

AIR INFILTRATION MEASUREMENTS IN A HOME USING A CONVENIENT PERFLUOROCARBON TRACER TECHNIQUE

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Using miniature perfluorocarbon tracer (PFT) sources and miniature passive samplers, both about the size of a cigarette, tests conducted in the laboratory and in a typical home successfully demonstrated the utility of the PFT kit as a means for implementing wide-scale infiltration measurements in homes. The PFT diffusion plug source, an elastomer containing a dissolved perfluorocarbon compound, was shown to emit vapors at the rate of about 0.1 to 5 nL/min, providing steady-state concentrations in a home of about 1 to 10 pL/L, i.e., parts per trillion by volume, when one source is deployed for each 300-500 ft³ (28-46 m³) of living space. The emission rate from the diffusion source was predictable, but its dependence on both temperature and time suggested the development of alternative approaches. One such alternative, a liquid PFT permeation source, had emission rates which could be tailored over the range 10-20 nL/min, were independent of age for as long as the liquid remained (~5 yr), and had significantly lower temperature dependence. A number of passive adsorption tube samplers performed reproducibly and identically (to within $\pm 2\%$ -3%) in laboratory tests. Together with a programmable tracer sampler, the miniature diffusion sources and samplers were deployed in a typical home; six PFT sources were uniformly deployed, three on each level of a two-story house. Multiple location sampling, as well as sampling in rooms with and without a miniature source, demonstrated that even in a house without forced-air circulation, a well-mixed modeling approach is justified. Analyses of the tracer samplers were performed back in the laboratory with an automatic electron-capture gas-chromatography system. The effects of the inside/outside temperature differential, as well as that of an open fireplace compared with a wood-burning stove, on the measured air infiltration rates were clearly demonstrated. Comparisons of the PFT tracer method with that of the SF₆ tracer decay approach showed the results of the two methods to be identical within experimental precision. With this miniature source and sampling tracer kit, infiltration rates in the range 0.2-5 air changes per hour can be measured over time-averaged periods of as little as 1 day up to several years.

Introduction

We spend a significant, if not the greatest, portion of our lives within an indoor environment, and thus the quality of that environment is at least as equally important as that of the outdoors. In characterizing our exposure to air pollutants in the indoor environment, it is also recognized that the extent and magnitude of exposure in homes and buildings is in part governed by the rate of air infiltration, the escape of heated air from a structure and its replacement by cold, ambient air (Harrje *et al.*, 1970; Sherman *et al.*, 1980), and the nature of the sources.

About one-third of the heat loss in buildings can be ascribed to air infiltration, the magnitude of which cannot be readily predicted but must be measured. At-

tempts to reduce heat loss through a reduction in air infiltration will unfortunately increase the occupant's exposure to internally generated pollutants. Thus, techniques for measuring the extent of air infiltration and, at the same time, the parameters affecting the magnitude of that exchange, such as temperature difference within and outside a house and wind speed and exposure (Malik, 1978; Shaw, 1981), must be available for researchers performing indoor air pollution or heat loss studies.

Two prominent techniques have been developed for the determination of air infiltration, one through the use of a conservative tracer (Malik, 1978; Harrje and Grot, 1977) and the other by a building pressurization technique (Sherman and Grimsrud, 1980). Both of these techniques use sophisticated sampling and/or measure-

ment instrumentation generally requiring a technical operator. The pressurization method uses a "blower door" assembly in place of the conventional door; as the blower input volume rate increases, pressure differential in a home is related to leakage crack areas and, ultimately, air infiltration rates. In addition, infrared scanners in the positive pressure mode can be used to locate major areas of leakage, such as the attic floor (Harrje *et al.*, 1979), and smoke sources near windows, electrical outlets, etc., will help to locate air leakage from the home (Harrje *et al.*, 1981). In the negative pressurization mode, cold air leakage in winter months can often be found by searching with one's hand. Several models have been applied to relating pressurization results with air infiltration rates obtained from tracer measurements (Shaw, 1981; Sherman and Grimsrud, 1980).

The tracer techniques generally provide the only direct measure of air infiltration in homes under actual living conditions. Several modes of operation have been used (Kronvall, 1981), the most prevalent being the tracer decay method (Bassett *et al.*, 1981; Hunt, 1980; Harrje *et al.*, 1979) and the steady-state tracer method (Harrje *et al.*, 1975; Condon *et al.*, 1980). In the former, a small amount of SF₆ is released in the home and the logarithmic decay of the concentration with time is directly proportional to the infiltration rate expressed as air changes per hour (ach). It is not necessary to know the exact amount of SF₆ released but the concentrations must be measured periodically (5–15 min) with a portable gas chromatograph (Harrje and Grot, 1978), or by collecting periodic samples in plastic bottles (Harrje *et al.*, 1979) or in syringes and air sampling bags (Grot, 1980) and analyzing on return to the laboratory. With this approach, air exchange rates are determined with data usually covering a period from 2 to 8 h.

In the steady-state tracer mode, previous systems have used elaborate mechanical systems to inject SF₆ on a periodic basis, determined by feedback from a gas chromatograph, and operated at a frequency designed to maintain the SF₆ concentration constant (Harrje *et al.*, 1975). A newer, similar approach used a micro-processor controller to automate the measurements further (Condon *et al.*, 1980). This method allows the determinations of infiltration rates to be measured over long periods of time and provides data on its variability with time. However, the systems are complex and expensive and require a trained operator to assure the best results.

A program was initiated at Brookhaven to develop a simple, inexpensive tracer kit; coined the Brookhaven National Laboratory Air Infiltration Measurement System (BNL/AIMS), it can be deployed by the average homeowner, so that a national survey of the range of the magnitude of air infiltration could be performed. Consequently, individual homeowners can determine the extent to which air infiltration is significant for their

homes. In addition, the technique in its simplest form is designed to provide an average air infiltration measurement coincident in time with the determination of average indoor air quality. Weekly, monthly, and shorter or longer periods can be averaged with this approach. With the use of available programmable tracer samplers, data on the short-term variability of the air infiltration rate can also be determined. This paper describes some of the tracer tools and presents some results of their use in indoor air infiltration measurements.

Tools for Tracer Infiltration Determination

The tracer system used in this study consisted of perfluorocarbon tracers (PFTs), i.e., a series of perfluorinated cyclic hydrocarbons, emitted from miniature sources; miniature passive as well as programmable samplers; and a laboratory gas chromatograph for the analysis of the samplers. The purpose of this section is to describe these tracer tools and to demonstrate their utility. The unique nature of the miniature source and sampler will provide a new, simple approach to infiltration measurements in homes.

When a tracer gas is released into a room, it generally mixes uniformly within 3 to 4 min (Condon *et al.*, 1980). It is quite safe to assume that the air in a house is well mixed, since the BNL/AIMS uses a miniature source in each room of a house and since sampling is integrated over no less than 20 min but usually hours or even weeks. Assuming that the house contains tracer sources emitting at known rates, a simple material balance yields

$$\frac{dV(t)}{dt} = R_s - R_i(t) \frac{V(t)}{V_h}, \quad (1)$$

where $V(t)$ is the volume (pL) of tracer gas present in the house at any time t (h), R_s is the tracer source rate (pL/h), $R_i(t)$ is the house infiltration rate which may vary with time (L/h), and V_h is the volume of the house (L). A constant tracer source rate is needed, as well as a means to measure the time-dependent tracer volume in the home, in order to determine the rate of air infiltration, $R_i(t)$.

The tracer source

A tracer technique for measuring long-range (greater than 500 km) transport and dispersion in the atmosphere (Ferber and Dietz, 1979; Ferber *et al.*, 1981) and for tagging explosives for detection of clandestine bombs (Senum *et al.*, 1980; Dietz and Senum, 1981), resulted in the development of a methodology for sensitively measuring PFTs at concentrations as low as 0.001 pL/L (Dietz *et al.*, 1978). Background concentrations of some of the PFTs are 0.030 pL/L for perfluorodimethylcyclo-

hexane (PDCH), 0.003 pL/L for perfluoromethylcyclohexane (PMCH), and less than 0.001 pL/L for perfluorodimethylcyclobutane (PDCB). The PFTs have been shown to be nontoxic in both inhalation and oral toxicity tests at concentrations in the percent range (Senum *et al.*, 1980). Thus, it is quite clear that home PFT concentrations of 1–10 pL/L are both safe and readily measurable.

During the development studies for applying PFTs to the tagging of explosives, extensive series of tests were conducted to determine the possibility of adsorption losses on common surfaces. A PFT source was placed within a towel, enclosed in a cloth suitcase, and placed in a dilution chamber. Concentrations within the dilution chamber as a function of time were compared to those generated when the source alone was placed in the chamber (Senum *et al.*, 1980). The results indicated no losses for the three PFTs. In preparation for the use of PFTs as atmosphere tracers, a 10-pL/L standard in air was exposed to tree branches with leaves, grass, and soil in a large plastic bag; no measurable loss of tracer could be discerned. Furthermore, under the steady-state conditions that will prevail in air infiltration tests, adsorption losses were shown to be negligible (Senum *et al.*, 1980).

The required source rate for a home infiltration test can be estimated from Eq. (1) for the steady-state conditions [$dV(t)/dt = 0$]. Thus, for one room in a house,

$$R_s = \frac{R_v(t)}{V_r} V_{ss} = (AC) V_{ss} = (AC) C_{ss} V_r, \quad (2)$$

where V_r is the volume of a room (typically 30,000 L), AC is the number of air changes in the house (typically 0.5 per hour), and C_{ss} is the desired steady-state concentration (1 pL/L or 0.001 nL/L). Substituting into Eq. (2) gives a needed source rate per room of 0.25 nL/min. To achieve a 10 pL/L concentration, the rate from the source must be 2.5 nL/min.

The tracer source for these tests was simply a fluoroelastomer plug impregnated with a known mass of a PFT and crimped within a metal shell (see Fig. 1). The dissolved PFT diffuses from the end of the plug at a known rate (Senum *et al.*, 1980) which is inversely proportional to the square root of time (see Fig. 2) for the emission of the first 50%–60% of the original amount present.

Since the emission rate decreases with time, it is important to choose an elastomer that gives the optimum lifetime for the source. Table 1 shows that for the fluoroelastomer, fluorosilicone 70, emission rates for the first 2 yr are in the desired range, but then the rate decreases more quickly. Table 2 shows that in Viton the emission rate is lower by about an order of magnitude, but still sufficient for infiltration measurements. The lifetime of the Viton source, however, is much greater than that for the fluorosilicone plug, lasting much

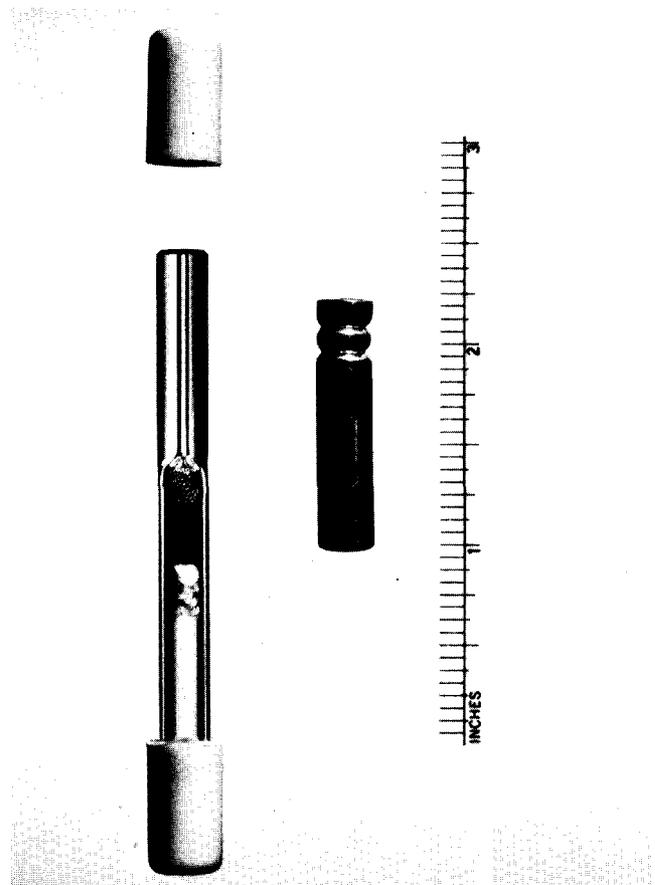


Fig. 1. Capillary adsorption tube sampler (CATS) on the left and PFT diffusion source on the right.

longer than 5 yr. Figure 3 shows that a 2-month-old source of PDCB in Viton has only lost 1.5% of its initial charge. Before any infiltration tests are performed, the emission rate from each source can be verified by passing a known rate (usually 1 L/min) of clean air over the source, mixing, and measuring the resulting concentration by gas chromatography.

The variability of the emission rate from these diffusion plug sources, based on gravimetric measurements for triplicate sources stored at each of three temperatures (Senum *et al.*, 1980), was, for a series of six different PFT/fluoroelastomer (Viton-type) combinations, within $\pm 6.4\%$ with a maximum of 11.6% and a minimum of 1.5%. Forty PDCH/fluorosilicone 70 diffusion plug sources were fabricated and the gravimetric data was used to compute the individual emission rates, which ranged from 3.5 to 4.9 nL/min, with an average of 4.03 ± 0.39 nL/min. Thus, individual calibration of the emission rate from each source should not be necessary, since the average emission rate from four to six sources used in a house should be within $\pm 7\%$ – 10% of the actual value.

In addition to the time dependence and the reproducibility of the diffusion source plug, its emission rate also varies with temperature. From the same Viton-type

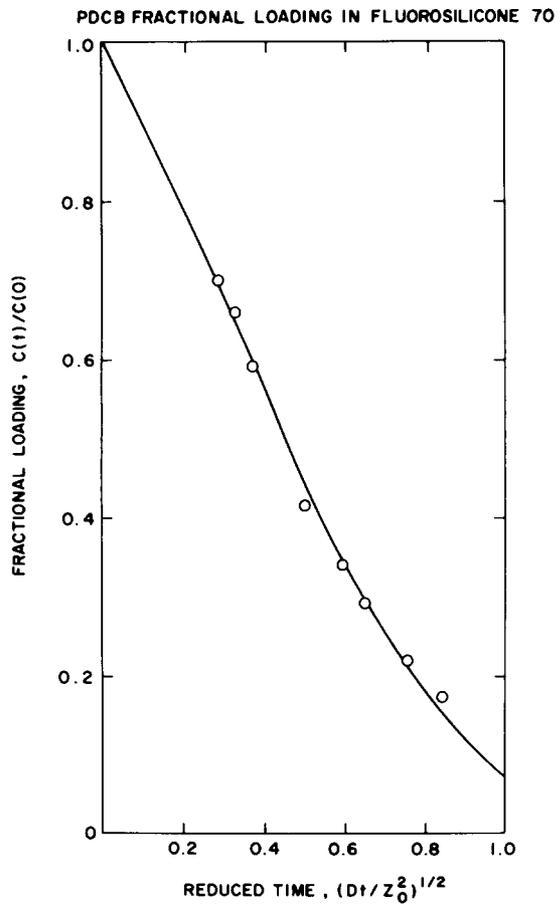


Fig. 2. Time-dependent loss of tracer loading.

data set above which had sources stored at room temperature ($\sim 22^\circ\text{C}$), 45°C , and 65°C , activation energies for the rate of emission varied from 11 to 14 kcal/mole. Thus, for a change in temperature from 20 to 23°C , the emission rate will increase 20% to 25%. The activation energy for the emission rate of PDCH from fluorosilicone 60 was only 3.0 kcal/mole, corresponding to a change of only 5% over the same 3°C change in temperature. Unfortunately, the emission rate dependence on source age is substantially more severe for the fluorosilicone 60 than for the Viton substrate. For the former, there is more than a sixfold decrease in emission rate for an age change of 0.5–2 yr (cf. Table 1), whereas for Viton the emission rate change is only a factor of 2 over the same age period (cf. Table 2).

To reduce these age and temperature effects, a series of permeation-type miniature sources was fabricated by

Table 1. PFT Source rate from fluorosilicone 70. (50 mg PFT per gram of F 70; 1 g source.)

PFT	Emission Rate (nL/min) at Various Ages (yr)			
	1/6	1/2	2	5
SF ₆	22.5	7.64	0.063	0.000
PDCB	7.0	3.87	0.597	0.014
PMCH	6.0	3.31	0.512	0.012

Table 2. PFT Source rate from Viton. (50 mg PFT per gram of Viton; 1 g source.)

PFT	Emission Rate (nL/min) at Various Ages (yr)			
	1/6	1/2	2	5
SF ₆	5.2	3.0	1.50	0.38
PDCB	0.37	0.21	0.106	0.048
PMCH	0.48	0.27	0.137	0.061
PDCH	0.43	0.25	0.123	0.056

placing the liquid PFT in the shell before crimping the elastomer plug in place. For liquid PDCH sources plugged with fluorosilicone 60, apparent activation energies ranged from 3.9 to 4.3 kcal/mole, which, for daily temperature fluctuations of 3°C , corresponded to emission rate changes of only 7% to 8%. Since it should be possible to know the room temperature to within better than 3°C , the maximum error due to temperature uncertainty in calculated infiltration or air exchange rates would be 8%. Another advantage of this permeation source is that the rate of emission is constant with time as long as any liquid remains. Source lifetimes of 5 yr or more are easily obtained, and typical emission rates for the permeation sources are in the range 5–20 nL/min, quite adequate for infiltration measurements.

Primarily diffusion plug-type sources were used in the air infiltration experiments reported in this paper; future studies will be conducted only with permeation-type sources. The utility of these miniature sources was best realized by deploying one source in each major room of a house, thereby assuring uniform mixing.

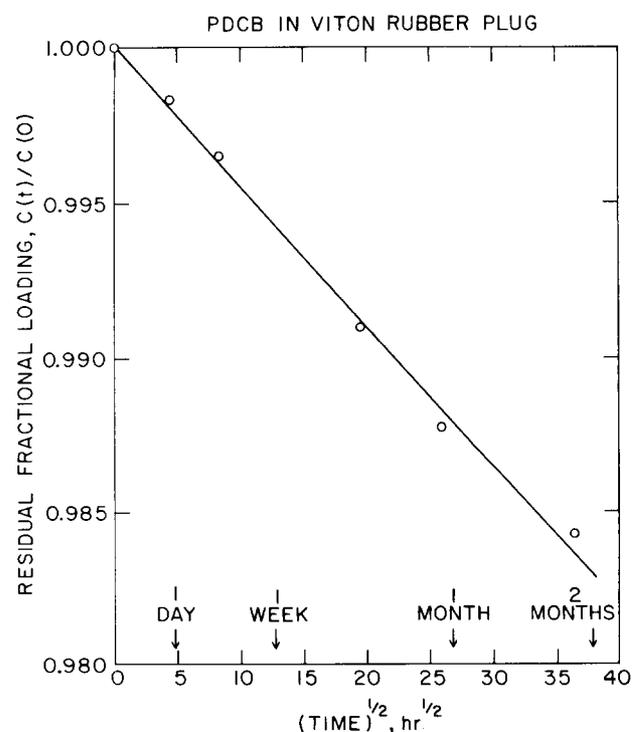


Fig. 3. Fraction of PDCB remaining at 2 months of age.

Tracer measurement

Based on the development and subsequent testing of the Brookhaven-modified laboratory electron capture gas chromatograph (Ferber *et al.*, 1981), the detectability of PFTs has been demonstrated to range from less than 0.01 pL to nearly 10,000 pL (cf. Fig. 4). This six-orders-of-magnitude capability provides the necessary dynamic range to sample PFTs in homes under almost any conceivable time interval or air infiltration rate.

Tracer samplers

To determine best an average infiltration rate, room air samples containing steady-state concentrations of a deliberately released tracer need to be collected over a period of weeks, perhaps one sample per week or one integrated sample per month.

The problem usually encountered with sampling for such extended periods of time is that very low flow rates are required, generally less than 0.1 mL/min, whereas most commercially available air monitoring sampling pumps are designed for flow rates no less than 0.5–1 mL/min. In addition, if a large number of homes and buildings are eventually to be monitored, the usual cost of the sampling pump alone may become significant. Thus, a passive sampling device would provide an efficient alternative to the pumping and cost considerations.

Passive sampler. A simple passive tracer sampler, coined

the capillary adsorption tube sampler (CATS), is shown in Fig. 1. The molecules of a dilute concentration of a gas in air diffuse through the uncapped length of capillary tubing at a rate which is proportional to the partial pressure driving force, that is, from the high concentration in the room at one end to the zero concentration in the adsorbent at the other end. This steady-state diffusion is defined by Sherwood and Pigford (1952) as

$$N_a = \frac{D_v P}{RTZ} y_a, \quad (3)$$

- where N_a = diffusion rate, g mole PFT/sec cm²;
- D_v = diffusion coefficient, cm²/sec;
- P = total pressure, atm;
- R = gas constant, 82.06 cm³ atm/°K mole;
- T = absolute temperature, °K;
- Z = length of capillary tube, cm;
- y_a = mole fraction of PFT in the room.

Equation (3) is applicable in all cases in which the concentration of the diffusing gas is small, that is, less than 0.01 mole fraction, which is certainly the case for PFTs in air.

As shown in Fig. 1, the whole CATS device is about 3 in. (7.6 cm) long and ¼ in. (6 mm) in diameter, that is, about the size of a cigarette. The tracer diffusing section is 1 in. (2.5 cm) long and 1 mm in diameter. About 50 mg of type XE-347 Ambersorb, a carbonaceous adsorbent made by Rohm and Haas, are packed in the mid-section, held in place by stainless steel screens and/or glass wool.

During use in the home, the plastic cap on the capillary end is removed and diffusion sampling commences. At the completion of sampling, a cap plug is placed on the diffusion tube end and the unit is mailed to the laboratory. In the laboratory, both ends of the tube are connected to the sampling valve of a chromatograph system and the PFT is thermally desorbed by a coiled nichrome wire resistive heater. Six CATS devices were fabricated and exposed to a mixed PFT standard for 202 min. As shown in Table 3, the ability to make all

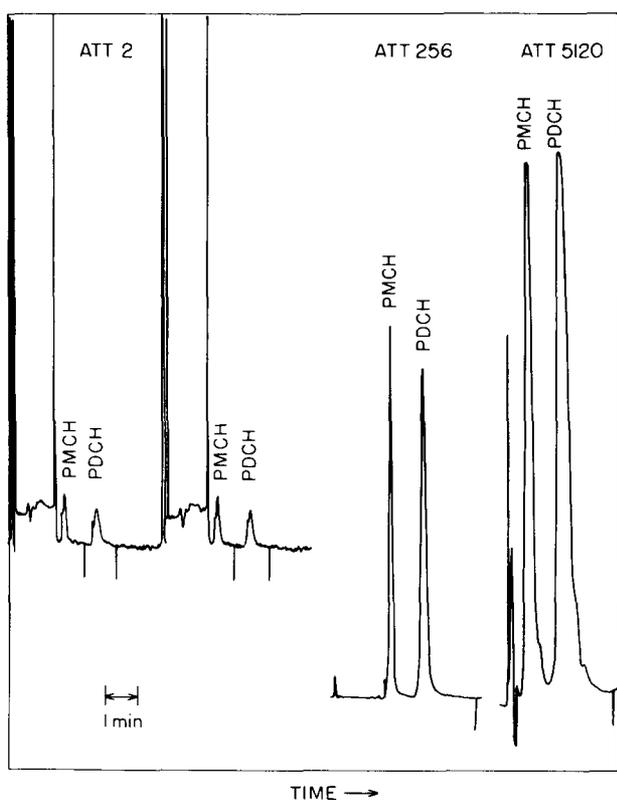


Fig. 4. Detection of two PFTs. On the left, 0.015 pL each; middle, 25 pL; right, 10,000 pL.

Table 3. Capillary adsorption tube sampler (CATS) measurement of PFT standard^a (Sampling duration: 202 min)

CATS No.	GC Area Response, 1000 counts		
	PDCB	PMCH	PDCH
1	b	b	976
2	890	141	1049
3	892	133	1027
4	870	135	1000
5	873	138	1008
6	889	133	1023
	883 ± 10 (± 1.1%)	136 ± 3 (± 2.2%)	1014 ± 25 (± 2.5%)

^aStandard contained 1 nL/L PDCB and PDCH and 0.14 nL/L PMCH.

^bMissed peak integration.

six tubes sample at the same rate was quite good, within about 2% to 3% overall.

The rate of diffusion sampling of PFTs by the CATS device can be calculated from Eq. (3) by substituting the appropriate values. The diffusion coefficients can be estimated by an empirical equation (Sherwood and Pigford, 1952); Table 4 lists the appropriate values for the three PFTs. Assuming a typical home temperature of 65 °F (291.7 °K), the diffusion rate is

$$N_{\text{PFT}} = 1.644 \times 10^{-17} D_v, \text{ g mole/cm}^2 \text{ sec pL/L}, \quad (4)$$

or a volume rate of

$$N_{\text{PFT}} = 3.95 \times 10^{-4} D_v, \text{ pL/cm}^2 \text{ sec pL/L}. \quad (5)$$

Since the cross-sectional area of the 1-mm capillary is 0.00785 cm² and the exposure time to the standard was 202 min, Eq. (5) can be used to calculate the expected volume of each tracer, as shown in Table 4. The agreement with the measured volume of tracer based on the average response area and the appropriate gas chromatograph response factor, *a*, was excellent (within a few percent).

In another test, the CATS tubes were exposed to 10-nL/L PMCH and PDCH standards for periods of 10 to 60 min. As shown in Fig. 5, the agreement between measured and calculated tracer volumes was again quite good. Thus, this passive sampler has been demonstrated to be a reliable sampling device. Using a typical *D_v* of 0.05 cm²/sec and the appropriate area in Eq. (5), the equivalent pumping rate is 0.0093 mL of air per minute.

Since the expected concentrations of PFTs in home infiltration studies is in the range 1–10 pL/L, Table 5 lists the sampling duration necessary for various measurable quantities of PFT to be collected, using both a 1- and 4-mm diam. capillary and assuming a 1-in. (2.5-cm) length. For example, for the 4-mm CATS exposed to 10 pL/L of a PFT, a detectable quantity can be collected in as little as 7 min, yet the tube could still be analyzed after exposure as long as 14 yr. Thus, the CATS sampler has a wide dynamic range for its sampling duration. Another positive aspect of CATS is that the sampling rate is not affected by changes in barometric pressure and is only dependent on the square root

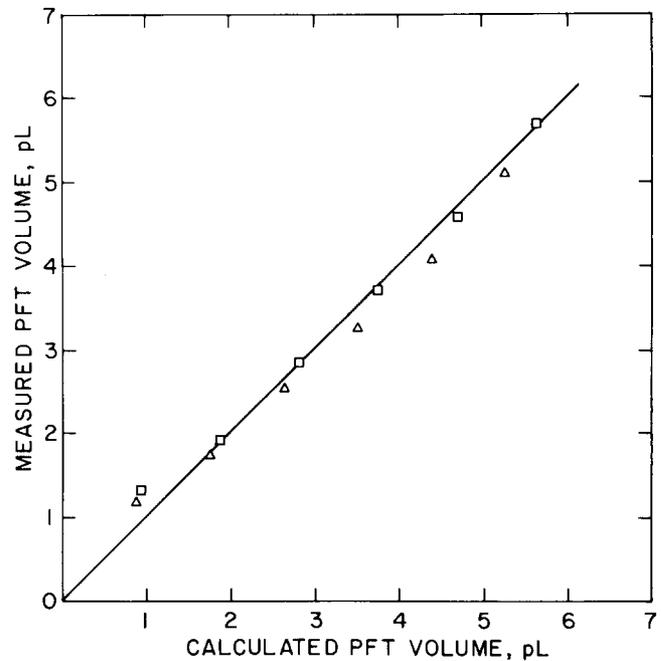


Fig. 5. Agreement between PFT concentrations measured by CATS and the calculated, expected levels; 1 pp 10⁸ standard of PMCH (squares) and PDCH (triangles) for 10–60 min.

of the absolute temperature. Thus, a 10 °C change in temperature would cause less than a 1% change in the sampling rate.

Programmable sampler. Another sampler, the programmable Brookhaven Atmospheric Tracer Sampler (BATS), is shown in Fig. 6. The unit is a battery-operated adsorption sampler containing 23 sampling tubes for collecting PFTs in air with an internal pump (Ferber *et al.*, 1981), and has been used extensively in a

Table 5. CATS range of sampling duration.

PFT Vol. (pL)	Sampling Duration			
	PFT = 1 pL/L		PFT = 10 pL/L	
	1 mm	4 mm	1 mm	4 mm
0.01	19 h	1.2 h	1.9 h	7.1 min
1	79 day	4.9 day	7.9 day	0.5 day
100	22 yr	1.4 yr	2.2 yr	49 day
10,000	—	—	—	14 yr

Table 4. CATS-measured versus calculated concentration. (Average response to 202 min of standard.^a)

PFT	<i>D_v</i> (cm ² /sec)	<i>a</i> (pL per 1000 counts)	Avg. Area (1000 counts)	<i>V</i> (pL) of PFT	
				Measured ^b	Calculated ^c
PDCB	0.0538	0.00228	883	2.01	2.02
PMCH	0.0505	0.00164	136	0.22	0.22
PDCH	0.0472	0.00188	1014	1.91	1.81

^aSee Table 3 for concentrations of standard.

^b $V_{\text{meas}} = a \times \text{Area}$.

^c $V_{\text{calc}} = 0.0376 D_v C_{\text{PFT}}$, where C_{PFT} is the concentration in the standard, in pL/L.

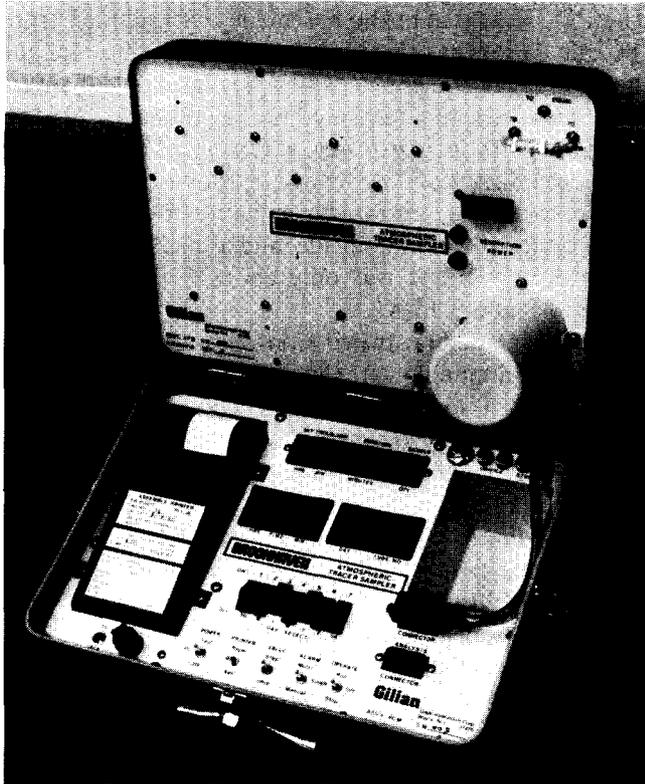


Fig. 6. The programmable Brookhaven Atmospheric Tracer Sampler (BATS).

number of long-range as well as complex terrain atmospheric tracer experiments. Its use in infiltration measurement in homes as well as the CATS devices will be demonstrated in the next section.

Infiltration Measurements in a House

Two tests were performed with the PFT tracer systems. In both cases, 6 PDCH diffusion sources were deployed in each major room of a two-story colonial house (~2000 ft² or 190 m² living area), three on each level. A seventh PDCH source was placed in the basement. One programmable BATS unit was deployed for sampling in both tests; in the second test, which covered 16 days, six CATS passive samplers were also used.

The PDCH sources were generally placed at about eye level on a piece of furniture in each room, near an outside wall to take advantage of the convective circulation from the hot water baseboard radiators. The samplers, on the other hand, were usually placed on a piece of furniture near an inside wall in order to assure mixing and to prevent direct sampling in the vicinity of the source.

Initial tracer release period (Tests 1 and 2)

The initial increase in tracer concentration—perfluorodimethylcyclohexane (PDCH) in these tests—as a function of time, is shown in Fig. 7 for Test 1. Since for short periods of 1–2 h the infiltration rate can be assumed

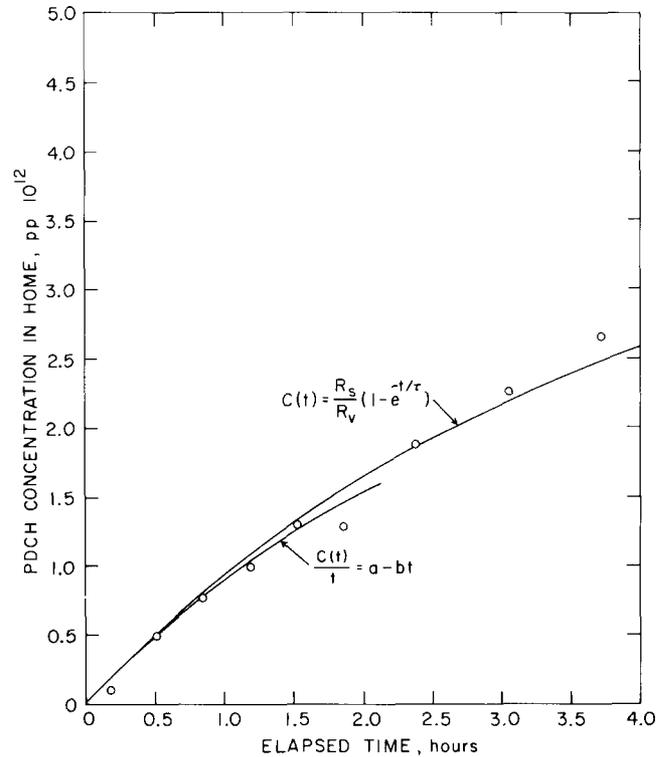


Fig. 7. Tracer concentration in home after placement of one PFT source in each room (pp 10¹² is equivalent to pL/L).

to be constant, Eq. (1) can be integrated, yielding, for an initial concentration of zero,

$$C(t) = \frac{R_s}{R_v}(1 - e^{-t/\tau}), \quad (6)$$

where $C(t)$ is the time dependent tracer concentrations (pL/L), that is, $V(t)/V_h$, and τ is the time for one complete change of air in the home (h), that is, V_h/R_v . Note that $1/\tau$ is equivalent to the familiar AC, the number of air changes per unit of time. Equation (6) can be expanded:

$$C(t) = \frac{R_s}{R_v} \left(\frac{t}{\tau} - \frac{1}{2} \frac{t^2}{\tau^2} + \frac{1}{6} \frac{t^3}{\tau^3} - \dots \right), \quad (7)$$

which, for short times ($t/\tau < 0.5$), can be approximated by just the first two terms of the expansion. Rearranging yields,

$$\frac{C(t)}{t} = \frac{R_s}{R_v\tau} \left(1 - \frac{t}{2\tau} \right) = a - bt, \quad (8)$$

which is the equation for a straight line. Since

$$a = \frac{R_s}{R_v\tau} = \frac{R_s}{V_h}, \quad (9)$$

Eq. (9) can be used to calculate the volume of the house from the intercept. Also, from Eq. (8),

$$\frac{a}{b} = 2\tau. \quad (10)$$

Thus, Eq. (10) can be used to calculate τ from the slope and the intercept.

To use data at a time greater than t/τ of 0.5, Appendix A shows that an adjusted $C(t)/t$ value can be used in Eq. (8) to give

$$\begin{aligned} \text{Adjusted } \frac{C(t)}{t} &= \frac{C(t)}{t} - a[(1 - e^{-t/\tau}) \frac{\tau}{t} + \frac{1}{2} \frac{t}{\tau} - 1] \\ &= a - bt. \end{aligned} \quad (11)$$

First, the early data are used in Eq. (8) to obtain estimates of a and τ , which in turn are then substituted in Eq. (11) to derive an adjusted $C(t)/t$, which is then plotted versus time to arrive at new estimates of a and τ .

The adjusted $C(t)/t$ data for both infiltration tests are plotted versus time in Fig. 8. Both tests result in straight lines converging at identical intercepts, which is to be expected since the intercept is related to the volume of the house, identical in both tests. The different slopes imply different infiltration rates for the two tests.

Using the slopes and intercepts from a least-mean-square fit of the data gives the results shown in Table 6.

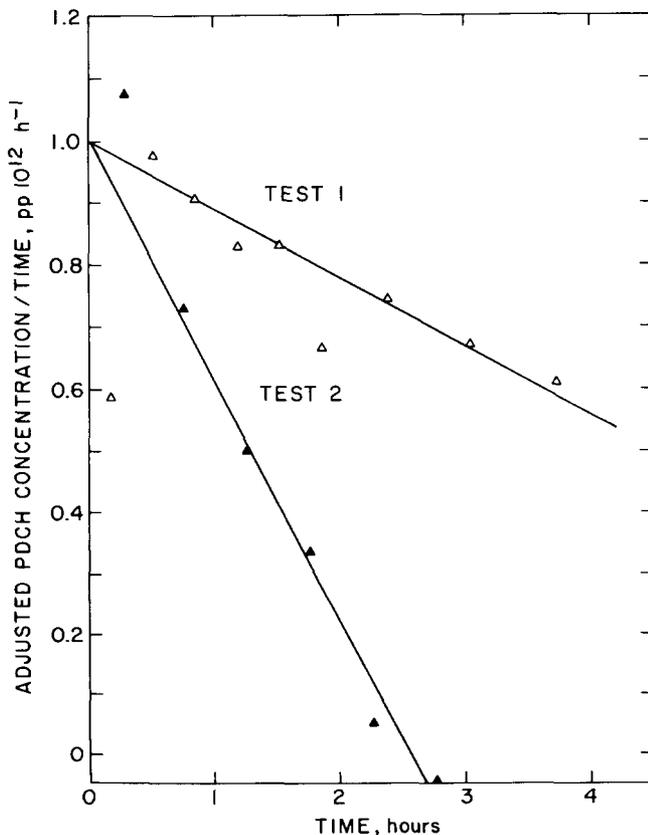


Fig. 8. Linearization of initial concentration function. Identical intercepts imply the same volume for the house; different slopes imply different infiltration rates (pp 10^{12} is equivalent to pL/L).

The PDCH source in the basement was excluded from the calculation in order to calculate the volume of the main floors of the house, which was in excellent agreement with the actual measured house volume of 505,000 L. The infiltration rates were different in the two tests by about a factor of 3.5. As will be shown later, the difference in infiltration rates is explained by the average temperature difference inside and outside the house, which during Test 1 was only about 3°C, but during Test 2 was about 15°C.

It has been demonstrated from these tests that by using a programmable sampler in conjunction with the miniature PFT sources, sufficient data can be collected in as little as 2–3 h to determine the infiltration rate as well as the volume of the house.

Extended duration tracer release

Over long periods of time, that is, many hours or days, the infiltration rate will not be constant. For example, it is known to vary quite markedly with temperature differential and with wind speed (Malik, 1978; Shaw, 1981). Equation (1) can be rearranged

$$\begin{aligned} R_v(t) &= \frac{V_h}{V(t)} (R_s - dV(t)/dt), \\ &= R_s \frac{V_h}{V(t)} \left(1 - \frac{dV(t)/dt}{R_s} \right), \\ &= \frac{R_s}{C(t)} \left(1 - \frac{dC(t)/dt}{R_s/V_h} \right). \end{aligned} \quad (12)$$

Considering programmable sampler data collected at midpoint times of t_1 , t_2 , and t_3 with concentrations (pL/L) of $C(t_1)$, $C(t_2)$, and $C(t_3)$, respectively, Eq. (12) can be written as

$$R_v(t_2) = \frac{R_s}{C(t_2)} \left(1 - \frac{dC(t_2)/dt}{R_s/V_h} \right), \quad (13)$$

where the slowly changing concentration with time can be estimated from the equation developed in Appendix B,

$$\frac{dC(t_2)}{dt} = \frac{C(t_3) - C(t_1)}{t_3 - t_1}. \quad (14)$$

If desired, the infiltration rate calculated from Eq. (13) can be converted to an air exchange rate simply by dividing by the volume of the house. The use of Eqs. (13) and (14) for extended-duration data is demonstrated for both tests.

Test 1. After about 18 h into the tracer test, a fire was started in the open fireplace. The effect was quite dramatic as shown in Fig. 9. The data in the figure are presented in Table 7; Eqs. (13) and (14) were used to calculate the infiltration rate at each sampling midtime.

Table 6. Volume of house and infiltration rate (calculated from best fits of Fig. 7).

Test	R_s (pL/min) ^a	V_h (L) ^b	τ (h)	AC (h ⁻¹)	R_v (L/min)
1	9100	544,000 ($\pm 2\%$)	4.5 ($\pm 15\%$)	0.22	2010
2	8500	503,000 ($\pm 6\%$)	1.28 ($\pm 14\%$)	0.78	6550

^aExcludes source in basement.

^bMeasured volume of house was 505,000 L.

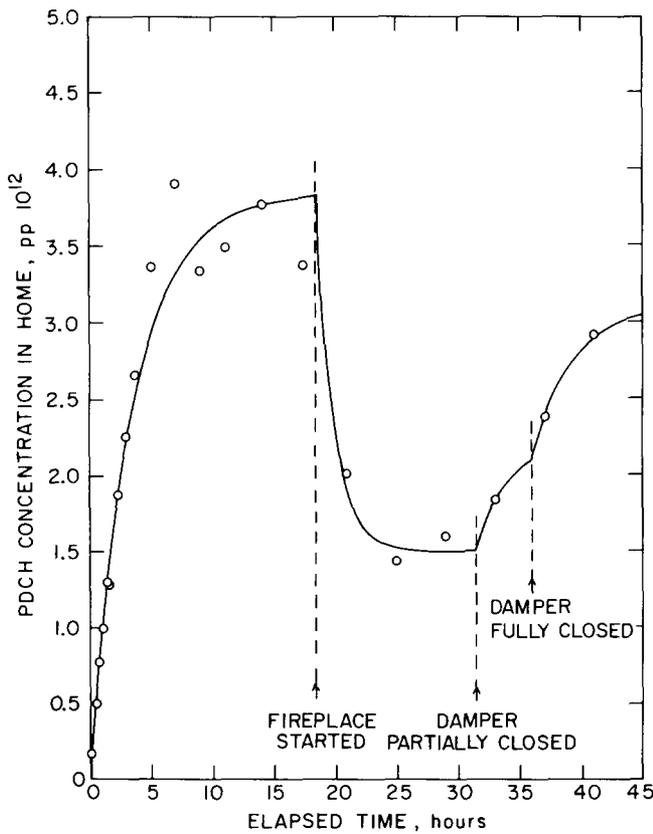


Fig. 9. Effect of starting an open fireplace on the steady-state PFT concentration during Test 1. Circles are data from BATS; line is theoretical best fit (pp 10¹² is equivalent to pL/L).

Before the fire was started, the average R_v for the first 16 h was 2380 L/min ($\pm 20\%$), in very good agreement with the initial 2-h estimate of 2010 L/min (cf. Table 6). For the next 13 h, with the fireplace lit, R_v was determined to be 5940 L/min ($\pm 11\%$), that is, 2.5 times higher with the open fireplace operating.

When the infiltration rate is constant for a period of time and then changes abruptly to another value for a further period, Eq. (1), which can be written as

$$\frac{dV(t)}{dt} = V_h \frac{dC(t)}{dt} = R_s - R_v C(t), \quad (15)$$

can be integrated over a constant ventilation rate (R_v) time period from t_0 to t_a to give

$$\ln \frac{\frac{R_s}{R_v} - C(t_0)}{\frac{R_s}{R_v} - C(t_a)} = \frac{R_v}{V_h} (t_a - t_0), \quad (16)$$

where $t_0 < t_a \leq t_1$. In the next time period, $t_1 < t_b \leq t_2$, the infiltration rate is assumed to be a different constant value, R'_v , resulting in the following expression:

Table 7. House infiltration test 1—extended duration (November 2-4, 1979).

Sample Period	t_m (h)	$C(t_m)$, Concentration PDCH (pL/L)	$R_v(t_2)$ (L/min)
2123-2203	3.73	2.66	
2205-0005	5.09	3.36	1679
0005-0205	7.09	3.91	2345
0205-0405	9.10	3.34	3007
0406-0606	11.10	3.49	2401
0606-1006	14.10	3.77	2453
1006-1300	17.45	3.37	3376
Fireplace start (1230)			
1300-1700	21.00	2.01	5716
1700-2100	25.01	1.44	6665
2100-0100	29.01	1.60	5427
Damper partly closed (0130)			
0101-0501	33.02	1.84	4392
0501-0901	37.02	2.38	3227
Damper fully closed (0900)			
0901-1301	41.03	2.92	

^aSample midtime.

$$\ln \frac{\frac{R_s}{R'_v} - C(t_1)}{\frac{R_s}{R'_v} - C(t_b)} = \frac{R'_v}{V_h} (t_b - t_1). \quad (17)$$

For the first period shown in Table 7, that is, from $t_0 = 0$ to $t_1 = 18.5$ h, which corresponded to the time when the fireplace started (1230), an average infiltration rate of 2380 L/min was used to compute the curve shown in Fig. 9. Then, over the next time period, from $t_1 = 18.5$ h and $C(t_1) = 3.82$ pL/L [calculated from Eq. (16)] to $t_2 = 31.51$ h (when the damper was partially closed), Eq. (17) was used with R'_v set equal to 6000 L/min, i.e., close to the average value shown in Table 7. As shown in Fig. 9, this procedure provides a good fit to the experimental data.

Test 2. The second test in the same home was conducted about 1 month later for a total period of about 17 days (November 30–December 17, 1979). Exactly 24 h after the sources had been placed in the house, six CATS passive samplers were also placed at various locations where they remained for about 15.5 days. For three weekend periods, a BATS programmable sampler was placed at various locations throughout the house.

It should be mentioned that between the first test and the second, a wood-burning stove insert was installed in the fireplace. The stove was used intermittently throughout the second test period, but, as shown in Fig.

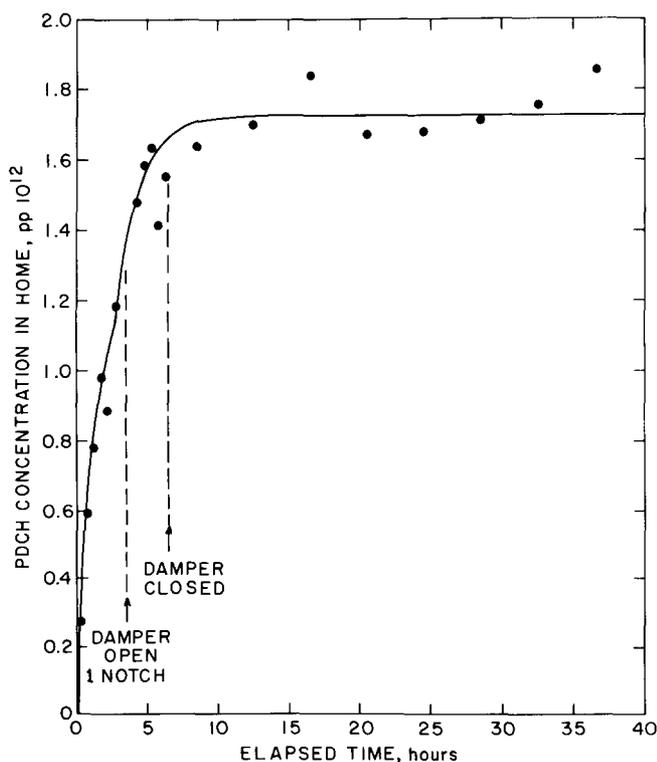


Fig. 10. Initial tracer concentration in home during Test 2 showing negligible effect of wood-burning stove on infiltration rate (pp 10^{12} is equivalent to pL/L).

10, which covers the first weekend period of the second test, the deliberate opening of the damper one position beyond its normal position appeared to have little or no effect on the measured infiltration rates. Using Eqs. (13) and (14), the infiltration rate was calculated for the five points when the damper was open one notch extra, as well as for the four points immediately after closing the damper (the fire was extinguished). The average infiltration rate before the damper was opened was 6550 ± 1500 L/min (taken from Table 6); after the damper was opened, 5255 ± 930 L/min; and after closing the damper and extinguishing the fire, 4930 ± 240 L/min. Thus, the fireplace operating with the stove insert does not appear to contribute to an increase in air infiltration. Certainly, the reduction in air flow through the stove is at least an order of magnitude less than through the open fireplace.

All of the data collected from the three BATS sampling periods is displayed in the lower portion of Fig. 11. Starting with the second weekend period, the BATS was moved to several locations to determine whether there was any variability in the tracer concentration due to sampling location. It must be pointed out that during these tests the temperature difference inside and outside the house varied from a low of 10°C to a high of 19°C as shown by the values in the upper portion of Fig. 11, which is a plot of the infiltration rates calculated from Eqs. (13) and (14). Thus, any attempt to examine differences in concentration or infiltration rate must consider both location and temperature difference.

Sampling was done at the floor level (f), table height (t), and near the ceiling (c) in the dining room as shown in Fig. 11. In general, taking the temperature differences into account, it appeared that the concentrations were highest near the ceiling and lowest near the floor. This is not overly surprising, since the general flow of air infiltrating into a house is caused by warm air rising and exfiltrating near the ceiling of rooms, being replaced by cool, outside air infiltrating near the floor level. By connecting the ceiling concentration points at 182 and 192 h with a straight line, and the table height values at 186 and 188 h with another line, it appears that the average PDCH concentration near the ceiling was 2.0 pL/L; at table height, 1.85 pL/L; and near the floor, 1.7 pL/L. Similarly, working with the data between 353 and 361 h gave 2.2, 1.9, and 1.7 pL/L, respectively, at the ceiling, table, and floor locations. This gradient may have existed in part because the PDCH source in the dining room was located about 2 ft from the ceiling and only 4 ft from the ceiling sampling location. The fact that the floor level concentration was only 15%–23% less than the ceiling concentration was actually good evidence for the assumption of well-mixed conditions in the rooms of the house.

The mixing from room to room was also evidenced by the data collected from 203 to 222 h. The upstairs hall did not contain a source, but its concentration (1.7

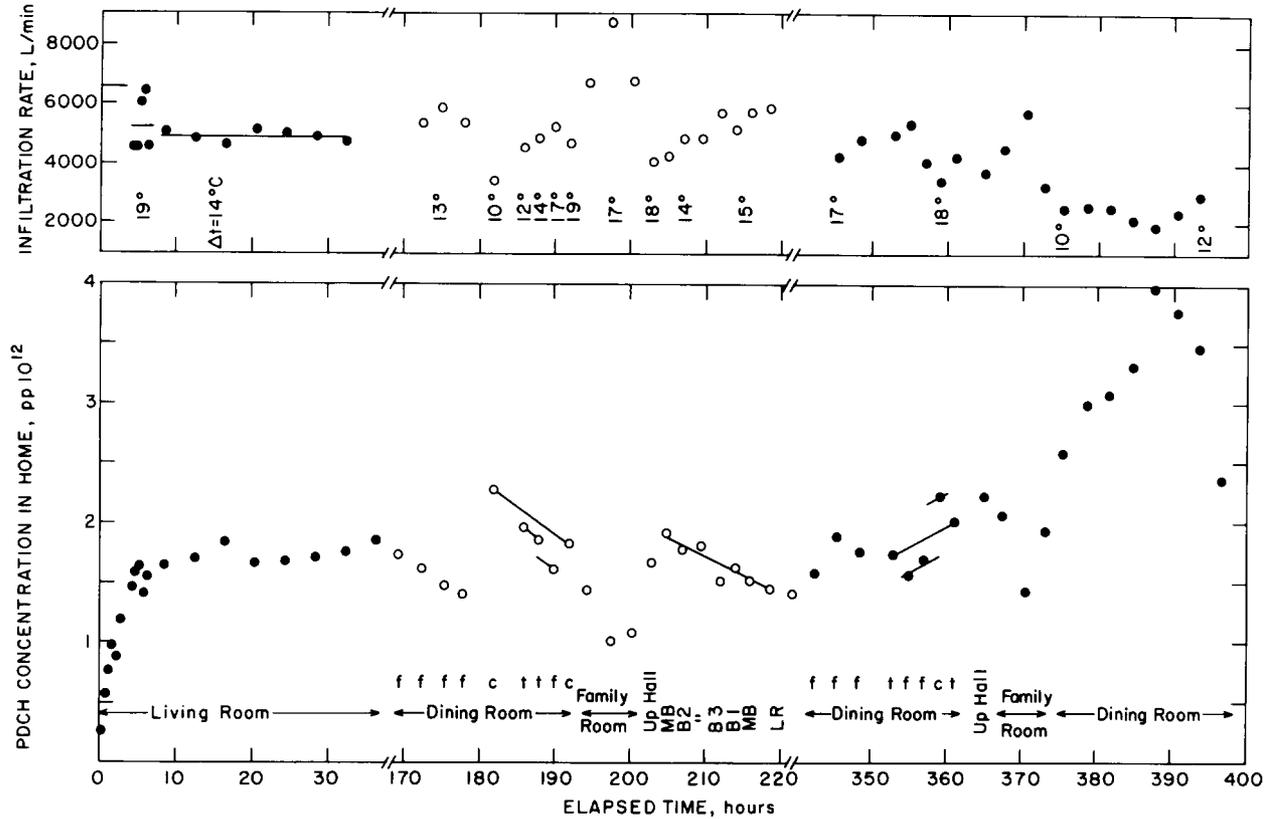


Fig. 11. Extended testing period (~16 days) showing steady-state PDCH concentration as a function of time (lower plot) and the calculated infiltration rate (upper plot), including the average inside/outside temperature difference (pp 10¹² is equivalent to pL/L).

pL/L) was only 8% less than the average 1.85 pL/L found in the master bedroom (MB) and bedroom 2, which did contain PDCH sources. Similarly, bedroom 3 did not contain a source but its concentration (1.5 pL/L) was only 9% less than the average between bedrooms 2 and 3, which did contain sources. Thus, it is apparent that placing a few PFT sources in each level of a house, even one without forced air circulation, provides a good approach to well-mixed, constant emission of tracer within a house.

Measurements made in the family room during both the second (194–201 h) and third (367–373 h) weekend sampling periods, confirmed that the infiltration rate was, on the average, higher (see upper portion of Fig. 11), and the concentrations lower than for the other areas of the house. This was not surprising for two reasons. First, the family room was the only room with three exposed outside walls (two to the outside and one to the unheated attached two-car garage). In addition, the room contained a sliding glass door. Both these factors contributed to a higher air infiltration rate. Second, because the family room contained the wood-burning fireplace insert, it was, on the average, about 5.6°C warmer than the rest of the house, also leading to a higher infiltration rate.

Effect of temperature on infiltration

For the last sampling period, from 340 to almost 400

h, the temperature differential between the home and the outside decreased from about 18°C to 10°C. As shown in Fig. 11, this caused the concentration of PDCH to increase, essentially doubling during this almost halving of the temperature differential. The result was that the calculated infiltration rates were reduced by about one-half. All the measurements in the dining room were made near the ceiling level during the period from 375–397 h, so the actual midlevel concentration may have been about 15%–25% lower, based on observations presented earlier.

The effect of temperature difference can be quantified by averaging the calculated infiltration rates for periods when the temperature difference was reasonably constant. This was done for several periods, as shown in Table 8, including both Tests 1 and 2. The first six entries are for measurements made in the main living areas such as the living and dining rooms, and the last two were for the family room, in which the temperature was generally 5–6°C warmer because of the wood-burning stove. Temperatures both inside and outside the house were noted and the average differences for the respective periods are shown in the table.

Although some researchers have demonstrated a logarithmic dependence of infiltration rate on temperature (Shaw, 1981), others have used a simple linear dependence (Malik, 1978), which we assumed. Table 8 shows the linear relationship found (see table footnote

Table 8. Infiltration rate versus temperature difference.

Date	Average Δt , ($^{\circ}\text{C}$) ^a	$R_i(t)$ (L/min)	
		Measured ^b	Calculated ^c
11/2/79	2.8	2380 \pm 470	2382
12/1/79	14.4	4920 \pm 190	4586
12/8/79	16.7	4880 \pm 780	5023
12/9/79	13.9	4960 \pm 640	4491
12/15/79	15.6	4370 \pm 640	4814
12/16/79	10.0	2350 \pm 300 ^d	3750
		Family Room	Calculated ^c
12/8/79	22.3	7404 \pm 1142	6087
12/16/79	15.6	4530 \pm 1235	4814

^aTemperature difference inside and outside the house.

^bMeasured in living or dining areas.

^c $R_i(t) = 1850 + 190\Delta t$; $r^2 = 0.89$.

^dUsed a value of 3000 since measurement was at ceiling level.

c) and gives the values calculated for comparison with the measured values. The correlation was fairly good (coefficient of determination of 0.89) for the 3–22 $^{\circ}\text{C}$ range in temperature differences.

This temperature correlation is used in the next section to compare measured infiltration rates from the CATS devices with the average degree days during the 15.6 days of sampling.

Results from CATS

Each of the six CATS tubes had been sampling the home air for a total of 374.2 h, or 15.6 days. Based on the verified rate of diffusion sampling for PDCH, it was calculated that the CATS devices sampled the equivalent of 197 mL of air. Thus, the average concentration of PDCH in the home at the various locations was calculated from the volume of tracer measured by the gas chromatograph and the volume of air sampled. As shown in Table 9, the concentrations were quite similar to those found by the programmable samplers.

At steady-state and for a constant infiltration rate, the change in concentration with time, $dC(t)/dt$, is

Table 9. CATS infiltration results: 1800 (12/1/79) to 0810 (12/17/79)^d; 223.3 $^{\circ}\text{C}$ days, avg. (14.9 $^{\circ}\text{C}$).

CATS No.	Location	Conc. PDCH (pL/L)	R_i (L/min)	
			Measured	Calculated ^b
1	Dining room	1.63	5157	
2	Living room (in)	2.07	4054	
3	Living room (out)	1.55	5426	
4	Family room ^a	1.33	6321	5745
5	Master bedroom	leak	leak	
6	Bedroom 3	1.99	4215	
	Average		4713 \pm 680 ^c	4674

^aFamily room was about 5.6 $^{\circ}\text{C}$ warmer than house average.

^b $R_i = 1850 + 190(\Delta t)$.

^cAverage excluding family room.

^dTotal sampling duration was 374.2 h (15.6 days).

zero. Thus, Eq. (13) implies that the infiltration rate can be calculated by dividing the total source rate, about 8400 pL/min, by the PDCH concentration. The resulting average measured infiltration rates are shown in Table 9. Excluding the family room value, the average for the house was 4713 L/min.

During this 15.6-day period, the number of degree days accumulated was a total of 223 $^{\circ}\text{C}$ days for an average of 14.9 $^{\circ}\text{C}$ days. Since the average temperature in the house was about 65 $^{\circ}\text{F}$ (18.3 $^{\circ}\text{C}$), the temperature base for the degree day measurement, the 14.9 $^{\circ}\text{C}$ can be assumed to be the average temperature difference between the inside and outside temperature during this period. Using the temperature effect correlation developed from the data in the previous section, the average calculated infiltration rate was 4674 L/min, in excellent agreement with that measured by the CATS device. Thus, simply by measuring the infiltration rate with a CATS device and recording the number of degree days, an infiltration rate normalized for temperature differences can be readily obtained.

In these tests, the wind speed was generally below 3 m/sec; below this value, wind speed did not generally play too significant a role on infiltration rate (Shaw, 1981).

The higher average temperature in the family room was also apparent from the higher average infiltration rate, and was also in good agreement with the expected calculated value. CATS No. 6 had been placed in bedroom 3, the one without a source. The average infiltration rate for that room was within one standard deviation of the average for all rooms, again substantiating that the mixing from room to room was quite adequate. In fact, even the spread between the two CATS located in the living room was within one standard deviation. CATS No. 2 was near a corner of two inside walls and No. 3 near a corner of two outside walls. Based on the relative locations in the living room, it was expected that the one near an outside wall would have an apparent higher infiltration rate.

PFT Method versus SF₆ Tracer Decay

Several SF₆ tracer decay experiments were conducted in the BNL Solar House while, simultaneously, PFT concentrations were being measured with the BATS programmable samplers. Between 8 and 10 L of air containing 85 ppm ($\mu\text{L/L}$) of SF₆ were released uniformly throughout both floors of the 2-story house; the logarithmic decay in concentration with time was monitored with the Brookhaven continuous tracer monitor (Dietz and Goodrich, 1980) for about 3 h.

Four PDCH permeation sources were deployed uniformly throughout the house, two placed downstairs and two upstairs. During each SF₆ decay test, the uniform, steady-state PDCH concentration was measured every 45–60 min with BATS units both up and

downstairs. In the first test, a fan was forcing circulation between the solar porch and the balance of the home; during the second test the next day, this fan was turned off.

Test 1 (March 23, 1982)

The three hourly averaged PDCH concentrations were 12.8 and 12.5 pL/L up and downstairs, respectively, within $\pm 5\%$. Since the total PDCH emission rate was 61.8 nL/min at the average house temperature of 24°C, the infiltration rate, R_v , was 61,800/12.7 or 4870 ± 400 L/min. Since the measured volume of the house was 511,000 L, including the solar porch, the number of air changes (AC) was

$$AC = \frac{R_v}{V_h} = \frac{4870 \times 60}{511,000} = 0.57 \pm 0.06 \text{ h}^{-1}.$$

About 9.8 L of air containing 85 ppm SF₆ were uniformly released into a portion of the house and the results were plotted according to

$$\ln C(t) = \ln C(t_0) - (AC)t. \quad (18)$$

By least-mean-square fit, $C(t_0)$ was found to be 2520 ± 56 pL/L and the air changes (AC) was 0.62 ± 0.01 h⁻¹, in excellent agreement with that found using the PFT method.

The doors to one bedroom, both bathrooms, and the closets were closed during the release and decay of the SF₆. The physical volume of the balance of the house into which the SF₆ was released was 391,000 L, including the porch. Since the volume of SF₆ released was 85 $\times 10^6$ pL/L times 9.81 L or 834 $\times 10^6$ pL, the available volume of the house can be calculated by dividing by the initial SF₆ tracer concentration; this gives 331,000 $\pm 25,000$ L, nearly identical to the physical volume.

Test 2 (March 24, 1982)

Four 45-min averaged PDCH concentrations of 15.6 and 16.5 pL/L up- and downstairs, respectively, were found when the porch fan was off (porch volume was not an active volume of the house). The total PDCH emission rate was 57.2 nL/min at the average house temperature of 21°C and, therefore, the infiltration rate was 3575 ± 400 L/min. Without the porch, the physical volume of the entire house was 451,000 L and the number of air changes was

$$AC = \frac{3575 \times 60}{451,000} = 0.47 \pm 0.05 \text{ h}^{-1}.$$

Only 7.8 L of 85 ppm SF₆ were released in this test, resulting in an initial concentration of 2282 ± 73 pL/L and a least-mean-square slope (AC) of 0.39 ± 0.02 h⁻¹, in good agreement with the PFT-derived value. Again, the initial concentration and the amount of SF₆ released

was used to calculate a house volume of 291,000 $\pm 24,000$ L, close to the physical volume of 331,000 L, which excluded that volume associated with rooms whose doors were closed.

As a result of these tests, it can be concluded that the results are essentially identical, within the precision of both tracer methods. Further details of this intercomparison will be provided subsequently.

Conclusions

The perfluorocarbon tracer (PFT) technique is a precise and reliable methodology for the determination of infiltration rates in homes. The present diffusion plug PFT source has an adequate emission rate and lifetime; the age of the PFT sources in these tests was about 230 days (7.5 months old). Since the emission rate of the diffusion plug source was both temperature and time dependent, PFT permeation-type sources were manufactured and evaluated. These sources have emission rates which are about an order-of-magnitude higher than the diffusion source, are not dependent on age, and have a tolerable dependence on temperature. For a 3°C change in temperature, the emission rate changes by only about 8%.

Two types of samplers for PFTs in home infiltration studies have been successfully deployed. The passive sampler, CATS, tested both in the laboratory and in home infiltration studies, was shown to have a predictable performance and to be reliable for obtaining average PFT concentrations in homes for periods of up to several weeks, accurately sampling concentrations as low as 1 pL/L. The programmable sampler, BATS, which has been laboratory and field tested for a number of atmospheric tracing experiments, was extremely useful for measuring the initial PFT concentration increase in a home and for studying the effects of weather and other variables on home infiltration rates. For example, a 2.5-fold higher infiltration rate when burning wood in an open fireplace was readily demonstrated. Essentially no further development of the sampling approach is needed.

In the first 2–4 h following deployment of PFT sources in a home, the programmable sampler provided the necessary measurements in order to calculate the volume of the house and the rate of air infiltration. Subsequent longer sampling periods with the BATS unit provided the necessary measurements for assessing the effect of the indoor/outdoor temperature difference on infiltration rates.

A 17-day infiltration test was successfully conducted to demonstrate the utility of these Brookhaven infiltration measurement tools. The sources provided sufficient, controlled tracer release to attain steady-state PFT concentrations of between 1 and 4 pL/L, which were readily measurable with the programmable and passive samplers. Periods of measurements with the

BATS programmable sampler were used to relate infiltration rate with the inside/outside temperature differential. It was shown that the 16-day average infiltration measurement obtained with a passive CATS sampler was essentially identical to that measured with the programmable sampler.

Uniform mixing of tracer within the home was demonstrated by making measurements near the floor and the ceiling of a room, as well as comparing PFT concentrations in rooms containing the PFT source with those not containing a source. These differences were no more than 20% to 25%, usually only about 10% to 15%. Rooms with more outside-wall exposure (for example, a typical family room adjacent to an attached garage) were shown to have higher infiltration rates than those with less wall exposure.

The PFT tracer method was successfully compared with two SF₆ tracer decay tests. Both methods yielded air exchange rates which were identical within the precision of the systems, that is, about ±15%.

The Brookhaven AIMS (air infiltration measurement system), a kit comprised of a miniature PFT source and miniature passive sampler, is an effective but simple approach to the implementation of wide-scale determinations of air infiltration using perfluorocarbon tracers.

APPENDIX A. Initial Tracer Release Period

A more explicit form of the linear dependence of $C(t)/t$ on time can be developed by rearranging Eq. (7):

$$\frac{C(t)}{t} = \frac{R_s}{R_v \tau} \left(1 - \frac{1}{2} \frac{t}{\tau} + \frac{1}{6} \frac{t^2}{\tau^2} - \dots \right), \quad (A1)$$

which, using the substitutions defined by Eq. (8) in the text, is equivalent to

$$\frac{C(t)}{t} = a - bt + a \left(\frac{1}{6} \frac{t^2}{\tau^2} - \dots \right) \quad (A2)$$

or

$$\frac{C(t)}{t} - a \left(\frac{1}{6} \frac{t^2}{\tau^2} - \dots \right) = a - bt. \quad (A3)$$

The terms in parentheses on the left are part of the exponential expansion

$$1 - e^{-t/\tau} = \frac{t}{\tau} \left(1 - \frac{1}{2} \frac{t}{\tau} + \frac{1}{6} \frac{t^2}{\tau^2} - \dots \right) \quad (A4)$$

or

$$(1 - e^{-t/\tau}) \frac{\tau}{t} + \frac{1}{2} \frac{t}{\tau} - 1 = \left(\frac{1}{6} \frac{t^2}{\tau^2} - \dots \right) \quad (A5)$$

Substituting Eq. (A5) into Eq. (A3) gives

$$\begin{aligned} \frac{C(t)}{t} &= a \left[(1 - e^{-t/\tau}) \frac{\tau}{t} + \frac{1}{2} \frac{t}{\tau} - 1 \right] \\ &= \text{adjusted} \frac{C(t)}{t} = a - bt. \end{aligned} \quad (A6)$$

APPENDIX B. Estimate of the Change in Concentration with Time

In order to solve Eq. (13), an estimate must be made for the derivative of the function approximating the variability of PFT concentration in a home with time. By differentiating Stirling's formula for a function expressed in terms of differences which are in the same horizontal line (Whittaker and Robinson, 1946), the derivative in Eq. (13) can be estimated by

$$\begin{aligned} \frac{dC(t_2)}{dt} &= \frac{1}{\Delta t} \left[\frac{\Delta C(t) + \Delta C(t - \Delta t)}{2} \right. \\ &\quad - \frac{\Delta^3 C(t - \Delta t) + \Delta^3 C(t - 2\Delta t)}{12} \\ &\quad \left. + \frac{\Delta^5 C(t - 2\Delta t) + \Delta^5 C(t - 3\Delta t)}{60} - \dots \right] \quad (B1) \end{aligned}$$

Assuming that only the first term is significant, Eq. (B1) becomes

$$\frac{dC(t_2)}{dt} = \frac{C(t_3) - C(t_2) + [C(t_2) - C(t_1)]}{2\Delta t}, \quad (B2)$$

which simplifies to

$$\frac{dC(t_2)}{dt} = \frac{C(t_3) - C(t_1)}{t_3 - t_1} \quad (B3)$$

for the case when the sampling time intervals are equal.

To demonstrate that the approximation of Eq. (B3) causes negligible error, data was used to calculate infiltration rates using the approximation as well as the first three terms [Eq.(B1)] in the expansion. As shown in Table B1, the infiltration rates calculated by the approximation were very close to those using Eq. (B1), generally within ±0.4%. Therefore, Eq. (B3), which is the slope of the line between the first and third data points, is a good estimate of the slope at the second data point.

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Table B1. Effect of simplified derivative on infiltration rate (December 1-2, 1979).

t_m (h)	$C(t_m)_3$ Concentration PDCH (pL/L)	Infiltration Rate, $R_s(t_2)$ (L/min)	
		Using Eq. (B1)	Using Eq. (B3)
4.54	1.451		
8.55	1.641		5020
12.56	1.701	4855	4873
16.56	1.841	4642	4632
20.57	1.674	5228	5178
24.57	1.681	5000	5032
28.58	1.714	4916	4913
32.58	1.756		4755
36.58	1.857		

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