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EVALUATION OF
PERCHLOROETHYLENE EMISSIONS
FROM DRY CLEANED FABRICS

Prepared for

Office of Toxic Substances

Prepared by

Air and Energy Engineering Research Laboratory Research Triangle Park NC 27711

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EVALUATION OF PERCHLOROETHYLENE EMISSIONS FROM DRY CLEANED FABRICS

bу

Bruce A. Tichenor Leslie E. Sparks Merrill D. Jackson

U.S. Environmental Protection Agency
Air and Energy Engineering Research Laboratory
Indoor Air Branch
Research Triangle Park, NC 27711

Prepared for:

U.S. Environmental Protection Agency Office of Research and Development Washington, DC 20460

Abstract

A study was conducted to evaluate the emissions of perchloroethylene* (perc) from dry cleaned fabrics to determine: a) how the introduction of fresh dry cleaning into a home affects the indoor concentration of perc, and b) the effectiveness of "airing out" for reducing perc emissions. Small chamber tests were conducted to determine perc emission characteristics for three fabrics at several temperatures and air exchange rates. Test house studies were conducted to determine the indoor concentration of perc due to the placement of dry cleaned clothing in the house. Based on the study results, and assuming that test conditions were representative of normal dry cleaning and consumer practices, the following conclusions were reached:

- 1) Emissions from freshly dry cleaned clothing cause elevated levels of perchloroethylene in residences.
- 2) For the three fabrics tested, "airing out" of dry cleaned clothing by consumers will not be effective in reducing perchloroethylene emissions.

It is emphasized that these conclusions are based on the results of the study reported. Significant variations in dry cleaning practices and/or in the mix of fabrics and clothing being cleaned could provide different results and conclusions.

Perchloroethylene is the common name for tetrachloroethylene; C2Cl4. It has a molecular weight of 166. This document reports concentrations in ug/m³ at standard conditions (0°C, 1 atmosphere). To convert to ppb, multiply by 0.135.

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ACKNOWLEDGMENT

The authors acknowledge the substantial contributions of the following individuals: Zhishi Guo for developing and executing the techniques used to analyze the small chamber data; Mark Mason and Michelle Plunket for conducting the small chamber tests; Susan Rasor for performing the IAQ test house experiments; and Glenn Smith for conducting the perc residuals evaluations. All of these individuals are employees of the Acurex Corp. and performed their work under EPA Contract No. 68-02-4701.

EXECUTIVE SUMMARY

The Indoor Air Branch (EPA's Air and Energy Engineering Research Laboratory -- EPA/AEERL) conducted a short term study to evaluate the emissions of perchloroethylene (perc) from dry cleaned fabrics. Specifically, the study was designed to answer two questions:

- A) To what extent does the residual perc in dry cleaned fabric increase the concentration of perc in residential environments?
- B) How effective is "airing out" in reducing indoor perc concentrations?

A study consisting of five components was conducted:

1) Fabric/Clothing Selection; 2) Emission Factor Determination
(Small Chamber Testing); 3) Evaluation of Perc Residuals (Solvent Extraction); 4) Indoor Air Quality (IAQ) Model Analysis; and

5) Evaluation of Indoor Concentrations (Test House).

The following results were achieved:

Emission Factors

Emission factors for perchloroethylene from dry cleaned fabrics were determined by testing in small environmental test chambers under controlled conditions. Evaluation of the data from these tests provided the following:

- A preliminary screening evaluation showed that wide variations in initial emission factor, R_{o} , and emission factor half-lives, t(1/2), occurred between different fabrics. Thus, the type of fabric is important in determining indoor emissions of perc from dry cleaned clothes.
- Based on the screening study and on the prevalence of fabrics used in dry cleaned clothing, three fabrics were selected for investigation: 55% polyester/45% wool; 100% wool; and 50% polyester/50% rayon.
- The air exchange rate showed no effect on the emission factor or decay rate for the three fabrics investigated. This suggests that the emissions are limited by the diffusion of perc within the fabric and are not controlled by evaporative processes. This also suggests that increasing the ventilation by airing out the clothes will not speed up the emission of perc.
- Since the three fabrics tested had emission factor halflives of about a day, airing the clothes out for a few hours before hanging them in the home will do little to reduce the indoor perc concentrations. For fabrics with faster perc decay rates, airing out may be more practical.

- Temperature had a major impact on the emission factors and decay rates. Increases in temperature caused higher initial emission factors and lower half-lives. Thus, exposing the clothing to higher temperatures prior to bringing them home shows promise as a means of reducing in-home exposure to perc.

Residuals

No acceptable data were developed on the perc residuals within the fabric. The solvent extraction procedure, using methylene chloride, failed to produce reliable results. A fully tested "standard method" is needed.

Indoor Concentrations

All the test house experiments showed that the introduction of dry cleaned clothing caused elevated levels of perc in the house. Differences in concentration between the tests were probably due to differences in the amount of perc retained at the dry cleaner.

Model Results

The IAQ model, using emissions data developed in the small chambers, predicted indoor perc concentrations which compared favorably with those measured in the test house. The effect of perc "sinks" in the test house was also demonstrated.

Conclusions

Based on the study results, and assuming that test conditions are representative of normal dry cleaning and consumer practices, the following conclusions are reached:

- 1) Emissions from freshly dry cleaned clothing cause elevated levels of perchloroethylene in residences.
- 2) For the three fabrics tested, "airing out" of dry cleaned clothing by consumers will not be effective in reducing perchloroethylene emissions.

It is emphasized that these conclusions are based on the results of the study reported herein. Significant variations in dry cleaning practices and/or in the mix of fabrics and clothing being cleaned could provide different results and conclusions.

1. INTRODUCTION

A. Study Objectives

The Indoor Air Branch (EPA's Air and Energy Engineering Research Laboratory -- EPA/AEERL) conducted a short term study to evaluate the emissions of perchloroethylene* (perc) from dry cleaned fabrics. Specifically, the study was designed to answer two questions:

- A) To what extent does the residual perc in dry cleaned fabric increase the concentration of perc in residential environments?
- B) How effective is "airing out" in reducing indoor perc concentrations?

B. Factors Affecting Emissions and Indoor Concentrations

A number of factors may affect the <u>amount of residual perc</u> in dry cleaned fabrics, including:

Type of fabric - Brodmann (1975) reports that the residual perc after cleaning varies widely between fabrics. In general, synthetic fabrics retained more perc than natural fabrics. Brodmann's evaluation of perc residuals immediately and 24 hours after treatment in a coin operated machine indicates:

Fabric Type	% Perc Retained** (Immediate)	% Perc Retained (after 24 hrs)
Arnel (triacetate)	0.80	0.41
Acetate (diacetate)	0.46	0.21
Polypropylene	0.82	0.20
Spun Dacron 54	0.18	0.12
Spun Dacron 64	0.07	0.07
Polyester Double Knit	0.09	0.05
Nylon 66	0.09	0.04

^{**%} of fabric weight

All other fabrics tested (Orlon, Acrilan, wool, Antron, cotton, and fiberglass) showed immediate residuals of < 0.02% and no detectable residuals after 24 hours.

Perchloroethylene is the common name for tetrachloroethylene; C2Cl4. It has a molecular weight of 166. This document reports concentrations in ug/m³ at standard conditions (0°C, 1 atmosphere). To convert to ppb, multiply by 0.135.

Type of treatment - Two types of dry cleaning machines are in use: "transfer" and "dry-to-dry." Data on the perc residuals from these two machines are very limited. Pressed fabric should have lower perc residuals than unpressed fabric.

Variability of treatment - Perc residuals may be affected by the drying cycle and the aeration step, as well as by such factors as age and condition of cleaning equipment, amount of material cleaned per load, operator technique, and solvent purity/age. While data on the effect of such factors are unavailable, such variables could cause differences in perc residuals between cleaning establishments and between loads at the same establishment.

Several additional factors are important with respect to the emission rate of perc from dry cleaned fabrics:

Environmental variables - Temperature, humidity, ventilation (air exchange), and the concentration of perc in the air may affect the rate at which residual perc is emitted from fabrics.

Type of fabric - The rate of perc emissions varies between fabric types. For example, Brodmann's data (shown above) indicate that Spun Dacron 64 retained 100% of the residual perc over a 24 hour period, Nylon 66 retained less than 50%, and polypropylene retained less than 25%.

Storage/handling parameters - A number of variables associated with in-home storage or handling of dry cleaned fabrics may affect the perc emission rate:

- Plastic storage bag retained or removed. Keeping the plastic bag on may reduce the rate of emission, but not the total emitted.
- The amount of dry cleaned fabric stored in a closet. The larger the amount of material stored in a given closet, the greater the total emissions. The emission factor (e.g., mg/m^2-hr) may be lower, however, due to the effect of vapor pressure suppression of evaporation. This would not occur if the emissions are limited by in-fabric diffusion.
- Pre-storage "airing out." Hanging the dry cleaned fabrics outdoors or in a well ventilated area prior to in-home storage may reduce indoor perc emission rates.
- Time since treatment. The amount of residual perc and subsequent emissions to the indoor environment will vary depending on the time between cleaning and placement in the home. Storage at the dry cleaners and transportation time will impact this variable.

Finally, several additional factors may affect the <u>indoor</u> <u>perc concentrations</u>:

Air exchange rate - The air exchange rate (amount of outside air infiltration) determines the dilution and flushing indoors. For a given amount of dry cleaning, the higher the air exchange rate, the lower the indoor perc concentration.

HVAC system - The operation of the HVAC (heating, ventilating, air conditioning) system in the home affects the mixing and movement of air. Air in residences is generally well-mixed when the HVAC fan is operating. This would cause the perconcentrations to be fairly consistent from room to room, except in the closet where the dry cleaning is stored.

Air movement - The amount of air movement between the closet and the adjoining room and between that room and the rest of the home will affect the perc concentrations throughout the residence. Factors such as HVAC operation and open or closed doors are important in affecting air movement.

<u>Sink effects</u> - Materials in the home may adsorb perc at higher concentrations and gradually release it over time. Such an effect would lower initial concentrations but extend the exposure time. Factors such as the amount of clothing in the closet (in addition to the dry cleaned items) could impact the sink effect.

C. Previous Studies

In addition to the study by Brodmann (1975) on perc residuals in dry cleaned fabrics, several other references deal with the issue of perc exposure from dry cleaning. Several industry sponsored articles are available dealing with safe handling of perc in dry cleaning establishments and methods for reducing occupational exposure (Fisher, 1976; HSIA, 1986; IFI, 1987). Fisher (1976) also provides data on perc concentrations measured in dry cleaning plants. Data on non-occupational perc exposure from dry cleaning are limited to studies conducted on alveolar (breath) air from people exposed to dry cleaners (Verberk and Scheffers, 1980; Wallace, 1988). The authors could find no published data on perc concentrations inside residences where freshly dry cleaned clothes were introduced.

2. STUDY DESIGN

In the design of any experimental program, constraints of time and resources limit the scope of the investigation. For the study described herein, these constraints were:

- Study completion by May 1988. (The study was initiated in March.)
- No additional resources.

Thus, the proposed study was limited to available resources and facilities of the Indoor Air Branch and was to be completed in a short time span. Given these constraints, it was obvious that a comprehensive analysis of perc emissions from dry cleaned fabrics which included consideration of all relevant variables (see above) was not possible. The following study plan was developed to best meet the study objectives within the imposed resource and time limits.

A study consisting of five components was conducted:

- Fabric/Clothing Selection;
- 2) Emission Factor Determination (Small Chamber Testing);
- 3) Evaluation of Perc Residuals (Solvent Extraction);
- 4) Indoor Air Quality (IAQ) Model Analysis; and
- 5) Evaluation of Indoor Concentrations (Test House).

A. Fabric/Clothing Selection

Since the available data on perc residuals in fabrics are over 10 years old, changes in types of fabrics and dry cleaning technologies may have occurred which would affect the perc emissions. Thus, prior to the selection of the test fabrics, a "screening study" was conducted. One yard (0.9 m) pieces of 12 fabrics were purchased and dry cleaned. Using fabric pieces instead of clothing allowed appropriate sizes to be cut for use in the small chambers. These samples were placed in small test chambers and preliminary data on perc emission rates were obtained. These data were then used to select three fabrics for further evaluation.

For the test house studies, clothing made of the three selected fabrics was used. A mix of clothing consisting of a man's suit, a woman's skirt, and two blouses was evaluated in the test house.

Treatment was by commercial cleaning at a single facility. The fabric samples and the clothing were handled in a manner consistent with normal dry cleaning operation, including pressing. Protective plastic bags from the cleaners enclosed all cleaned material prior to testing.

B. Emission Factor Determination (Small Chamber Testing)

Small environmental research chambers (six 53-L chambers; and two 166-L chambers) were used to develop data on emission factors for perc from dry cleaned fabrics. Three fabrics (see above) were evaluated. The effect of air exchange (air changes per hour [ACH]) on emission factors was investigated for each fabric to determine the effect of "airing out." The effect of temperature on emission factors was also evaluated. All testing was conducted with a water vapor content equivalent to a 50% relative humidity at 20°C . This test program was designed to provide emission factors for perc (ug/m²-hr), information on the rate of decay of the emissions for the three fabrics, and information on the effects of air exchange and temperature.

C. Evaluation of Perc Residuals (Solvent Extraction)

Testing was conducted to determine the amount of perc within the fabrics. The method selected involved solvent extraction using methylene chloride followed by gas chromatography. Residual testing was conducted before and after emissions testing in the small chambers and the test house.

D. <u>IAQ Model Analysis</u>

The chamber emission factor data were used in an IAQ model to predict expected perc concentrations in the test house based on available data on the air exchange and air movement in the test house. These results were used to design the test house experiments. In addition the model enabled an evaluation of the "sink" effect.

E. Evaluation of Indoor Concentrations (Test House)

Based on the results of the chamber tests and subsequent model analyses, four 2-week test house experiments were conducted. Each test consisted of 1 week of testing followed by 1 week of data evaluation and house "airing" in preparation for the next test. Indoor air samples were collected at three

locations in the house: a) the closet where the clothing was hung, b) the adjacent bedroom, and c) the den. Sampling frequency was determined based on the IAQ model analysis of the small chamber data. A set of background samples were collected prior to each test.

The following four tests were conducted:

- Test A Cleaned clothes hung in empty closet in corner bedroom. Plastic wrap removed. HVAC system on. Closet door closed. All other interior doors open.
- Test B Same as Test A, except plastic wrap not removed.
- Test C Same as Test A, except clothing "aired out" for 4 hours prior to being hung in the closet.
- Test D Replicate of Test A.

F. Quality Assurance/Quality Control

The following QA/QC procedures were designed for the study to ensure the production of data with known, acceptable quality.

1. Calibration Audit for Perchloroethylene

A calibration audit was performed for the analysis of perchloroethylene by gas chromatography (GC). Two audit gas cylinders (one high and one low concentration) were obtained for analysis by the GC in the chamber laboratory and the GC at the test house. The following goals applied: accuracy, \pm 20%; precision, \pm 15%.

2. Standard Rotation

A perchloroethylene standard rotation procedure was implemented between the chamber laboratory and the laboratory where perchloroethylene residual was analyzed. One perc standard from each laboratory was analyzed by the other. The objective for analytical accuracy was $\pm 20\%$; for precision, $\pm 15\%$.

3. Chamber Laboratory

QC measures for the chamber laboratory included documentation and control of the variability of sample size, test chamber temperature, relative humidity, air flow, and precision and accuracy of measurements of the perchloroethylene concentration. Variabilities of sample size, temperature, relative humidity, and air flow were determined for each test. The QA/QC goals for the above parameters were \pm 5%, \pm 1°C, \pm 15%, and \pm 5%, respectively.

The allowable range for the recovery of the internal standard, present in every sample, was \pm 20%. The goal for the relative standard deviation (RSD) between duplicate samples was < 10%.

4. Residual Analysis

For GC calibration, five standard perchloroethylene solutions were prepared, spanning the range of perc concentrations in the fabric sample extracts; for each standard, the goal for the RSD for quadruplicate injections was < 15%.

Duplicate fabric samples were used; for each sample duplicate extract injections were made; the goal for the RSD between duplicate injections was $\leq 15\%$.

In each automatic sampler run (6 - 10 samples), two standards were included, one before and one after the samples. For the run to be accepted, the standards must have a relative error less than 10% from known concentration.

5. Test House

The gas chromatograph for perchloroethylene analysis was calibrated every morning of each sampling day prior to sampling. The GC performance was checked every 3 hours during the sampling day by injecting at least two liquid perc standards with different concentrations.

Duplicate samples were taken from each sampling location. The QA/QC objective for the precision of the duplicate samples was \pm 15%.

3. EXPERIMENTAL PROCEDURES

A. Fabric Treatment

All fabrics (bolt material and clothing) were cleaned at a local dry cleaning establishment. The cleaner used a dry-to-dry machine with a 35 lb (16 kg) capacity operating at 140°F (60°C). Both utility and legger presses were available, with steam traps at 300°F (150°C). The cleaned fabrics were picked up from the cleaners within 1 hour of being cleaned.

B. Small Chamber Testing

1. Facilities and Equipment

AEERL's Indoor Air Source Characterization Laboratory contains two chamber test systems: one with a pair of 166 L test chambers, the other with six 53 L chambers. Each system consists of the following components: a clean air conditioning and delivery system, an incubator containing the environmental test chambers, sampling manifolds, and sample collection adsorbers using Tenax and charcoal. A permeation system for quality The environmental variables are monitored control is included. and controlled by a microcomputer. Organic analyses are conducted by thermal desorption, concentration via purge and trap, and gas chromatography (GC) using flame ionization detection (FID). A separate microcomputer provides GC data analysis. All data are input to spreadsheets for further analysis (Tichenor and Mason, 1988). Figure 1 is a schematic drawing of the 166-L chamber system; Figure 2 shows the 53-L system in less detail.

2. Testing/Measurement Methods

Within 1 hour of being picked up at the dry cleaners, fabrics were cut to size, hung on wire racks, and placed in the test chambers. The first sample was collected within 30 minutes of the start of testing. Several samples were collected on the first day; a sampling frequency of one per day was continued until the end of the test period. Most tests were concluded within 5 days.

Samples were collected by pulling a portion of the chamber air stream through tandem glass cartridges filled with Tenax and Tenax/charcoal sorbents. Sampling was conducted at 100 cm³/min. Sampling time varied from 5 to 100 minutes providing sample volumes of 0.5 to 10 L. The sorbent cartridges were thermally desorbed at 220°C to the Tenax/charcoal concentrator column of a purge-and-trap unit. The concentrator column was rapidly heated, and the collected compounds were desorbed to the analytical column of a gas chromatograph (GC). Perchloroethylene was identified by retention time and quantified by FID response.

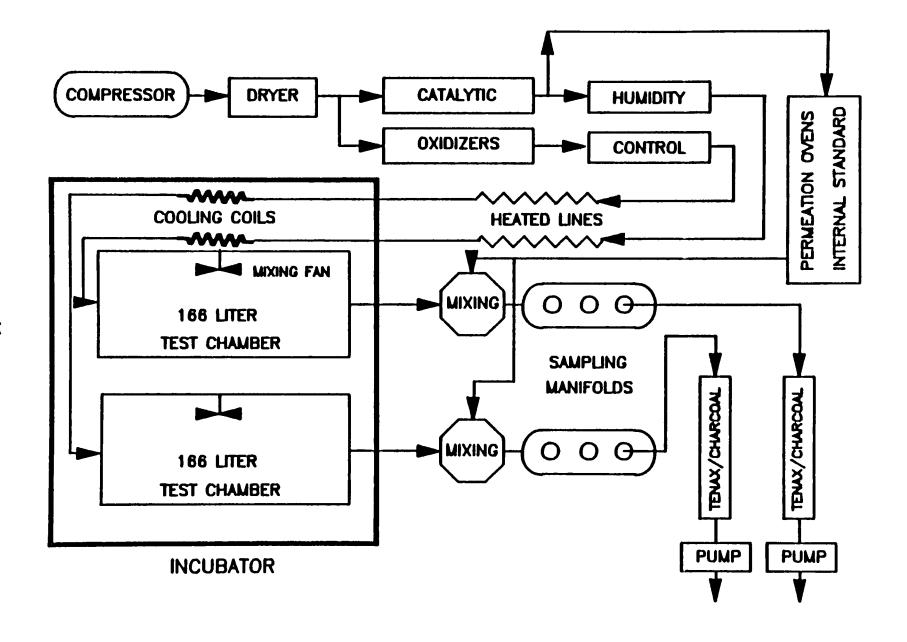


Figure 1. Small chamber (166-L) emissions testing facility.

Figure 2. Small chamber (53-L) emissions testing facility.

The GC was calibrated by loading Tenax sample cartridges with known amounts of perc. Liquid standards were prepared by serial dilution of a gravimetrically determined primary standard. Two microliters of stock solutions, ranging in concentration from 16 to 1600 ng/ul, were injected through the hot port of the purge-and-trap concentrator. Volatilized compounds were swept by a helium purge to the Tenax sample cartridges. The cartridges were then analyzed in the same manner as the chamber samples. A linear least squares fit of the FID response (in area counts) provided a response factor which was used to convert FID response to mass units.

C. Residual Analyses

1. Fabric Preparation

Upon receipt of the fabric (bolt material or clothing) to be sampled, sections of cloth were cut out and placed in brown-glass screw-cap jars with Teflon seals. The lids were sealed with Teflon tape, and the jars were stored at 4°C.

Before cutting samples to be extracted, the jars were warmed to room temperature. Two rectangular samples were cut from each fabric, about 3 by 10 cm (approx. 0.5 g). The actual size of the samples was determined by the thickness of the cloth. Thicker cloth required smaller samples to reduce crowding in the vial. The dimensions of the fabric were measured to the nearest millimeter, the samples were folded in half lengthwise, rolled, and placed in pre-weighed 15 ml vials. The vials were capped and weighed, and the mass of the fabric calculated.

For spiked samples, a drop of tetrachloroethylene was added and the vials weighed again.

2. Extraction Procedure

Five milliliters of methylene chloride (dichloromethane) was added by volumetric pipette to the vials containing the fabric samples. This quantity of solvent was sufficient to cover the sample completely. The vials were tightly capped, placed in an ultrasonic bath, and sonicated for 1 hour, during which time the temperature of the bath rose from 25 to 40°C. The vials were remove and allowed to cool to room temperature, and the solvent was decanted into 7.5 ml vials. The vials were capped, sealed with Teflon tape, and stored at 4°C.

3. Analysis

Aliquots of the sample extract were placed in 0.75 ml vials and sealed. The vials were loaded into an automatic sampler (preceded and followed by known concentration QC samples). The samples were analyzed for perchloroethylene by GC with an FID.

D. IAQ Test House

1. Facilities and Equipment

A single-story frame house with three bedrooms, two baths, den, kitchen, and a living/dining combination is leased for use as an IAQ test house (Figure 3). The house is 8 years old, has a forced-air gas heating/electric air conditioning system, and is insulated to be energy efficient (Jackson, et al., 1987). A garage along one end of the house provides a convenient instrument room. Available instruments included a GC with ECD (electron capture detector). This instrument was used for determination of perc concentrations.

2. Testing/Measurement Methods

A standard set of clothing consisting of a woman's wool skirt, two polyester/rayon blouses, and a man's two-piece wool blend suit was dry cleaned in a commercial facility. The clothes were transported to the house in the standard plastic bag provided by the dry cleaners. The clothes were placed in the closet of the corner bedroom and the closet doors were closed. All other interior doors were open. The house was closed, and the sampling initiated within 15 minutes. The house was maintained at a temperature of 20°C (68°F). The HVAC fan was operated in the normal mode, and the fan operating times were recorded.

Samples were collected at three locations for each test:
a) the closet in which the clothes were placed, b) the corner
bedroom (adjacent to the closet), and c) the den. Samples were
collected by gastight syringes for immediate injection into the
GC. Samples were taken in the center of each room at a height of
160 cm from the floor. Samples were also collected at heights of
15 and 198 cm in the closet to check for stratification. A
system was design to allow sampling while the closet doors
remained closed. This system also returned the air to the closet
to prevent the loss of perchloroethylene by dilution.

The air exchange rate for the house was determined on the first day of each test by use of SF6 tracer gas. The gas was released at the start of the each run and collected in Tedlar bags at hourly intervals until eight samples were taken. The SF6 was analyzed by GC with ECD.

After each test, all the windows in the house were opened and the house was allowed to air out for a minimum of 4 days and then was reclosed for 4 hours before the start of the next run. A background check was performed to ensure that the the perc from the previous run was below detection limits. The detection limit for the sampling and analysis system used in the study was 1 ug/m^3 .

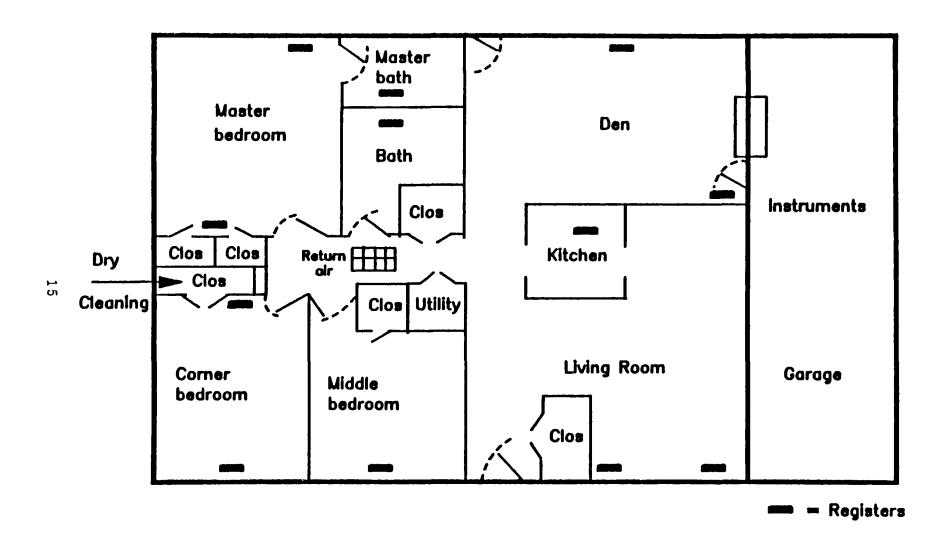


Figure 3. IAQ test house.

4. RESULTS

A. Chamber Tests

1. Data Analysis

Models have been developed to analyze the results of the chamber tests in order to provide emission rates (Dunn and Tichenor, 1988). The simplest model (i.e., neglecting sink and vapor pressure effects) assumes: a) the chambers are ideal continuous stirred tank reactors (CSTRs), and b) the change in emission factor can be approximated by a first order decay, as shown in Equation (1):

$$R = R_0 e^{-kt} \tag{1}$$

Where, $R = Emission factor, mg/m^2-hr$

 $R_o = Initial emission factor, mg/m^2-hr$

e = Natural log base

k = First order rate constant, hr-1

t = Time, hr

The mass balance for the chamber over a small time increment, dt, is:

Change in mass = Mass emitted - Mass leaving chamber

This can be express as:

$$VdC = AR_0 e^{-k\tau} dt - QCdt$$
 (2)

Where, $V = Chamber volume, m^3$

C = Chamber concentration, mg/m³

 $A = Area of source, m^2$

Q = Flow through chamber, m³/hr

Equation (2) can be rearranged:

$$dC/dt + (Q/V)C = (A/V)R_0 e^{-kt}$$
(3)

Equation (3) is a linear, non-homogeneous differential equation. Given that C = 0 when t = 0, the solution to Equation (3) is:

$$C = AR_o \left(e^{-kt} - e^{-Nt}\right) / V(N - k) \tag{4}$$

Where, N = Air exchange rate, hr^{-1} , and is equal to Q/V

Using a non-linear regression curve fit routine, implemented on a microcomputer, values of R_0 and k can be obtained by fitting the concentration vs. time data from the chambers to Equation (4). To conduct such analyses, initial estimates of R_0 and k are required. A good initial estimate of k is:

(5)

Where, tmax is the time of maximum concentration, Cmax.

Equation (5) is obtained by substituting C [Equation (4)] into Equation (3) and setting dC/dt = 0 at t = tmax. Once an estimate of k is achieved from Equation (5), R_0 can be estimated from Equation (4). Figure 4 illustrates the curve fitting process for a polyester/rayon fabric chamber test at an air exchange rate of $1 \ hr^{-1}$; the solid line is the "best fit" of Equation (4), and the data points are shown as solid squares.

All of the test runs were analyzed using this procedure, and the results (R_0 and k) for perc are presented in Tables 1 and 2. Values for R_0/k are also presented; they represent the total available emissions (or source strength) per unit area for the material being tested. Total emissions are estimated by integrating Equation (1) from time zero to infinity. Finally, the half-life of the emission factor, t(1/2), is also provided; t(1/2) is the amount of time required for the emission factor to be reduced by 50%.

2. Fabric Selection

A preliminary screening study was conducted on 12 different fabrics (cleaned/unpressed). The fabrics were investigated in the 53-L test chambers under the following conditions: air exchange rate = $1.0~hr^{-1}$, temperature = 20° C, relative humidity = 50%, and sample area (one side) = $0.168~m^{2}$. The sample area was selected to provide a chamber loading (area of sample/volume of chamber) similar to what was expected in the test house closet.

It is emphasized that the results from this preliminary screening study are useful in a qualitative sense for comparing the emission characteristics of the fabrics tested. Only one short term test was conducted on each fabric, and only two or three data points were collected. Thus, the results of the curve fit procedure described above should be used with caution. These results are presented in Table 1.

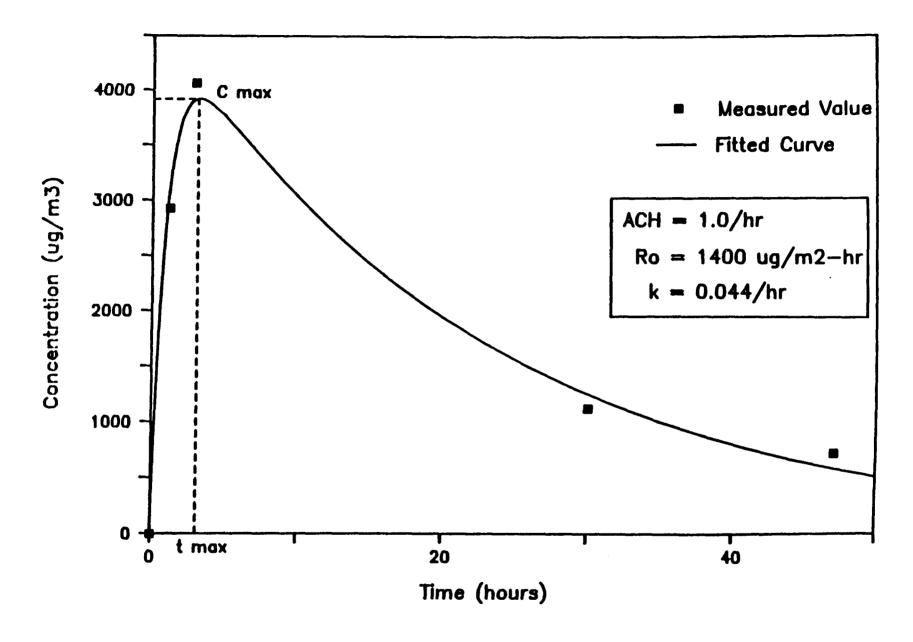


Figure 4. Perc emissions from dry cleaned polyester/rayon (modeling of small chamber data).

Table 1. Perc Emissions from Various Fabrics

Fabric		Ro m²-hr)	k (hr-1)	R_o/k (mg/m ²)	t(1/2) (hr)
50% Polyester/50%	Rayon*	220	0.030	7.3	23
Rayon	•	55	0.034	1.6	20
Polyester Knit		430	0.031	14	23
Acetate		6700	0.009	740	77
Acrylic Knit		56	0.039	1.4	18
Wool Blend (I)*		1200	0.033	36	21
Wool Blend (II)		990	0.080	12	8.
Cotton		440	0.140	3.1	5.
Linen		570	0.076	7.5	9.
65% Polyester/35%	Cotton	350	0.210	1.7	3.
85% Rayon/15% Flax		180	0.150	1.2	4.
Silk		(No	data - S	Sampling	errors

^{*}Selected for further testing.

The results (Table 1) show that the amount of perc held on fabric surfaces varies greatly depending on fabric type. The total amount of perc initially available for emission (R_0/k) ranges from 1.2 to 740 mg/m². Also, some fabrics hold perc longer; t(1/2) values range from 3.3 to 77 hours. Again, the reader is cautioned to use these results only in a qualitative way to compare one fabric to another. Use of these data to calculate emissions should be avoided.

Based on these preliminary results and an evaluation of the prevalence of materials used in clothing on the market that is normally dry cleaned, the following three fabrics were selected as the test materials:

- A <u>55% Polyester 45% Wool Blend</u>. This fabric had a relatively high emission rate and is widely used in men's suits.
- A 50% Polyester 50% Rayon Blend. This fabric had a relatively low emission rate and is widely used in women's clothing.
- A 100% Wool. Wool was not evaluated, but it is widely used and commonly sent to dry cleaners.

The highest emitter, acetate, was not selected due to its low volume use in dry cleaned clothing.

3. Effect of Air Exchange Rate

The three selected fabrics (cleaned/pressed) were investigated in the 53-L test chambers under the following conditions: air exchange rates = 0.25, 1.0, and 2.0 hr⁻¹; temperature = 20° C; relative humidity = 50%; and sample area (one side) = 0.168 m^2 . The results are shown in Table 2.

Fabric	Air Exchange (hr-1)	Ro (ug/m²-hr)	k (hr-1)	Ro/k (mg/m²)	t(1/2) (hr)
Polyester/	0.25	1500	0.028	54	25
Wool	1.0	2400	0.045	54	15
	2.0	800	0.028	29	25
100% Wool	0.25	930	0.041	23	17
	1.0	1200	0.028	43	25
	2.0	800	0.052	15	13
Polyester/	0.25	560	0.022	26	32
Rayon	1.0	1100	0.038	28	18
•	2.0	470	0.027	17	26

Table 2. Effect of Air Exchange Rate on Perc Emissions

The results provided in Table 2 show variations of R_0 , k, R_0/k , and t(1/2) for each fabric at the three air exchange rates tested. Some of this variation is due to experimental error; estimates of the sampling and analysis errors are presented in Appendix A. Another source of the variations is the amount of perc retained at the time the fabrics were picked up from the dry cleaners. Four separate dry cleaning "loads" were required over a 5 week period to complete the tests reported in Table 2. Whatever the cause of the variations, the data do not suggest that higher air exchange rates have a significant effect on the rate of decay of the emission rate, k, or on the half-life of the emission factor, t(1/2).

4. Effect of Temperature

Two fabrics, 55% polyester/45% wool and 100% wool, were selected to evaluate the effect of temperature on the emissions of perc from dry cleaned fabrics. Tests were conducted in the 166-L chambers under the following conditions: temperatures = 30, 35, 40, and 45°C; air exchange rate = 1.0 hr⁻¹; and sample area = $0.5~\text{m}^2$. The humidity control system was set to supply water vapor at a rate equivalent to 50% relative humidity at 20°C . The results for Ro and t(1/2), along with the previous data for 20°C , are shown in Table 3.

Table 3, Effect of Temperature on Perc Emissions

Fabric	Temperature (°C)	Ro (ug/m²-hr)	t(1/2) (hr)
Polyester/	20	2400	15
Wool	30	8600	4.5
	35	5400	2.9
	40	9200	2.5
	45	16000	1.8
100% Wool	20	1200	25
	30	3100	7.1
	35	3200	1.3
	40	8100	0.6
	45	14000	0.5

Table 3 shows that temperature has a major effect on the emission characteristics of perc. Increasing temperatures caused higher initial emission factors, R_0 , and lower emission factor half-lives, t(1/2).

B. Perc Residuals

The data on the residual perc determined by solvent extraction using methylene chloride are not reported herein. Several problems were encountered: 1) mechanical breakdowns of the automatic sampler caused extensive delays in analyzing the extracts, and no data are available on how such delays might affect the final results; 2) the recoveries of spiked perc from samples of different materials were quite variable and often too low (some variability in recoveries for samples of the same material was also observed); 3) the amount of residual perc extracted was inconsistent with values determined from the chamber data; and 4) the amount of residual perc from the clothing used in the test house experiments (immediately after cleaning vs. after removal from the test house) showed little relationship to the actual amount of perc emitted in the test house as determined by a mass balance using measured concentrations.

The reasons for the failure of the solvent extraction procedure to provide useful data are unclear. A search of the literature did not uncover a "standard method" for extracting perc from dry cleaned fabrics, although solvent extraction by methylene chloride is mentioned by Brodmann (1975). Subsequent to initiating the study, discussions with representatives of the dry cleaning industry suggested that extraction using carbon disulfide is more common.

C. Indoor Concentrations (Test House)

Evaluations of the impact of dry cleaned clothes on indoor concentrations of perc were conducted in the IAQ test house. For each test, the following clothing was dry cleaned and brought to the test house:

- A two piece man's suit (55% polyester/45% wool blend),
- A woman's skirt (100% wool), fully lined (acetate), and
- Two women's blouses (50% polyester/50% rayon).

The total area of this mix of clothing, including linings, padding, pockets, and accounting for seam overlap, is $8.6~\text{m}^2$.

Four tests were conducted as described in Section 3.D:

- a) "Bag Off" the plastic bag was removed prior to placing the clothes in the closet (March 22 31);
- b) "Bag On" the plastic bag was not removed (April 6 13);
- c) "Aired Out" the plastic bag was removed, and the clothes were hung in an open carport for 4 hours prior to being placed in the closet (April 18 - 26);
- d) A repeat of the first "Bag Off" test (May 2 10).

The daily sampling frequency for the four tests is shown in Table 4. Early in each test, 24 hour coverage of the test house was provided. After the first couple of days, sampling occurred during normal working hours. Limited sampling occurred on weekends.

The results of the test house evaluations are shown in Table 5 and in Figures 5 - 11. Figures 5 - 7 show the average daily concentrations for all four tests as measured in the closet, bedroom, and den, respectively. The closet values are the average of the three sampling elevations, since no consistent stratification was observed in the closet. Figures 8 - 11 provide the average daily values for the bedroom and den for the four tests. Note that, in all cases, the level of perc dropped to near or below the detection limit within a day after the clothing was removed from the test house.

The results show two consistent patterns: a) on any given day, the concentrations are highest in the closet, followed by the bedroom, with the den having the lowest concentrations; and b) the concentrations in all rooms generally decrease over time.

Table 4. IAQ Test House Sampling Frequency -- Perc Tests

Test	Day	No. of Samples	Sampling Interval (hr)
Bag Off (1)	1 2 3 4 5 6 7 8 9*	16 13 4 4 1 1 1 4 5 3	0.5 1.5 2.25 2.25 24 24 1.25 1.5 1.5
Bag On	1 2 3 4 5 6 7* 8	15 15 4 3 3 6 6	1.25 1.0 2.0 4.0 4.0 0.75 0.5
Aired Out	1 2 3 4 5 6 7 8* 9	8 25 20 5 1 1 1 3	0.75 1.0 1.0 1.8 24 24 24 24
Bag Off (2)	1 2 3 4 5 6 7 8*	12 26 17 5 5 1 2	1.0 1.0 1.0 1.0 24 6.0 24

^{*}Clothes removed from house.

Table 5. IAQ Test House Results

Average Perc Concentration (ug/m³)

Test	Day	Closet	Bedroom	Den
Bag Off	1	521	60	36
(1)	2	325	58	29
	3	310	57	32
		273	46	25
	4 5 6	258	45	26
	6	158	25	13
	7	125	15	8
	8	52	14	6
	9*	13	9	6
	10	ND	ND	ND
Bag On	1	1240	139	37
	2	498	61	32
	3 4	438	44	25
	4	331	32	16
	5	307	31	18
	6	393	27	14
	7*	290	18	9
	8	ND	ND	ND
Aired Out		2900	195	79
	2	2740	124	83
	2 3 4	425	50	24
	4	371	55	33
	5	393	43	26
	6	307	37	27
	7	720	85	40
	8*	146	24	15
	9	3	3	
Bag Off	1	1420	119	51
(2)	2	745	104	46
	2 3	606	52	28
	4	353	39	26
	5	218	22	15
	6	557	16	16
	7	170	19	12
	8*	168	11	5
	9	ND	ND	ND

^{*}Clothes removed from house; ND = Not Detected.



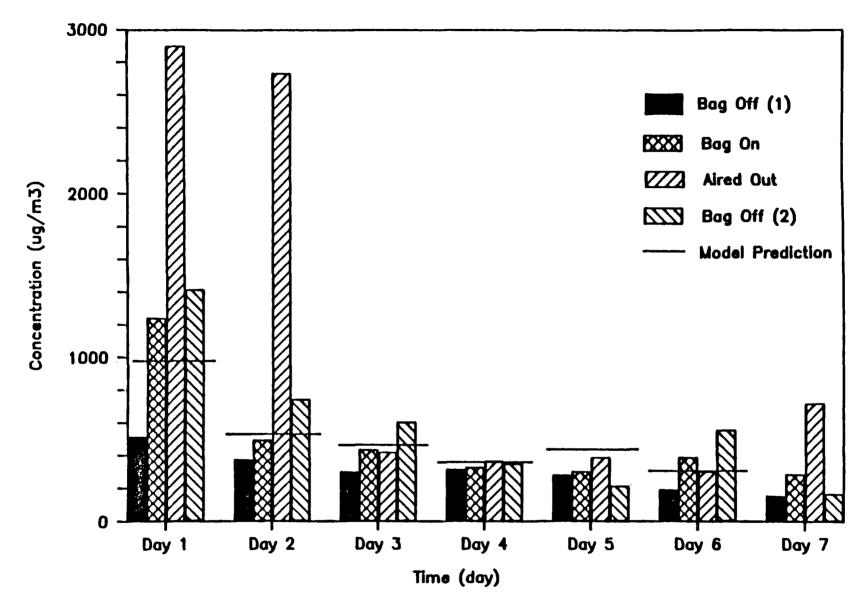


Figure 5. Perchloroethylene in closet.



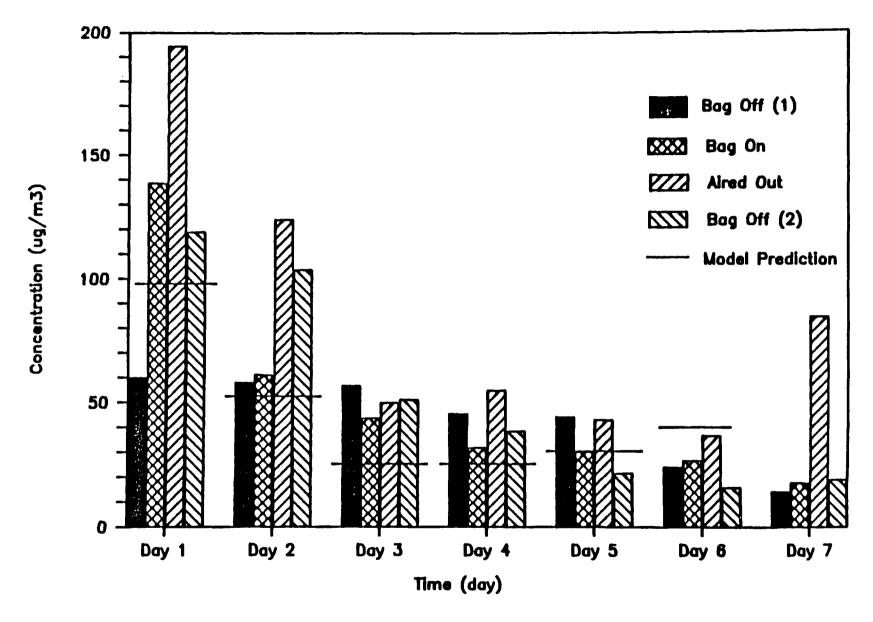


Figure 6. Perchloroethylene in bedroom.

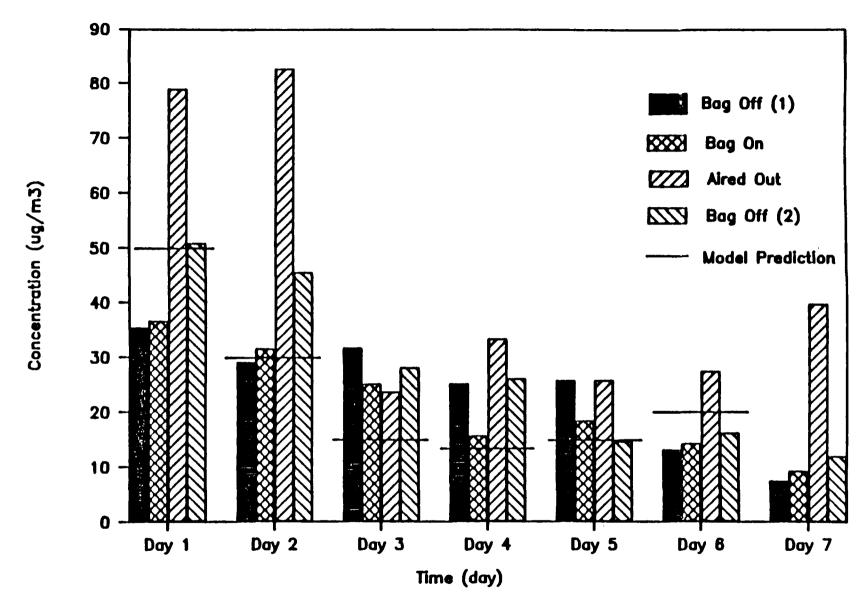


Figure 7. Perchloroethylene in den.

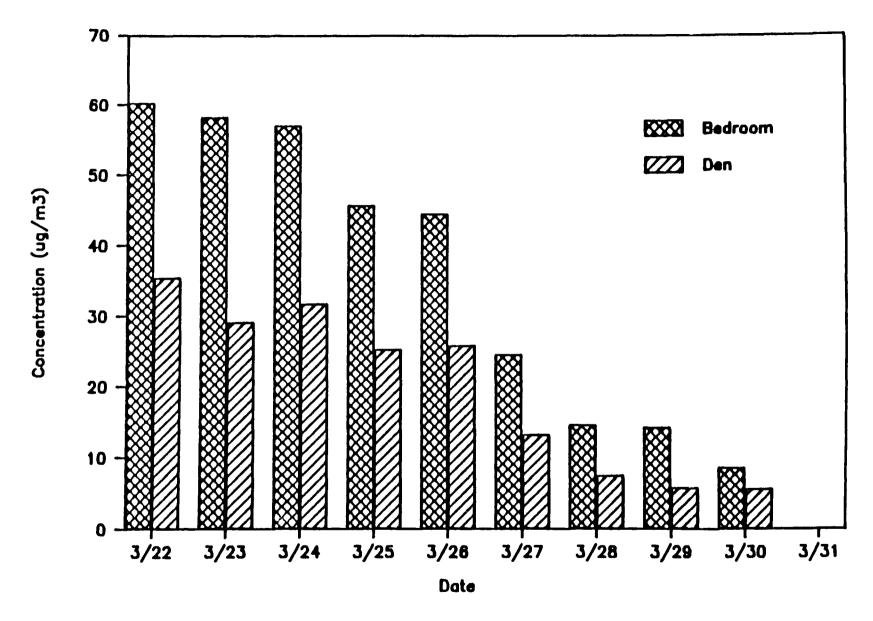


Figure 8. Perc in bedroom and den -- bag off (test 1).

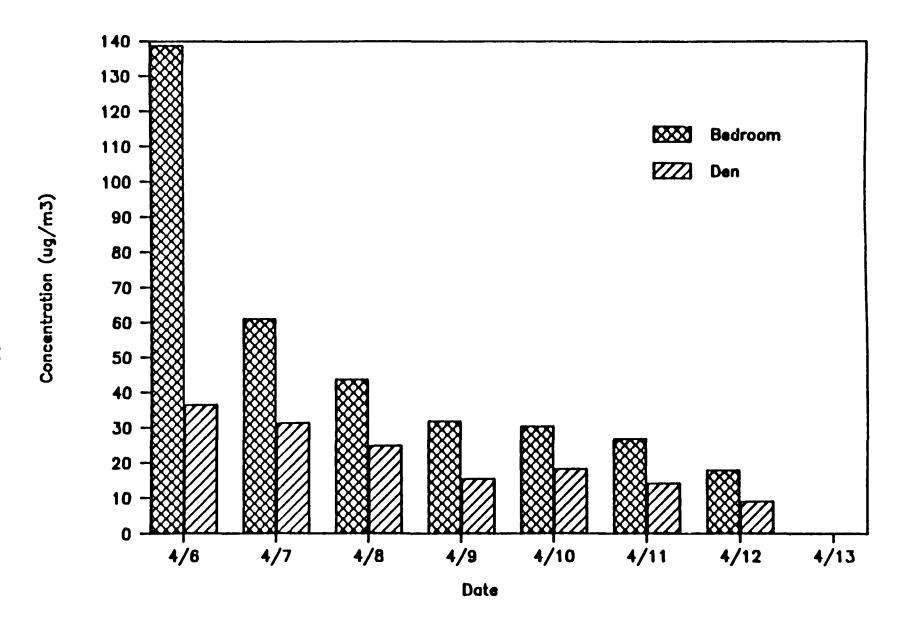


Figure 9. Perc in bedroom and den -- bag on.

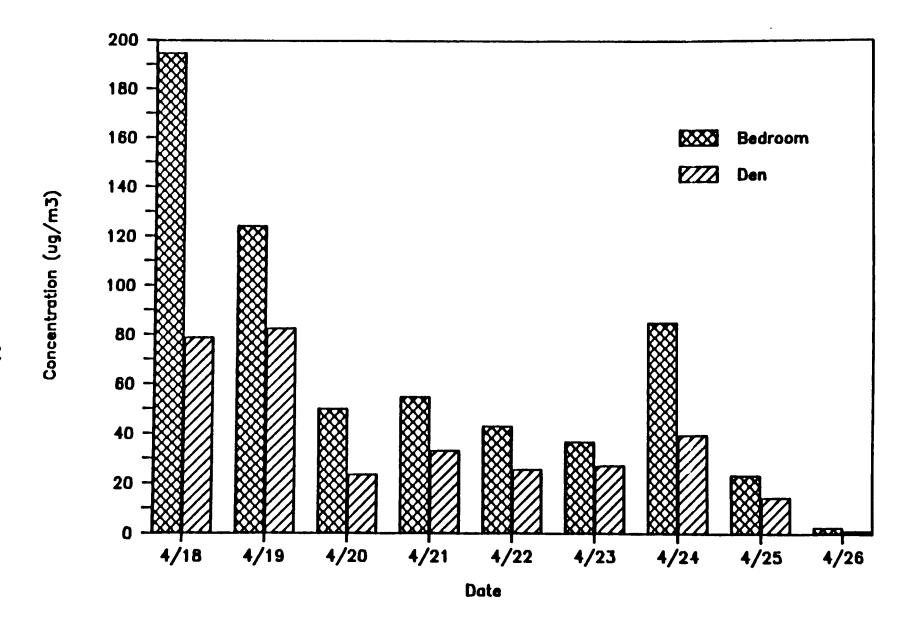


Figure 10. Perc in bedroom and den -- aired out.

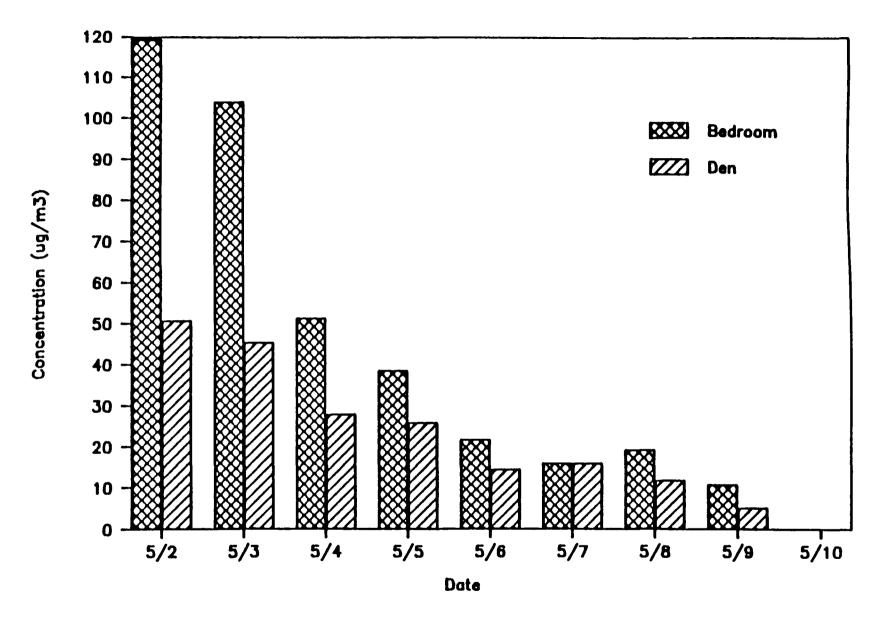


Figure 11. Perc in bedroom and den -- bag off (test 2).

A wide variation in perc concentrations was observed between the four tests for the first couple of days of each experiment. Since one would not expect that keeping the bag on or airing out the clothes would cause increases in emissions, these differences are not believed to be due to the experimental variables. Rather, it is assumed that the differences are due to the amount of perc retained in the clothes at the dry cleaners. At any rate, under all test conditions, elevated levels of perc were measured in the test house when freshly dry cleaned clothing was placed in the closet. Also, the perc dropped to near or below the detection limit after the clothes were removed from the house.

D. IAQ Model Analysis

The EPA IAQ model (Sparks, et al., 1988) was used to determine the consistency of the test house and chamber data. The model was also used to estimate sink effects.

1. Model description

The model estimates the effects of heating, ventilating, and air conditioning (HVAC), air cleaning, room to room air movement, and natural ventilation on pollutant concentrations. The model has been used in a similar fashion with experiments conducted in the EPA test house using moth crystal cakes composed of para-dichlorobenzene (Sparks, et al., 1988). The agreement between small chamber emission factors, model predictions, and test house data was very good. Predicted weight loss of the moth crystal cakes was within 5% of the measured weight loss. Predicted room concentrations of para-dichlorobenzene was within 10% of the measured values.

The EPA IAQ model is based on conducting mass balances of pollutant and air flow between multiple well-mixed model rooms. This model was selected because data from the EPA test house showed that pollutant concentrations within a room do not vary significantly with position in the room (Jackson, et al., 1987).

The model uses a menu-driven "fill in the form" data-input user interface. This interface is easy to use and is self prompting. The user is able to change the input parameters quickly and easily, and several conditions can be rapidly analyzed. The results of the model calculations are displayed as plots of concentration versus time for the various rooms. The plots require that a graphics adapter and color monitor be installed on the computer.

2. Source Terms

The source term used to model the perc emission was based on the small chamber data and is in the form:

$$E(t) = R_0 e^{-kt} A \tag{6}$$

Where, E(t) = Emission rate, ug/hr, at time t

 R_0 = Initial emission factor, ug/m^2-hr

k = First order rate constant, hr-1

A = Area of clothes, m^2

For the test house studies, $R_0 = 1600 \text{ ug/m}^2 - \text{hr}$, $k = 0.03 \text{ hr}^{-1}$, and $A = 8.6 \text{ m}^2$. These values were obtained by using a weighted average of the chamber test results based on the measured areas of the fabric types for the clothing used in the test house experiments.

3. Sink Terms

A re-emitting sink was used in the perc modeling. The rate going to the sink was assumed to be:

$$R_s = k_s C_r A_s \tag{7}$$

Where, Rs = Rate to the sink, ug/hr

ks = Constant, m/hr

 $C_r = Concentration in the room, ug/m³$

 $A_s = Area of the sink, m^2$

The value of k_8 was estimated using data from a special perc experiment, data from moth crystal cake experiments, and a theoretical analysis of mass transfer to walls in a well stirred reactor. The value used for k_8 was 0.35 m/hr.

The emission rate from the sink, E_{s} (ug/hr), was assumed to be:

$$E_8 = k_e M_s A_s (C_r - C_c), \text{ when } C_r > C_c$$
 (8)

$$E_s = 0$$
, when $C_r \le C_c$ (9)

Where, ke = Emission constant, m/ug-hr

Ms = Mass collected on the sink, ug

Cc = Critical concentration, ug/m³

When $C_r > C_c$, emissions are possible, and when $C_r \le C_c$, emissions are not possible. k_e was estimated to be 5 m/ug-hr, and C_c was estimated to be 40 ug/m³.

4. Results

The results of the initial model runs with no sink effects did not provide good agreement with the measured test house perc concentrations. The predicted concentrations were too high, and the predicted curves did not show the changes in slope noted in the experimental data. The likely explanation for the differences between predicted and measured concentrations is the existence of a sink effect.

Model runs were then made with a re-emitting sink as described above. The model results for days 1 - 6, using a re-emitting sink, are shown in Figures 5 - 7. Note that the agreement between the model predictions and the measured data is good both in magnitude and in the shape of the decay curve. Also note that the model predictions, as well as the measured data, show plateaus of nearly constant perc concentration, including slight increases, for days 3 through 6. These plateaus and small increases in perc concentration are caused by re-emissions from the sink, because, without a re-emitting sink, the concentrations would have continued to decay.

The main effects of the re-emitting sink are:

- 1. Initial peak concentrations are reduced as perc goes to the sink; and
- 2. The rate of concentration decay is slowed as perc is re-emitted from the sink, and some increases in concentration can also be observed.

5. DISCUSSION AND CONCLUSIONS

A. Emission Factors

Emission factors for perchloroethylene from dry cleaned fabrics were determined by testing in small environmental test chambers under controlled conditions. Evaluation of the data from these tests provides the following results:

- A preliminary screening evaluation showed that wide variations in initial emission factor, Ro, and emission factor half-lives, t(1/2), occurred between different fabrics. Thus, the type of fabric is important in determining indoor emissions of perc from dry cleaned clothes.
- Based on the screening study and on the prevalence of fabrics used in dry cleaned clothing, three fabrics were selected for investigation: 55% polyester/45% wool; 100% wool; and 50% polyester/50% rayon.
- The air exchange rate showed no effect on the emission factor or decay rate for the three fabrics investigated. This suggests that the emissions are limited by the diffusion of perc within the fabric and are not controlled by evaporative processes. This also suggests that increasing the ventilation by airing out the clothes will not speed up the emission of perc.
- Since the three fabrics tested had emission factor halflives of about a day, airing the clothes out for a few hours before hanging them in the home will do little to reduce the indoor perc concentrations.
- For fabrics with faster perc decay rates, airing out may be more practical. The percent emitted during airing out is calculated by:

% Emitted during airing = (Amount emitted/Total available)100%

The Amount emitted equals the integral of the emission rate function [Equation (1)] over the time aired out:

Amount emitted =
$$(R_0/k)(1 - e^{-ak})$$
 (10)

Where, a = airing out time (hr)

As defined above, the Total available = R_0/k , thus:

% Emitted during airing =
$$(1 - e^{-ak})100\%$$
 (11)

The effect of airing out is illustrated in Figure 12 which shows the percent of perc emitted during airing out as a function of the time aired out for a wide range of decay rates (k). Note that, for the decay rates determined for the three fabrics

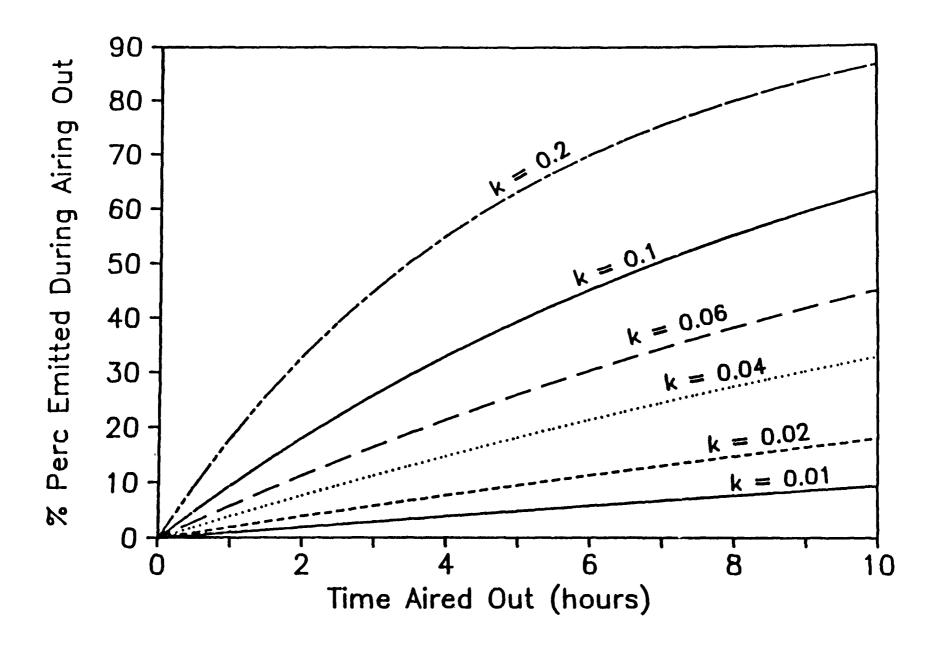


Figure 12. Effect of airing out.

investigated in this study (from Table 2, k ranges from 0.022 to 0.052 hr⁻¹), the percent of perc emitted over a 4 hour airing out period would range from 8 to 19%; for an 8 hour period, the range would be 16 to 34%.

- Temperature had a major impact on the emission factors and decay rates. Increases in temperature caused higher initial emission factors and lower half-lives. Thus, exposing the clothing to higher temperatures prior to bringing them home shows promise as a means of reducing in-home exposure to perc.

B. Residuals

No acceptable data were developed on the perc residuals within the fabric. The solvent extraction procedure, using methylene chloride, failed to produce reliable results. A fully tested "standard method" is needed.

C. Indoor Concentrations

All the test house experiments showed that the introduction of dry cleaned clothing caused elevated levels of perc in the house. Differences in concentration between the tests were probably due to differences in the amount of perc retained at the dry cleaner.

D. Model Results

The IAQ model, using emissions data developed in the small chambers, predicted indoor perc concentrations which compared favorably with those measured in the test house. The effect of perc "sinks" in the test house was also demonstrated.

E. Conclusions

Based on the study results, and assuming that test conditions are representative of normal dry cleaning and consumer practices, the following conclusions are reached:

- 1) Emissions from freshly dry cleaned clothing cause elevated levels of perchloroethylene in residences.
- 2) For the three fabrics tested, "airing out" of dry cleaned clothing by consumers will not be effective in reducing perchloroethylene emissions.

It is emphasized that these conclusions are based on the results of the study reported herein. Significant variations in dry cleaning practices and/or in the mix of fabrics and clothing being cleaned could provide different results and conclusions.

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APPENDIX A. QUALITY ASSURANCE/QUALITY CONTROL RESULTS

As discussed in Section 2.F, <u>Quality Assurance/Quality Control</u>, a number of QA/QC steps were implemented in the conduct of the study; the results are reported below. Accuracy (or bias) is reported as the average deviation from the true value:

Accuracy =
$$[(m - x_0)/x_0]100\%$$
 (A-1)

Where, m is the mean, and xo is the true value.

Precision is reported as Relative Standard Deviation (RSD). RSD (also called the Coefficient of Variation) is calculated by:

$$RSD = (s/m)100\%$$
 (A-2)

Where, s is an estimate of the standard deviation, and m is the mean.

1. Calibration Audit for Perchloroethylene

Two audit gas cylinders were obtained and analyzed for perchloroethylene in the chamber laboratory and at the test house. The results of the analyses are shown in Tables A-1 and A-2. In all cases, the goals of 20% accuracy and 15% precision were met.

Table A-1. Audit Gas Analysis -- Chamber Laboratory

Reported Conc. (ppb)	Number of Analyses	Measured Conc. (ppb)	Accuracy (%)	Precision (%)
400	2	452	12.8	1.0
10.9	3	9.4	13.8	11.2

Table A-2. Audit Gas Analysis -- Test House

Reported Conc. (ppb)	Number of Analyses	Measured Conc. (ppb)	Accuracy (%)	Precision (%)
400	7	370	7.5	6.7
10.9	4	10.0	8.3	2.3

2. Standard Rotation

A perchloroethylene standard rotation procedure was implemented between the chamber laboratory and the laboratory that conducted the residual analysis. One liquid perchloroethylene standard from each laboratory was analyzed by the other laboratory. The results are shown in Table A-3. The QC goals of 20% accuracy and 15% precision were met.

Table A-3. Perc Standards Rotated between Residuals Laboratory and Chamber Laboratory

Reported Conc. (mg/ml)	Number of Analyses	Measured Conc. (mg/ml)	Accuracy (%)	Precision (%)
0.325*	4	0.270	16.9	3.1
0.158**	2	0.157	0.6	3.1

^{*}Prepared by residuals laboratory; analyzed by chamber laboratory.

3. QC for Chamber Laboratory

Variability of sample size, determined by comparing the mass of each fabric used in the individual experiments with the average mass in all tests, ranged from 1.2 to 4.2% for each fabric. The average test chamber temperature was controlled to within \pm 1°C of the setpoint for all but one experiment, which exceeded the expected range by 0.4°C. Relative humidity was controlled to within \pm 15% of the setpoint for all experiments. Uncertainty of chamber air flow, determined from the difference in measured flow at the beginning and end of each experiment, was

^{**}Prepared by chamber laboratory; analyzed by residuals laboratory.

 \pm 4%. Recovery of the internal standard, present in every sample, averaged 103 \pm 9.8% for 270 samples, meeting the goal of 20%. The precision of duplicate samples averaged 7.4 \pm 4.7% for 39 duplicate pairs; the goal was 10%.

Estimates of the variability of the R_0 , k, and t(1/2) for all tests conducted at 20°C are shown in Tables A-4, A-5, and A-6, where the mean and standard deviation for each of these parameters is given. The variability indicated in these tables includes load to load variation at the dry cleaners, experimental error, and statistical errors associated with the curve fitting of the data.

Table A-4. Variability in R_0 , k, and t(1/2) for Polyester/Wool

Air Ex.	No. of Samples	R _o (ug/m²-hr)	k (hr-1)	t(1/2) (hr)
0.25	6	1507 ± 529	0.0277 ± 0.0067	26.5 <u>+</u> 6
1.0	5 1	2412 ± 495 798	0.0453 <u>+</u> 0.0123 0.0278	16.3 <u>+</u> 3.9 24.9

Table A-5. Variability in R_0 , k, and t(1/2) for 100% Wool

Air Ex. (hr-1)	No. of Samples	Ro (ug/m²-hr)	(hr-1)	t(1/2) (hr)
0.25	2	933 <u>+</u> 95	0.0410 <u>+</u> 0.0170	20.4 <u>+</u> 8.4
1.0	4 2	1186 ± 539 2021 ± 18	$\begin{array}{c} 0.0277 \pm 0.0095 \\ 0.0385 \pm 0.0035 \end{array}$	$\begin{array}{c} 25.5 \pm 3.7 \\ 18.7 \pm 0.9 \end{array}$

Table A-6. Variability in Ro, k, and t(1/2) for Polyester/Rayon

Air Ex. (hr-1)	No. of	Ro	k	t(1/2)
	Samples	(ug/m²-hr)	(hr-1)	(hr)
0.25	2	562 ± 32	$\begin{array}{c} 0.0220 \pm 0.0060 \\ 0.0380 \pm 0.0041 \\ 0.0274 \end{array}$	34.0 ± 9.3
1.0	2	1072 ± 254		18.5 ± 2.0
2.0	1	474		25.3

4. QC for Test House

The GC used for perchloroethylene analysis was calibrated every morning before any samples were taken, and its performance was checked every 3 hours during the day by injecting two liquid perchloroethylene standards.

Duplicate samples were taken from each sampling location. A total of 69 pairs of duplicate samples were taken. The precision of the duplicate samples averaged $5.6 \pm 5.7\%$. The precision of 7 of the 69 duplicate pairs exceeded the QC goal of 15%, which occurred during the first sampling in the house and may depict levels of perchloroethylene rising due to the recent introduction of the clothing into the house. These samples had a slight difference in collection time.

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AUTHOR(S)	8. PERFORMING ORGANIZATION REPORT
Bruce A. Tichenor, Leslie E. Sparks, and	ı
Merrill D. Jackson	W ELEMENT NO.
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6. ABSTRACT The report gives re	roethylene
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cleaning into a house affects	effectiveness
of "airing out" for reducing	e conducted t
determine perc emission ch	emperatures
and air exchange rates. Tes	ne the indoor
concentration of perc due to	ie house.
Based on study results, and	entative of
normal dry cleaning and cor	s were rea-
ched: emissions from fresh	ls of per-
chloroethylene in residence	out" of dry
cleaned clothing by consume	proethylene
emissions. Significant varia	he mix of fab
rics and clothing being clea	clusions.

7. KEY WORDS AND DOCUMENT ANALYSIS				
a. DESCRIPTORS	b. OENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group		
Pollution	Pollution Control	13B		
Tetrachloroethylene	Stationary Sources-	06C,07C		
Emission	Perchloroethylene	14G		
Fabrics	Indoor Air	llE		
Dry Cleaning		13H		
Atmosphere Contamination Control		06K		
3. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report) Unclassified	21. NO. OF PAGES		
Release to Public	20. SECURITY CLASS (This page) Unclassified	22. PRICE		