Evaluating Sources of Indoor Air Pollution

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Evaluation of indoor air pollution problems requires an understanding of the relationship between sources, air movement, and outdoor air exchange. Research is underway to investigate these relationships. A three-phase program is being implemented: 1) Environmental chambers are used to provide source emission factors for specific indoor pollutants; 2) An IAQ (Indoor Air Quality) model has been developed to calculate indoor pollutant concentrations based on chamber emissions data and the air exchange and air movement within the indoor environment; and 3) An IAQ test house is used to conduct experiments to evaluate the model results. Examples are provided to show how this coordinated approach can be used to evaluate specific sources of indoor air pollution. Two sources are examined: 1) para-dichlorobenzene emissions from solid moth repellant; and 2) particle emissions from unvented kerosene heaters.

The evaluation process for both sources followed the three-phase approach discussed above. Para-dichlorobenzene emission factors were determined by small chamber testing at EPA's Air and Energy Engineering Research Laboratory. Particle emission factors for the kerosene heaters were developed in large chambers at the J. B. Pierce Foundation Laboratory. Both sources were subsequently evaluated in EPA's IAQ test house. The IAQ model predictions showed good agreement with the test house measurements when appropriate values were provided for source emissions, outside air exchange, in-house air movement, and deposition on "sink" surfaces.

Evaluation of indoor air pollution problems requires an understanding of several factors, including:

- the source of the indoor pollutants;
- emission characteristics of the source;
- air exchange between the building and the outdoors;
- air movement within the building;
- interaction of the pollutant with surfaces within the building (i.e., sink effects); and
- chemical or physical interactions affecting the pollutant concentration.

Knowledge of these factors would allow investigators of "sick buildings" to perform a systematic assessment of possible sources within the building and to determine their potential for causing the levels of pollutants measured within the building.

A three-phase program, illustrated by Figure 1, is being implemented by scientists and engineers in the Indoor Air Branch of EPA's Air and Energy Engineering Research Laboratory in Research Triangle Park, North Carolina:

1) Environmental chambers are used to provide source emission factors for specific indoor pollutants;

2) An IAQ (Indoor Air Quality) model has been developed to calculate indoor pollutant concentrations based on chamber emission data, air movement within the indoor environment, and outdoor air exchange; and

3) An IAQ test house is used to conduct experiments to evaluate the model results.

This paper shows how this coordinated approach can be used to evaluate specific sources of indoor air pollution. Pollutants from two sources are examined to illustrate the approach: 1) para-dichlorobenzene emissions from moth crystal cakes; and 2) particulate emissions from unvented kerosene heaters.

IAQ Model

The IAQ model estimates the effect of heating, ventilating, and air conditioning (HVAC) systems, outdoor air exchange, room-to-room air movement, sources, and sinks on indoor air pollutant concentrations.[1] The model's present configuration allows for up to ten rooms and assumes that air in all rooms is completely mixed. It runs on IBM PCs (and compatibles) and provides a menu-driven user interface. Comparisons with other models are reported elsewhere.[1]

The pollutant concentration in a room is calculated by a mass balance of the various pollutant flows. For a single room:

Mass in − Mass out + Mass produced
    − Mass removed by sinks
        = Mass accumulated in room

The analysis can be extended to multiple rooms by writing a system of equations for each room. The amount of air entering a room from all sources (the HVAC system, outdoors, and other rooms) must equal the amount of air leaving the room.

The various mass balances discussed above can be used to write a set of linear differential equations. These equations can be solved using many techniques. The simplest technique is the Euler method. The equation for Room 1 becomes:

$$dC_1 = dt/V_1(C_2Q_{2-1} + C_0Q_{0-1} - C_1Q_{1-2} - C_1Q_{1-0} + S_1 - R_1) \tag{1}$$

Likewise, for Room 2 the following equation applies:

$$dC_2 = dt/V_2(C_1Q_{1-2} + C_0Q_{0-2} - C_2Q_{2-1} - C_2Q_{2-0} + S_2 - R_2) \tag{2}$$

Implications

The techniques and results presented in "Evaluating Sources of Indoor Air Pollution" show how emission factors developed from chamber tests can be used to predict concentrations of indoor air pollutants. A user-friendly Indoor Air Quality model running on a personal computer can be used by non-experts to investigate the effects of pollutant sources in indoor environments. Such analyses are very useful in evaluating and comparing products and materials with respect to their environmental impact. The approach could also be used to prescribe an emission rate limit for indoor sources based on an "acceptable" indoor pollutant concentration level for occupant health and comfort.
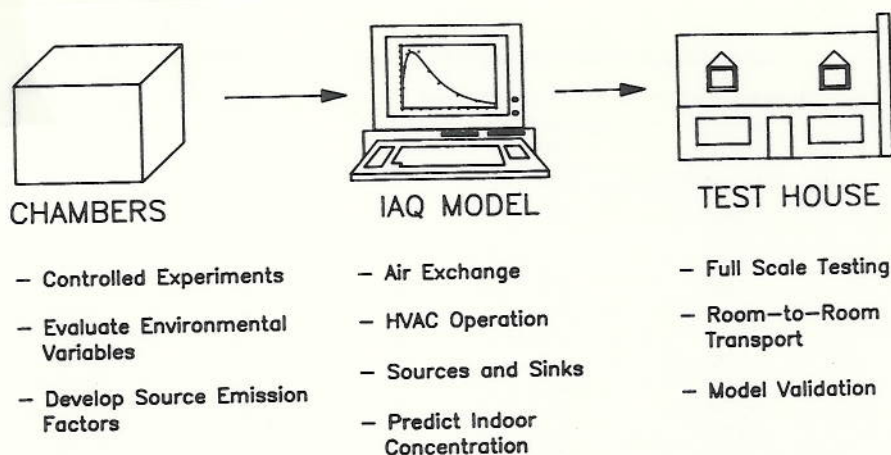
Figure 1. Three-phase IAQ research approach for evaluation of indoor air pollution sources.

where, C_1 and C_2 are the concentrations in Rooms 1 and 2, t is time, V_1 and V_2 are the volumes of Rooms 1 and 2, Q_{1-2} is the volumetric flow rate from Room 1 to Room 2, Q_{0-1} and Q_{0-2} are the volumetric flow rates from the outdoors into Rooms 1 and 2, Q_{2-1} is the volumetric flow rate from Room 2 to Room 1, Q_{1-0} and Q_{2-0} are the volumetric flow rates from Rooms 1 and 2 to the outdoors, S_1 and S_2 are the source terms for Rooms 1 and 2, and R_1 and R_2 are the removal (sink) terms for Rooms 1 and 2. The initial conditions are: at $t = 0$, $C_1 = C_{i1}$, and $C_2 = C_{i2}$ where C_i is the initial concentration. The concentration at any time t can be found by stepping through the solution in small time steps, dt.

An important feature of the model is its ability to simulate variations in building air flows. The air flows generated by a building's air handling system are generally several times larger than natural air flows. Thus when an HVAC system is on, the building air flows are dominated by the HVAC system. Air flow patterns in a building with the air handling system on may be significantly different from air flow patterns in the same building with the air handling system off. For example, many one story houses have a single return inlet for the air handling system. When the air handling system is on, air flow is dominated by the flow to the return inlet. When the air handling system is off, air flow is less directed. The on/off behavior of the air handling system is modeled by allowing two air flow patterns to exist in the building: one is active when the air handling system is on, and the other when the air handling system is off. The model switches between these two patterns depending on the state of the air handling system. The state of the air handling system (on or off) is determined by a random number generator designed to ensure that the air handling system is on for a specified fraction of each hour. The air handling system may switch from on to off and back several times in an hour. This random switching appears to provide good modeling of actual air handling system behavior.

Moth Crystal Cake Emissions (Para-dichlorobenzene)

Small Chamber Tests

Small environmental test chambers were used to determine the emission rate of para-dichlorobenzene from moth repellant.[2] The test facility used in these tests is shown schematically in Figure 2 and consists of a clean air conditioning and delivery system, an incubator containing two 166-liter stainless steel environmental test chambers, and associated sampling manifolds. A permeation system for quality control is included. Temperature, humidity, and air exchange rate are monitored and controlled by a microcomputer. Organic analyses are conducted by gas chromatography (GC) using flame ionization detectors (FID).[3-5]

Half a cake of moth repellant (surface area = 55 cm²) was used for each test to simulate the manufacturer's recommended usage. Environmental parameters were chosen to reflect typical residential conditions. Frequent sampling of the chamber exhaust during the initial stages of the test was required to observe the rapidly changing concentrations, followed by a reduction in frequency as the concentration leveled off. Once the analyst felt that the concentration was stabilized, sampling was discontinued.

The moth repellant was a relatively pure chemical compound, para-dichlorobenzene. An internal standard, hexane, was added to the sample stream through a port in a 1-liter glass mixing flask just downstream of the test chamber. Samples were taken from a glass manifold connected to the mixing flask. Sampling occurred by drawing a portion of the air stream through a heated valve (175° C) equipped with dual calibrated sampling loops or by drawing a sample from the manifold with a 5-cc gastight syringe. In each case, the sample was injected into a 2.4-m (8 ft) long by 2-mm (0.1 in.) ID stainless steel gas chromatograph (GC) column packed with 1 percent SP-1000 on Carbopack B (60/80 mesh). Data were collected on a microprocessor-based data station running software developed specifically for GC. The output signal from the FID was digitized and stored for further processing.

Table I presents the results of the moth crystal cake chamber experiments. The last column provides the emission factor for para-dichlorobenzene.
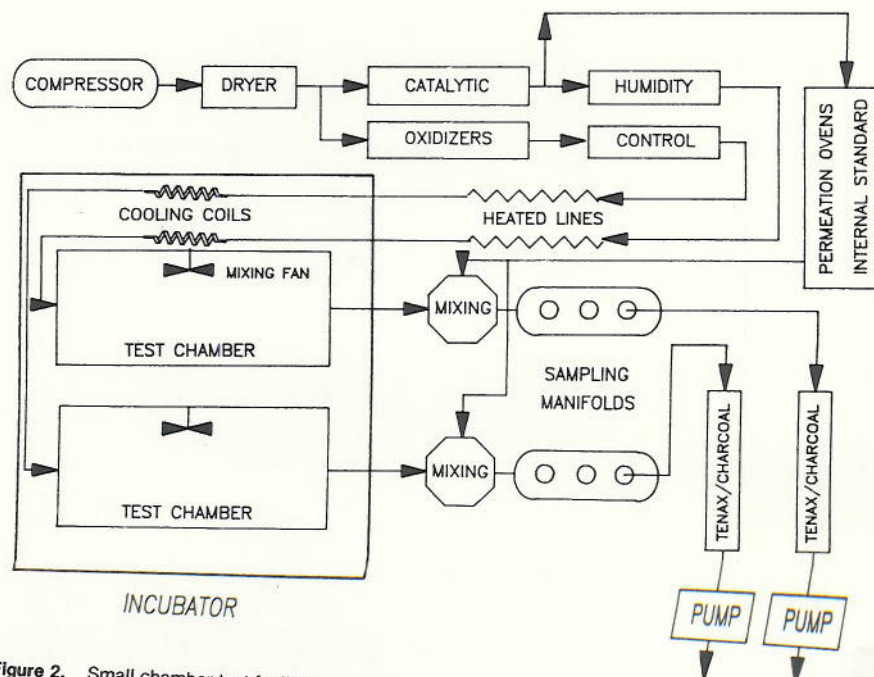

Figure 2. Small chamber test facility.

data on para-dichlorobenzene concentration and the average emission factor can be used to estimate the sink effect. The analysis is based on treating the house as a single chamber. The mass balance for the entire house at any time, t, gives:

$$dc/dt = E(t) - Qc_t \qquad (3)$$

where, dc/dt is the change of para-dichlorobenzene concentration in the house as a function of time, $E(t)$ is the emission rate (which may be a function of time), Q is the airflow through the house, and c_t is the concentration in the house at time t. The equation can be rearranged to allow calculation of $E(t)$:

$$E(t) = dc/dt + Qc_t \qquad (4)$$

An effective emission rate, $Ee(t)$, was obtained by smoothing the concentration data using a robust regression analysis[7] and then calculating dc/dt from the smooth curve. $Ee(t)$ was then calculated for various times The average effective emission rate, calculated from the $Ee(t)$ curve, was 480 mg/h. This compares with an average emission rate of 800 mg/h based on the weight lost by the moth crystals during the experiment (i.e., 1.4 mg/cm²-h × 570 cm²). The difference between the emission rate calculated from the weight loss and the effective emission rate calculated from the house concentration data is assumed to be the sink term. Thus, 40 percent of the emissions [(800–480)/800 = 0.4] go to the sink.

Initial model runs with the sink term and estimates of the room-to-room airflows were in reasonable agreement with the measurements for all locations except the closet. The predicted closet concentrations were high by a factor of 13. Also, the den and master bedroom concentrations were somewhat low relative to the corner bedroom concentration. Additional model runs showed that the flow from the closet to the corner bedroom was the key unknown parameter for determining the para-dichlorobenzene concentrations in the house.

Therefore, experiments were conducted to define the airflows in the test house and to estimate the type of mixing. During these experiments, the air-handling system flows were measured. Flow visualization studies to determine the nature of the in-room and room-to-room mixing were conducted with neutral density balloons and with neutral density helium bubbles. The measured flows of the air-handling system are shown in Table III. The balloon and bubble experiments showed that, even with the air-handling system off, considerable mixing existed between rooms. These experiments also indicated that there was a substantial airflow into and out of the closet. Finally, the visualization studies indicated that

there was flow between the closet and the hallway and between the closet and the master bedroom.

Hot-wire anemometer measurements were made of the airflow velocities through the openings around the closet door. These measurements showed that the airflow into and out of the closet was between 4 and 9 m³/h.

Model calculations were performed with the input data from the experimental studies. The input data for the model run are shown in Table IV. The model calculations were stopped after several hours of simulated time because steady-state was reached. The results of this run are shown in Table V. The agreement between the model and the measured data is excellent for all rooms. The measured values shown in Table V are the average concentrations for each room for the 4 days of sampling over the 11 days when the moth crystal cakes were in the closet.

Unvented Kerosene Heater Emissions (Particles)

Large Chamber Tests

The kerosene heater characterization was conducted in the large scale 34 m³ environmental test chamber at the J. B. Pierce Foundation Laboratory, New Haven, Connecticut.[8] Twelve heaters were borrowed from the Consumer Product Safety Commission: five radiant (R), three convective (C), and four double burner designs, including three combination convective/radiant (C/R), and one radiant/radiant (R/R). The R/R design also included a ceramic catalytic disk above the concentric radiant elements.

The test group included both new

and used heaters of the same type produced by the same manufacturer. Except for a single comparison run, all heaters were fueled with the same 1-K kerosene fuel; its composition had been previously determined by an independent testing laboratory. No effort was made to maltune any of the heaters to obtain "worst case" scenarios.

The characterization strategy consisted of two phases. Phase I was a general screening step. In this phase, each heater was fueled outside the chamber and subsequently placed in the chamber on a Potter scale to measure fuel consumption. The heater and the chamber were allowed to equilibrate for 0.5 hour while the mixing rate was established and chamber background levels for key parameters were measured. The heater was then fired up and run continuously for 6 hours. Dur-

ing the run, the heater was cycled through the range of fuel consumption/operational settings (i.e., high, low, and normal) that are expected to occur during normal residential use. The actual control settings were determined visually based on the manufacturer's recommendations included with each heater. At the end of 6 hours, the heater was shut off but left in the chamber while the decay of CO_2, particles, and combustion gasses was measured.

The particle size distribution was measured using an electrical aerosol analyzer, a condensation nuclei counter, and an optical particle counter. Integrated mass samples were collected using eight portable pumps distributed around the chamber sampling at 2 liters/min and an Andersen

Table II. Test house results for para-dichlorobenzene emissions from moth crystal cakes.

Day	Closet (mg/m³)	Corner bedroom (mg/m³)	Master bedroom (mg/m³)	Den (mg/m³)
4	107	4.72	3.49	3.84
6	53.6	4.41	3.50	3.30
8	70.9	5.51	4.18	3.80
11	63.0	5.61	4.23	4.02
12	5.43	2.11	2.07	1.80
14	1.53	0.90	0.98	Lost
16	1.30	0.52	0.65	0.45
18	0.97	0.42	0.55	0.45

Table IV. IAQ model input data—moth crystal cake analysis.

Variable	Value
Emission factor	1.4 mg/cm²-h
Air exchange with outdoors	0.35 ACH
Air exchange between closet and corner bedroom	4 m³/h
Air circulation due to HAC system	Table III
Sink term	40 percent

Table III. Test house HAC system flows.

Room	Measured flow from HAC register (m³/hr)
Den	679
Middle bedroom	38
Corner bedroom	278
Master bedroom	280

PM–10 sampler. The sampler was modified to include an XAD–2 sorbent cartridge for the collection and measurement of semivolatiles. The particles were collected on Teflon-backed glass fiber (Pallflex) filters for gravimetric analysis and bioassay. Volatiles were collected in Summa canisters for GC/MS analysis. Measurements of NO_x, SO_x, CO, CO_2, and total hydrocarbons were recorded using continuous monitors. The decay of CO_2 at the end of a run was used to calculate the chamber ventilation rate as well as to provide a measure of reactivity of the other parameters. Only the particle data are reported in this paper.

Table V. Comparison of measured test house concentrations of para-dichlorobenzene with IAQ model predictions.

Room	Measured value (mg/m^3)	Predicted value (mg/m^3)
Closet	74	74
Corner bedroom	5.1	5.7
Master bedroom	3.9	4.0
Den	3.7	4.0

The chamber was operated in a flow-through mode using outdoor air conditioned to provide the proper temperature and humidity. For each chamber run, the pollutant generation rate was calculated by:

$$G = (Ceq - Co) \times (Qv + Qa + Qr)$$
$$(5)$$

where, G is the generation rate (mg/h), Ceq is the steady state chamber concentration (mg/m^3), Co is the inlet (outdoor air) concentration (mg/m^3), Qv is the chamber flow rate (m^3/h), Qa is the removal rate by surfaces (i.e., sinks), and Qr is the effective removal rate of contaminant by collection method used. The units for Qa and Qr are equivalent cubic meters per hour of outdoor air. Qa was determined by evaluating the difference in the decay rate of the pollutant compared to the decay rate of CO_2, which was assumed to have no sink effect.

The emission factor for the heater was then calculated as:

$$EF = (G/F) \times 1000 \qquad (6)$$

where, EF is the emission factor ($\mu g/g$ of fuel) and F is the fuel consumption rate (g/h). The emission factor can also be expressed in micrograms per kilojoule if the fuel consumption is measured in kilojoules per hour.

Four heaters were chosen for the intensive Phase II characterization based on pre-established criteria designed to select the heater from each category with the largest particulate mass production. These heaters were prepared for testing as in the Phase I screening,

Table VI. Particle emission factors for Phase I and II characterization of unvented kerosene space heaters.

Heater type	Phase I ($\mu g/g$)	Phase I ($\mu g/kJ$)	Phase II ($\mu g/g$)	Phase II ($\mu g/kJ$)
C	55	1.3	38	0.9
C/R	47	1.1	42	1.0
R	58	1.3	45	1.0
R/R	218	5.0	376	8.7

Note: Phase I emission factors based on cyclic operation over 6 h.
Phase II emission factors based on steady operation over 12 h.

and the same parameters were measured. However, in Phase II they were operated for 12 h at a normal flame setting, rather than cycled.

The particle emission factors based on the portable pump samples for both the Phase I and II characterizations are contained in Table VI. All results in Table VI are based on a single test, except for the Phase II tests of the C/R, R, and R/R heaters where three tests were conducted. Taken as a whole, the particulate data available to date indicate that unvented kerosene space heaters are a strong source of indoor respirable suspended particles. Particle size distribution measurements indicate that the particles produced by all kerosene heaters are less than 2.0 μm in diameter, with the peak volume occurring around 0.15 μm. The largest mass production occurs during startup. Total mass concentrations ranged from 73 to 721 $\mu g/m^3$. The greatest mass emissions were produced by the multiple radiant design that incorporated the catalyst. Since only one heater of this design was tested, it is not known whether the high levels are a function of design or are related to the heater's usage history.

Test House Experiments

Two heaters were taken from the intensive Phase II characterization and placed in the test house for further characterization. These heaters, one convective/radiant (C/R) and one radiant/radiant (R/R), were chosen because they represented the newer generation of kerosene heaters and included the highest emitter of particles.

The heater to be tested was placed in the den. One window was opened 2 in. (5 cm) to provide outdoor air as recom-

mended by the heater manufacturer. The air exchange rate of the house under these conditions was 0.5 ACH based on SF_6 measurements. The HAC system was not operated during the test. Particles were sampled in the den, in the corner bedroom, and outside the house. The samples were collected on Pallflex filters using 4 cfm (113 liter/min) samplers. Filters were changed every 6 h and the total sampling time was 12 h.

The test house results are shown in Table VII. The R/R heater produced about 4 times as much particulate as the C/R heater, which is consistent with the emission rate data obtained in the chamber studies.

IAQ Model Analysis

The general strategy for modeling the kerosene heater experiments was the same as used for the moth crystal studies. Initial model runs were made with the emission factors ($\mu g/kJ$) developed in the chamber studies and the fuel consumption data from the test house experiments. The outdoor air exchange was assumed to be 0.5 ACH. Thermal gradients in the house due to heater operation were assumed to cause the air in the house to mix internally five times per hour. Room-to-room air flows were calculated on this basis.

The results of the initial model runs were in fair agreement with the experimental data. However, all predicted concentrations were higher than the measured concentrations which suggests a particle sink. An estimate of the sink effect was obtained by reviewing indoor particulate work by Offermann et al.[9] They conducted chamber studies and measured the sink effects due to the chamber. For the particle diame-

Table VII. Test house results—particle emissions from unvented kerosene heaters.

Test	Location	Sample size (n)	Average conc. ($\mu g/m^3$)	Standard dev. ($\mu g/m^3$)	Range ($\mu g/m^3$)
C/R htr	Outdoors	6	28.4	19.1	12.3–65.9
	Den	6	70.9	17.3	46.4–91.6
	Bedroom	6	37.9	20.1	13.1–65.2
R/R htr	Outdoors	6	29.1	7.1	17.8–36.8
	Den	6	288.3	42.1	225.0–366.8
	Bedroom	6	252.0	52.9	181.7–330.7

Table VIII. Comparison of test house particle concentrations with IAQ model predictions.

Heater type	Location	Measured value ($\mu g/m^3$)	Predicted value ($\mu g/m^3$)
C/R	Den	71	60
	Bedroom	38	52
R/R	Den	288	285
	Bedroom	252	235

ters of interest in the kerosene heater study, their data indicated that the sink should collect between 10 and 20 percent of the emitted particulate matter. A sink factor of 20 percent was used in subsequent model runs. This sink factor was also consistent with the results of the chamber studies conducted at J. B. Pierce Foundation.

Table VIII presents the results of the model predictions. The predictions agree closely with the measured values for the R/R heater and are within 1 standard deviation (based on sampling) for the C/R heater.

Conclusions

A three-phase approach employing environmental chambers, IAQ models, and test house experiments is effective in linking sources of indoor pollutants to measured concentrations. Emission factors developed in test chambers can be used to evaluate full scale indoor environments. A PC-based IAQ model has been developed that can accurately predict indoor concentrations of specific pollutants under controlled condi-

tions in a test house. The model is also useful in examining the effect of pollutant sinks and variations in ventilation parameters. Further validation of the approach is required for other source types, including solvent-based materials and aerosol products.

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