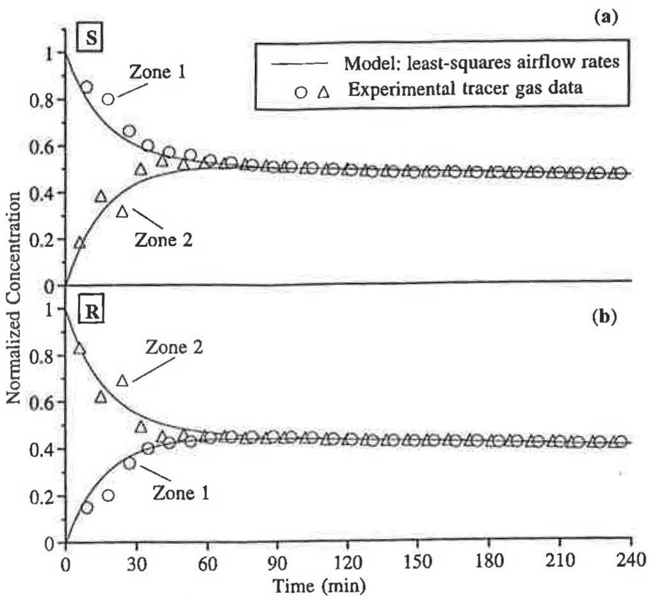

Fig. 7 Comparison between model predictions and measurements for the INFILT experiment: (a) SF_6 concentrations (denoted by S) and (b) R13B1 concentrations (denoted by R). Mopdel predictions are plotted using parameters estimated by the least-squares minimization employing a power function for data standard deviation

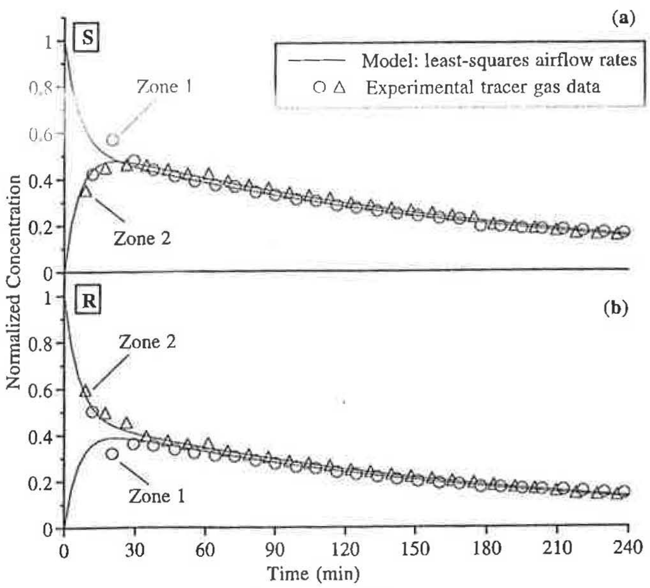

Fig. 8 Same as Figure 7, for the VENT experiment

dispersion period, and airflow rates were substantially uncontrolled.

Prior to the beginning of each experiment, the door connecting zones 1 and 2 was closed and the wall fans were operated for 5–10 minutes to promote air mixing. Then, a small volume of tracer gas was released in each of the zones by injection through a copper tube: 25–30 cm^3 of 17.6% SF_6 (in helium) into zone 1 and 10–15 cm^3 of R13B1 into zone 2. The wall fans were operated for an additional 5–10 minutes to thoroughly mix the tracer gas. The fans were then turned off and the door connecting the two zones was remotely opened. Tracer gas concentrations were monitored for four hours using a GC with an ECD. Room air was drawn at 9-minute intervals through norprene sample lines positioned in the center of the room, 1.6 m above the floor.

We conducted a total of six experiments under various ventilation conditions (both natural and mechanical). Two illustrative experiments are presented here. One (denoted INFILT) was conducted under low infiltration conditions. The second (denoted VENT) was conducted with increased local ventilation supplied through a high-efficiency-particulate-air (HEPA) filter from outside to zone 1 at floor level through 0.1-m ducting. The supply airflow rate was approximately 20 $m^3\ h^{-1}$, as determined with an orifice plate. During VENT, the exhaust hood duct located in zone 1 was opened to allow excess air to flow outside.

Results

We applied the least-squares minimization to the INFILT and VENT experiments assuming that the data standard deviation was constant. Analysis of the residuals, however, revealed large non-random values during the first hour of data (Figure 6). Consequently, we modified the minimization to incorporate a non-constant data standard deviation: we fit power functions to the residuals in the SF_6 and R13B1 data to construct an estimate of the data standard deviation as a function of time. Figure 6 shows the residuals and the power function model for R13B1 in the two zones of the VENT experiment. We carried out the modified minimization using the square of the power functions for the weighting factor in equation (13). The absolute relative difference in estimated parameters from the minimization using constant standard deviations versus using the power function model ranged from 1–25%, with a mean difference of 4% for the INFILT experiment and 10% for the VENT experiment.

Table 3 Results of least-squares minimization analysis for INFILT and VENT experiments[a]

Parameters	INFILT	VENT
F_{21} ($m^3\ h^{-1}$)	60±2	154±17
F_{12} ($m^3\ h^{-1}$)	59±2	163±18
F_{10} ($m^3\ h^{-1}$)	2.4±1.3	10±5
F_{20} ($m^3\ h^{-1}$)	0.001±0.001	11±5
F_{01} ($m^3\ h^{-1}$)	1.6±0.2	19±1.4
F_{02} ($m^3\ h^{-1}$)	0.8±0.2	2±1.3
C_{S_0} (ppb_v)	101±1	129±2
C_{R_0} (ppb_v)	428±6	365±8

[a] The power function model was used to represent the data standard deviation; parameters are best estimate±standard deviation as determined by Monte Carlo simulation

From plots of the tracer gas data, overlaid with the model-predicted concentration profiles, it can be seen that good fits were obtained (Figures 7 and 8). The KS tests for goodness-of-fit indicated that there were no statistically significant (significance level: 0.05) differences between observed and modeled values (see Table 1).

The estimated airflow rates and their standard deviations, determined by Monte Carlo simulation, are summarized in Table 3. The interzonal flow rates have low fractional uncertainty under both experimental conditions, suggesting that these flows are a strong determinant of the behavior of tracer gases in the system.

Discussion

We have developed a nonlinear least-squares minimization method to estimate interzonal airflow rates in a two-zone building from two-tracer gas decay experiments. One attractive feature of this method is that it uses information from all of the measurement data to determine the airflow rates. In validation experiments, we found that the airflow rates could be inferred with a mean accuracy of $\pm 8\%$.

When applying the nonlinear least-squares minimization technique in practice, accuracy may be poorer than in these controlled experiments due to the many sources of error associated with multizone tracer gas measurements. Statistical fluctuations due to measurement error can be averaged out with enough data, or predicted based on previous experience. Incomplete mixing of tracer gas into the zones can be a major source of error, as much as 12–18% (Sandberg, 1987). Residual analysis for the uncontrolled experiments suggested that the mixing time for our system was roughly one hour. The residuals were large during the first hour of the experiments (Figure 6); after the first hour, the values were comparable to our well-mixed validation experiments. Previous studies on the mixing rate of pollutants in a single zone, conducted in zone 1 of the same research house, showed that the mixing time for quiescent conditions was of the order of one hour and that factors such as forced airflow and internal heating could reduce the mixing time by an order of magnitude (Baughman et al., 1994; Drescher et al., 1995). Although mixing patterns in multizone enclosures include mixing between zones in addition to mixing within zones, these mixing time measurements support our inference that incomplete mixing contributed most significantly to the error in the data from the uncontrolled experiments.

To mitigate the effects of mixing error, we modeled the data standard deviation with a power function. Among the four sets of data that we analyzed (two experiments, two tracers), the exponential parameter in the power function varied over a narrow range: $0.5<\beta<0.8$. This finding again supports the notion that despite differing experimental conditions, the main source of data error was similar among uncontrolled experiments and most likely due to incomplete mixing.

In addition to incomplete mixing, there are several other issues to consider. The sampling of tracer gas should be frequent enough that there are ample data for model fitting, particularly during the early phase when concentration changes are most rapid. The mechanics of solving the system of mathematical equations used in this method (equations (9)–(12)) may prove difficult. In some cases, the system that must be solved to determine the theoretical airflow profiles will be ill-conditioned; that is, some of the elements of the system matrix are so close to zero that inversion results in numbers approaching infinity. This problem can arise when the airflow rates between zones are large. Although the interzonal airflow rates for our VENT experiment were large, we did not encounter this problem. A very powerful set of techniques, known as singular value decomposition, can be applied to such systems (Press et al., 1990). Previous researchers have alternatively solved this problem by collapsing zones with large interzonal flows into a single zone (Nagda et al., 1995).

The temporal stability of airflows is another important factor. Researchers have suggested that multiple tracer gas methods accommodate changing interzonal airflows better than methods using single tracer gases (Sherman, 1990b; AIVC, 1991). The method of O'Neill and Crawford (1990) is able to track flow rate parameters in a system that vary slowly with time. The method we describe in this paper is only applicable to data collected during periods with relatively constant air flows due to the formulation of the well-mixed two-zone building model. Applying the minimization technique to data gathered from a system with time-varying flows may result in airflow rate predictions that yield the best overall fit to the data but do not necessarily represent the true mean values. Sandberg (1987) explored the accuracy of tracer gas techniques for predicting the mean flows in a time-varying system. He concluded that errors caused by varying flow rates could be disregarded compared to other errors, provided that there were no oscillations with a time period greater than one-third of the mean nominal time constant (inverse of the air-exchange rate). In addition, experimental measurements in Sandberg's study showed that most oscillations in a naturally ventilated multizone house were of a high frequency nature (on the order of minutes) and would therefore not affect accuracy. Applying these results to our work, oscillations with a period of greater than 60 minutes and 9 hours, respectively, would have affected the accuracy of our VENT and INFILT experiments. The impact of time-varying flow rates on tracer gas techniques merits future exploration.

It is feasible to extend the minimization method described in this paper to determine airflow rates in buildings with more than two zones. A mathematical description of the tracer gas concentration profiles in the multiple zones must be written similar to equations (9)–(12). For each additional zone, an additional tracer gas is needed resulting in n^2 equations for an n-zone building. Once this set of equations is written, they must be solved either numerically or analytically. If an analytical solution is written, then the minimization scheme described in this paper can be used directly. If the equations must be solved numerically, the minimization method would require small changes: the governing equations will need to be solved with each iteration that generates a new set of parameter values. More rigorously, given a set of tracer gas data, where C_{ij} is the datum associated with t_i for zone j, and an initial set of parameters $\vec{a}^0$, the coupled system of differential equations is numerically solved for the values $C_j(t_i,\vec{a}^0)$. A X^2 is computed by equation (13). Successive parameter estimations are made according to the LM method. When the parameter estimation results in a new X^2 that is less than the previous value, the parameter estimates are updated to their new, more optimal values. At this point, the differential equations must be solved again for the values $C_j(t_i,\vec{a}^k)$ using the new parameters $\vec{a}^k$. Iterations are repeated until convergence is reached. Given present computational capabilities, coupling a numerical differential equation solver with the minimization procedure is a relatively straightforward exercise.

In summary, least-squares nonlinear minimization is an effective procedure for interpreting data from multizone tracer gas decay. Our approach to the specific two-zone, two-tracer problem, relative to the eigenvalue, integral, and differential methods, produced a more robust tool for studying ventilation and indoor air quality in cases in which a well-mixed single reactor model of the indoor environment fails and estimates of interzonal airflows are needed. An improved understanding of multizonal airflow is important because of the errors inherent in using the conventional well-mixed single reactor model in nonideal situations.

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