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AN INDOOR AIR QUALITY STUDY OF FORTY EAST TENNESSEE HOMES

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1. EXECUTIVE SUMMARY

This study of indoor air quality in 40 homes in East Tennessee lasted nearly one year. It represents one of the first attempts to monitor the levels and temporal fluctuations of a wide range of chemical pollutants over an extended period of time and in a wide selection of housing stock. Improved understanding of contemporary indoor air quality includes interrelationships of indoor pollutants with types of housing, geographical location, consumer products in the homes, the lifestyles of the occupants, and seasonally related weather changes. These are complex interactions, but they must be taken into account in any comprehensive assessment of indoor air quality. Due to the limited sample size and limited geographical location of the study, care should be used in extrapolating results from this small study to general statements on indoor air quality.

The following important pollutants were measured during the study:

1. formaldehyde - Passive membrane samplers and a pumped molecular sieve technique, both developed at Oak Ridge National Laboratory (ORNL), were used for routine measurements. [Quality control measurements were performed with a refrigerated sampling system and with the National Institute for Safety and Health (NIOSH) technique.]
2. combustion gases - Nitrogen oxides were measured with a chemiluminescence analyzer, carbon monoxide was monitored with an electrochemical analyzer, and carbon dioxide was measured with an infrared analyzer.
3. volatile organics - A portable photoionization gas chromatograph (GC) and Tenax sorbent traps, followed by thermal desorption and GC or gas chromotography/mass spectrometry (GC/MS) analysis, were used in the study to measure volatile organics.
4. particulate material - A ten-stage cascade impactor was used to monitor the particulate levels in the homes.
5. radon - Passive track etch detectors were used in all of the study homes to obtain a three-month integrated exposure. A Wrenn chamber with hourly readout was used in a subset of homes to measure short-term variations in radon concentration.

The formaldehyde concentrations were generally higher in newer homes than in older homes. In houses less than 5 years old, levels of formaldehyde were elevated about twofold over the levels in houses of greater age ($P < 0.01$). The highest concentrations of formaldehyde tended to occur during hot and humid summer weather. The sources of indoor formaldehyde are probably the formaldehyde resins found predominantly in structural materials, insulation, and furnishings. The generation and release of formaldehyde from such products are known to increase with higher temperature and humidity.

In over half of the homes measured, the concentration of formaldehyde exceeded the American Society of Heating, Refrigeration, and Air Conditioning Engineers (ASHRAE) 0.1-ppm ceiling level on at least one occasion. In contrast, mean monthly concentrations of formaldehyde outdoors were always less than 0.03 ppm. The highest concentrations of formaldehyde (up to 0.4 ppm) were experienced in an energy-efficient house with a polyethylene vapor barrier. This house was less than three years old.

Comparative measurements in the laboratory were made with the different active and passive monitoring systems used in the study. The methods compared included a refrigerated impinger sampler, a CEA 555 analyzer, the modified NIOSH procedure, the pumped molecular sieve technique, and passive membrane samplers. At a concentration of 0.3 ppm, all of the techniques gave adequate accuracy and precision. For short-term measurements in the field, the molecular sieve method was more sensitive and more convenient to use than the modified NIOSH procedure.

Volatile hydrocarbons of C_7 and above were collected using a Tenax trap and subsequently analyzed by GC and GC/MS. Seventeen compounds were quantified routinely in the indoor air of all of the houses and in several samples of outdoor air. Of these compounds the most volatile were toluene, ethyl benzene, and xylenes, and the least volatile were 2-methylnaphthalene and the $C_{14\text{-}16}$ alkanes. The indoor air contained a ten-fold or higher concentration of organic vapors than the outdoor air contained. This was the case for organic compounds of all degrees of volatility measured in the study. The two compounds with the highest concentrations were toluene and xylene. Chemicals such as dichlorobenzenes and siloxanes were occasionally encountered. These two types of compounds are contained in mothballs and furniture polish, which are sources that might be found in homes.

Measurements of more highly volatile organic compounds were obtained with a portable photoionization GC. These measurements revealed gasoline fumes in all rooms of several houses. The instrument response increased with approach to the garage, usually attached, or to storage vessels containing gasoline or used motor oil. An operating car in the garage produced exhaust fumes containing uncombusted gasoline, which were drawn quickly into adjoining rooms. Lesser amounts of gasoline vapors were also measured outdoors. Cleaning, polishing, and painting operations introduced additional volatile organic compounds into room air. The identification, concentration, and persistence of most of these latter compounds have yet to be determined.

The concentrations of gases associated with the products of combustion were generally low during the warm seasons of the year. Carbon monoxide and NO_x were less than 2 and 0.02 ppm, respectively, except at a few houses located near busy thoroughfares or when automobiles were operated inside attached garages. In winter, the operation of unvented combustion devices (gas cooking stoves and kerosene space heaters) caused marked and transient increases in the levels of CO_x and NO_x. Carbon monoxide was monitored using an electrochemical device, and NO_x levels were monitored with a chemiluminescence analyzer.

The mean concentration of particulates measured during the warm seasons was 32 $\mu g/m^3$ of total suspended particulates and 24 $\mu g/m^3$ of respirable suspended particulates. Similiar levels were measured during the winter in a subset of homes without combustion sources. Particulate levels were measured using a 10-stage cascade impactor with a near real-time display of concentration.

The approximate guideline for the average annual concentration of radon (4 pCi/L) was exceeded in many homes. Long-term measurements of radon during the winter season showed that the guideline was exceeded in 30% of the houses. The higher levels of radon were found in houses built on permeable dolomite ridges. The mean radon levels in these houses was 4.4 pCi/L in the living area and 6.3 pCi/L in basements. Houses located on less permeable clay sediment in the valleys had a mean level of radon of 1.7 pCi/L in the living area and 3.3 pCi/L in basements. It is speculated that the porous dolomite serves as a conduit for radon gas produced in underlying uranium-containing shale deposits.

The rate of exchange between outdoor and indoor air in closed-up houses was found to be generally low. The mean value was 0.39 air changes per hour during the first phase of the study (internal fans were used to ensure good tracer mixing). When the duct fan was running in houses with central heating, ventilation, and air conditioning (HVAC) systems, the mean air exchange rate nearly doubled to 0.72 air changes per hour. Leaks in the duct system exterior to the living space are most likely responsible for this effect. This is a finding of considerable importance in the context of energy conservation, weatherization, and ventilation, and further investigations should be performed to determine if this is unique to this particular region or common elsewhere.

A number of special studies were carried out during the course of this research. The most extensive of these were measurements of formaldehyde made in three houses, primarily during the scoping phase of the study, that reflect seasonal and diurnal variations in concentrations and the effectiveness of various abatement techniques. Seasonal fluctuations of formaldehyde were as much as an order of magnitude. These changes were brought about by variations in the weather, the operation of the HVAC system, and the lifestyle of the occupants. Diurnal fluctuations were measured in three houses. Changes in concentration of formaldehyde were as much as twofold. Increasing the ventilation rate produced a less than inversely proportionate decrease in the concentration of formaldehyde. This behavior is consistent with formaldehyde sources whose emission rates are dependent on the airborne concentration of formaldehyde. Another type of remedial action, the operation of an air cleaner with a small charcoal filter, was completely ineffective in reducing the level of formaldehyde.

This recently completed, year-long indoor air quality study in 40 homes in East Tennessee represents a considerable addition to the existing data base on residential indoor air quality. Several important conclusions can be drawn from the preliminary data analysis. The general findings are:

1. Formaldehyde levels in the study homes frequently exceeded 0.1 ppm.
2. Formaldehyde levels were highest in the newer homes (<5 years old).
3. Formaldehyde levels can fluctuate twofold to tenfold or more, both diurnally and seasonally.
4. Radon levels in the study homes frequently exceeded 4 pCi/L.
5. Radon levels correlated strongly with house location.
6. Organic pollutant levels were at least an order of magnitude higher indoors than outdoors.
7. Combustion sources (especially unvented) increased (at least a transient increase) levels of CO_x, NO_x, and particulates.
8. Air exchange rates were increased nearly twofold by operation of the HVAC central air circulation fan.

More definite correlations and conclusions are anticipated as the extensive data bases are analyzed for correlations of pollutant levels with housing characteristics and meteorological conditions.

Several areas where further research and development work is needed to support indoor air quality activities have been identified during the course of this study.

1. New and improved portable instrumentation capable of automatic recording of time-varying concentrations of low levels of pollutants is required to allow more intensive monitoring.
2. Source identification and experimental characterization (including environmental chamber tests) are needed for various pollutants, especially organic compounds.
3. Additional passive integrating monitors for chronic exposure monitoring should be field tested.
4. Rigorous laboratory and field intercomparisons of monitoring devices are necessary.
5. Pollutant emission rates from various sources need to be determined and pollutant concentrations in homes need to be modeled and the models validated experimentally.
6. Before and after measurements of indoor air quality impact due to energy conservation (and alternative fuels) measures should be performed.
7. Remedial actions should be evaluated and improved based on a better understanding of source behavior.
8. Feedback should be provided to building designers and manufacturers of consumer products on the consequence of various products on indoor air quality. Suggestions for alternative materials and practices should be made as appropriate.
9. Similar studies should be conducted in different geographical areas and in a statistical sampling of homes from the national population.

2. INTRODUCTION

The seriousness of indoor air pollution remains to be adequately answered. Relatively short-term studies of a limited number of indoor air pollutants in a small number of houses have previously been the norm.[1,2] Such studies provide fragmentary evidence about the extents, magnitudes, and interrelationships between pollutants and their dependencies upon types of housing and lifestyles. This study, covering about one year, of 40 East Tennessee homes was intended to rectify some of the inadequacies of these less comprehensive studies. Additional studies in other areas of the nation will be required to verify these findings before they are considered conclusive.

During the past two decades concern over the relationship between air quality and health has focused on outdoor and workplace environments. Ambient outdoor air quality gained widespread attention—and the resources to address the problem—with the establishment of the Environmental Protection Agency (EPA). Consequent strides have been made in improving the ambient air quality, especially in major metropolitan areas. Similar attention has been given to the air quality associated with the industrial workplace through the establishment of the Occupational Safety and Health Administration (OSHA) and its research arm, the National Institute of Occupational Safety and Health (NIOSH). However, no such mandate exists for the evaluation of residential indoor air quality, although many of the potential sources of indoor air pollutants fall under the jurisdiction of the Consumer Product Safety Commission (CPSC).

It is gradually being recognized, since the indoor levels of a large number of pollutants are higher than outdoor levels, that the total pollutant exposure of a population could provide a more meaningful correlation with adverse health effects than just outdoor exposure. This may be especially true for chronic illnesses that often manifest symptoms long after the initial exposures begin. With Americans spending about 65% of their time in residential indoor environments,[3] and many of the more susceptible individuals of the population (such as the infirm, young children, and the elderly) spending all or almost all of their time indoors,[4] the residential indoor air quality cannot be ignored in evaluating the impact of air quality on human health.

Additional factors are increasing the importance of indoor air quality. Post-1973 residential construction practices and retrofit measures have generally attempted to reduce the air exchange rate between indoor and outdoor air and, consequently, the energy loss required to heat or cool this air. A lower air exchange rate tends to increase the concentration of pollutants that have indoor sources.[5] Additionally, specific conservation measures may themselves contribute to the source of indoor pollutants. Examples of such practices include insulating with urea-formaldehyde foam insulation (UFFI), which can release formaldehyde, and the use of rockbeds for solar heat storage, which may release radon into the indoor environment.

With the reduced air exchange rates brought about by conservation, there is additional concern over the consequence of the introduction of consumer products that may emit pollutants into the home. The use of synthetic materials, many of which contain organic chemicals, has increased dramatically since World War II. Pressed-wood products, which are used extensively in the construction of dwellings, are examples of consumer products that can have a significant impact on indoor air quality. The bonding materials, especially urea-formaldehyde resins, used in the manufacture of pressed-wood products contain formaldehyde, which may be released over several years into the living environment.

Mobile homes are an example of how the use of such products can cause a substantial deterioration in indoor air quality.[1]

Although increased attention is being given to indoor air quality, as evidenced by an increase in relevant publications, committee reports, and reviews,[1,2,4-10] there is still a paucity of quantitative data on residential levels of both organic and inorganic pollutants. To extend the data base available to researchers evaluating the potential health risk due to indoor air pollutants, the U.S. Consumer Product Safety Commission (CPSC) and the U.S. Department of Energy (DOE) supported a study by Oak Ridge National Laboratory (ORNL) of 40 homes in the Oak Ridge/Knoxville area of East Tennessee. This work is the first study to monitor such a wide range of indoor pollutants in a sizable number of homes with repetitive measurements over the course of a year. The objectives of the study were to quantify the levels of formaldehyde, CO, NO_x, particulates, and radon, as well as selected volatile organic pollutants. In addition, information on air exchange rates, meteorological conditions, and structural and consumer products use was collected to assess the correlation of pollutant levels with these parameters. Field measurements were conducted in the 40 homes from April 1982 through February 1983 so that measurements could be conducted during each season of the year. Scoping measurements in six houses were conducted prior to the beginning of the larger study. During the winter phase of the study, emphasis was placed on a comparison of homes with internal combustion devices (gas stoves, wood stoves, unvented space heaters, etc.) and homes without such combustion devices.

3. STUDY DESIGN AND IMPLEMENTATION

The population size for the Indoor Air Quality Study was selected based on a compromise between the need for a statistically large sample size and the limitation of available resources. A population size of 40 homes was deemed appropriate. A cross section of houses in areas categorized as industrial urban, residential urban, residential semiurban, and rural was desired. This cross section would span the spectrum of outdoor air qualities. However, due to practical considerations homes in Oak Ridge and west Knoxville, representing essentially a semiurban population, were monitored. Homes spanning the age spectrum from new to older than 15 years were included. A distribution of various heating and cooking sources and home construction materials, especially insulation type, was also sought. The homes studied used electricity as the primary source of energy since such homes predominate in this area. A detailed survey of each home was performed and included in the data base for subsequent analysis.

The pollutants selected for monitoring during the study were chosen on the basis of their potential exposure levels and probable health effects concern. The pollutants chosen for monitoring were: formaldehyde, volatile organics, combustion gases (CO_x, NO_x), particulates, and radon. Measurements were made of air exchange rates and of meteorological conditions for subsequent correlation with the indoor air quality.

The data obtained from the house survey, the pollutant measurements, the air exchange rates, and the meteorological conditions were maintained in a large Statistical Analysis System (SAS)[11] data base prepared to allow easy access to the data for analysis. Correlations and relationships of the various parameters can easily be evaluated using this system. The entire data base is available on a 9-track magnetic tape.

3.1 House Selection

The selection of the homes to participate in the study included solicitation of volunteers, evaluation of respondent characteristics relative to the desired characteristics, and resolicitation to obtain additional volunteers in certain areas. Education of the homeowners on various aspects of the study was provided.

The industrial urban area in and around central Knoxville was excluded from the study because the 1982 World's Fair was held there from May through October, 1982, and the anticipated crowds and traffic congestion were judged to be too serious a detriment in conjunction with the long travel distance (about 50 miles round trip). Houses in outlying rural areas also presented serious logistics problems and were excluded because the time and effort required to visit and monitor them could not be met within the budget.

3.1.1 Solicitation of volunteers

The selection of homes for the study was restricted to residential semiurban areas of Oak Ridge and west Knoxville. During the selection process, houses that were clustered together geographically were given preference. On a given day, whenever a number of houses were being visited for the dropping off and/or picking up of monitoring devices, it was particularly cost-effective if the houses to be visited were located reasonably close to each other.

The primary vehicle for solicitation of volunteers was a number of articles in local newspapers. In addition to people reading and responding to these articles, knowledge of the impending study spread by word of mouth. Approximately 150 people offered to join the study.

In spite of the gratifying numerical response, some important categories of houses were lacking. For example, by the end of March, only three houses were available that were less than 5 years old. Responses were predominantly from established residents of the area who were living in houses 15 years old or older. The lack of response from owners of new houses was due in part to the severe recession in new house construction. This problem was rectified by a last minute effort to spread the word of our need by neighbor-to-neighbor contacts and by the article of March 30 in the *Oak Ridger* newspaper. In oral communications with new homeowners it was learned that many new homeowners' free time and interest are taken up with establishing the home and grounds and/or in rearing small children, which resulted in a lower response from this group. The final number of houses less than 5 years old was 18.

Another imbalance was the lack of homes that used gas for cooking. The historical availability from the Tennessee Valley Authority (TVA) of relatively cheap electric power is the reason for the heavy bias towards electric cooking.

Several characteristics were used in the final selection of homes for participation in the study. There was a desire to have about 25% of the homes with urea-formaldehyde foam insulation. The design also sought to include a large number of new homes. Another factor that influenced the final selection was a desire to have a range of heating and cooking sources represented in the study. A final, but important, factor was the location of the homes. Efficient use of resources required some clustering of homes. The final selection of homes was not considered to be biased with homeowners who had specific health complaints related to poor indoor air quality. Section 5.1 describes the characteristics of the selected homes.

The homeowner identity and specific location of the homes in the Indoor Air Quality Study were maintained in confidence. The house identification number used in data reporting is based on the number assigned when the homeowner volunteered for the study; consequently the number may be greater than 40. Figure 3.1 shows the general location of the houses selected in Oak Ridge and west Knoxville.

3.1.2 Homeowner briefing

On the evening of April 1, 1982, a meeting was held with prospective homeowner participants in the auditorium of the Museum of Science and Energy in Oak Ridge. The objectives were to give a briefing about the nature of indoor air pollution and energy conservation, describe the activities and purpose of the impending field study, and answer questions from the audience. These proceedings were handled by Drs. R. B. Gammage and D. C. Parzyck. Also in attendance were the members of the field monitoring teams, who had assembled the equipment that was to be used in the houses during the study. These instruments were operated at the end of the meeting so that people were aware of their size and noise levels.

About 70 people attended the meeting, representing 37 residences. The audience was enthusiastic and asked many good questions both from the floor and in later one-to-one

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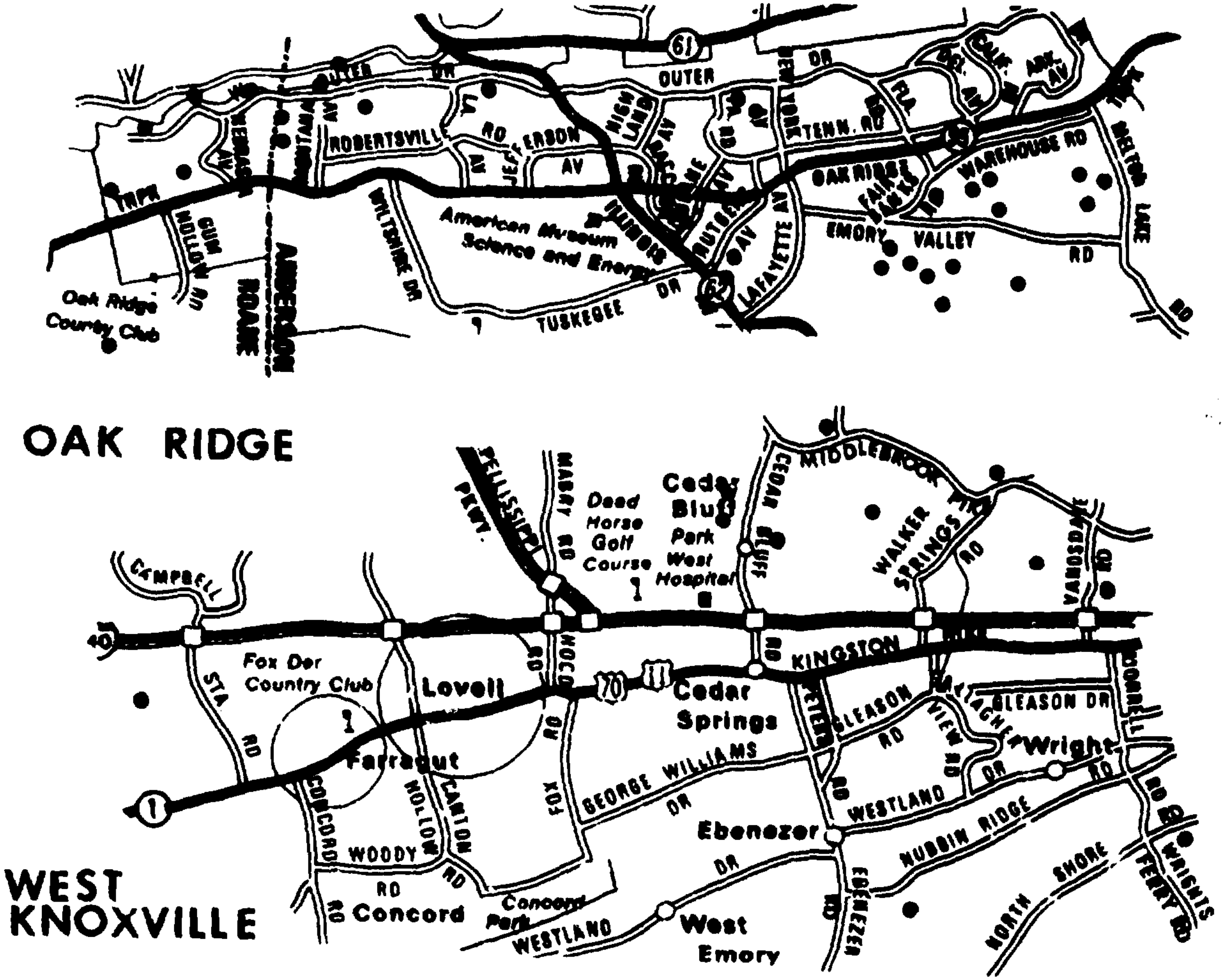

Fig. 3.1 Partial maps of Oak Ridge and Knoxville. Dots indicate location of study houses.

discussions. The issue of greatest concern to most of the attendees was not having their homes included in the study.

Upon submission of the Final Report to CPSC, another meeting with the homeowners is planned. Gratitude will be extended to the homeowners for their assistance and forebearance. The general results of the study will be discussed and the results of measurements in particular houses will be made available to the respective occupants. A copy of the final report will be provided to any homeowner who requests one.

3.2 Scoping and Special Studies

A lapse of 8 months occurred between initiation of the original contract and the start of the main phase of the study. During this period, newly developed monitors were tested more thoroughly under both laboratory and field conditions. As a result, their reliability, accuracy, and precision became substantially better. Scoping studies were carried out in six houses. The logistics of house visitations were examined and modified. Instrumental and sample collection inadequacies were measured and rectified. Unexpected and important findings led to new or modified activities being incorporated into the monitoring protocol. A series of special studies not included in the original work statement were identified and conducted either before or in parallel with the main phase of the study. A few specific examples will be given to illuminate each category.

The extended scoping phase proved beneficial for making improvements to the design and operation of the passive, semipermeable membrane monitors for formaldehyde. Improved cleanliness increased the operating lifetime of the detectors. After repetitive use (about six exposures in the field), however, a dramatic drop in the sampling rate was often experienced. Once the phenomenon was recognized, the passive formaldehyde monitors were routinely discarded after six field uses. It was also discovered that reliability and precision were improved if the detectors were stored containing water, so that the semipermeable membrane did not become dry.

The preliminary house measurements revealed other information about airborne formaldehyde concentrations. Upon closing up a house (#4), the formaldehyde concentrations increased nearly to maximum levels within three-quarters of an hour. Consequently, a close-up period of 1 h was chosen for establishing a near maximum formaldehyde concentration prior to starting a formaldehyde collection via the amended NIOSH procedure. It was also determined that the location of the passive formaldehyde monitors in a room had little effect on the measured concentration, provided they were not placed close to or on top of a formaldehyde emitting surface.

Several logistical problems became evident during the early phase. Without extending the collection of the passive formaldehyde monitors (exposed for 24 h) into the weekend, field exposures could only be conducted for an effective 4 days per week (depositing monitors Monday through Thursday and picking up monitors Tuesday through Friday). Maintenance of twice monthly visitations to each of the 40 houses required that detectors be deposited or picked up at 10 houses on 3 days of the week and at 5 houses on 2 days of the week. This operation required a field team of two people (Team A). Despite best intentions, this schedule inevitably developed flaws. There was last minute unavailability of houses for monitoring because of guests or homeowner absences and public holidays for the survey team. It was impossible to maintain a rigorous and uninterrupted schedule of twice-monthly measurements of formaldehyde in the 40 houses over the whole of the 9 months; thus some measurements were missed.

A collection problem was experienced initially in the trapping of airborne organic species on 2.5 g of Tenax sorbent. At the originally chosen pumping rate of 1.5 L/min of air over a 24-h period, too much organic matter of moderate volatility was being collected. Consequently, no more than 1 mg of Tenax could be used for the GC analysis without saturating the response of the detector. Weighing a small amount of Tenax introduced a substantial error, an error that was subsequently reflected in the quantitative analysis of individual organic compounds. The problem was greatly reduced by decreasing the rate of air flow and the collection period to allow 60 mg of Tenax to be analyzed. This collection regime was tailored for the monitoring of organic compounds of medium and low volatility (range of C_6 to C_{12}) such as styrene, dichlorobenzene, and naphthalene. Unfortunately, these parameters were unsuitable for quantitatively analyzing highly volatile organic compounds, such as the chlorinated methanes and ethylenes, because of breakthrough.

On February 18, 1982, a trial full-scale monitoring of pollutant levels inside house #7 was conducted by survey Team B (two persons). The purpose was to uncover problems in logistics and instrumentation. Measurements were made of CO, NO_x, particulates, volatile organics (Photovac portable GC), formaldehyde, air exchange rate (ach), and meteorological parameters. Other than the general experience gained from this exercise, the unexpected and important finding was made that the air exchange rate was greatly increased (fourfold on this occasion) by operation of the heating, ventilation, and air conditioning (HVAC) air circulation fan. This led to fan-on and fan-off measurements of the air exchange rate being made in all houses with central HVAC systems.

Another important finding was that the changes in concentration of formaldehyde were less than inversely proportional to the air exchange rate. This result led to more extensive and special studies of the relationship between concentration of formaldehyde and air exchange rate.

Other unexpected and protocol-modifying findings were the relatively high levels of radon measured by the Wrenn chambers in a few ridge-top houses. Consequently, all houses were monitored with passive, integrating radon monitors.

In addition to the twice-monthly measurements of formaldehyde concentration, the need was perceived for the measurement of more finely resolved temporal measurements of formaldehyde concentrations. The resultant special studies of diurnal and seasonal fluctuations, and of the effects of varying ventilation rate and air cleaning, were conducted largely during the scoping period. These results have been published.[12]

3.3 Authorizations

Before the main phase of the 40-house field study began, the following authorizations were obtained:

I. Approval of study protocol by CPSC Liason Officer.

An approved study protocol was a prerequisite and required iterations before final approval. See Section 3.4 for a discussion of the protocol.

II. Approval from the Committee on Human Studies, Oak Ridge Associated Universities and Oak Ridge National Laboratory.

An application (Ident. No. ORNL-7) for the use of humans as experimental subjects was submitted on October 2, 1981. Questions by the Committee were focused on the injection of Freon-12 into the houses during measurement of air exchange rates. It was agreed that the occupants should vacate their home for the 1-2 hours necessary to close up the house and make the measurements. The maximum concentration of Freon-12 was estimated at 20 ppm. (The OSHA TLV for 8-h continuous exposure is 1000 ppm.) Carbon dioxide was to be used if any homeowner objected to the use of Freon. After a final meeting to appear before the committee and answer further questions, approval for conduct of the study was granted on October 29, 1981. Formal signed documentation was not received, however, until April 6, 1982.

III. Approval to start the study from the Office of Management and Budget (OMB).

Approval by OMB was delayed until the middle of February 1982. The approval number for the project was OMB-3041-0010. This authorization was obtained by CPSC officials.

IV. Homeowner's Agreement signed by each participant.

The Homeowners' Agreement (included in Appendix A) was approved by CPSC and was signed by each homeowner (both spouses, where appropriate) and the UCCND representative, Dr. Herman Postma, the ORNL Director.

3.4 Study Protocol

The final approved study protocol is included as Appendix A of this report. Further changes to the protocol were the result of a meeting with CPSC Health Sciences staff in October 1982. The primary modification was to extend the monitoring activities through January and February 1983 when the effect of combustion sources on indoor air quality would be assessed.

The field monitoring portion of the Indoor Air Quality Study consisted of several phases. One phase monitored the formaldehyde levels in the 40 homes on a regular basis from April 1, 1982, until December 15, 1982, to evaluate the variation of levels as a function of environmental conditions. Measurements were made in three rooms per house on a twice per month basis during the first two months of the study. The monitoring frequency was then reduced to one visit per month and one room per house in those houses where formaldehyde levels were below 0.1 ppm and where the variation between room readings was not large. Occupant behavior (such as opening and closing windows) was recorded but not modified during these measurements.

A second component of the study consisted of single-day visits to each of the 40 homes in the study. These visits (typically 6 h of monitoring) focused on measurements of formaldehyde, nitrogen oxides, carbon monoxide, and particulates. Measurements of air exchange rates and meteorological conditions were also conducted. The warm weather phase of this component was conducted from April 1, 1982, until July 16, 1982. The monitored house was closed one hour before and during the course of these measurements to reflect pollutant levels due primarily to indoor sources.

Volatile organic monitoring using Tenax sorbent was conducted from April until December in this study. No modification to the homeowner's lifestyle was made during this 24-h measurement.

Radon monitoring with hourly printout of radon levels was conducted periodically from the beginning of the study until January of 1983. This monitoring procedure would typically place the measurement chambers in a home for a period of about one week. No modifications of lifestyle were required of the home occupants.

Monitoring protocols were modified for the winter phase of the study to concentrate on a comparison between homes with combustion devices (fireplaces, wood stoves, space heaters, etc.) to homes without such devices. A subset consisting of 20 houses was selected (10 control houses and 10 combustion device houses) for single-day measurements during the winter monitoring period. Table 3.1 lists the houses selected for the winter study and the dates visits were made. The ten homes selected as control homes are also listed along with the date visited. Carbon dioxide and sulfur dioxide measurements were added to the list of pollutants monitored. Due to instrument malfunction, the SO_2 measurements were not meaningful. Table 3.2 lists the houses selected for winter organic sampling with Tenax sorbent and the dates of the sampling. More extensive radon measurements were also conducted during the winter phase of the study.

3.5 Quality Assurance

Throughout the study, an active internal quality assurance program (described in Section 4) was in effect. In addition, two other quality assurance exercises were carried out. On June 24-25, 1982, a visit to ORNL was made by two CPSC liaison officials, Drs. K. C. Gupta and W. Porter. They observed and audited laboratory and field activities. Included was a visit to house #21 to observe firsthand the measurements conducted by both A and B survey teams. Also at this time, a measurement was made of the specific leakage area using a blower door fan. Advice was rendered for improved measurement of the sampling rates of passive formaldehyde monitors. A protocol was jointly agreed upon for laboratory validation of each of the five formaldehyde monitoring techniques being used in the study.

The indoor air quality study was also audited on August 2, 1982, under ORNL's internal quality assurance program. The auditing officer, in his written report, gave a favorable report on the quality assurance aspects of the study and stated that there were no quality-related failures or nonconformances.

Table 3.1. Houses monitored by Team B during the winter season with combustion sources identified

House number	Dates of visits	Combustion source(s)	Comment
24	01-17-83 02-03-83	Wood stove	Stove operated both visits, monitors 2 m from stove
39	01-18-83 02-09-83	Wood/coal stove	Stove operated both visits, smoking problem with stove on second visit, heavy[a] cigarette smoking both visits
46	01-06-83 01-24-83	Fireplace	Operated second visit only, heavy[a] cigarette smoking first visit, moderate[b] cigarette smoking second visit
54	01-26-83 02-07-83	Wood stove	Operated both visits
55	01-31-83 02-08-83	Wood stove	Operated both visits
58	01-20-83 02-02-83	Gas cooking, fireplace	Gas stove operated briefly during both visits, fireplace not operated
62	01-07-83 01-27-83	Fireplace	Operated second visit only
65	01-25-83 02-11-83	Wood stove	Operated both visits
68	01-28-83 02-10-83	Kerosene space heater	Operated both visits
83	01-03-83 02-01-83	Gas cooking	Gas stove operated briefly during first visit
2	12-13-82		Control home
5	12-14-82		Control home
12	12-17-83		Control home
14	12-16-83		Control home
26	01-11-83		Control home
36	12-15-82		Control home
37[c]	12-15-83		Control home
70	01-19-83		Control home
79	12-09-82		Control home
80	01-12-82		Control home
82	01-05-83		Control home

[a] Two or more people smoking two or three cigarettes each during visit.

[b] One person smoking more than one cigarette during visit.

[c] Not visited by Team B. Monitored for organics (Tenax sample) only.

Table 3.2. Combustion source houses monitored for organics using the Tenax sampler (winter season)

House number	Dates of Tenax sampling[a]	Combustion source(s)	Comment
24	01-26-83	Wood stove	Tenax sampler placed in room with stove[b]
39	02-11-83	Wood/coal stove	Tenax sampler not placed in room with stove[b]
46	02-02-83	Fireplace	Fireplace not operated during sampling time
54	02-10-83	Wood stove	Tenax sampler placed in room with stove[b]
55	02-02-83	Wood stove	Tenax sampler not placed in room with stove (stove operated entire sampling time)
58	02-08-83	Fireplace, gas cooking	No combustion source operated during sampling time
62	01-18-83	Fireplace	Tenax sampler placed in room with fireplace (fireplace operated 30 minutes during sampling time)
65	01-31-83[c]	Stove	Tenax sampler placed in room with stove[b]
68	02-08-83	Kerosene heater	Tenax sampler placed in room with space heater (space heater operated 2 hours during sampling time)
83	02-10-83	Gas cooking	Tenax sampler not placed in kitchen (gas stove operated 30 minutes during sampling time)
5	12-14-82		Control home
14	12-16-82		Control home
26	01-11-83		Control home
37	12-15-82		Control home
70	01-18-83[d]		Control home

[a] Sampling time typically 6-h.

[b] Stove operated during entire sampling period.

[c] 02-11-83: outside sample.

[d] 01-19-83: outside sample.

ABSTRACT

Over a one-year period, measurements of indoor air pollutants (CO_x, NO_x, formaldehyde, volatile organics, particulates, and radon) were made in 40 homes in East Tennessee. The houses were of various ages with different types of insulation and heating. Over one-half of the houses exceeded the ASHRAE indoor ceiling guideline of 0.1 ppm for formaldehyde on at least one occasion. Over the duration of the study, older houses averaged 0.04 ppm of formaldehyde while houses less than 5 years old averaged 0.08 ppm ($P < 0.01$). The highest concentration of formaldehyde measured was 0.4 ppm in a new home. Diurnal and seasonal fluctuations in levels of formaldehyde in some homes were as much as twofold and tenfold, respectively. The highest levels of formaldehyde were usually recorded during summer months. The concentration in indoor air of various organics was at least tenfold higher than in outdoor air. Carbon monoxide and nitrogen oxides were usually <2 and <0.02 ppm, respectively, except when gas stoves or kerosene space heaters were operating, or when a car was running in the garage. In 30% of the houses, the annual indoor guideline for radon, 4 pCi/L, was exceeded. The mean radon level in houses built on the ridgelines was 4.4 pCi/L, while houses located in the valleys had a mean level of 1.7 pCi/L ($P < 0.01$). The factor having the most impact on infiltration was operation of the central duct fan of the heating, ventilation, and air conditioning system. The mean rate of air exchange increased from 0.39 to 0.74 h^{-1} when the duct fan was operated (measurements prior to December 1982). This report presents the study design and implementation, describes the monitoring protocols, and provides a complete set of the data collected during the project.

4. MATERIAL AND METHODS

This section provides a description of the equipment and experimental protocols used in the Indoor Air Quality Study. When different equipment and/or procedures were used in different phases of the study, these deviations will be noted.

4.1 Formaldehyde Monitoring

Measurements of the levels of formaldehyde in the air were made using a variety of techniques. These measurements included 24-h integrated samples, 15-min short-term samples, and various quality control tests. The passive membrane samplers and the pumped molecular sieve samplers were developed under an earlier CPSC- and DOE-supported effort aimed toward developing sensitive, convenient, and cost-effective monitoring techniques for measuring formaldehyde at low concentrations. Laboratory and field intercomparisons were also conducted.

4.1.1 Passive membrane samplers

Passive membrane dosimeters[13] developed at ORNL were used for sampling formaldehyde concentrations over a 24-h period. The device consists of a semipermeable dimethylsilicone membrane 0.0025 cm (1 mil) thick (General Electric, Schenectady, NY) over a 15.2-cm^2 opening in a small polyethylene container. The membrane controls the sampling rate and contains the 5 mL of water sorbent used in the sampler. One-foot-square sheets of membrane were obtained, and approximately 50 samplers were produced from each sheet. The glue used to attach the membrane to the container was tested to ensure that it did not contribute to the measured formaldehyde levels. The devices can be reused several times before becoming visibly dirty and producing erratic sampling rates. The samplers do exhibit a slight face velocity effect when operated in very still environments or in high velocity environments.[13]

Figure 4.1 shows the samplers being collected in one of the study homes. Also shown are the slightly larger transportation containers used to keep the samplers clean. Initially, the samplers were scheduled to be taken to and from the field in these transportation containers with the sorbent water in the samplers. However, concern over the effectiveness of the seal in preventing sampling during transportation led to the practice of adding the water to the sampler in the field. At the end of the 24-h sampling period the sorbent was removed and the water-formaldehyde sample returned to the laboratory in a sealed glass vial for analysis. Laboratory analysis was conducted on the same day that samples were collected to minimize any changes that may occur during storage.

The analysis was performed using the pararosaniline colorimetric method as modified.[14] Using a 24-h sampling period and the modified pararosaniline analysis, a detection limit of approximately 25 ppb of formaldehyde in the air was obtained. Blanks (unexposed dosimeters) were prepared each day to provide a measure of quality control. Figure 4.2 summarizes the performance of the passive samplers based on a large number of laboratory tests. A sampling rate of 0.13 μg/mL per ppm-h was used for data analysis unless otherwise noted. See Sect. 4.1.3.3 for details on the calibration facility.

4.1.2 Pumped molecular sieve samplers

Short-term formaldehyde concentration measurements were made using the pumped molecular sieve method.[15] The device consists of a 5-g cartridge of 13X molecular sieve sorbent through which the sampled air is pumped at 2 L/min for 15 min. A DuPont P2500 or P4000 pump with feedback flow control was used to draw air through the sorbent. Figure 4.3 shows this technique in use at one of the study homes.

The sieve sorbent was transported to and from the study houses in sealed glass containers tested to ensure that the formaldehyde content in the sieve neither increased nor decreased over extended time periods. The formaldehyde was extracted from the sieve sorbent using a simple 20-min distilled-deionized water rinse, and the subsequent formaldehyde-water sample was analyzed using the modified pararosaniline method. With this sampling and analysis procedure, a detection limit of approximately 25 ppb was obtained for formaldehyde vapor concentration.

Newly purchased molecular sieves were conditioned prior to use by flowing distilled water through the sieves for about 2 days. The molecular sieve sorbent can be reused up to 5 times with this cleaning procedure before there is physical deterioration of the sieve. Prior to use, the sieves are dried for more than 20 min at about 150°C and sealed in medicine vials with rubber septa for transport to the field. Analyses of unexposed blank sieves were made each day as a quality control measure.

ORNL PHOTO 0737-82

Fig. 4.1 Member of Team A placing passive samplers in a house.

4.1.3 Quality control measurements

Both laboratory and field quality control measurements were conducted throughout the study. Long-term and short-term comparisons were made between the various monitoring techniques.

4.1.3.1 NIOSH impinger/chromotropic acid procedure - The NIOSH procedure[16] designed for occupational monitoring and frequently used for assessing formaldehyde levels in residential environments was used to obtain comparisons with the other methods and as a short-term reference method. The procedure consisted of a double impinger sampling train with 20 mL of 1% sodium bisulphite solution in each impinger. Air was pumped through the impingers at 1 L/min for 1 h using a DuPont P4000 pump. Subsequent analysis was performed using the chromotropic acid analysis method. The detection limit for this procedure was approximately 60 ppb,[13] making it of only marginal applicability in many of the study homes.

4.1.3.2 LBL refrigerated sampler - A refrigerated sampler on loan from Lawrence Berkeley Laboratory (LBL) served as a reference sampler to compare with the passive membrane samplers used for 24-h measurements. This unit consists of four sampling trains, each train containing a series of two water impingers and a critical orifice for flow control. An integral pump maintains sufficient pressure drop across the critical orifices to give a constant air flow, and the entire sampling train is maintained at a refrigerated temperature to minimize evaporation from the impingers and to reduce any effects of temperature-dependent collection efficiencies. Each impinger contains 25 mL of water, and the two impingers are pooled prior to analysis by the pararosaniline procedure. This unit is suitable for long-term (24 h) unattended sampling and is transportable but requires two people for transport and is relatively noisy for overnight use.

4.1.3.3 Calibration facility - The calibration of all passive and active formaldehyde sampling units was accomplished using a dynamic generation apparatus. A schematic of

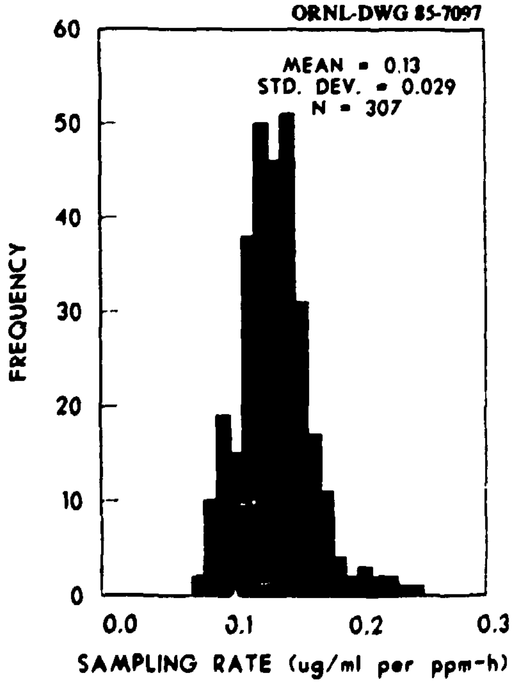

Fig. 4.2 Sampling rates measured for passive membrane samplers.

the apparatus, which summarizes the operational characteristics of the system, is shown in Fig. 4.4. Dilute formalin solution is injected into a multiply-filtered, heated air stream to supply formaldehyde vapor of known concentration and humidity to a 0.2 m^3 Teflon-lined chamber. The injection rate is controlled by a Sage Instruments Model 341A syringe pump. The dilution air flow rate is controlled using Matheson mass flow control valves. The water vapor concentration can be controlled by both the temperature of the water bubbler and the relative amount of air passing through and bypassing the water bubbler. The chamber smoothes any intermittent fluctuations in the formaldehyde concentration produced by the generation apparatus. This generation system was used to produce formaldehyde concentrations in the range of 0.005 to 10 ppm. Regular cleaning of the chamber helped keep wall loss effects to a minimum.

The exposure chamber was also equipped with a microcomputer-controlled turntable that could be adjusted to various speeds. This device allowed simulation of various face velocities for use in evaluating the response of passive monitors.

ORNL PHOTO 0736-82

Fig. 4.3 Demonstration of pumped molecular sieve technique.

4.1.4 Intercomparison measurements

In addition to the two ORNL-developed sampling methods and the two previously mentioned quality control methods, an intercomparison between these methods and several alternative sampling procedures was conducted. Two other passive samplers and the CEA 555 continuous formaldehyde monitor were briefly evaluated. The 3M and Air Quality Research (AQR) passive samplers were received from CPSC and returned to the vendor for analysis. No control was maintained over the age of the passive samplers prior to testing. Both laboratory and field intercomparisons were conducted. Insufficient resources were available to adaquately evaluate problems associated with the performance of these devices.

4.2 Combustion Gases Monitoring

The oxides of nitrogen and carbon were measured during one-day visits to the study houses. Instrumental methods providing real-time response were used; thus no subsequent laboratory analysis was required.

4.2.1 Nitrogen oxides

Concentrations of nitrogen oxides were measured using a Monitor Labs Nitrogen Oxides Analyzer (Model 8840), a gas-phase chemiluminescence device capable of performing continuous dry analysis of nitric oxide (NO), total nitrogen oxides (NO_x), and, by subtraction, nitrogen dioxide (NO_2) assuming that the total NO_x results from NO and NO_2. Real-time analysis is made possible by dual reaction cells and detectors—a separate cell for the NO and NO_x channels.

Instrument operation is based on the chemiluminescence of an activated species of NO_2 produced by the chemical reaction between NO and ozone (O_3). As the chemiluminescence reaction is between O_3 and NO, a direct reading of the NO concentration is obtained. For the NO_x measurement, the NO_2 is reduced chemically to NO by a catalytic molybdenum converter prior to entry into the NO_x reaction cell where the resulting total NO concentration is measured. A photomultiplier tube with each channel measures the intensity of the radiation emitted as the activated NO_2 species reverts to a lower energy state. The signals are amplified and provided as voltage outputs.

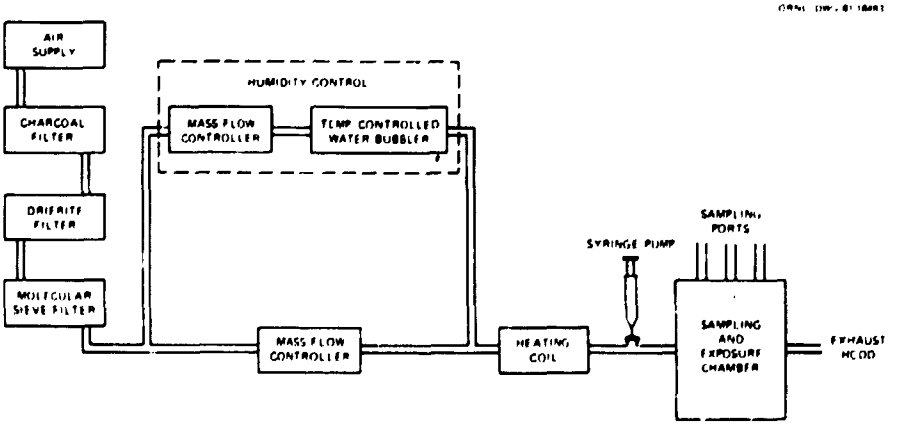

Fig. 4.4 Diagram of formaldehyde test atmosphere generation apparatus.

Accurate air flow control is maintained with critical orifices and a vacuum pump. The minimum detectable concentration is approximately 2 ppb. Calibration of the nitrogen oxides analyzer was conducted using a Monitor Labs 8500 calibrator system with standard NO and NO_2 gas mixtures.

4.2.2 Carbon monoxide

Carbon monoxide concentrations were measured by electrochemical detectors. During the summer phase of the study, a General Electric SPE Carbon Monoxide Detector (Model 15ECS3C01) was used. This instrument is a self-contained direct-reading unit that continually displays ambient levels of CO in ppm on a LCD readout. Sampling is active with ambient air drawn into the detector through a selective filter by an integral air sampling pump. The sample is discharged to a hydrated, solid polymer electrolyte sensor cell where the CO is oxidized, producing a voltage output proportional to the CO level. The solid polymer electrolyte is in the form of a plastic sheet. The lower detectable limit for this detector is about 1 ppm.

Monitoring of CO levels during the winter phase was conducted with an Interscan Series 5000 Dosimeter, an electrochemical voltametric detector with continuous sampling of the environment under passive, diffusion-controlled conditions. Carbon monoxide is detected by a gas diffusion type electrocatalytic sensing electrode. The gas molecules diffuse into the device and are absorbed onto the electrode, where they are electrochemically oxidized at an appropriate electrode potential. This electrochemical reaction generates an electric current directly proportional to the CO concentration. A detection limit of 2.5 ppm was observed for this monitor. Calibration of both detector types involved zero checks with high purity air and span setting with carbon monoxide gas standards in the 50 ppm range.

4.2.3 Carbon dioxide

Carbon dioxide measurements were made using a MIRAN-80 single-beam infrared analyzer. Wavelength scans were performed between 3.90 μm and 4.30 μm. The maximum absorbance corresponding to the CO_2 absorption wavelength and a baseline value were obtained from these scans. Through the use of a calibration plot of absorbance vs concentration prepared from a known standard, and dilution of this standard, levels of carbon dioxide were determined for a subset of the houses visited.

4.3 Volatile Organic Measurements

Volatile organic compounds were monitored using two different techniques. One technique used a sorbent collection procedure with subsequent laboratory analysis by high resolution GC and/or GC/MS, while the other technique used a portable GC capable of providing on-site analysis. The two techniques complement each other in that the former measures the more volatile compounds while the latter measures the less volatile compounds.

4.3.1 Tenax sorbent traps

Volatile organic pollutants were measured with Tenax-GC as a sorbent.[17] Cartridges containing 2 to 2.5 g of sorbent were used at a sampling rate of approximately 1 L/min (flow rates ranged from 0.61 to 1.80 L/min as experience was gained with loading and breakthrough). DuPont P4000 pumps were used to flow air through the sampling cartridges. During the first phase of the study, sampling was conducted for approximately 24 h. Sampling time was decreased to 6 h during the winter phase to reduce breakthrough of the more volatile compounds (benzene and lighter) and to complete collection during one working day.

Subsequent analysis was performed using GC and/or GC/MS.[18] Quantitative results were obtained for 17 of the compounds most frequently found in the homes. The 17 compounds were selected partly on the basis of certainty of identification and availability of authentic standards. A typical detection limit for this sampling and analysis procedure is about 1 $\mu g/m^3$.

4.3.2 Photovac GC

Detection of volatile organic vapors in several homes was effected by use of a Photovac 10A10 portable photoionization gas chromatograph (Photovac, Inc.). The 10A10 is a gas-only system with the chromatographic column operating at ambient temperature conditions. The column used was the standard column supplied with the instrument, a 4-ft SE-30 56 on 60-80 mesh Chromosorb G. Typically, the sample volume of indoor air injected was 1 mL, with a carrier (ultrapure air) flow rate of 15 mL/min. Output was connected to a Hewlett-Packard 7004B X-Y recorder. Because insufficient work has been done in calibration of this instrument for qualitative and quantitative analysis, this portion of the study was conducted primarily as reference for future investigation. Nevertheless, chromatograms obtained may be used as a "fingerprint" of their respective houses for comparisons of gross organic levels. Lower limits of detection vary with the individual compounds being measured, but are generally between 0.1 and 20 ppb. Sensivities vary in the general order: chlorinated ethylenes > aromatics > oxygenated solvents > chlorinated alkanes > saturated hydrocarbons.

4.4 Particulate Material

Particulate levels were measured using the model PC-2 Quartz Crystal Microbalance cascade impactor (California Measurements, Inc.) designed for the measurement of mass concentration and size distribution of fine particles suspended in air. The PC-2 is composed of a microprocessor control unit and a detachable sensing stack which, in the field, was fitted to a tripod for stability and height adjustment. The sensing stack of the instrument operates at a flow rate of 240 mL/min and has ten stages that separate aerosol samples into ten fractions between 25 and 0.05 μm. Piezoelectric quartz crystal sensors are used in each stage as mass monitors to provide real-time mass collection data. Ten frequency signals, one emanating from each stage, are fed into a microprocessor-based control unit where they are stored and processed to yield calculations of mass concentration in units of mg/m^3. As operated, the PC-2 provided repeated sets of data in 15-min intervals during the summer phase of the study and in intervals of 5-min during the winter months. The instrument can detect aerosol samples in a concentration range between 10 $\mu g/m^3$ and 60 mg/m^3. In a control test performed in the laboratory environment, the particulate analyzer exhibited good stability. The short-term (a few hours) standard deviation of the ten channels was generally less than 0.1%, while the longer-term (overnight) variation was generally less than 0.5%.

4.5 Radon Monitoring

Radon levels were monitored during the study using two different techniques. An active technique was used to obtain temporal fluctuations of radon levels in selected homes, and a passive technique was used to determine levels in all 40 study homes.

4.5.1 Wrenn chambers

Measurements for radon (^{222}Rn) were performed in a number of homes using an ORNL-developed instrument based on a design which depends upon the diffusion of radon gas through a polyurethane foam layer.[19] The foam serves to trap the particulate radon daughters and allows only radon to pass into a hemispherical chamber. A hemispherical light pipe coated with zinc sulfide scintillator and covered with aluminum to provide a conducting surface constitutes the alpha detector in this instrument. The alpha decay pulses from a photomultiplier are amplified, fed into an energy threshold selection circuit, and then to a counter-timer. The threshold discriminator rejects all but alpha pulses. The electronics control unit can support two monitoring chambers. Radon levels are automatically calculated based on the decay counts and recorded at fixed time intervals to allow continuous operation. As operated, concentration of radon (pCi/L) was obtained once per hour over periods lasting about one week. A detection limit of 0.25 pCi/L was determined.

4.5.2 Track etch monitors

During the winter phase of the study, Terradex track etch radon detectors[20] were placed in all of the 40 homes. These detectors are completely passive and are intended for use in relatively long-term monitoring. Fashioned from a light, plastic cup, the radon monitors are based upon plastic nuclear track detectors that are sensitive to the alpha radiation from decaying ^{222}Rn atoms. When alpha particles penetrate the track detectors, they produce radiation damage tracks that are retained for long periods of time. A filter installed over the mouth of the cup prevents entry of radon daughters and dust. After the detectors are returned from the field, analysis by the vendor (Terradex Corporation) involves enlargement of the damage tracks by a chemical etching technique. The tracks are then counted microscopically. The reading of the detector is in tracks per unit area and is proportional to radon exposure. Six detectors were placed per home (three in an upper-level room, three in a basement or lower level room) with three additional detectors being employed for outdoor monitoring in protective canisters at six of the homes to represent the study area. The period of measurement was approximately 3 months, from late November and early December to early March.

4.6 Air Exchange Rates

Air exchange rates were obtained by monitoring the concentration decay of Freon-12 gas after injection into the houses. Portable floor fans were used to mix the tracer during measurement periods. During the course of the measurement interior doors in the home were opened, while exterior doors and windows were closed. No attempt was made to quantify the efficiency of mixing between the various rooms with the portable fans off. The term "air exchange" as used in this report assumes a unity "mixing factor" due to our efforts to ensure good tracer mixing.

5. RESULTS

The experimental results of the Indoor Air Quality Study are included as Appendix B of this report. This appendix is provided on microfiche in the pocket on the inside back cover. The data bases listed in the microfiche have been provided to the CPSC Project Officer on a 9-track magnetic tape. The tape is a standard label 1600-bpi magnetic tape with the following format characteristics:

record length = IBM standard label default
record format = IBM standard label default
maximum block size = 13030 bytes
data set name = (see Table 5.1 for data set names)

The information is in a format suitable for input as data sets to the Statistical Analysis System (SAS). The variables included in each data set are detailed in the remainder of this section. Written comments on the results are confined to examples of the appropriate data set and to summary information in this section. Table 5.1 lists the data sets available and references the appropriate appendix. An indication of the number of observations in each data set is also provided.

There are several variables common to more than one data set. In general, these variables can be used to correlate observations in different data sets with respect to space and/or time. Table 5.2 provides a list of names and definitions for these common variables.

5.1 House Attributes

Two data sets describe selected attributes of each house that might affect indoor air quality. One data set, ORNL.XHSE, is described in Appendix B.1.1. These data derive from answers to a questionnaire provided to the occupants of each residence. Table 5.3 lists the attributes, categorical classes, and the associated variable names in the data set. Table 5.4 lists the percentage of the houses falling in each categorical class for the nonquantitative variables. Table 5.5 provides the mean and range for the quantitative variables. See Section 5.9 for air exchange rate data set.

5.2 Formaldehyde Measurements

Approximately 7000 individual formaldehyde measurements were made during the course of this study using the passive membrane sampler and the pumped molecular sieve technique. Measurements were conducted during the spring, summer, and fall using the passive samplers. Pumped samples were collected during the intensive one-day visits. Details of the measurement methods are presented in Section 4. The sampling strategy is presented in Section 3. Only summary results of the measurements will be presented in this section.

5.2.1 Passive formaldehyde sampler results

Based on 307 measurements using 154 samplers conducted in our calibration chamber during the course of this study, the sampling rate of the ORNL-developed passive membrane samplers was 0.13 g/mL formaldehyde in solution per ppm-h of exposure.

Table 5.1. Data set names and appendices containing the data

Data set name	Number of observations	Appendix
ORNL.XHSE	41	B.1.1
ORNL.XPASS	1601	B.2.1
ORNL.SSIV	40	B.2.2
ORNL.WSIV	240	B.2.3
ORNL.SNIOSH	40	B.2.4
ORNL.SORG	85	B.3.1
ORNL.WORG	17	B.3.2
ORNL.SCO	138	B.4.1
ORNL.WCO	355	B.4.2
ORNL.SNOX	39	B.5.1
ORNL.WNOX	2739	B.5.2
ORNL.SPART	475	B.6.1
ORNL.WPART	929	B.6.2
ORNL.WCO2	104	B.7.1
ORNL.SRDN	15	B.8.1
ORNL.WRDN	95	B.8.2
ORNL.XVNT	126	B.9.1
ORNL.XWOSA	442	B.10.1
ORNL.XWOSB	40	B.10.2
ORNL.XWNOAA	348	B.10.3
ORNL.XPRECIP	123	B.10.4

Table 5.2. Variables common to most data sets

Name	Definition
HSENO	Identification number for each house
DATE	Date of measurement in alphanumeric form
DAY	Date of measurement in Julian form
TIME	Time of measurement expressed as minutes since midnight
PLACE	"IN" or "OUT" indicating general location of measurement
RM	Specific room in which measurements were made

Table 5.3. House attributes and associated variables

Variable	Description of variable	Categorical classes
HSENO	House number	-
Y1	Group number	-
Y3	f house	Quantitative (y)
Y4	r of occupants	Quantitative (persons)
Y5	Cigarette smoking rate	Quantitative (packs/wk)
Y6	Living area	Quantitative (ft^2)
Y7	Type of house	1(1½ Stories); 2(Split Foyer); 3(2 Stories); 4(Rancher); 5(Tri Level)
Y8	Type of construction	1(Cemesto); 2(Frame); 3(Frame and Brick/Block); 4(Block or Brick)
Y9	Basement	1(None); 2(Finished); 3(Unfinished)
Y10	Garage	1(None); 2(Carport); 3(Attached); 4(Unattached)
Y11	Air conditioning	1(None); 2(Central); 3(Window)
Y12	Primary source of heating	1(Gas); 2(Electric); 3(Other)
Y13	Wood-burning	1(None); 2(Fireplace/used less than once per week); 3(Fireplace/used more than once per week); 4(Stove used as primary heat source); 5(Stove used as secondary heat source)
Y14	Space heater	1(None); 2(Electric); 3(Gas or Kerosene)
Y15	Cooking fuel	1(Gas); 2(Electric); 3(Other)
Y16	Wall insulation	1(None); 2(UFFI); 3(Fiberglass); 4(Other)
Y17	Type of UFFI	1(None); 2(Prefit); 3(Retrofit)
Y18	Age of UFFI	Quantitative (y)
Y19	Attic insulation	1(None); 2(Cellulose); 3(Fiberglass); 4(Rock Wool); 5(Other)
Y20	Vapor barrier	1(None); 2(Yes); 3(Unknown)
Y21	Air cleaner	1(None); 2(Room); 3(House)
Y22	Humidifier	1(None); 2(Room); 3(House)
Y23	Electrostatic precipitator	1(None); 2(Room); 3(House)
Y24	Ridge/Valley indicator	1(Valley); 2(Ridge)

Table 5.4. Categorical classification of study houses

Attribute	Percentage in various classes[a]				
Type of house	1½ Story 7.3	Split foyer 17.1	2 Story 19.5	Rancher 36.6	Tri-level 19.5
Type of construction	Cemesto 4.9	Frame 19.5	Frame&block/brick 70.7		Block or brick 4.9
Basement	None 41.5	Finished 41.5	Unfinished 17.1		
Garage	None 12.2	Carport 9.8	Attached 78.0	Unattached 0.0	
Air conditioning	None 4.9	Central 85.4	Window 9.8		
Primary heating fuel	Gas 24.4	Electric 75.6			
Wood burning	None 2.4	Fpl.[b] (<1/wk) 26.8	Fpl.[b] (>1/wk) 41.5	Stove(pri.) 14.6	Stove(sec.) 14.6
Space heater	None 87.8	Electric 9.8	Kerosene or gas 2.4		
Cooking fuel	Gas 4.9	Electric 95.1			
Wall insulation	None 7.3	UFFI [illegible].8	Fiberglass 53.7	Other 12.2	
Type of UFFI	None 73.2	Prefit 17.1	Retrofit 9.8		
Attic insulation	None 4.9	Cellulose 22.0	Fiberglass 51.2	Rock wool 12.2	Other 9.8
Vapor barrier	None 12.2	Yes 75.6	Unknown 12.2		
Air cleaner	None 90.2	Room 9.8	House 0.0		
Humidifier	None 73.2	Room 14.6	House 12.2		
Electrostatic precipitator	None 80.5	Room 4.9	House 14.6		
Ridge/Valley indicator	Valley 66	Ridge 34			

[a] Percentages may not add to 100% due to rounding.

[b] Fireplace.

Table 5.5. Quantitative description of study houses

Attribute	Mean	Range
Age of house (y)	11.9	0.1 - 45
Number of occupants	3.2	1 - 5
Cigarette smoking (packs/wk)	0.9	0 - 10
Living area (sq ft)	2439.7	750 - 3400
Age of UFFI (averaged over UFFI houses only) (y)	3.4	2 - 4.5

Analysis of variance in this data set suggests that the variance component due to sampler variability is about 2.8 x 10^{-4} (g mL^{-1} ppm^{-1} $h^{-1})^2$. Based on the analysis of this extensive data set, the estimated standard deviation associated with a single use of one of these passive samplers is about ±25%. The monitors were stored containing water between uses to condition the membranes. After repeated use (typically 5 or 6 times) the monitors had to be replaced because the variability in sampling rates increased.

Approximately 6000 passive membrane sampler measurements were conducted in 1601 room and outdoor visits from April 1, 1982, until December 15, 1982. Appendix B.2.1 contains the results of these measurements. Table 5.6 lists and defines the variables found in data set ORNL.XPASS. Results that indicated formaldehyde concentration below the detection limit of 25 ppb were entered in the data set as zero. These zero values should be treated by subsequent investigators in an appropriate manner. Subsequent calculations in this report based on this data set treat values below the detection limit as being equal to one-half the detection limit. Other data sets are handled similarly.

The mean and standard deviation of the 1149 room means in ORNL.XPASS are 78 and 82 ppb. The room means range from <25 to 634 ppb. Outdoor levels were typically below the detection limit of 25 ppb. If a less than detection limit is assigned a value of 12.5 ppb, then the mean outdoor level of the collected data was 16 ±18 ppb.

5.2.2 Pumped sieve results

During the intensive one-day monitoring visits, formaldehyde levels in the air were determined by pumping known volumes of air through molecular sieves and measuring the amount of adsorbed formaldehyde. These measurements were taken concurrently with measurements of other pollutants. During the first phase of the study, four consecutive 15-min samples were taken. Additional sampling was conducted during the winter phase to include periods when combustion sources were operating. The summer data are found in data set ORNL.SSIV and the winter data in ORNL.WSIV. The data are listed in Appendices B.2.2 and B.2.3, respectively. Table 5.7 lists the variables in these data sets. Outdoor samples were not taken with this technique.

5.2.3 Chromotropic acid results

The NIOSH recommended method of measuring formaldehyde, which involves chromotropic acid, was also used in this study to provide a reference to the frequently used industrial hygiene method of formaldehyde measurement. These data are found in data set ORNL.SNIOSH, and Appendix B.2.4 includes a listing of the data. Table 5.7 lists

Table 5.6. Variables in ORNL.XPASS

Variable	Definition
DATE	See Table 5.2
DAY	See Table 5.2
HSENO	See Table 5.2
RM	See Table 5.2
PPB	Formaldehyde level (ppb)
PPB_ERR	Standard error (ppb)

Table 5.7. Variables in ORNL.SSIV, ORNL.WSIV, and ORNL.SNIOSH

Variable	Definition
DATE	See Table 5.2
DAY	See Table 5.2
HSENO	See Table 5.2
PPB	Formaldehyde level (ppb)
PPB_ER[a]	Standard error (ppb)

[a] Not in ORNL.WSIV.

and defines the variables in this data set. The mean and standard deviation of the 34 data points are 49 ± 40 ppb. Values less than the detection limit of 60 ppb were averaged as 30 ppb in this calculation.

5.3 Organics

During both the summer and winter phases of the study, known volumes of air were pumped through Tenax sieve, and the absorbed organic species were analyzed. Seventeen individual species were quantified. Data sets ORNL.SORG and ORNL.WORG give the summer and winter data. In Table 5.8 the variables used and their definitions are listed while Appendices B.3.1 and B.3.2 list the summer and winter data. Table 5.9 provides means and ranges for the levels of each chemical during both seasons.

5.4 Carbon Monoxide Measurement

Carbon monoxide was measured during both the summer and winter phases of the study. Summer data are in data set ORNL.SCO, and winter data are in ORNL.WCO. These data are listed in Appendices B.4.1 and B.4.2, respectively. Table 5.10 lists and defines the variables in these data sets. The summer mean and range were 1.73 (0-27) ppm (N=376) while for noncombustion source homes in the winter the mean and range were 1.80 (0-10) ppm (N=58). Summary statistics for combustion source homes are not reported since data were generally collected during staged combustion episodes. These data should be appropriately analyzed by subsequent investigators. The mean outdoor level was 3.3 (0-6) ppm (N=17) during the summer. Reliable outdoor winter measurements could not be made due to the temperature sensitivity of the monitoring instrument.

Table 5.8. Variables in ORNL.SORG and ORNL.WORG

Variable	Definition
DATE	See Table 5.2
DAY	See Table 5.2
HSENO	See Table 5.2
PLACE	See Table 5.2
METHOD	Analytical method (GC or GC/MS)
Y1	Toluene ($\mu g/m^3$)
Y2	Ethyl Benzene ($\mu g/m^3$)
Y3	Xylene(meta and para) ($\mu g/m^3$)
Y4	Nonane ($\mu g/m^3$)
Y5	Cumene ($\mu g/m^3$)
Y6	Benzaldehyde ($\mu g/m^3$)[a]
Y7	Mesitylene ($\mu g/m^3$)
Y8	Decane ($\mu g/m^3$)
Y9	Limonene ($\mu g/m^3$)
Y10	Undecane ($\mu g/m^3$)
Y11	Naphthalene ($\mu g/m^3$)
Y12	Dodecane ($\mu g/m^3$)
Y13	2-Methylnaphthalene ($\mu g/m^3$)
Y14	Tridecane ($\mu g/m^3$)
Y15	Tetradecane ($\mu g/m^3$)
Y16	Pentadecane ($\mu g/m^3$)
Y17	Hexadecane ($\mu g/m^3$)
Y18	Benzene ($\mu g/m^3$)
Y19	Flag indicating that additional compounds were quantified

[a] Incomplete resolution - benzaldehyde and/or C_3-benzenes.

5.5 Measurements of Nitrogen Oxides

During the summer both indoor and outdoor levels of nitric oxide and of total nitrogen oxides were determined using a real-time monitor and a strip chart recorder. The raw data were analyzed, and results were put into data set ORNL.SNOX. Appendix B.5.1 lists those results. Table 5.11 lists and defines the variables in the data set. The means and ranges were:

	Indoor	Outdoor
NO	5.3 (0-50) ppb (N=35)	2.8 (0-37) ppb (N=37)
NO_x	12.9 (0-75) ppb (N=38)	11.3 (0-72) ppb (N=38)

For the winter phase of the study, a data logger was used to record the measurements. For this reason, data set ORNL.WNOX includes many more observations than ORNL.SNOX. Table 5.12 lists and defines the variables in the data set, and Appendix B.5.2 lists the data. The mean and range of indoor levels recorded in noncombustion source homes were 6.2 (0-82) ppb (N=120) for NO and 9.4 (0-86) ppb (N=120) for NO_x. As was the case for carbon monoxide, summary statistics will not be reported for combustion source homes.

Table 5.9. Levels of selected organic compounds ($\mu g/m^3$)

Chemical	BP[a] (°C)	Winter			Summer		
		N	Mean	Maximum		Mean	Maximum
Toluene	110	17	27.2	58	85	61.7	655
Ethyl benzene	136	14	4.4	12	84	10.5	161
Xylene(m,p)	139	17	16.6	54	84	44.2	697
Nonane(C_9)	151	14	10.6	98	82	6.4	36
Cumene	152	7	2.2	7	68	1.3	13
Benzaldehyde[b]	180	11	17.8	124	24	20.7	106
Mesitylene	165	3	1.4	2	80	6.7	39
Decane(C_{10})	174	14	12.4	39	81	8.9	81
Limonene($C_{10}H_{16}$)	178	13	10.3	24	77	20.8	167
Undecane(C_{11})	196	14	8.2	38	81	12.0	115
Naphthalene	218	12	9.4	75	80	17.5	675
Dodecane(C_{12})	216	15	4.2	10	84	13.7	675
2-Methylnaphthalene	241	8	2.5	7	47	2.6	17
Tridecane(C_{13})	236	14	2.3	5	85	16.5	113
Tetradecane(C_{14}	254	15	3.2	5	85	7.5	75
Pentadecane(C_{15})	271	14	2.8	5	84	1.9	12
Hexadecane(C_{16})	287	4	3.7	5	81	3.8	21

[a] Boiling point.

[b] Incomplete resolution - benzaldehyde and/or C_3-benzenes. Benzaldehyde may be any artifact of the sampling method.

5.6 Particulate Measurements

Particles in various size ranges were measured, and both total suspended particulate material and respirable suspended particulate material were recorded. The data are in ORNL.SPART and ORNL.WPART for the summer and winter phases, respectively. These data are listed in Appendices B.6.1 and B.6.2, respectively, and Table 5.13 lists and defines the variables used. The means and ranges for measurements made in all houses were:

	TSP	RSP
Summer indoor	31.7 (0-277) g/m^3 (N=410)	23.9 (0-233) g/m^3 (N=410)
Summer outdoor	36.9 (0-185) g/m^3 (N=340)	33.1 (0-184) g/m^3 (N=340)

The mean winter indoor TSP level in ten noncombustion source houses was 30.0 (3-111) g/m^3 (N=115), while the mean RSP level was 21.1 (3-55) g/m^3 (N=115). Summary statistics for combustion source houses are not reported due to the episodic nature of the data.

Table 5.10. Variables in ORNL.SCO and ORNL.WCO

Variable	Definition
DATE	See Table 5.2
DAY	See Table 5.2
HSENO	See Table 5.2
PLACE	See Table 5.2
TIME	See Table 5.2
PPM	Carbon monoxide level (ppm)

Table 5.11. Variables in ORNL.SNOX

Variable	Definition
DATE	See Table 5.2
DAY	See Table 5.2
HSENO	See Table 5.2
NO_I_AV	Average (by inspection) indoor level of NO (ppb)
NO_I_MN	Minimum indoor level of NO (ppb)
NO_I_MAX	Maximum indoor level of NO (ppb)
NO_O_AV	Average (by inspection) outdoor level of NO (ppb)
NO_O_MN	Minimum outdoor level of NO (ppb)
NO_O_MX	Maximum outdoor level of NO (ppb)
NOX_I_AV	Average (by inspection) indoor level of NO_x (ppb)
NOX_I_MN	Minimum indoor level of NO_x (ppb)
NOX_I_MX	Maximum indoor level of NO_x (ppb)
NOX_O_AV	Average (by inspection) outdoor level of NO_x (ppb)
NOX_O_MN	Minimum outdoor level of NO_x (ppb)
NOX_O_MX	Maximum outdoor level of NO_x (ppb)

Table 5.12. Variables in ORNL.WNOX

Variable	Definition
DATE	See Table 5.2
DAY	See Table 5.2
HSENO	See Table 5.2
PLACE	See Table 5.2
TIME	See Table 5.2
TYPE	Indication of which pollutant (NO or NO$_x$)
PPB	Pollutant level (ppb)
PPB_ERR	Standard error (ppb)

Table 5.13. Variables in ORNL.SPART and ORNL.WPART

Variable	Definition
DATE	See Table 5.2
DAY	See Table 5.2
HSENO	See Table 5.2
TIME	See Table 5.2
RSP_IN	Indoor level of respirable suspended particles ($\mu g/m^3$)
RSP_OUT[a]	Outdoor level of respirable suspended particles ($\mu g/m^3$)
TSP_IN	Indoor level of total suspended particles ($\mu g/m^3$)
TSP_OUT[a]	Outdoor level of total suspended particles ($\mu g/m^3$)

[a] Not in ORNL.WPART.

5.7 Measurements of Carbon Dioxide

During the winter phase of the study, carbon dioxide was measured in ten combustion and three noncombustion homes. These data are reported in ORNL.WCO2, and Appendix B.7.1 lists the data. Table 5.14 lists and defines the variables used. The mean and range in noncombustion homes were: 595 (392-794) ppm (N=6). The outdoor ambient concentration of carbon dioxide is about 400 ppm.[4] Summary statistics for combustion source houses are not listed.

5.8 Radon

During the study, radon was measured in some of the homes using Wrenn chambers. The data are in the data set ORNL.SRDN, and Table 5.15 lists the variables and their definitions. Appendix B.8.1 contains a listing of that data. The mean radon level for this group of houses was 4.4 (0-33) pCi/L (N=15).

Table 5.14. Variables in ORNL.WCO2

Variable	Definition
DATE	See Table 5.2
DAY	See Table 5.2
HSENO	See Table 5.2
TIME	See Table 5.2
CMT	Comment concerning any unusual event
PPM	Carbon dioxide level (ppm)

During the winter, radon was measured in all houses using track etch monitors. The data for room and outdoor averages are in the data set ORNL.WRDN. The variables and their definitions are listed in Table 5.16, and Appendix B.8.2 lists the data. The mean and range of the winter data were 2.8 (0.2-17.4) pCi/L (N=95).

5.9 Air Exchange Rate Measurements

Air exchange rate measurements were conducted during the intensive one-day monitoring visits to the houses. (All measurements were made with floor fans operating to ensure that there was good mixing of the tracer.) These data are in the data set named ORNL.XVNT and are presented in Appendix B.9.1. Table 5.17 lists and defines the variables in ORNL.XVNT. The mean and range of the air exchange rates measured before 12/01/82 are 0.74 (0.42-1.98) for fan-on measurements (N=31) and 0.39 (0.18-1.47) for fan-off measurements (N=35). The mean and range of the exchange rates measured after this date are 0.98 (0.31-1.57) for fan-on measurements (N=15) and 0.65 (0.11-1.21) for fan-off measurements (N=18).

5.10 Weather Monitoring

Weather conditions around a building are known to affect air exchange and indoor air quality. Three general measures of weather were recorded: (1) on-site weather data, (2) NOAA weather data, and (3) local precipitation data.

5.10.1 On-site measurements

Throughout the study, team members made on-site meteorological measurements. Those data associated with the monthly visits to obtain 24-h formaldehyde levels are recorded in the data set ORNL.XWOSA. Table 5.18 lists the variables used and their definitions. Appendix B.10.1 lists the data. During the intensive one-day visits, team members measured meteorological conditions as well as air exchange rates. These data are recorded in ORNL.XWOSB and in Appendix B.10.2; Table 5.19 lists the variables used and their definitions.

Table 5.15. Variables in ORNL.SRDN

Variable	Definition
DATE	See Table 5.2
DAY	See Table 5.2
HSENO	See Table 5.2
CODE	Location of measurement[a]
N_DAYS	Duration of operation (days)
N	Number of measurements
MN_PCL	Average radon level (10^{-12} Ci/L)
SEM	Standard error (10^{-12} Ci/L)
MAX	Maximum radon level (10^{-12} Ci/L)
MIN	Minimum radon level (10^{-12} Ci/L)

[a] GAR = garage; TAG = totally above ground;
PBG = partially below ground

Table 5.16. Variables on ORNL.WRDN

Variable	Definition
HSENO	See Table 5.2
RM	See Table 5.2
CMT	Comment describing location of measurement
PICOCI_L	Mean radon level (10^{-12} Ci/L)

Table 5.17. Variables in ORNL.XVNT

Variable	Definition
DATE	See Table 5.2
DAY	See Table 5.2
FAN	Indication of whether central fan is on
HSENO	See Table 5.2
EX_ACH	Air exchange rate (h^{-1})
ERR_ACH	Standard error (h^{-1})

Table 5.18. Variables in ORNL.XWOS

Variable	Definition
DATE	See Table 5.2
DAY	See Table 5.2
HSENO	See Table 5.2
TIME	See Table 5.2
CODE	Weather condition[a]
T_MN_I	Minimum indoor temperature (°C)
T_MX_I	Maximum indoor temperature (°C)
T_MN_O	Minimum outdoor temperature (°C)
T_MX_O	Maximum outdoor temperature (°C)
RH_MN	Minimum relative humidity
RH_MX	Maximum relative humidity

[a] 1 = Sunny; 02 = Rain; 03 = Snow; 04 = Cloudy

Table 5.19. Variables in ORNL.XWOS2

Variable	Definition
DATE	See Table 5.2
DAY	See Table 5.2
HSENO	See Table 5.2
T_BEG	Beginning time of measurements
T_END	Ending time of measurements
DIR	Wind direction
T_MX_I	Maximum indoor temperature (°C)
T_MN-I	Minimum indoor temperature (°C)
T_MX_0	Maximum outdoor temperature (°C)
T_MN_O	Minimum temperature (°C)
T_AV_O	Average outdoor temperature (°C)
RH_I	Indoor relative humidity
RH_MX_O	Maximum outdoor relative humidity
RH_MN_O	Minimum outdoor relative humidity
RH_AV_O	Average outdoor relative humidity
PRECIP	Precipitation (mm)
W_SPD_MX	Maximum wind speed (m/s)
W_SPD_MN	Minimum wind speed (m/s)
W_SPD_AV	Average wind speed (m/s)

5.10.2 NOAA measurements

The official U.S. weather data reported at the Illinois Avenue site in Oak Ridge were recorded in the data set ORNL.XWNOAA. Table 5.20 lists the variables used and their definitions while Appendix B.10.3 provides a listing of the data.

5.10.3 Precipitation

Local precipitation as reported by volunteers is recorded in the data set ORNL.XPRECIP. Table 5.21 lists the variables and their definitions. Appendix B.10.4 lists the data.

Table 5.20. Variables in ORNL.XWNOAA

Variable	Definition
DATE	See Table 5.2
DAY	See Table 5.2
T_MX	Maximum temperature (°C)
T_MN	Minimum temperature (°C)
T_AV	Average temperature (°C)
T_DEL	Deviation from normal temperature (°C)
PRECIP	Precipitation (mm)
SKY_CVR	Sky cover[a]
BAR_MX	Maximum barometric pressure (mm of Hg)
BAR_MN	Minimum barometric pressure (mm of Hg)
DEW_PT	Dew point (°C)

[a] Clear = 0-3; Partly Cloudy = 4-7; Cloudy = 8-10

Table 5.21. Variables in ORNL.XPRECIP

Variable	Definition
DATE	See Table 5.2
DAY	See Table 5.2
HSENO	See Table 5.2
PRECIP	Precipitation (mm)

5.11 Results Not Available Via Electronic Media

Certain of the results of the study have not been included on the magnetic data tape or in Appendix B. Generally, these data were excluded for one of two reasons. Some data as collected were not particularly amenable to computer analysis. Chromatograms obtained with the portable Photovac GC are an example. Other results consist of relatively small data sets and/or do not directly relate to the basic measurements being made in the houses. Examples of this include various laboratory and field calibrations that may be of interest to the reader.

5.11.1 Laboratory intercomparisons of formaldehyde monitoring methods

Laboratory intercomparisons of formaldehyde monitoring methods were conducted in detail on four separate occasions. Table 5.22 lists the dates and experimental conditions of the laboratory intercomparisons. The results of the first intercomparison are presented in Table 5.23. The remaining intercomparison results are presented in Tables 5.24-5.26.

5.11.2 Field intercomparisons of formaldehyde monitoring methods

Table 5.27 lists the formaldehyde results from the field measurements made with the LBL refrigerated sampler, which was used as a reference, and the results from the ORNL passive samplers. Field intercomparisons of several passive formaldehyde samplers were made in conjunction with a subset of the refrigerated impinger measurements, and these results are presented in Table 5.28.

Table 5.22. Conditions for laboratory intercomparisons of formaldehyde monitors

Intercomparison	Concentration	Duration	Internal fan
1 (07-23-82)	300 ppb	5 h	on
2 (11-03-82)	300 ppb	5 h	on
3 (11-03-82)	300 ppb	24 h	on
4 (12-07-82)	300 ppb	24 h	on

Table 5.23. Results of a 5-h formaldehyde exposure (July) at a nominal 300-ppb concentration

Method	N	Mean	Standard deviation	Minimum value	Maximum value	Std. error of mean
CEA	6	271	11	255	282	5
LBL	3	298	8	291	306	4
NIOSH	2	302	35	277	327	25
PASSIVE[a]	4	273	31	231	304	15
SIEVE	5	283	26	260	322	12

[a]Using a standard sampling rate of 0.13 $\frac{g/mL}{ppm \cdot h}$.

5.11.3 Fluctuations of formaldehyde levels

Detailed measurements of temporal fluctuations of formaldehyde concentrations were made during the scoping phase of the study.[12] Figure 5.1 shows the diurnal fluctuations in levels in house #2 and house #3. Figure 5.2 shows similar data for house #7. These data were obtained with either the pumped molecular sieve technology or a CEA 555 formaldehyde monitor at intervals as frequent as 15 min. No modifications were made to the houses other than to ensure that all windows were closed. The effect of opening the windows in a corner room of house #3 on the measured formaldehyde levels is shown in Fig. 5.3. Further effects on the level of formaldehyde due to changing air exchange rates are shown in Fig. 5.4 for house #7. Finally, the effect of a small charcoal-based air cleaner on the formaldehyde levels in house #7 is presented in Fig. 5.5.

5.11.4 Volatile organic measurements with a portable photoionization GC

Figures 5.6-5.7 illustrate the type of field results obtained with the Photovac portable GC. These are examples from over one hundred chromatograms taken during the study. Further measurements are being conducted under another contract (CPSC-IAG-82-1300) with emphasis on organic sources and levels in houses. Additional results from the collected chromatograms may be presented with the results of that study.

Table 5.24. Results of a 5-h formaldehyde exposure (November) at a nominal 300-ppb concentration

Method	N	Measured concentration (ppb)
Refrigerated sampler	3	350 ± 10
Pumped molecular sieve	6	300 ± 15
NIOSH impinger	3	420 ± 35
CEA 555	5	305 ± 10
Passive membrane (ORNL)	3	300 ± 10
Mean response (excluding NIOSH)		315 ± 25

Table 5.25. Results of a 24-h formaldehyde exposure test (November) at 300 ppb

Method	Measured concentration (ppb)	Comment
CEA 555	315 ± 10	(avg. of 5 readings during working hours)
Pumped molecular sieve	290 ± 10	(avg. of 8 15-min samples during working hours)
Refrigerated sampler	335 ± 10	(24-h samples)
Passive membrane (ORNL)	340 ± 50	(24-h samples)
AQR passive	180 ± 40	(24-h samples)
3M passive	20	(24-h samples)
Mean response (excluding AQR and 3M)	320 ± 25	

5.11.5 Air leakage via blower door measurements

Air leakage measurements under pressurized conditions were made in house #21. A blower door was installed and obvious leaks such as fireplace and ductwork openings were temporarily sealed prior to testing. An exchange rate of 7.1 h^{-1} was measured at a pressure of 50 Pa. The extrapolated specific leakage area at 4 Pa was 3.1 cm^2/m^2 for the house. This ratio indicates the equivalent area of a single hole for air leakage per unit area of floor space. Blower door measurements were not conducted in any other of the study houses.

Table 5.26. Results of a 24-h formaldehyde exposure test (December) at 300 ppb

Method	Measured concentration (ppb)	Comment
Pumped molecular sieve	275 ± 25	(avg. of 9 15-min samples during working hours)
Refrigerated sampler	400 ± 10	(24-h samples)
Passive membrane (ORNL)	190 ± 50	(24-h samples)
AQR passive	290 ± 40	(24-h samples)
3M passive	20	(24-h samples)
Mean response (excluding 3M passive)	290 ± 85	

Table 5.27. Formaldehyde concentrations (ppb) measured using the LBL refrigerated sampler and the ORNL passive samplers

House Number	Date	LBL sampler	ORNL passive
10	05-05-82	25 ± 5	<25
7	05-07-82	265 ± 5	245 ± 65
27	05-07-82	45 ± 5	<25
34	05-27-82	330 ± 5	333 ± 60
39	06-15-82	25 ± 5	<25
81	06-17-82	250 ± 25	365 ± 80
5	08-13-82	180 ± 20	220 ± 20
3	08-25-82	310 ± 30	<25[a]
4	09-08-82	60 ± 10	30
2	09-09-82	70 ± 10	30
7	12-03-82	380 ± 10	265 ± 30

[a]Subsequent sampling yielded 360 ± 20 ppb. See text for discussion.

Table 5.28. Results of field intercomparisons of various formaldehyde sampling devices

Monitor	House Number				
	2	3	4	5	7
LBL reference	0.07 ± 0.01	0.31 ± 0.03	0.06 ± 0.01	0.18 ± 0.02	0.38 ± 0.01
AQR passive	0.12 ± 0.03	0.30 ± 0.01	0.09 ± 0.05	0.19 ± 0.03	0.42 ± 0.06
3M passive	0.05 ± 0.02	0.16 ± 0.01	0.03 ± 0.01	0.13 ± 0.01	0.17 ± 0.02
ORNL passive	<0.03	<0.03 (0.36 ± 0.02)[a]	0.03 ± 0.01	0.22 ± 0.02	0.27 ± 0.03

[a]Repeat measurement during following week.

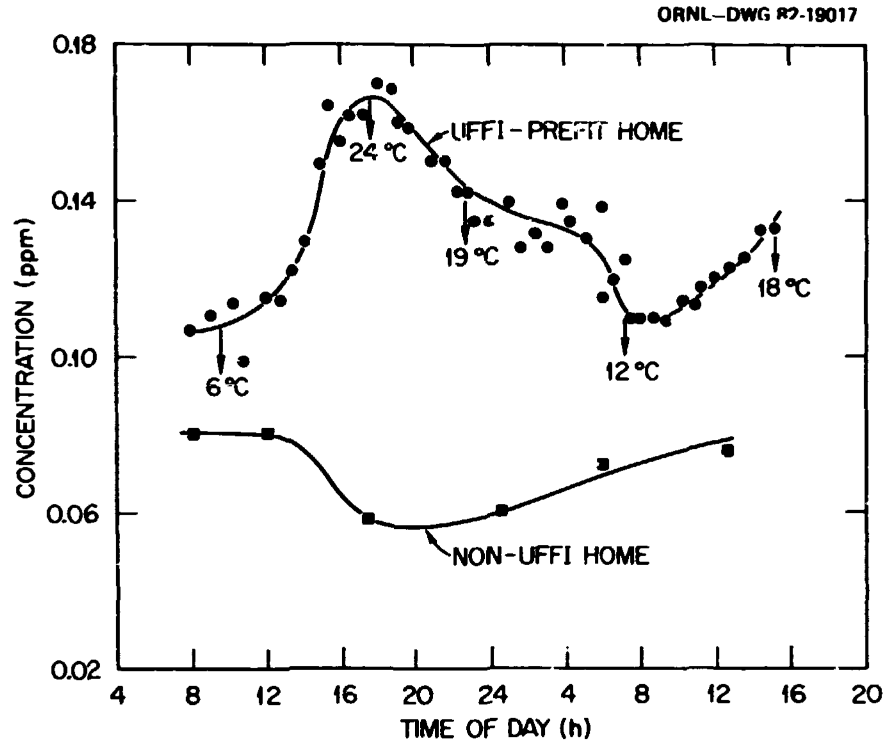

Fig. 5.1 Diurnal fluctuations in formaldehyde levels inside a 3-year-old UFFI-prefit house and a 10-year-old non-UFFI house; the outdoor temperatures are indicated.

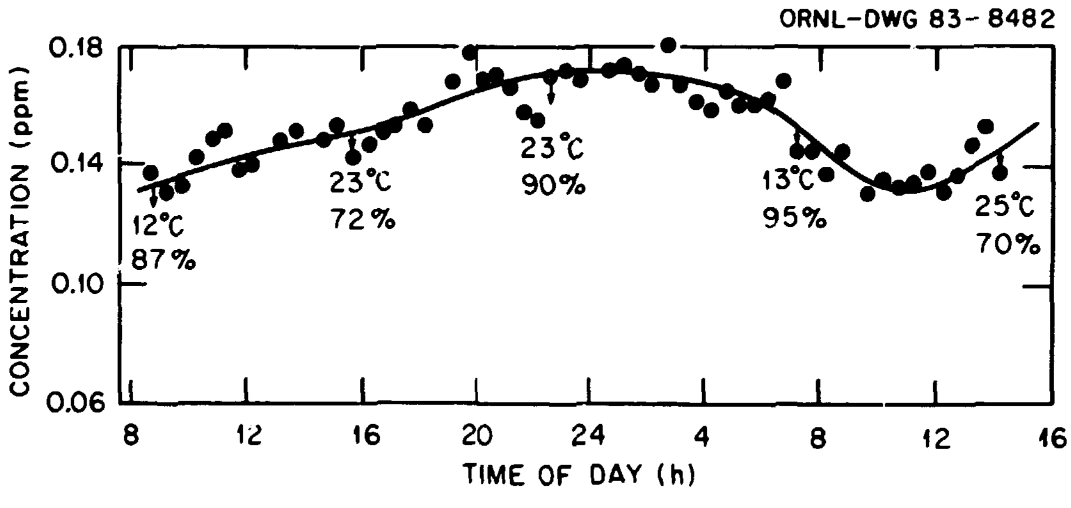

Fig. 5.2 Diurnal fluctuations in formaldehyde levels inside a 2-year-old energy-efficient, non-UFFI house; the outdoor temperatures and humidities are indicated.

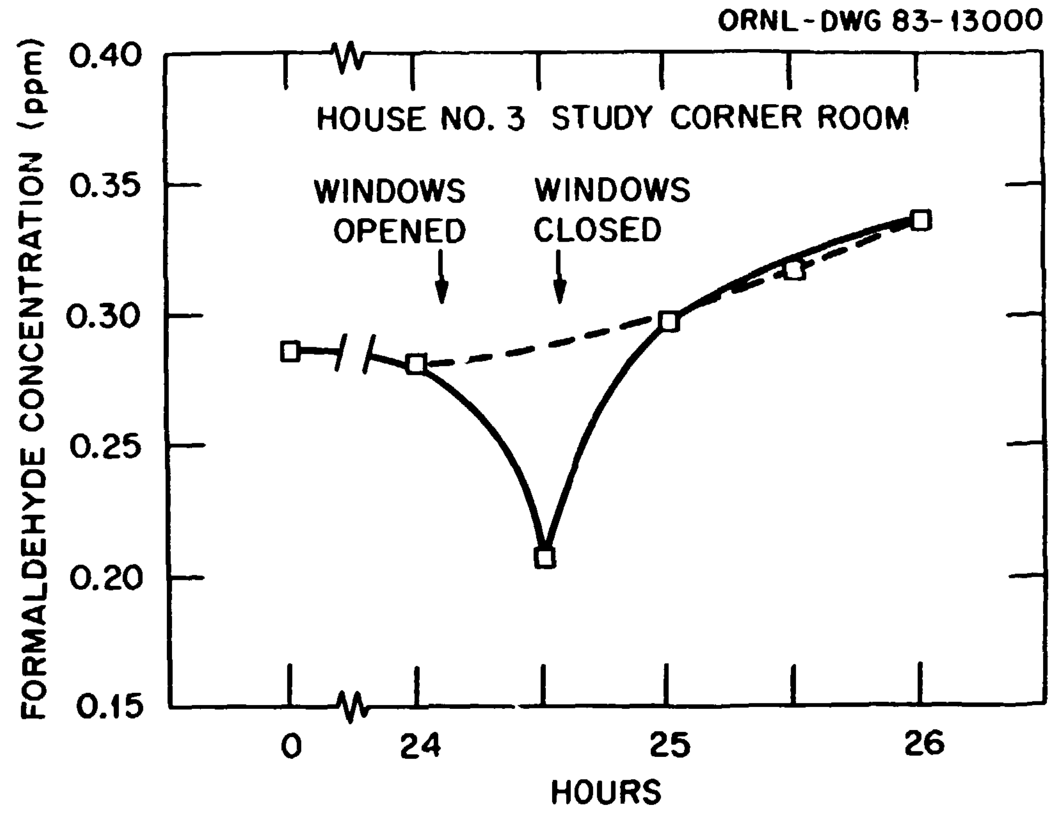

Fig. 5.3 Effect of opening windows in a corner room of house #3 on formaldehyde levels. Dashed line is presumed behavior with windows closed.

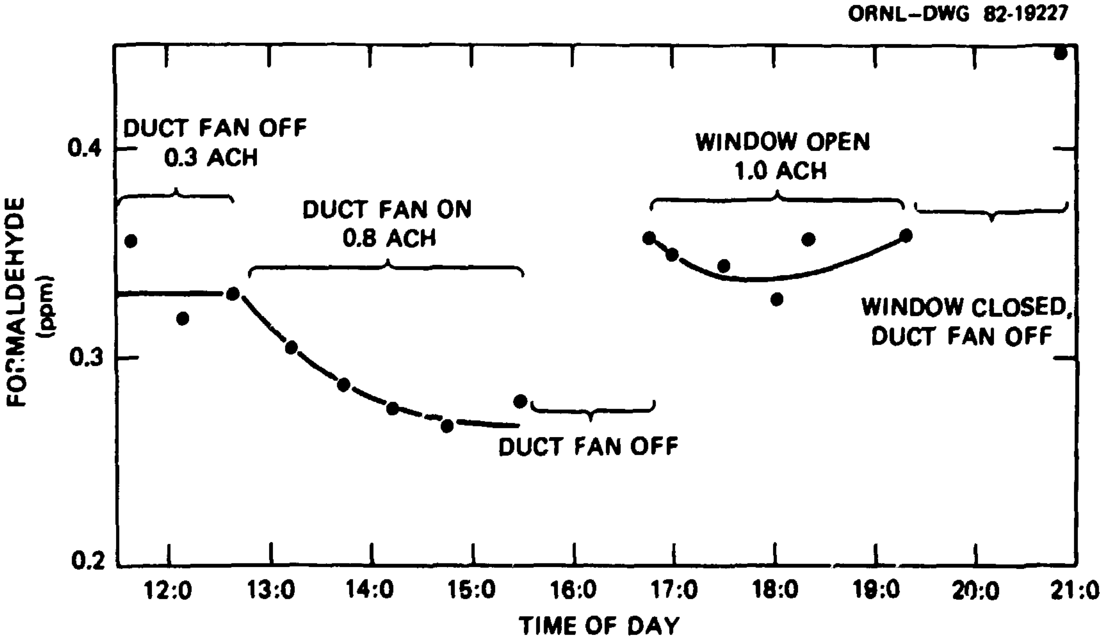

Fig. 5.4 Influence of air infiltration on the formaldehyde concentration inside a 2-year-old energy-efficient house during a warm day in May.

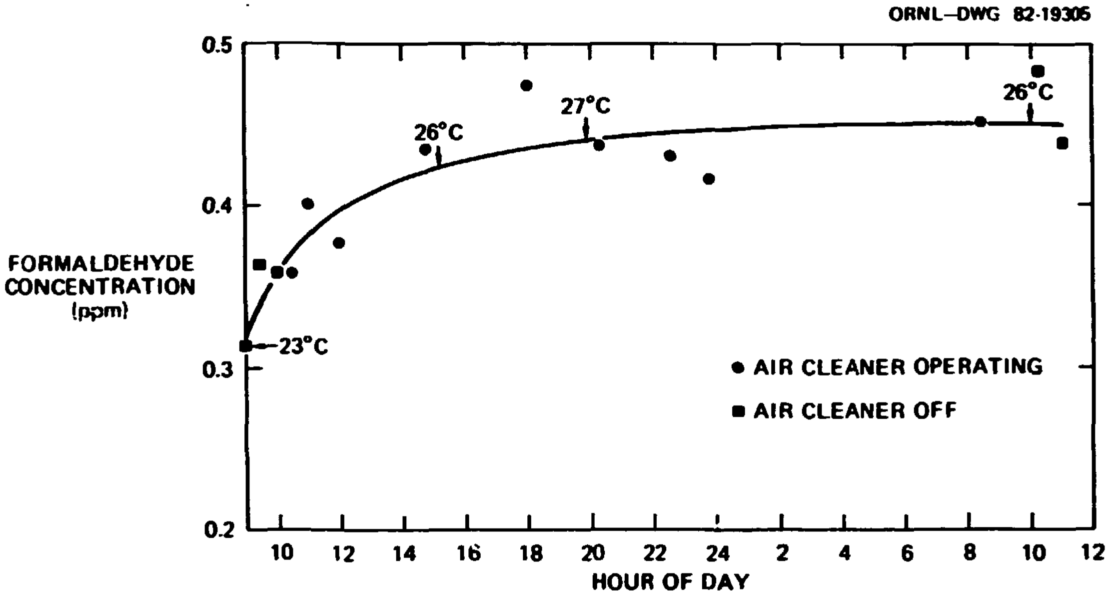

Fig. 5.5 Formaldehyde concentration measured during operation of a charcoal-based air cleaner inside a closed-off room; the indoor temperatures are indicated.

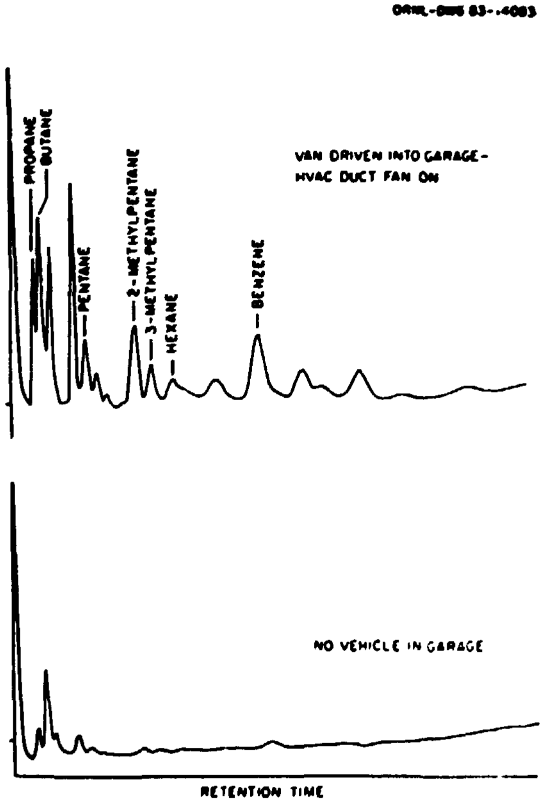

Fig. 5.6 Photovac chromatograms for house #2.

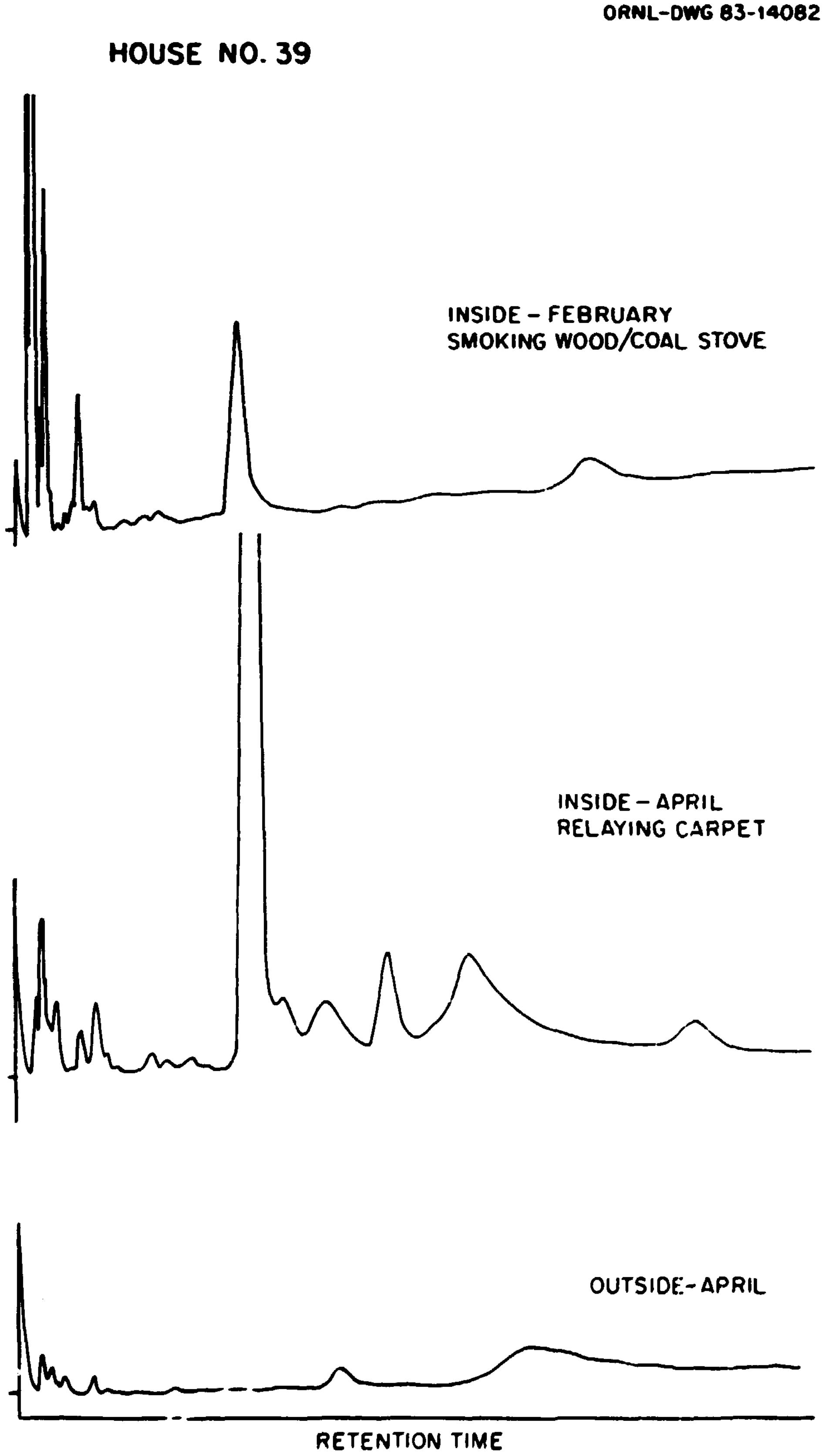

Fig. 5.7 Photovac chromatograms for house #39.

A Wilks MIRAN-80 single-beam infrared spectrometer was used to monitor the decrease in Freon concentration with time. Absorbance at 9.258 μm was measured, which is directly proportional to the concentration of Freon present in the instrument sampling cell. An integral air pump drew ambient air through the sample cell continuously at a flow rate of 9.5 L/min. A mathematical model was used to obtain the best fit of the absorbance vs time to give the rate of air exchange. In homes having a central HVAC system, an air exchange rate was determined for a period when the central fan was on and for a period when the fan was off.

4.7 Meteorological Data

On every visit, indoor and outdoor temperature and relative humidity were recorded on a hygrothermograph for the duration of the measurement period. Additionally, during the summer phase, maximum-minimum thermometers recorded the temperature range for each visit. Wind speed and direction were monitored by a T. M. Young model 12102D light-chopper-type cup anemometer and a Young model 12302 wind vane. Transducers and chart recorders were powered by a 12-V automobile battery. Unfortunately, instrumentation problems encountered due to freezing temperatures precluded measurement of wind parameters during most of the winter phase of the study. Additional meteorological data were obtained from the NOAA weather station on Illinois Avenue in Oak Ridge.

4.8 Data Logging

During the winter phase of the study a Commodore PET series 2001 personal computer was used as a data logger to monitor outputs of the particulate analyzer, the nitrogen oxides analyzer, and the carbon monoxide monitor. Appropriate software was written to allow selection of sampling frequency, to control switching of monitoring between instruments, and to record data on a cassette tape in a form suitable for direct input into the data base computer. Continuous sets of data were generated by the data logger for the above-mentioned devices in 5-min intervals over the sampling period. During the summer phase of the study, prior to implementation of the data logger, data were recorded from the particulate analyzer on paper tape printed at 15-min intervals. Output of the nitrogen oxides analyzer during the summer was directed to a 10-V strip chart recorder with periodic switching of channels to record NO and NO_x levels. Summer measurements of carbon monoxide were recorded intermittently by hand every 30 minutes over the 6-7-h sampling period.

6. DATA ANALYSIS AND DISCUSSION

The data collected during the course of this study represent a large amount of information. Due to resource constraints, a limited analysis of these data was completed and presented in this report as preliminary findings. Additional analysis will include such items as the correlation of various housing characteristics and meteorological conditions with the various pollutant data sets. We hope to be able to perform more detailed analyses subsequently.

6.1 Formaldehyde Measurements

The nearly 7000 measurements made during this study represent the most intensive investigations of formaldehyde levels in residential environments. Preliminary analysis of these measurements indicate several definite trends including generally decreasing concentrations with the age of the houses and higher levels during summer. It should be noted that detailed analyses with temperature, humidity, air exchange rates, and other house variables have not been completed. These are examples of the additional analyses that will be forthcoming. Following is a brief discussion of the work currently completed.

6.1.1 Distribution of formaldehyde levels

Figure 6.1 shows the distribution of the 1150 room formaldehyde concentration measurements. When these values are averaged over all rooms and all visits to yield a mean value per house over the course of this study, 32 of the house averages are less than 0.1 ppm. One house had an average greater than 0.2 ppm, and seven houses had averages greater than 0.1 ppm and less than 0.2 ppm. On at least one occasion 60% of the houses had a measured concentration of greater than 0.1 ppm in at least one room. The monthly means of the outdoor measurements were less than 0.025 ppm for all months.

6.1.2 House age and formaldehyde levels

Table 6.1 and Fig. 6.2 show the mean formaldehyde levels as a function of the age of the house. There is a significant ($P < 0.01$) correlation between the house age and the mean level of formaldehyde. The newer homes seem to reflect the higher emission rates of newer formaldehyde-emitting materials. This observation is consistent with the use of formaldehyde-resin-containing materials in the construction and furnishing of homes.[21]

6.1.3 Seasonal variation of formaldehyde levels

A seasonal trend toward higher concentrations in the summer is indicated by the analysis shown in Fig. 6.3, which plots the monthly averages of all readings. There is an approximately threefold difference between the levels in August and the levels in October. This observation is consistent with the behavior of many formaldehyde-containing products that have higher emission rates with increased temperature and humidity.[22,23] The lower levels in the fall of the year are also consistent with lower temperatures and, perhaps, with opening of windows for natural ventilation during pleasant weather. October was also the driest month of the study. Table 6.1 also indicates that the highest mean concentrations were generally observed in the summer and the lowest mean concentrations in the fall (however, the study does not comprise winter measurements).

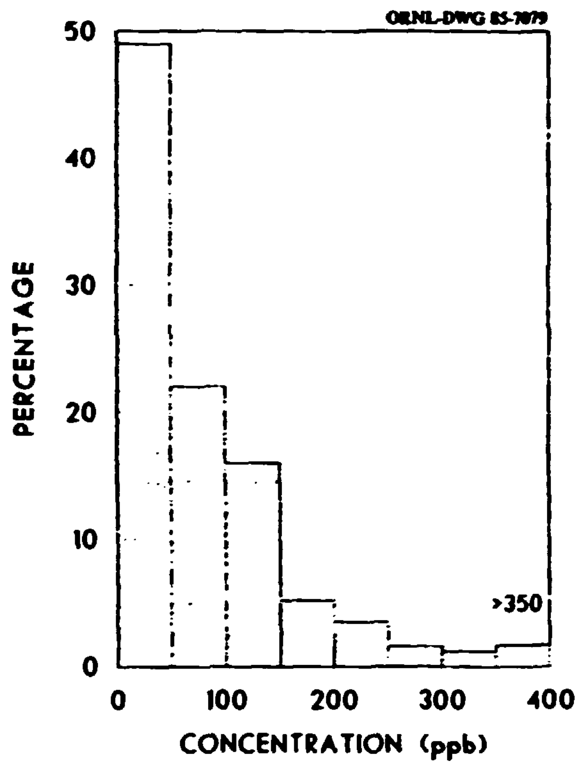

Fig. 6.1 Distribution of formaldehyde concentrations averaged over all rooms and all visits.

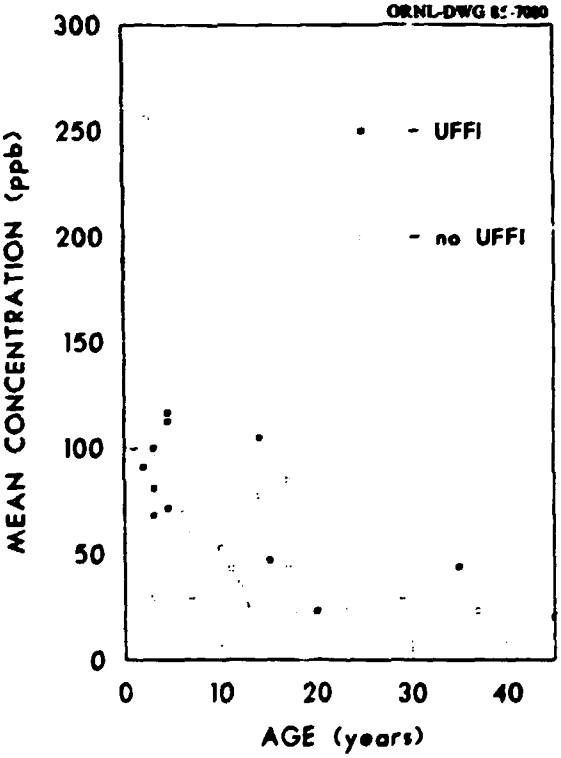

Fig. 6.2 Mean monthly formaldehyde levels for all homes in the study.

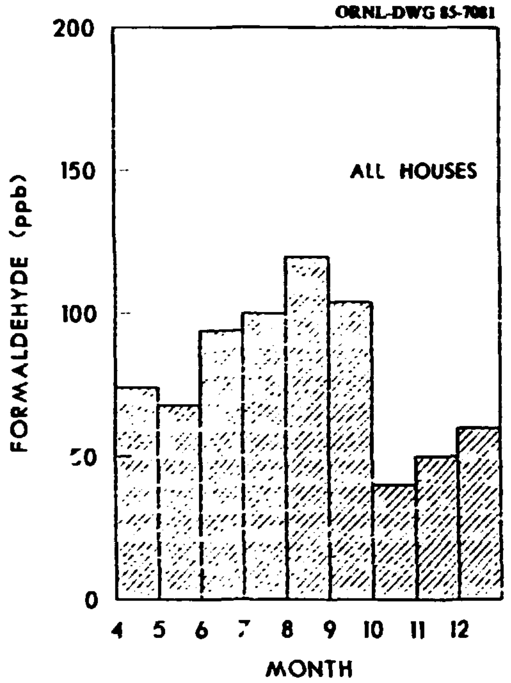

Fig. 6.3 Monthly formaldehyde levels in a new house (#81).

Table 6.1. Mean formaldehyde concentrations (ppb) as a function of age and season (Outdoor means are less than 25 ppb)

Age of house	Season	$\bar{x}^a$	s^a	m^a	n^a
all	all	62	77	5903	40
0-5 years	all	84	91	3210	18
5-15 years	all	42	42	1211	11
older	all	32	42	1482	11
0-5 years	spring	87	93	1210	
	summer	111	102	1069	
	fall	47	55	931	
5-15 years	spring	43	40	626	
	summer	49	48	326	
	fall	34	35	259	
older	spring	36	51	757	
	summer	29	37	341	
	fall	26	23	384	
all	spring	62	76	2593	
	summer	83	91	1736	
	fall	40	47	1574	

[a] x = mean concentration; s = standard deviation; m = number of measurements; n = number of homes

6.1.4 Houses with high formaldehyde levels

Figure 6.4 shows the monthly formaldehyde levels in a new house that was occupied for the first time at the beginning of the study. Initial concentrations in this home were greater than 0.3 ppm. The variation in the subsequent monthly values is presumably due to factors such as material aging, environmental parameters (temperature, humidity), and occupant behavior. Further analysis will investigate these factors.

Another house that had elevated formaldehyde levels was an energy efficient home. A 6-mil thick polyethylene vapor barrier covered the entire surface of the floors, walls, and ceilings and was lapped 6 in. at all joints. The polyethylene was placed between the subflooring and the main flooring (which was particleboard) and between the fiberglass insulation and the gypsum board on the walls and ceiling. This barrier appeared to effectively trap the formaldehyde emissions into the living space. This house was relatively small (1000 ft^2 living space) and may have had a high emitting surface-to-volume ratio. Emissions from pressed-wood products in the construction materials and furnishings of this home are most likely responsible for the elevated levels. A plot of the levels observed during the study in this home is shown in Fig. 6.5. The levels measured were consistently greater than 0.1 ppm and frequently greater than 0.3 ppm. The highest levels observed during the study were in this house.

6.1.5 Comparison of homes insulated with UFFI to homes without UFFI

Figure 6.2 indicates the mean formaldehyde levels versus the age of the house with the presence or absence of UFFI indicated. For the four houses with retrofit UFFI (UFFI houses older than 10 years in Fig. 6.2), the age of the UFFI was 2-4 years, the same as in prefit newer homes (age 2-5 years). The formaldehyde levels are higher in newer UFFI homes than older UFFI homes. House age (as a surrogate for other formaldehyde emission) may be an important factor to consider when comparing the formaldehyde levels in UFFI and non-UFFI homes. Table 6.2 shows the results obtained by comparing UFFI vs non-UFFI in various age groups. Also shown is the number of houses in each group. The mean level for non-UFFI houses is shown for houses between two and five years of age and for all non-UFFI houses. Six of the 11 non-UFFI houses were less than two years old.

Recall from Fig. 6.2 that the formaldehyde levels in three non-UFFI houses less than three years old are significantly higher than the levels in the other houses. Two of the three houses were discussed in Section 6.1.4. These houses strongly influence the means for new non-UFFI houses. Five of the 11 non-UFFI houses less than five years old had mean formaldehyde levels less than 0.05 ppm. The degree to which this subset of houses represents the larger population of new houses is unknown.

6.1.6 Room-to-room variations in formaldehyde levels

Two major groups of factors may influence formaldehyde levels in a particular room of a house. One group varies from day to day and principally includes meteorological factors such as humidity, temperature, and wind speed. Another group varies from room to room and includes factors such as local loading of emitting source material. Table 6.3 summarizes a two-way analysis of variance which tested a null hypothesis that concentrations do not vary room to room or day to day. Additional analysis revealed no significant room by day interaction. The results show substantial variation among days. This is indeed consistent with what is known about environmental effects on formaldehyde

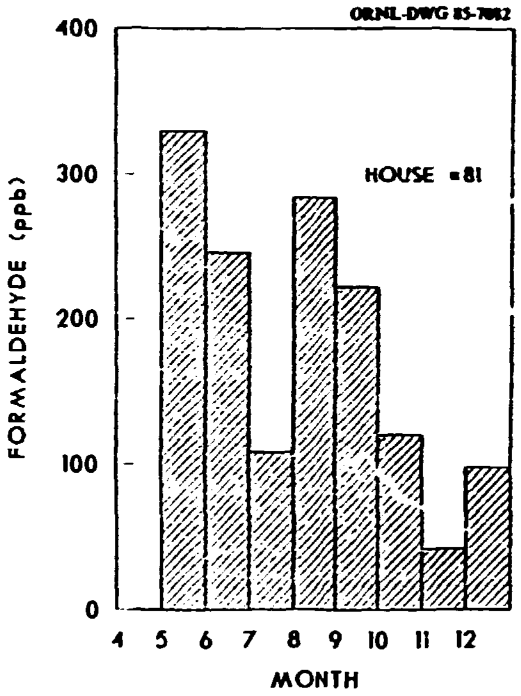

Fig. 6.4 Monthly formaldehyde levels in an energy-efficient house (#7).

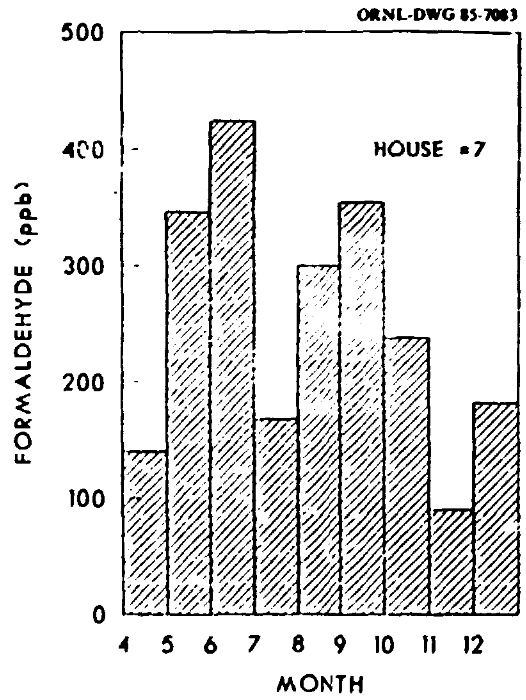

Fig. 6.5 Mean formaldehyde levels as a function of age (UFFI and non-UFFI indicated).

emissions from materials containing urea-formaldehyde resins. Room-to-room variation is much less evident and is only statistically evident when all the data are analyzed. This indicates that the room to room variability is less than other sources of variability in most houses.

6.1.7 One time versus multiple measurements

One of the important questions regarding formaldehyde monitoring is the validity of taking a single point-in-time (usually one hour) measurement as representative of the long-term average exposure level in a house. A comparison can be made of the formaldehyde levels obtained by the short-term molecular sieve technique and the 24-h passive monitor. Figure 6.6 shows the relationship between the short-term formaldehyde levels as measured by Team B during the summer phase using pumped molecular sieve samplers and the mean levels obtained by Team A with the 24-h passive sampler on the day closest to the day of the sieve measurements. A least-squares analysis yields a slope through the origin of 0.96 ± 0.07 and a correlation coefficient (r) of 0.94. Most comparisons were between measurements taken during visits less than one week apart, and all visits were within one month of each other. Although the relationship between the 15-min measurements and the 24-h measurements is relatively good when taken over a large number of measurements, the relationship between individual measurements is less predictable. This can be seen by noting that several values in Fig. 6.6 deviate from the solid line by 50 ppb or more. This observation is not surprising due to the significant time-dependent variations that can occur and the fact that a single measurement has a precision of about 25%.

6.1.8 Temporal fluctuations in formaldehyde levels

As was shown in Fig. 5.1 and Fig. 5.2, diurnal measurements made in a UFFI house revealed fluctuations in formaldehyde levels as much as 50%. Note that the variations in formaldehyde concentrations in the houses with higher levels seem to correlate with outdoor temperature changes. A single measurement made in this house at 8 o'clock in the morning would be substantially different than a measurement at 4 o'clock in the afternoon.

6.1.9 Formaldehyde levels in combustion source houses versus control houses

Table 6.4 compares molecular sieve measurements made during the summer phase with similar measurements made during the winter phase of the study. The formaldehyde levels are not significantly different in houses with combustion sources than in homes without such sources. As noted earlier, the levels in the homes measured were lower in winter than they were in summer.

6.1.10 Quality control measurements

Analysis of the results of the field quality control measurements made in conjunction with the routine measurements in a subset of houses confirms that, within analytical and statistical limitations, the various techniques are indicating similar levels of formaldehyde. Short-term and long-term quality control measurements will be discussed separately.

6.1.10.1 NIOSH impinger/chromotropic acid results - The modified NIOSH procedure was used as a reference method with which to compare the results of the

Table 6.2. Formaldehyde levels (ppb) observed in houses with and without UFFI

Age of house	Type of house	No. of houses	Mean concentration (ppb)	Range (ppb)
2-5 years	UFFI-prefit	7	90	68 - 117
15-35 years (UFFI age: 2-4 years)	UFFI-retrofit	4	55	23 - 105
2-5 years	non-UFFI	5	115	30 - 255
all	non-UFFI	29	60	20 - 255

Table 6.3. Analysis of variance of between-room and between-day differences in formaldehyde levels[a]

Type of wall insulation	Age of house	Number of houses	Number of observations	Day variation		Room variation	
				F	P	F	P
UFFI[b] (prefit)	<5 yrs	7	347	7.46	<0.01	1.35	0.25
non-UFFI	<5 yrs	11	293	10.65	<0.01	2.17	0.06
UFFI (retrofit)	>5 yrs	4	163	28.68	<0.01	1.11	0.35
non-UFFI	>5 yrs	18	344	3.59	<0.01	0.48	0.79
all	all	40	1147	5.76	<0.01	4.13	<0.01

[a] P = probability of difference due to chance.

[b] UFFI = urea-formaldehyde foam insulation.

Table 6.4. Comparison of summer and winter formaldehyde levels as measured by the molecular sieve method

House subset	No. of homes	Summer visit	Winter visit
Combustion source	10	52 (0 - 110) ppb	31 (0 - 120) ppb
Noncombustion control	10	60 (18 - 147) ppb	26 (9 - 119) ppb
Other	21	91 (18 - 250) ppb	

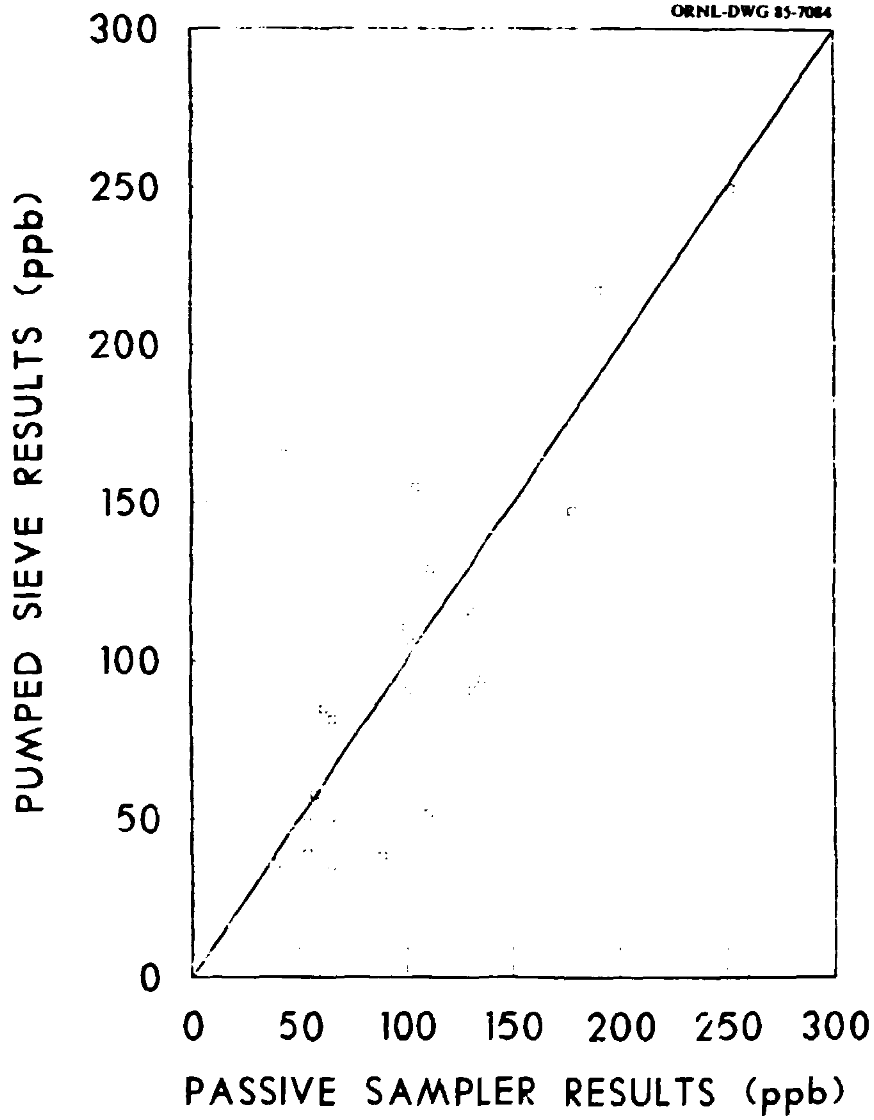

Fig. 6.6 Comparison of mean formaldehyde concentrations measured using the 24-h passive samplers and the short-term pumped sieve technique (line for reference only).

pumped molecular sieve technique. The latter technique was preferred for routine use for the reasons outlined in the methods section. Figure 6.7 shows the correlation between results greater than 0.06 ppm obtained by the modified NIOSH procedure and by the pumped molecular sieve technique based on measurements during the summer phase of the study. Four 15-min molecular sieve samples were typically taken during the 1-h impinger collection period. The solid line in the figure shows, for reference, a 1:1 correlation between the formaldehyde concentrations measured by these two techniques. A least-squares analysis yielded a slope through the origin of 1.16 ± 0.15 and a correlation coefficient (r) of 0.94.

The primary disadvantage of the NIOSH method in monitoring formaldehyde levels in residential environments is the procedure's lack of sensitivity. The detection limit of about 0.06 ppm,[13] for a sampling period of one hour, is higher than desirable for this type of study. Additional disadvantages are the inconvenience posed by the liquid collection medium and the necessity of dealing with concentrated acid during the analysis. The pumped molecular sieve method offers distinct advantages in sensitivity (resulting in a short 15-min sampling time and a lower detection limit of 0.025 ppm) and convenience. Excellent stability of the collected formaldehyde on the molecular sieve allows sample storage prior to analysis.

6.1.10.2 LBL refrigerated sampler results - The refrigerated sampler allowed comparisons to be made between the 24-h passive membrane monitors and an active sampling technique over the same period. By refrigerating the collection medium and having larger volumes of water, the LBL sampler provided such a quality control technique and was used as the reference technique with which to compare the passive samplers. Table 6.5 presents the comparison of the formaldehyde concentrations obtained by the two techniques during the course of the study. If only values greater than 75 ppb are analyzed and a zero slope is assumed, the relative response of the passive samplers to the refrigerated sampler was 0.97 ± 0.13 with a correlation coefficient (r) of 0.93. These measurements, therefore, indicate that the results obtained by the passive membrane monitors are a good measure of the formaldehyde concentrations in homes at concentrations above the quantitation limit.

6.1.11 Intercomparison measurements

The intercomparison measurements made in the field and laboratory both confirmed the general validity of all the tested formaldehyde monitoring techniques and pointed out occasional specific problems associated with various methods.

An analysis of the five-method intercomparison conducted under controlled laboratory conditions during July is presented in Table 6.6. The mean value of each of the five methods is within 10% of the laboratory-generated formaldehyde concentration of 0.3 ppm. The relative standard deviation of the NIOSH, the ORNL passive sampler, and the molecular sieve results is about 10%, while the relative standard deviation for the CEA and the LBL refrigerated unit results is about 4%. This test represents a carefully prepared and monitored intercomparison of these five methods. In order to obtain this quality of intercomparison, attention must be paid to the formaldehyde generation system, including minimization of all possible loss mechanisms, and to the mixing within the exposure chamber to avoid concentration gradients.

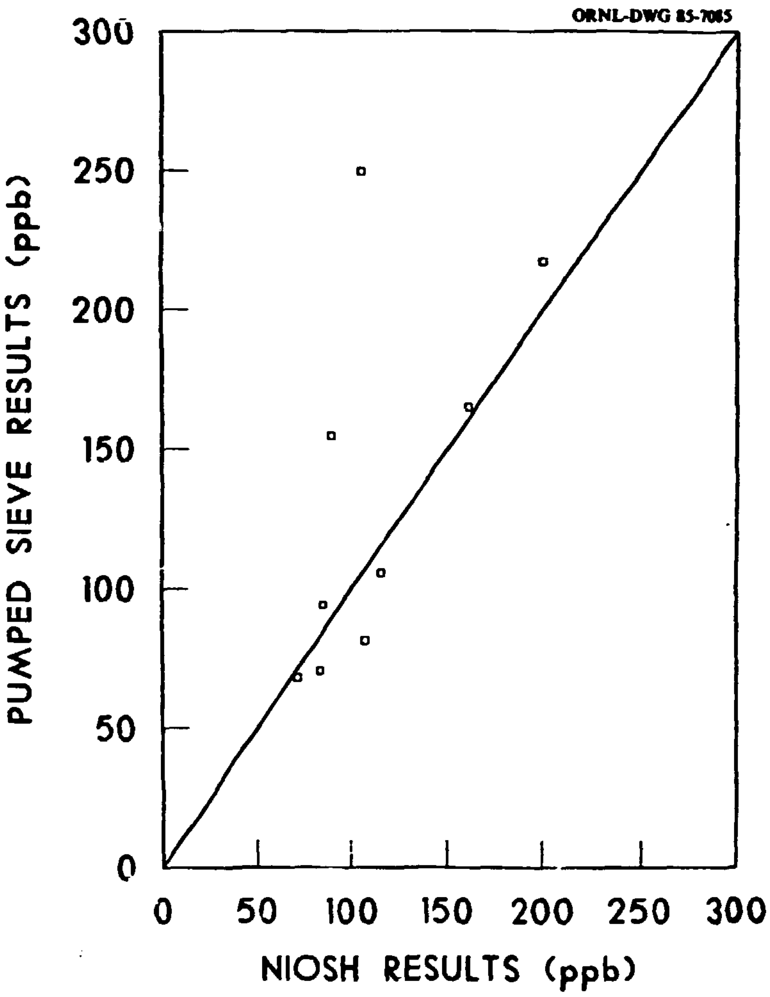

Fig. 6.7 Comparison of formaldehyde levels obtained using the modified NIOSH procedure and the pumped sieve technique (line for reference only).

Table 6.5. Comparison of formaldehyde levels (ppb) obtained with the LBL refrigerated sampler and with the ORNL passive samplers

House no.	Date	LBL results	Passive sampler results
10	05/05/82	25 ± 5	<25
7	05/07/82	265 ± 5	246 ± 65
27	05/07/82	45 ± 5	<25
34	05/27/82	330 ± 5	333 ± 63
39	06/15/82	25 ± 5	<25
81	06/17/82	250 ± 25	365 ± 78[a]
5	08/13/82	180 ± 20	220 ± 20
3	08/25/82	310 ± 30	<25
4	09/08/82	60 ± 10	30 ± 10
7	12/03/82	380 ± 10	265 ± 30

[a] LBL placed in dining room while passive samplers were in kitchen.

Table 6.6. Results of a July intercomparison of various formaldehyde sampling devices exposed to 300 ppb

Method	N	Measured Concentration (ppb)
Refrigerated sampler (LBL)	3	298 ± 8
Pumped molecular sieve (SIEVE)	5	283 ± 26
NIOSH impinger (NIOSH)	2	302 ± 35
CEA 555 monitor (CEA)	6	271 ± 11
Passive membrane (ORNL)	4	273 ± 31
Mean response		285 ± 15

The results of the November 5-h laboratory intercomparison (Table 5.24) were once again in good agreement with the expected value with the exception of higher than expected NIOSH results (140% of generated value). No explanation was found for this result.

The 24-h intercomparison, also conducted during November, included commercial passive monitors (3M and Air Quality Research) in addition to the five normally tested methods (see Table 5.25). Unfortunately, the commercial AQR monitors used in this test were four months old and beyond the vendor's suggested shelf life of three months. This presumably contributed to the erratic response of these devices. The laboratory intercomparison was conducted in an atmosphere of less than 30% relative humidity. It was later learned that the 3M devices require a higher humidity atmosphere to function properly. The results of the four other techniques tested were within 15% of the generated concentration.

A final 24-h laboratory intercomparison (Table 5.26) was conducted during December. Once again the 3M monitor gave a null response; however, the result for the AQR monitor (obtained at the same time as those tested in November) was in excellent agreement with the expected value. The LBL refrigerated sampler response and the ORNL passive monitors deviated about 0.1 ppm in opposite directions from the generated value; this deviation remains unexplained.

Results of the field intercomparison (Table 5.28) of passive monitors with the reference refrigerated impinger system indicated some variability between the different techniques under field conditions. The intercomparisons conducted in five houses generally indicated that the AQR monitor results were higher than the LBL reference method while the 3M results were low consistent with a recognized potential for lower response due to potential face velocity and humidity effects.[24] The AQR monitors have been reported to have a slight positive bias. The ORNL passive monitors were generally lower than the LBL reference method. In house #3 (August 22) the ORNL passive membrane monitor gave a null reading. It was later learned that old samplers were used during this week due to a shortage of membrane for fabricating new monitors. A repeat measurement later in the month with new ORNL passive monitors gave a value of 0.36 ppm. In a larger comparison of ORNL passive monitors with the LBL reference method there was reasonable correlation (Section 6.1.10.2). Due to the limited number of observations for intercomparison of the three passive monitors, these results are not conclusive and an extensive field evaluation is needed.

The problems experienced with the passive formaldehyde monitors can be summarized as follows. The unexposed AQR devices used as controls gave high blank readings (frequently >0.05 ppm equivalent), even when within their recommended shelf life. These blank readings were a high percentage (42-88%) of the readings obtained from the detectors exposed in the field. These high blank readings contributed to the difficulty in obtaining reliable formaldehyde measurements. It should be stressed, however, that if the detectors had been exposed for seven days, as recommended by the vendor, the total amount of formaldehyde collected would be considerably larger relative to the blank values. The 3M devices that were used as blanks gave low readings, usually equivalent to 0.01 ppm or less. The levels of formaldehyde measured in the field, however, were generally low compared to the results of the refrigerated impinger unit. These results are consistent with reduced sampling rates at low face velocities. A modified device has been constructed by 3M specifically for area monitoring where low face velocities are expected. These new units were not tested during this study. The ORNL passive monitors failed on one occasion by giving a zero reading in house #3, which had a concentration of 0.3 ppm. The cause was associated with aging of the membranes, which had developed a dirty appearance as the result of six prior exposures in the field over the course of two months. A repeat measurement in this house a few weeks later using new passive ORNL monitors gave a mean concentration of 0.36 ppm.

6.2 Combustion Gases

Combustion gases during the summer phase of the study were relatively low compared with guidelines for ambient air quality set by EPA and were about the same as outdoor levels. Levels that were much above the background levels were associated either with specific episodes, such as operating an automobile in a basement garage, or with elevated outdoor levels associated with an adjacent busy street. This is consistent with combustion sources not generally being operated during this time of the year. During visits in the winter heating season, short-term elevated levels were associated with operation of specific combustion sources. Selected examples of the temporal variation in pollutant levels will be presented for each pollutant, as well as a discussion of results for the summer vs winter (both combustion source and control houses) measurements. For obvious reasons, care must be exercised in comparing average summer levels with the average winter levels in homes with unvented combustion sources. Much of the winter data should be considered as resulting from episodic events and are not necessarily to be interpreted as mean exposure levels.

6.2.1 Nitrogen oxides

The levels of nitrogen oxides in the homes were generally below 25 ppb during the summer. Outdoor levels were also below this value with the exception of higher levels near a busy thoroughfare in west Knoxville where the outdoor levels ranged between 0.01 and 0.065 ppm. The mean indoor level in the two house located near the thoroughfare were 0.03 ppm of NO and 0.06 ppm of NO_x. Elevated levels were associated with the operation of an automobile in a basement garage. The NO_x concentration in the bedroom above the garage increased from 0.005 ppm to 0.07 ppm after the engine was operated for a period of 3 min. The central circulation fan of the HVAC was operating during this period and seemed to rapidly spread the exhaust pollutant through the house.

During the winter visits to house #58, specific investigation of the emission of NO_x associated with the operation of a gas stove was conducted. Figure 6.8 shows the increase in NO and NO_x measured in a dining room adjacent to the kitchen. The air was monitored about 5 m from the gas stove. The exhaust fan over the stove was not operated during these measurements. The windows and doors of the house were closed during this period. On January 20, two top burners were operated for 30 min on high heat. The sequence of operation on February 2 was as follows. One burner was operated for 15 min on the high setting to boil water. This burner was then turned to a low setting and a second burner operated on maximum heating to boil water for 15 min. Both burners were then turned off. During the second visit to house #58, a portable floor fan was operated in the kitchen. This resulted in better mixing of air and a smoother time variation in the levels of NO_x. Peak values of NO_x 200-300 ppb greater than the mean summer values were measured. Elevated levels of NO_x due to gas stove operation are consistent with the results obtained by other researchers.[1]

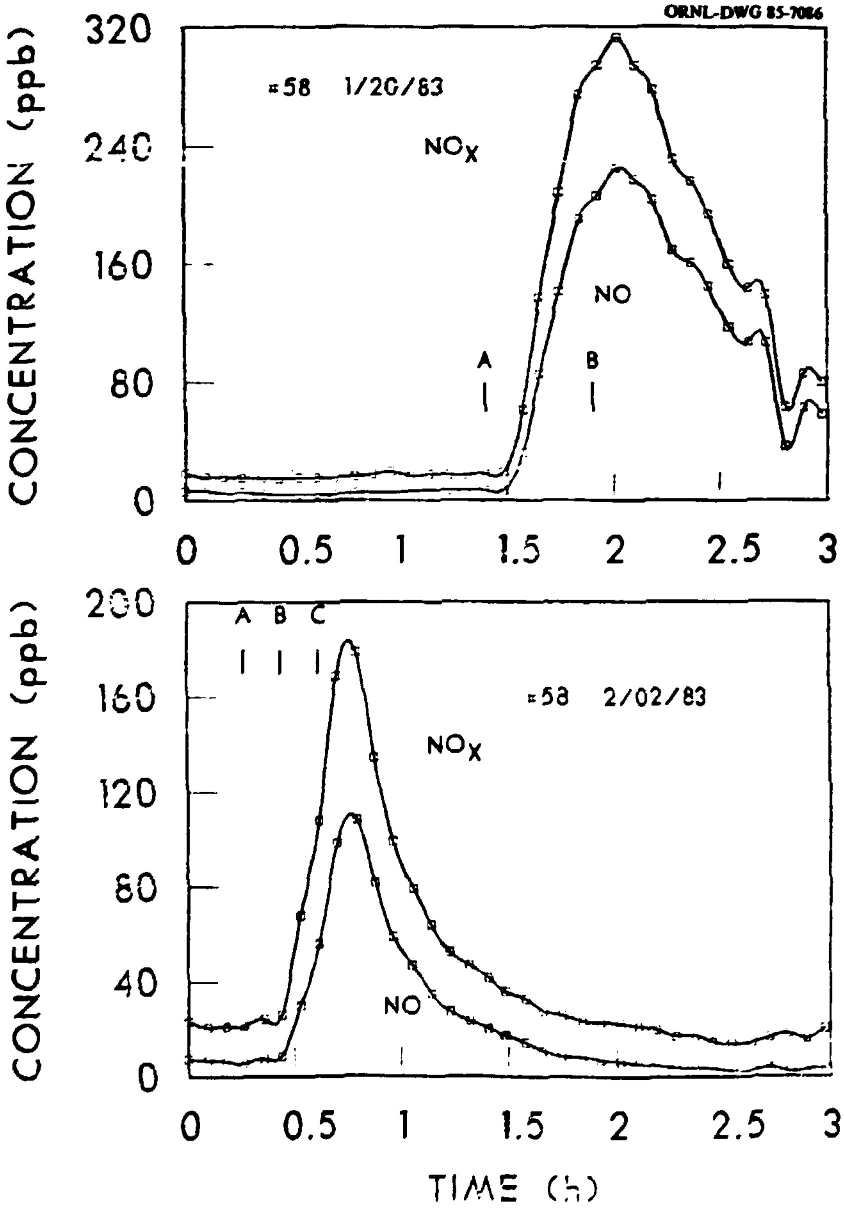

Fig. 6.8 Effect of operating a gas stove (house #58) on NO_x concentrations.

Increased levels of NO_x were also observed in house #68 with a 7000-BTU kerosene space heater as a supplemental source of heat. The monitoring equipment was located about 1.5 m from the space heater. The increase in NO_x levels that occurred during the operation of the space heater is shown in Fig. 6.9. During the January 28 visit to house #68, the heater was ignited shortly after the sampling team arrived. On the February 10 visit, the unit was already operating upon arrival and continued to operate throughout this second sampling period. This particular unvented space heater elevates the indoor levels of NO_x to a peak that is 60-80 pbb greater than the mean summer levels.

6.2.2 Carbon monoxide

The CO levels measured during the summer phase were generally near the detection limit of about 1 ppm. Exceptions include two houses (#65 and #70) in west Knoxville located near a busy road intersection. The outdoor levels of CO were higher (up to 6 ppm) near these homes than outside homes removed from busy streets where outdoor levels were near or below the detection limit. Another example of elevated CO levels indoors is associated with the operation of an automobile in a basement garage. The CO concentration in the bedroom above increased from <1 ppm to 17 ppm after the engine was operated for a period of 3 min. The central circulation fan of the HVAC was operating during this period and seemed to rapidly pull the garage air into the duct system and spread the exhaust pollutants through the house.

The CO monitor used during the winter measurements proved to be quite sensitive to temperature changes, and significant effort was required to obtain stable measurements. Of the readings that were valid, levels of CO were not greatly increased during the winter measurement period. Unfortunately, the CO monitor was not functioning properly during the specific combustion episodes monitored in house #58 (gas stove) and in house #68 (unvented kerosene heater).

6.2.3 Carbon dioxide

Carbon dioxide levels were generally higher in the houses with combustion sources than in the three control houses in which CO_2 was monitored. The mean concentration in the control homes was about 600 ppm, while the concentrations in some of the combustion houses were as high as 2000 ppm. The measured outdoor concentration was about 350 ppm.

There were several examples of significantly elevated CO_2 levels due to unvented indoor combustion. Figure 6.10 shows the increase in CO_2 concentration in house #58 on January 20, 1983, due to operation of the gas stove as described above. Figure 6.11 shows the increase in the CO_2 levels due to the operation of an unvented kerosene heater in house #68 on January 28, 1983. The level observed during this test approaches the 2500 ppm guideline for indoor air quality.[1]

6.3 Volatile Organics

The concentration of volatile organics was always substantially higher indoors than outdoors, usually by one order of magnitude or more. Figure 6.12 shows an example of chromatograms of an indoor and an outdoor sample collected with Tenax sorbent. Only the organic compounds with lower volatility than benzene were quantitatively retained in the trap. The large volume of air sampled through the Tenax cartridges resulted in the

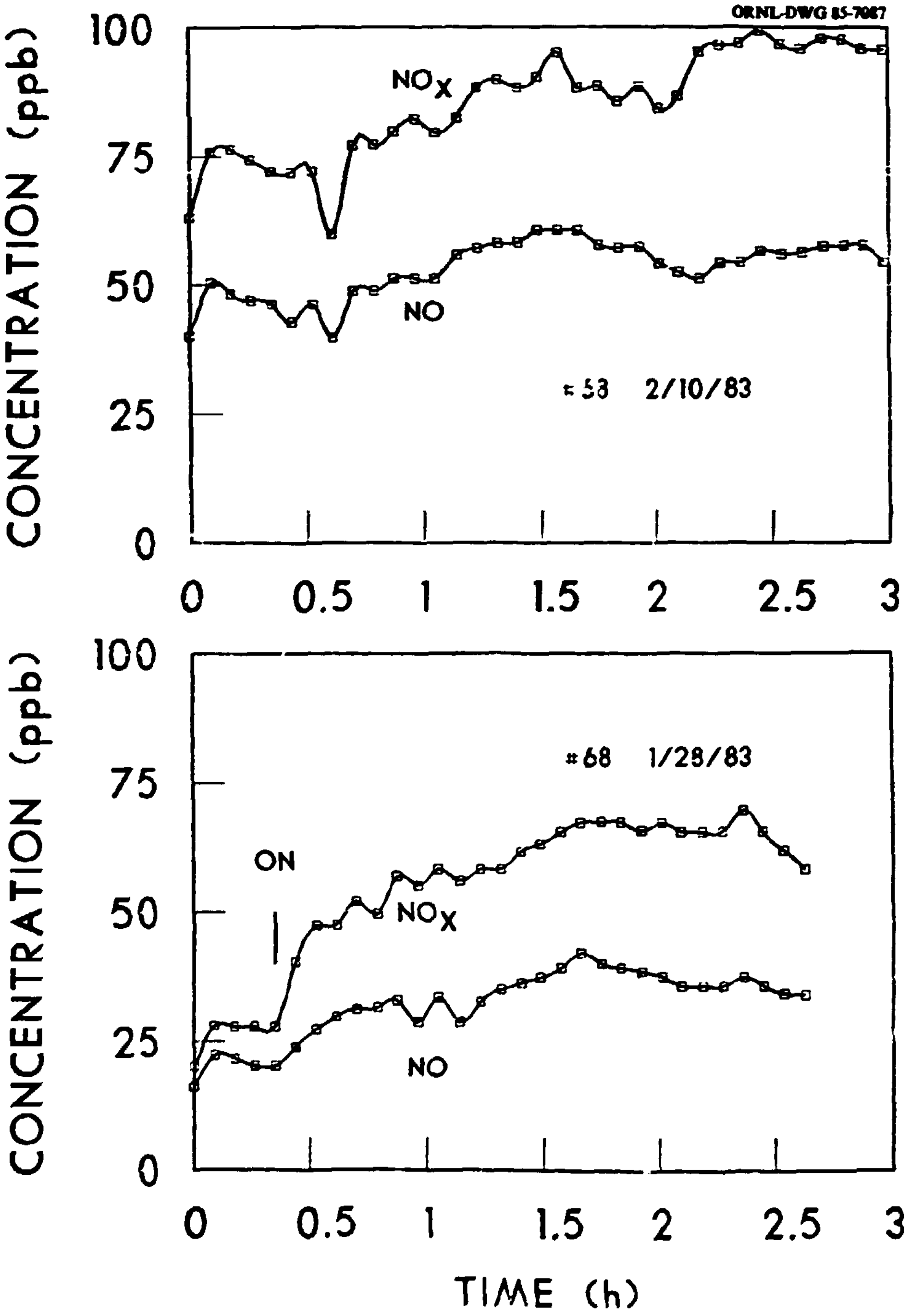

Fig. 6.9 Effect of operating a kerosene space heater (house #68) on NO_x concentrations.

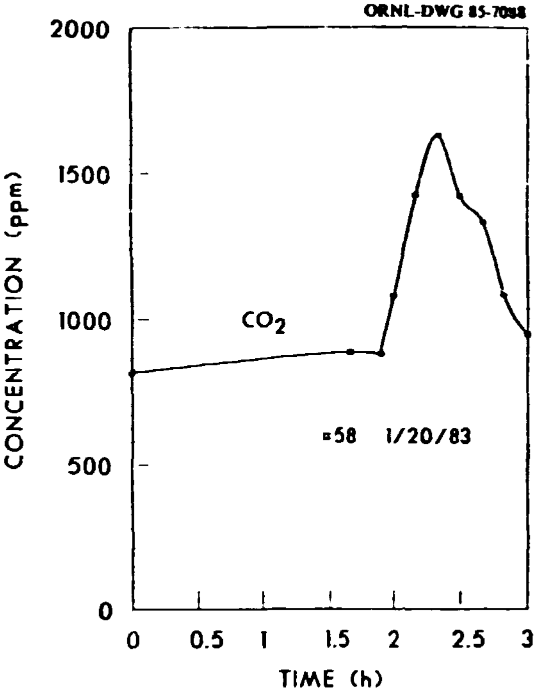

Fig. 6.10 Effect of operating a gas stove (house #58) on CO_2 concentrations.

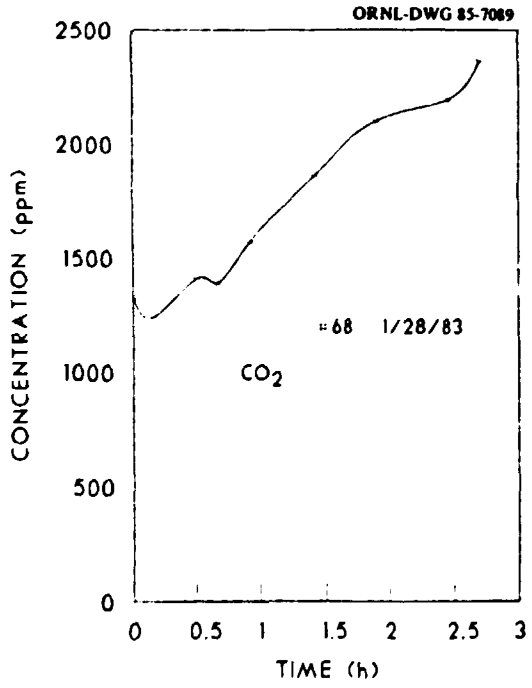

Fig. 6.11 Effect of operating a kerosene space heater (house #68) on CO_2 concentrations.

ORNL-DWG 85-7090

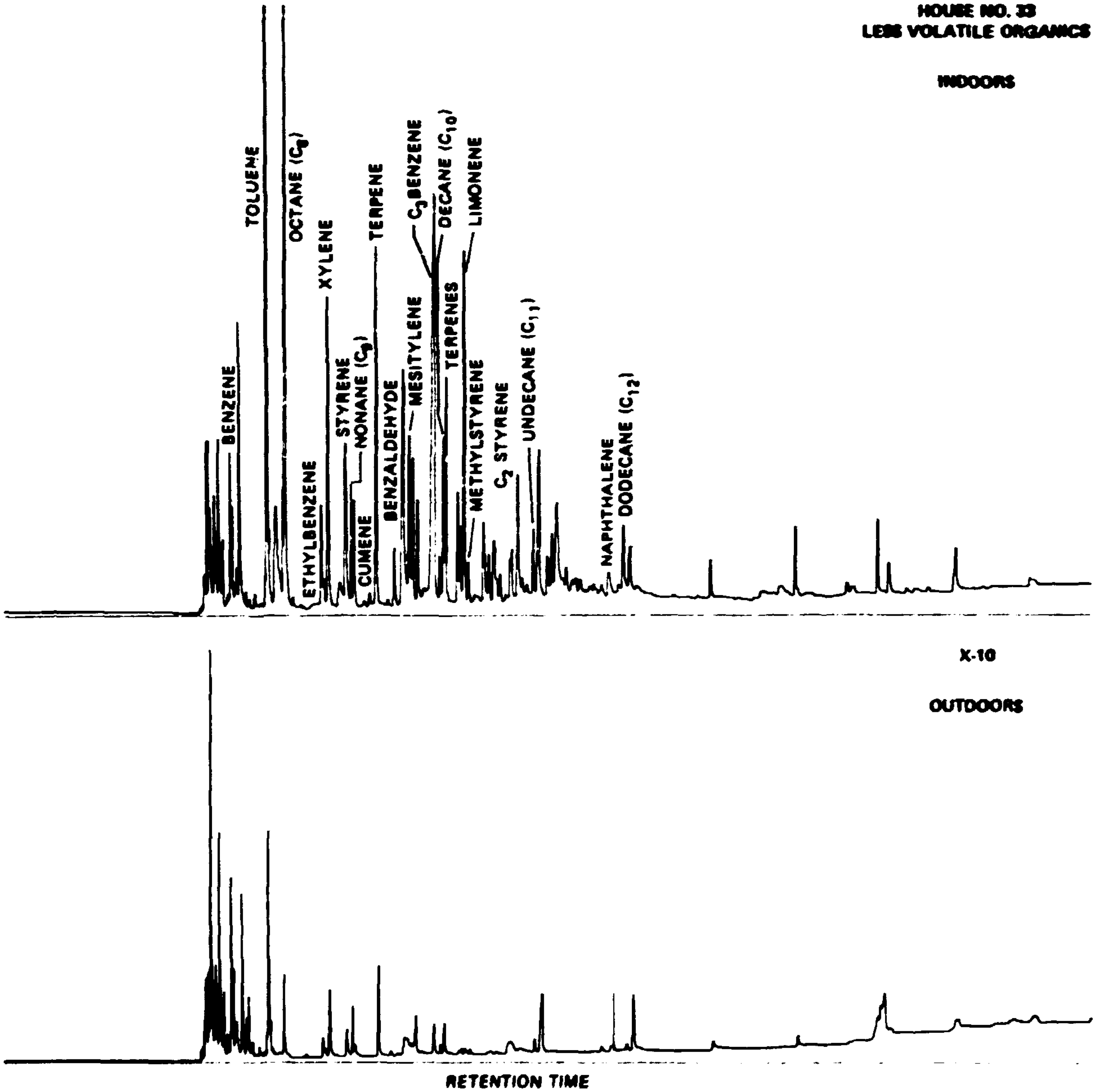

Fig. 6.12 Chromatograms illustrating the difference between indoor and outdoor organic content.

breakthrough of all highly volatile organic pollutants. Thus a separate contract has been established with ORNL by the CPSC to identify and quantify the highly volatile organic compounds in indoor air.

6.3.1 Tenax sorbent results

The chromatograms shown in Fig. 6.12 illustrate the difference between organic content of an indoor air sample and an outdoor air sample. The compounds listed in Table 5.9 were routinely quantified with the sampling and analytical methodology available for the study. This methodology is not suitable for the more volatile compounds. Of the compounds routinely quantified during the summer phase of the study, toluene and xylene were generally found at levels greater than 20 $\mu g/m^3$. Benzene was frequently present but was not quantified due to sample breakthrough. Some houses contained C_4- and C_5-benzenes, tetrachloroethylene, indane, and C_1- and C_2-indanes, naphthalene, and 1- and 2-methylnaphthalenes, plus some terpenes, phthalates, and siloxanes. House #65 contained a relatively large amount of dichlorobenzene and naphthalene (5000 $\mu g/m^3$ and 675 $\mu g/m^3$, respectively) that seemed to be associated with the visual observation of the liberal use of mothballs in that home.

Table 6.7 compares the summer and winter levels in combustion source houses and in the noncombustion source control houses. The results indicate similar levels of essentially all compounds in both subsets of homes measured during the winter. The approximate twofold higher total mean concentration during the summer months is consistent with increased vapor pressures resulting from increased temperatures.

Recently Molhave[21] has listed the ten most frequently encountered and the ten most abundant compounds in the air around a variety of building materials. Each of these ten most frequently encountered compounds (toluene, o- and m-xylene, 1,2,4- and 1,2,5-trimethyl benzene, ethyl benzene, n-propyl benzene, n-decane, n-undecane, and n-nonane) was frequently encountered in our study of homes in East Tennessee. The three compounds that Molhave lists as occurring in the highest concentrations are toluene, m-xylene, and the $C_{10}H_{16}$ terpene (limonene). For our study also, toluene and xylene were the most abundant organic compounds collected on Tenax. Limonene also was one of the more abundant compounds in the group of 17 compounds listed in Table 5.9.

6.3.2 Photovac GC results

Traces of gasoline vapor (identified on the basis of similarity of chromatograms with the chromatogram of gasoline headspace vapors) were ubiquitous to the indoor air of the 40 homes (see Figs. 5.6-7 for example). This circumstance is attributed to the storage of petroleum-based products and the operation of automobiles in a garage usually attached directly to the house. Air samples outside the houses also frequently contained trace levels of gasoline vapors. Sometimes the concentration of gasoline vapors indoors became significant [several tenths of a ppm of equivalent benzene (defined as nonspecific response obtained from the instrument that was calibrated with benzene vapors) as measured with a total organic monitor with a photoionization detector]. This was particularly the case if an automobile engine was operated inside the garage and the exhaust fumes were drawn into the house through the HVAC duct system. Motor oil or gasoline containers commonly stored in the garage, workshop, or basement contributed to the ubiquitous presence of gasoline vapors inside the houses.

Table 6.7. Comparison of summer and winter mean concentrations ($\mu g/m^3$) of organics collected on Tenax and analyzed by gas chromatography

Compound	Summer		Winter	
	Combustion	Control	Combustion	Control
benzene	[a]	[a]	5 ± 4	5 ± 4
toluene	43 ± 34	69 ± 74	30 ± 14	34 ± 19
ethyl benzene	8 ± 10	11 ± 13	5 ± 3	5 ± 4
m-xylene/p-xylene	32 ± 41	45 ± 55	17 ± 14	14 ± 4
nonane	7 ± 4	5 ± 6	5 ± 4	3 ± 2
cumene	2 ± 2	1 ± 2	2 ± 2	3 ± 3
benzaldehyde[b]	26 ± 29	24 ± 37	30 ± 52	6 ± 2
mesitylene	12 ± 12	6 ± 8	1 ± 1	-
decane	10 ± 8	6 ± 8	13 ± 12	11 ± 11
limonene	14 ± 19	30 ± 50	13 ± 8	3 ± 3
undecane	16 ± 19	6 ± 8	10 ± 12	7 ± 4
napthalene	46 ± 162	9 ± 11	13 ± 27	3 ± 2
dodecane	40 ± 150	4 ± 8	5 ± 4	2 ± 2
2-methylnaphthalene	7 ± 6	2 ± 2	2 ± 1	1 ± 1
tridecane	25 ± 40	15 ± 22	2 ± 1	2 ± 1
tetradecane	7 ± 7	4 ± 4	3 ± 1	3 ± 1
pentadecane	2 ± 3	2 ± 2	3 ± 1	2 ± 1
hexadecane	3 ± 3	3 ± 5	4 ± 2	
sum	300 ± 318	242 ± 216	163 ± 116	104 ± 42

[a] Benzene concentrations were not quantified because of breakthrough.

[b] Benzaldehyde and/or C_3-benzenes. May be an artifact of sampling method.

Additional and mostly unidentified peaks were associated with persistently emitting sources, a variety of organic species associated with transient operations such as from carpet relaying (see Fig. 5.7), or smoke leaking from a stove burning a mixture of wood and coal (Fig. 5.7).

The Photovac GC has yet to be developed to its full potential. Better control is needed of the column temperature and the rate of gas flow. Current inadequate control of these parameters causes highly variable retention times. Compound identification is very difficult unless an internal standard(s) is injected concurrently with the indoor air sample. Positive identifications have thus far been made only for benzene and toluene, two compounds encountered in most houses.

6.4 Particulate Material

The level of RSP (particulates less than 3.2 μm aerodynamic diameter) averaged over 34 houses during the summer phase of the study was 29 ± 14 $\mu g/m^3$, with the maximum average concentration for a single house being 68 $\mu g/m^3$. The mean level of TSP (particulates less than 25 μm aerodynamic diameter) was 38 ± 17 $\mu g/m^3$, with the maximum average concentration measured being 88 $\mu g/m^3$. The mean outdoor levels were 30 ± 30 $\mu g/m^3$ and 34 ± 29 $\mu g/m^3$, respectively, for the RSP and TSP. Measurements obtained in six of the houses during the early part of the study were not valid due to overloading of the crystals and were not included in the analysis. Few of the houses in this study had occupants who were smokers (>1 pack per week). Of the houses with smokers, the smokers were generally not present during the summer when the particle analyzer was operating.

Table 6.8 shows a comparison of the particulate levels obtained in the winter control homes and the combustion source homes with the corresponding levels obtained during the summer visits. The mean levels of RSP and TSP are 27 ± 20 $\mu g/m^3$ and 36 ± 26 $\mu g/m^3$, respectively, in the control homes during the summer and 19 ± 10 $\mu g/m^3$ and 26 ± 15 $\mu g/m^3$ during the winter period. The summer readings in the combustion source homes are 23 ± 32 $\mu g/m^3$ and 26 ± 36 $\mu g/m^3$, while during the winter phase the mean values are 37 ± 46 $\mu g/m^3$ and 44 ± 48 $\mu g/m^3$. Figure 6.13 shows the increase in particulate concentration when the kerosene space heater was operated in house #68. An increase is also noted in Fig. 6.14 when the wood/coal stove (house #39) was in operation. It was observed upon arrival of the monitoring team that the stove was not operating correctly; smoke was clearly visible in most rooms of the house. Soon after the measurement period began, the problem was corrected, and the smoke slowly dissipated. A decrease in particulate levels can be seen in Fig. 6.14.

Table 6.8. Comparison of summer and winter particulate levels ($\mu g/m^3$)

House Number	RSP		TSP	
	Summer	Winter	Summer	Winter
2[a]	15 ± 6	16 ± 8	22 ± 8	31 ± 14
5	30 ± 9	14 ± 5	36 ± 12	17 ± 5
12	11 ± 10	16 ± 5	14 ± 11	20 ± 7
14	35 ± 20	27 ± 10	42 ± 26	33 ± 13
26	44 ± 20	19 ± 7	52 ± 26	29 ± 8
36	29 ± 17	10 ± 6	34 ± 17	12 ± 7
70	30 ± 13	24 ± 9	47 ± 22	42 ± 21
79	41 ± 35	18 ± 3	50 ± 38	32 ± 8
80	28 ± 22	9 ± 5	42 ± 36	14 ± 6
82	15 ± 9	40 ± 6	27 ± 23	44 ± 6
24[b]	32 ± 54	19 ± 7	39 ± 58	24 ± 9
39	.	96 ± 97	.	111 ± 98
46	.	106 ± 51	.	115 ± 52
54	44 ± 30	23 ± 9	47 ± 36	27 ± 10
55	29 ± 22	19 ± 19	32 ± 26	21 ± 20
58	24 ± 11	15 ± 8	28 ± 16	19 ± 10
62	46 ± 39	46 ± [illegible]	51 ± 48	52 ± 33
65	25 ± 10	12 ± 9	26 ± 9	16 ± 10
68	.	29 ± 24	.	42 ± 26
83	.	19 ± 5	.	27 ± 11

[a] First ten houses are control houses.

[b] Last ten houses are combustion source houses.

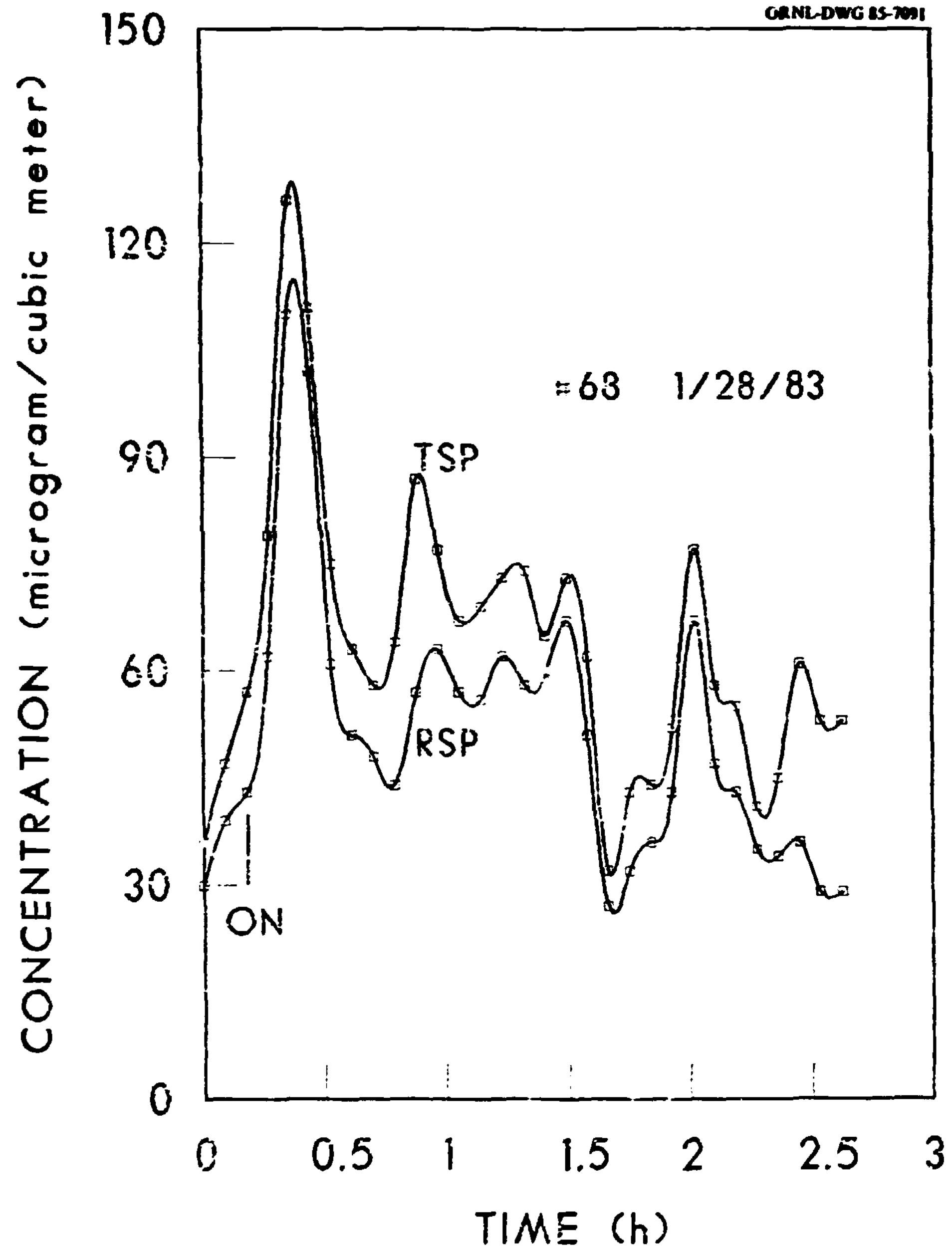

Fig. 6.13 Effect of operating a kerosene space heater (house #68) on particulate levels.

No chemical analysis of the particulate matter in the study homes was conducted. The measured levels in most of the homes were below the EPA outdoor standard of 75 $\mu g/m^3$ yearly average.[1]

6.5 Radon Monitoring

Significant differences were observed between the radon levels in the various houses in the study. These differences, ranging from <1 pCi/L to >10 pCi/L, were measured both by the short-term, active monitor and by the long-term, passive monitors. Levels exceeded various indoor guidelines of about 4 pCi/L[1] in about one-third of the 40 homes. A possible explanation for the elevated levels appears to be the location of the houses relative to geological structures.[25] A large fraction of the homes with higher levels of radon are located on a dolomite ridge, which appears to offer a low resistance conduit from uranium-bearing shale. The general area is noted for bituminous shale which can be a source of radon emission. These data illustrate the large differences in indoor levels of radon that can occur even within a local area. To aid in the interpretation of the data, the houses have been classified as "ridge" or "valley" based primarily on the monitoring team's subjective observations at the site of each house.

6.5.1 Wrenn chambers

Table 6.9 shows the differences in radon levels in the ten homes in which the Wrenn chambers were operated. Also listed is the corresponding value obtained with the passive monitors for these same ten houses during the winter. Generally, quite good agreement is obtained between the two methods. Diurnal fluctuations were observed in some of the monitored houses. Figure 6.15 illustrates one of the more extreme fluctuations observed. The maximum concentration occurs about 7 AM each day. No satisfactory explanation for this behavior has been found. This house does not have deep well water which could be a source of radon. Interestingly, this house is not identified as a "ridge" house, yet it has rather high (>4 pCi/L) levels of radon.

6.5.2 Track etch monitors

The measured values for the three dosimeters in each room were found not to be significantly different from each other ($P < 0.05$) and were averaged to form a single room value for subsequent analysis. Figure 6.16 shows the distribution of radon levels between "ridge" houses and "valley" houses. In 8 of 13 houses on the "ridge," radon levels are above 4 pCi/L, while only 4 of 26 houses in the "valley" are above this level. Table 6.10 lists the measured radon levels in each of the 40 homes, indicating the levels in partially underground rooms and noting if this portion of the house is finished and regularly occupied. The outdoor radon levels averaged less than 0.5 pCi/L and were not significantly different for the "ridge" and "valley" houses.

6.6 Air Exchange Rates

The most striking result of the air exchange rate measurements is the difference between the exchange rate when the HVAC circulation fan is operating and when the fan is turned off. This difference was not due to poor tracer mixing as portable floor fans were used at all times to ensure good mixing. Figure 6.17 shows the different distributions obtained for the two cases. The geometric mean of the fan-on data is 0.79 h^{-1} and of the fan-off data is 0.44 h^{-1}. [The chi-square value ($P < 0.001$) obtained for the log normal fit indicates that this function is a likely distribution for the air exchange rates. The arithmetic means for the fan-on and fan-off operation is 0.74 h^{-1} and 0.39 h^{-1}, respectively.] Table 6.11 shows the reproducibility obtained for the fan-on rates for homes with at least three visits.

The likely reason for the much higher air exchange rates when the central fan is operating is that air leaks into and from the duct system. Exchange with outside air is likely to occur in ducting located in crawl spaces, garages, and heat pumps located outside the house. The pressure drop created in the duct system going to the fan and the increased pressure after the fan provides the driving force for the air leakage. In individual houses, inspection revealed air escaping from around the service panels of heat-

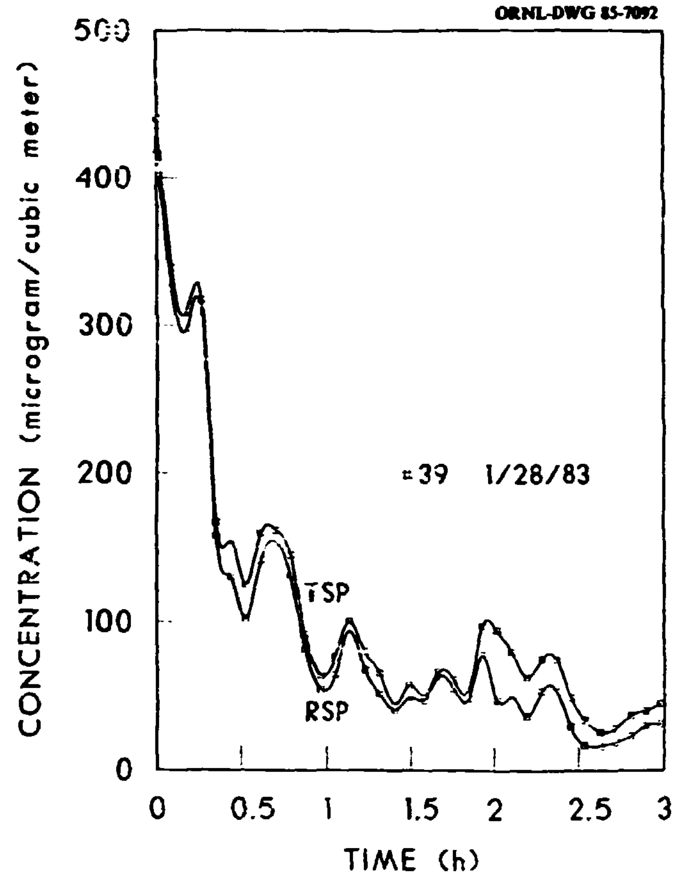

Fig. 6.14 Effect of operating a wood/coal stove (house #39) on particulate levels.

Table 6.9. Mean radon levels (pCi/L) in houses where both active and passive measurements were made

House no.	Start date for Wrenn chambers	Radon concentration Wrenn chambers	Radon concentration Terradex monitors	House location
2	04-13-82	3.8 ± 1.3	3.5	Ridge
		(5.3 ± 1.2)[a]	(5.7)[a]	
82	04-05-82	8.0 ± 1.0	13.2	Ridge
		(18.0 ± 6.0)[a]	(17.4)[a]	
4	04-20-82	10.0 ± 3.0	12.0	Ridge
14	01-18-82	4.7 ± 2.0	3.8	Ridge
7	02-17-82	0.8 ± 0.5	0.8	Valley
11	03-05-82	0.7 ± 0.3	1.0	Valley
27	05-25-82	1.0 ± 0.5	1.9	Valley
64	06-10-82	1.2 ± 0.7	3.3	Valley
70	05-05-82	3.3 ± 2.3	7.8	Valley
81	10-15-82	0.7 ± 0.2	0.8	Valley

[a] Values in parentheses are basement measurements.

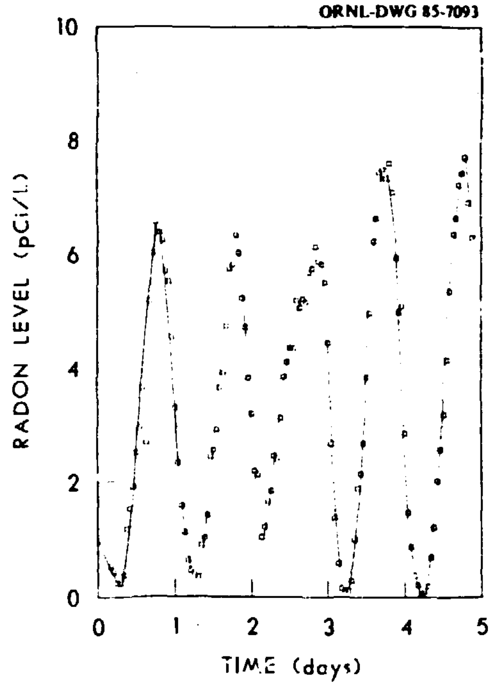

Fig. 6.15 Diurnal fluctuations in radon levels in house #70.

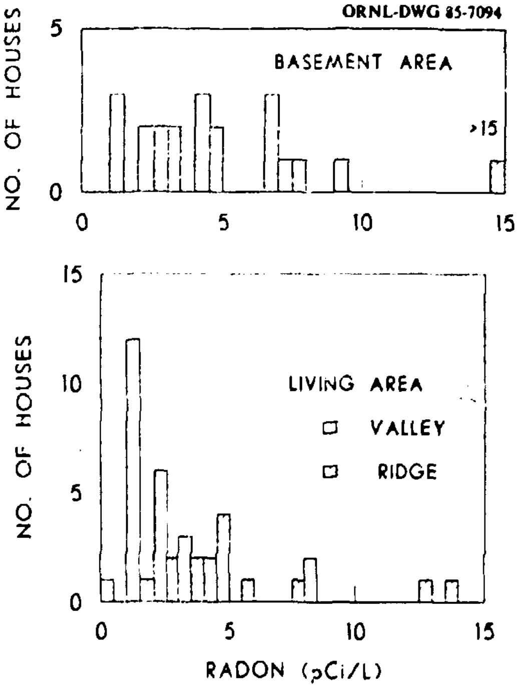

Fig. 6.16 Distribution of radon levels with "ridge" vs "valley" houses indicated

Table 6.10. Radon levels measured with Terradex track etch monitors [levels in partially underground (including basements) living space indicated in parentheses]

House No.	Radon level (pCi/L)	
2	3.49	(5.68)
3	1.90	
4	11.99	(6.90)
5	1.81	
7	0.76	
8	1.62	
9	2.62	
10	3.78	(2.48)
11	1.04	(1.76)
12	4.09	(5.57)
14	3.85	(6.23)
15	5.16	(8.19)
21	3.96	(4.12)
23	0.57	(0.96)
24	1.15	
26	0.60	
27	1.90	(2.69)
29	1.03	(1.31)
33	0.79	
34	1.59	(2.01)
36	0.57	
37	1.68	(1.81)
39	3.37	(4.16)
41	6.65	(4.35)
46	3.61	
54	1.38	(2.70)
55	3.77	(4.13)
58	0.96	(1.38)
61	1.60	
62	1.77	
64	3.28	(4.28)
65	7.98	(7.31)
68	3.20	
70	7.80	(8.54)
75	0.78	
79	0.40	
80	1.39	
81	0.85	
82	13.22	(17.41)

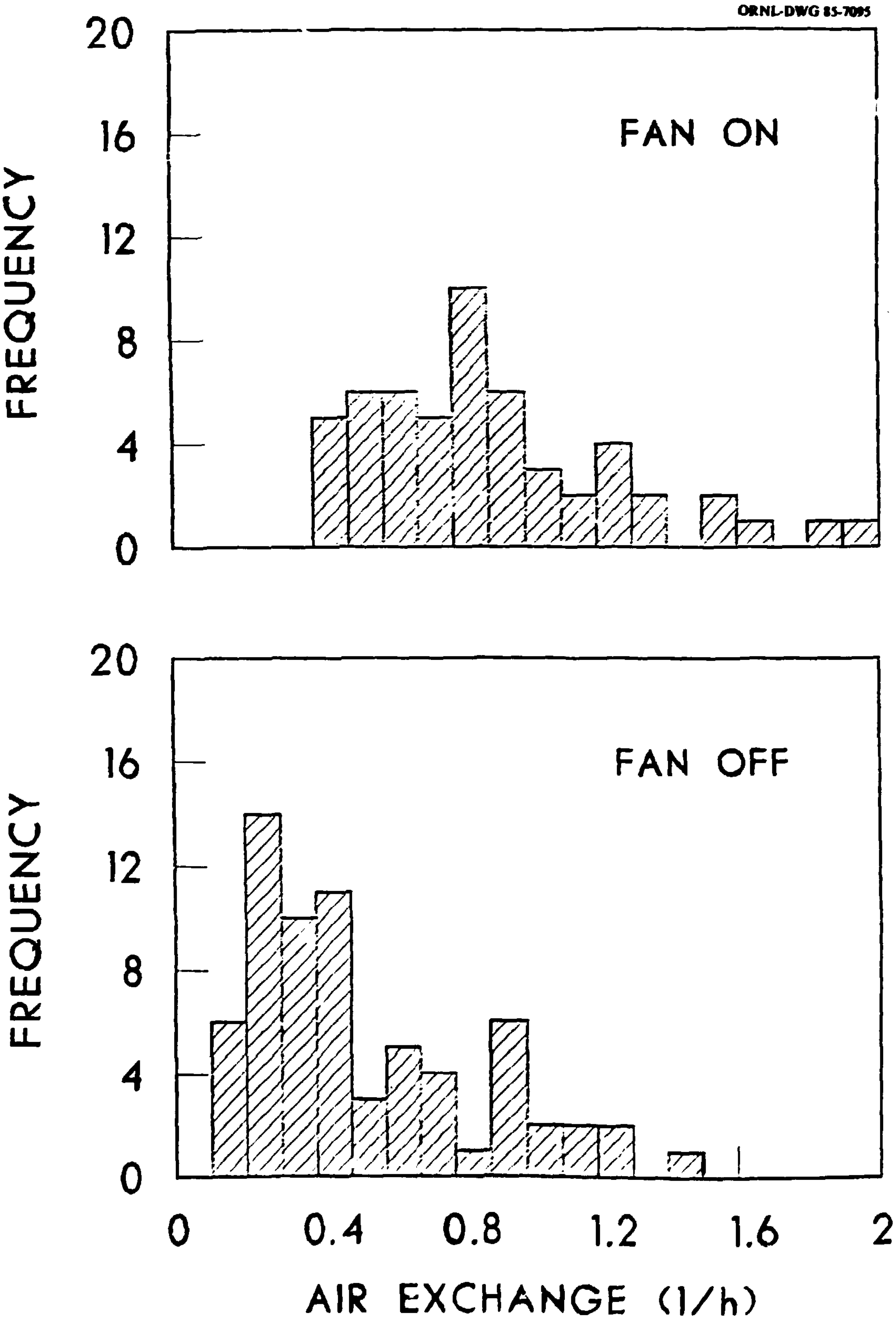

Fig. 6.17 Distribution of air exchange rates for fan-on and fan-off measurements.

Table 6.11. Air exchange rates (ACH) in study houses in which measurements were made on three different dates

House No.	Date	ACH (h^{-1})		Date	ACH (h^{-1})		Date	ACH (h^{-1})		Mean ACH (h^{-1})	
		Fan Off	Fan On		Fan Off	Fan On		Fan Off	Fan On	Fan Off	Fan On
2	04/13/82	-	0.51	09/09/82	0.31	0.65	12/13/82	0.48	0.81	0.39	0.66
5	04/22/82	0.50	-	08/13/82	0.22	0.45	12/14/82	0.18	0.42	0.30	0.43
39	04/27/82	0.27	-	01/18/83	1.12	-	02/09/83	0.60	-	0.66	-
46	06/02/82	0.34	-	01/06/83	0.51	-	01/24/83	1.47	-	0.77	-
54	07/14/82	0.45	-	01/26/83	0.97	0.89	02/07/83	0.98	1.21	0.80	1.05
55	06/22/82	0.28	0.31	01/31/83	0.35	-	02/08/83	0.39	0.51	0.34	0.41
58	11/12/82	0.95	1.56	01/20/83	0.71	1.27	02/02/83	1.08	1.23	0.98	1.31
62	05/06/82	0.27	1.57	01/07/83	0.75	1.69	01/27/83	1.28	1.98	0.76	1.75
65	05/27/82	0.26	0.32	01/25/83	0.41	0.96	02/11/83	0.31	1.05	0.33	0.78
68	06/09/82	0.83	1.34	01/28/83	-	0.77	02/10/83	0.40	0.83	1.55	0.98
82	04/07/82	-	0.46	11/10/82	0.24	0.51	01/05/83	0.50	0.96	0.37	0.64
83	11/05/82	0.60	1.07	01/03/83	0.96	1.84	02/01/83	0.65	0.96	0.74	1.29

pump units located outside the house. Inside attached garages examples were recorded of air containing automobile exhaust gases being pulled into the duct system, which is normally under negative pressure as air is being pulled back to the heat/cooling system in some of the houses.

These findings provide important implications to homeowners and researchers concerned with weatherization to reduce leakage and conserve energy. Another obviously important finding (Section 6.3.2) is that polluted air in garages can be drawn into HVAC duct systems and rapidly dispersed to the living space of the house. The air exchange data base and the several pollutant data bases offer a significant opportunity to evaluate correlations with air exchange rates and levels of various pollutants, especially within the same home. This evaluation has yet to be conducted and will, it is hoped, be completed in the near future.

6.7 Meteorological Data

Table 6.12 shows the monthly averages for temperatures as recorded by the Oak Ridge office of the NOAA. Also presented are the monthly totals for the precipitation measured at the weather station on Illinois Avenue. The spring and early summer were drier and cooler than normal. Hot and humid weather did not arrive until August. Winter weather was late in arriving and milder than normal.

The meteorological data obtained during the study provides an opportunity to evaluate correlations between various weather-related parameters and the several pollutants monitored. Resource limitations have prevented this analysis from being conducted within the main portion of this study; however, it is anticipated that support can be found to complete this assessment.

Table 6.12. Monthly averages for temperatures and monthly totals for precipitation as recorded by the Oak Ridge Office of the NOAA

Month	Average measured temp. (°C)	Average historical temp (°C)	Total measured precip. (mm)	Total historical precip. (mm)
April	11.6 ± 4.6	14.9 ± 1.9	71	109
May	20.8 ± 3.1	19.3 ± 1.3	73	106
June	22.2 ± 2.0	23.5 ± 1.0	55	105
July	25.4 ± 1.3	25.0 ± 0.0	184	135
August	23.3 ± 1.7	24.5 ± 0.6	117	96
September	19.6 ± 3.9	21 ± 1.6	150	92
October	14.4 ± 5.1	14.9 ± 2.0	49	75
November	8.7 ± 5.1	8.4 ± 1.8	198	116
December	6.3 ± 6.9	3.9 ± 0.7	183	141
January	2.0 ± 3.1	2.6 ± 0.3	44	137
February (through 02/15/83)	1.0 ± 3.6	3.4 ± 0.3	91	121

7. CONCLUSIONS

The recently completed, year-long Indoor Air Quality Study in 40 homes in East Tennessee represents an important addition to the existing data base on residential indoor air quality. The extensive measurements made of formaldehyde levels especially represent a unique data base. This report has presented the data obtained during the study and some preliminary analysis of the data. Several important preliminary conclusions can be drawn from the data analyzed to date. These conclusions may need to be confirmed by additional analysis of this data base or by additional studies. The major conclusions are listed below:

1. Formaldehyde levels in the study homes frequently exceeded 0.1 ppm. Approximately 30% of the homes exceeded 0.2 ppm in one room on one or more visits while about 60% exceeded 0.1 in one room on one or more visits.
2. Formaldehyde levels were highest in the newer homes. Mean levels for houses older than 5 years did not exceed 0.1 ppm unless the house contained UFFI as a retrofit or other major sources of formaldehyde. Both UFFI and non-UFFI houses show a decrease in mean formaldehyde levels with age.
3. Formaldehyde levels can fluctuate considerably, both diurnally and seasonally. In the limited diurnal measurements taken, variations of 50% or more were observed between the minimum and maximum levels. Variations of tenfold or more occurred in some houses over the course of the study, probably due to environmental dependence of sources, product aging, and occupant lifestyle.
4. Organic pollutant levels were at least an order of magnitude higher indoors than outdoors. Attached garages appeared to be a frequent source of the highly volatile organics, which are being further investigated and quantified in a subset of houses. Higher mean levels in summer than winter were observed for many of the quantified organics.
5. Combustion sources (especially unvented sources such as kerosene heaters and gas stoves) substantially increased levels of CO_x, NO_x, and particulates in some cases. Short-term peaks of more than 300 ppb of NO_x and more than 2000 ppm of CO_2 were measured during operation of the devices.
6. Radon levels in the study homes frequently exceeded 4 pCi/L. Approximately 30% of the homes measured had levels greater than this value.
7. Radon levels correlated strongly with house location. Houses that were located on the ridge surrounding Oak Ridge were most frequently associated with elevated levels.
8. Air exchange rates were increased nearly twofold by operation of the HVAC central air circulation fan. The air exchange rate ranged from 0.1 to 2.0 h^{-1}, with a mean value of 0.86 h^{-1} when the fan was on and 0.53 h^{-1} when the fan was off or there was no central fan in the house. (These means include both winter and summer data.)

8. RECOMMENDATIONS

Several areas where additional research and development work is needed to support indoor air quality activities have been identified during the course of this study.

1. New and improved portable instrumentation capable of automatic recording of time-varying concentrations of low levels of pollutants is required.
2. Source identification and experimental characterization (including environmental chamber tests) are needed for various pollutants, especially organic compounds.
3. Additional passive integrating monitors for chronic exposure monitoring should be field tested.
4. Rigorous laboratory and field intercomparisons of monitoring devices are necessary.
5. Emission rates and pollutant concentrations need to be modeled and the models validated experimentally.
6. Before and after measurements of indoor air quality impact due to energy conservation (and alternative fuels) measures should be performed.
7. Remedial actions should be evaluated and improved, based on a better understanding of source behavior.
8. Feedback should be provided to building designers and manufacturers of consumer products on the consequence of various products on indoor air quality. Suggestions for alternative materials and practices should be made.
9. Similar studies should be conducted in other geographical locations and in a statistical sampling of houses representing national housing stock.

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APPENDIX A

PROTOCOL FOR MEASURING INDOOR AIR POLLUTANTS IN OAK RIDGE AREA HOMES

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OBJECTIVES AND BACKGROUND

Over the past several years, increased energy costs have intensified the need for home weatherization. As this has occurred, concern has grown regarding the level of indoor air pollutants associated with reduced air-infiltration rates. The effect of increased levels of indoor pollutants on human occupants is of some concern, since the mutagenic or carcinogenic effects of some of these pollutants are only now being determined and certain individuals may demonstrate an allergic response. To date a comprehensive program for measuring the full spectrum of indoor pollutants has not been developed. Such a comprehensive program must be developed before it will be possible to effectively assess potential health risk.

The purposes of this study are to: a) identify the various components of indoor air pollution and house products that are the sources of some of these pollutants, b) quantitate the levels and the fluctuations therein of selected pollutants such as organics, CO_x, SO_2, NO_x, and particulates in typical indoor environments, and c) evaluate and, where necessary, introduce new developmental instruments or techniques that can be used on a routine basis for measurements of indoor air pollutants. An attempt will also be made to assess the correlation of the pollutant levels with the age of the home, supplemental energy sources, air exchange rate, outdoor meteorological conditions, structural features, and consumer product use (types of home insulation, air cleaning systems, furniture, furnishings, etc.).

PROPOSED PROTOCOL

A brief report on the details of the Consumer Product Safety Commission (CPSC) indoor environment study has appeared on February 25, 1982, and March 30, 1982, in the local newspaper, the Oak Ridger, and through the Union Carbide Corporation Nuclear Division Newsletter (December 3, 1981, and March 4, 1982). An article has also appeared in the Knoxville News-Sentinel on March 20, 1982. The name and telephone number of the project leader(s) were listed and those interested in participating in the study were encouraged to contact the identified individual(s). A house survey form was sent to approximately 100 individuals who responded to the published articles. This survey form is included as Attachment 1 and requests information from potential participants on the structure and layout of their home, type of heating employed, type of cooking stove used, insulating materials employed, and the type of air cleaning system (if any) used.

From the initial mailing of 100 survey forms, 40 homes have been selected for the study. The forty homes were selected so as to provide a statistically significant sampling of homes by age, modes of cooking and heating, and types of insulation. Factors such as socio-economic levels, proximity to busy thoroughfares, ease of transportation by the monitoring teams to and from the home, recent home improvements, permanence of residence, pets, children, and the anticipated degree of cooperation from the homeowners over the course of the study were considered. The forty homeowners were mailed a copy of the Homeowner's

Agreement (Attachment 2) stating their willingness to participate in the study and the obligations of the investigators to the homeowner. Receipt of the completed Homeowner's Agreement signified formal participation in the study.

The pollutant monitoring program is to be carried out in forty houses with specific sets of characteristics. Major selection criteria are listed below along with the number of homes that we sought to identify within each category. The distribution of home characteristics is as follows:

Old (10+ yrs)	New (0-2 yrs)	Electric[1]	Gas[2]	Wood/Coal[3]
10		10		
10			10	
10				10
	10	10		
	10		10	
	10			10

[1] Primarily electric heating and cooking.
[2] Gas cooking and/or gas-type space heating.
[3] Wood stoves used primarily for heating although other forms of heating may be present. Coal may also be used at times. This category, therefore, will overlap the electric and gas numbers.

The major difference will be between old and new houses. Other secondary parameters used in the selection process include: 1) weatherization measures, 2) use of supplementary energy sources, 3) type of insulation, and 4) air cleaning systems employed. It was initially quite impossible to predict exactly the number of homes that would fall into each category. This was possible only after return of the initial survey forms. The above distribution reflects the optimum case that would permit us to draw conclusions about the contribution of each factor to the total air pollution situation.

The homes were selected by location according to the following criteria:

- Semi-rural environment (Oak Ridge, TN, population 25,000)
- Semi-urban environment (Knoxville, TN, population 250,000)

It was determined after examination of the initial mailing of 100 survey forms that there existed three large groupings of homes. These were east Oak Ridge (15 homes), west Oak Ridge (15 homes), and west Knoxville (10 homes). The differences in setting permits us to draw conclusions about indoor pollutants associated with urban vs small-town life styles. From a practical standpoint, an attempt was made to choose homes within a reasonable distance of one another. Travel time and transportation becomes increasingly important when one considers the number of measurements to be taken. Homes owned by laboratory personnel offer special benefits from the standpoint of degree of participation and interest in energy-conservation measures.

Each home in the study will be surveyed according to two separate monitoring schemes. The homes will be visited for one-day intervals four times during the study. The one-day visits will occur so that each house will be sampled once during the spring, summer, fall, and winter seasons. The second monitoring scheme involves the sampling for formaldehyde levels in each home on a bi-weekly basis. The details of the the two monitoring schemes are presented in the following pages. Specifics about the routine methodology can be found in Appendix A. A list of the major instruments used in the study is given in Appendix B. Detailed analytical procedures will be presented in Appendix C.

One-Day Surveys:

The surveys conducted once during each season of the study will focus on measurements of formaldehyde, general organics, nitrogen-monoxide and -dioxide, sulfur dioxide, particulates, carbon-monoxide and -dioxide, and radon. Measurements of air-infiltration rate and outdoor meteorological conditions will also be made on the occasion of each visit. At the outset of each one-day visit the home will be closed up for two hours. This two-hour period will allow build-up of ambient pollutant levels and permit measurement of air infiltration rates. The air-infiltration rates will be measured through use of a tracer gas, Freon-12. The concentration of the tracer gas will be followed over time with an infrared analyzer. During the course of the air-infiltration measurement, interior doors in the home will be opened while the exterior doors and windows will be closed. At the same time that the air-infiltration measurements are being taken, the meteorological data will also be collected. These measurements will include temperature, relative humidity, and wind direction and speed. The measurements will be continued throughout the day-long monitoring period and will be checked against official data obtained from the National Oceanic and Atmospheric Administration (NOAA). After the air exchange measurement is complete we will use, in just one season of the year, a door blower fan to measure ventilation rates for the home.

Pollutant-specific measurements during the day-long monitoring period will be carried out in the main living or family room, preferentially on the same level as the kitchen, according to the following outlines:

> Formaldehyde. While the house is closed for the air-infiltration measurement, one room of the house will be measured by two monitoring techniques for formaldehyde build-up. The first method will involve pumped molecular

sieve measurements. The second technique is the standard National Institute of Occupational Safety and Health-(NIOSH-) approved single/double impinger method utilizing 1% sodium bisulphite solution as the collecting agent with subsequent colorimetric analysis using chromatropic acid. Use of two monitoring techniques will provide a measure of quality control.

Organics. Ancillary sampling of organics will be conducted utilizing a portable gas chromatograph with detection by photoionization. An attempt will be made to locate specific sources of key organic pollutants identified on Tenax sorbent. At a later time we expect to expand this project to include a major study of the sources of these pollutants. This will require some major laboratory work in the categorization of a large number of fingerprints from these substances to enable identification of the complicted spectra that are normally obtained. With increasing temperature the spectra obtained tends to expand, especially at the higher retention times. Also, variability in the flow rate can affect the measured spectra. Therefore, careful calibration of the unit is necessary because of these fluctuations.

NO_x. Indoor levels of NO and NO_2 will be monitored in a single room of the home using a chemiluminescent monitor. Measurements will be taken to obtain peak and average concentrations for the entire one-day monitoring period. This monitoring period will encompass those times of the day when generation of NO_x from routine activities is likely to be at a maximum, e.g., during operation of gas stoves, space heaters, or from cigarette smoking. Measurements will also be made outdoors specifically during those periods of the day when NO_x concentrations are at their maximum.

SO_2. Measurements of SO_2 will be performed in selected residences which use space heaters fueled by coal or kerosene. A DUVAS (Derivative UV-Absorption Spectrometer) will be used for this purpose. Measurements while the space heaters are in use will be made.

Particulates. A measurement of ambient levels of particulates will be made in one room of the home with a particle-size analyzer. Eight-hour peak and average concentrations will be measured within the home and a measurement of particulate loading in the outdoor environment will be made. A ten-stage cascade impactor, quartz crystal microbalance will be used to separate and measure particles down to 0.05 μm. The 8-hour sampling period will lessen the possibility of overloading the crystal microbalance and reducing measurement accuracy.

CO_x. Measurements of indoor levels of carbon monoxide will be made using 4 small electrochemical dosimeters. These measurements will be taken every 30-minutes for an 8-hour period. The measurements will be taken in several rooms of the home and in the outdoor environment, upwind of the house. Multiple measurements are planned to pinpoint specific sources of carbon monoxide such as a gas stove located in the kitchen.

Carbon dioxide will also be measured using an infrared analyzer. This will be done after the air exchange measurements are performed.

Radon. The levels of radon within selected homes, about ten in number, will be measured using two modified Wrenn chambers. Monitoring will be carried out over a four-day survey period. It is likely that one chamber will be placed in the crawl space or basement beneath the home and the other chamber will be located in the main living area. These measurements will be performed in fewer than forty residences because of the extended survey period required for the ingrowth of radon daughters.

The types of measurements to be performed by this survey team in a given time period over the course of the study are given below:

Measurement	Number/Home	Number/Survey	Total Number*
One-day Survey			
Formaldehyde			
Double Impinger	2	8	320
Molecular Sieve	4	160	140
Organics			
Portable gas chromatograph	as appropriate		
NO, NO_2			
Chemiluminescence	1	40	160
SO_2			
Absorption Spectrometer	as appropriate		
Particulate			
Cascade Impactor	1	40	160

Measurement	Number/Home	Number/Survey	Total Number*
<u>One-day Survey</u> (continued)			
CO			
Electrochemical	3	120	480
CO_2			
Infrared Analyzer	1	40	160
Radon			
Wrenn Chamber	1	10**	40

*Comprises one complete survey (40 homes) during each of four seasons. This number excludes outside measurements and any blanks.

**In ten selected homes.

Routine Formaldehyde Surveys

The second monitoring scheme involves bi-weekly monitoring of formaldehyde at each of the forty homes. This second monitoring scheme has been developed to provide an assessment of the variation in formaldehyde levels as a function of environmental conditions. On Monday through Thursday of each work week, 15 passive samplers will be placed in 5 of the 40 houses within the study. The samplers will be exposed for a 24-hour period and picked up the next day for evaluation. These passive samplers are plastic cups containing water and are covered by a semi-permeable membrane. After exposure, the contents of the dosimeter will be removed, if necessary stored in disposable glass tubes in a refrigerator, and within 24 hours analyzed by the pararosaniline colorimetric method. During the course of a week, 20 homes will be surveyed. Four passive dosimeters will be placed in each of three rooms within the home and an additional three dosimeters will be placed outside the home to measure outdoor levels. Blanks (unexposed dosimeters) will be prepared each day to provide a measure of quality control. Additional efforts will be made to determine the presence of any long-term variations in the sampling rates of individual dosimeters.

The Lawrence Berkeley Laboratory (LBL) Reference Method will also be used to check on the reliability of the passive samplers for quality assurance. At this time we will also test the 3M Brand Formaldehyde Monitor 3750 and the DuPont Formaldehyde Badges, Series II, Type C-60 to be provided by the Consumer Product Safety Commission.

On each of the formaldehyde monitoring surveys, indoor and outdoor maximum and minimum temperatures will be recorded as well as any other meteorological conditions that might effect the levels of formaldehyde within the home such as wind conditions. These meteorological data, as well as the formaldehyde monitoring data, will be summarized in the format shown as Attachment 3. Weather conditions will also be checked against official data from the National Oceanic and Atmospheric Administration (NOAA).

This monitoring team will perform the following types of measurements over the course of the study:

Measurement	Number/Home	Number/Time Period	Total Number*
Routine Formaldehyde Surveys			
Formaldehyde			
Passive Samplers	15	300/week	16,000
LBL Reference Method	1	2/month	24
3M	4	8/month	96
DuPont	4	8/month	96

Measurement	Number/Home	Number/Time Period	Total Number*
Organics			
Tenax	1	40/survey	160

*Does not include blank passive samplers or outside measurements of organics. This assumes a 12-month study with one complete survey during each of the four seasons.

The measurements of organics will be carried out using Tenax collection and subsequent analysis by gas chromatograph/mass spectrometry (GC/MS). The measurement will be made in one room of the house. An outdoor measurement will be made at the same time. An attempt will be made for the sake of economy to minimize the number of outdoor samples by choosing a single sample location centrally located to a number of homes in the study. Qualitative identification will be made of each of the organics found in the GC/MS analysis and selected critical organics will be chosen for quantification based on consideration of high ambient levels or high relative toxicity. The Tenax sampler will be operated for a 24-hour period.

HOUSE SURVEY FORM

Date ______________ Name of Participant ______________________

Telephone Numbers: (home) __________ (work) __________ Participant No. ____

Home Address ____

Own ☐ Rent ☐ How long at Residence ______________ years.

Number of Occupants ________________________________

Type of Home:

☐ Basement Rancher ☐ Split Foyer

☐ Story and One-Half ☐ Other ______________

Construction:

☐ Cemesto ☐ Frame/Brick

☐ Frame ☐ Block

Age ______ yrs. Living Area ______ sq. ft. Non-Living Area ______ sq. ft.

Garage:

☐ Attached ☐ Detached ☐ None

Basement:

☐ yes ☐ no

Heating:

☐ Heat Pump ☐ Gas ☐ Oil Heat

☐ Kerosene ☐ Wood Burning Stove ☐ Other __________

☐ Electric Heat ☐ Gas Space Heater

Air Cleaning Machine ☐ No ☐ Yes

Electrostatic Precipitator ☐ No ☐ Yes

Cooking:

☐ Electric ☐ Gas

Humidifier:

☐ Yes ☐ No

☐ Central ☐ Room Which Room(s) ____________

Insulation:

☐ Asbestos ☐ Fiberglass ☐ Styrofoam

☐ Cellulose ☐ Urea-Formaldehyde Foam (UFF)

If UFF:

☐ Retrofit

☐ New

☐ Age of Insulation ______________________________________

Vapor Barrier:

☐ Inside Wall ☐ Outside Wall ☐ None

Kitchen:

Size of Room ________________ sq. ft.

Carpeted ☐ Yes ☐ No Age __________ Type ______________

Paneled ☐ Yes ☐ No Age __________

Cabinets — Particleboard ______________ sq. ft.

Windows (number) ______________

Living Room:

Size of Room ________________ sq. ft.

Carpeted ☐ Yes ☐ No Age __________ Type ______________

Paneled ☐ Yes ☐ No Age __________

Particleboard ________________ sq. ft.

Windows (number) ______________

Study or Bedroom:

Size of Room _____ ____________ sq. ft.

Carpeted ☐ Yes ☐ No Age __________ Type ______________

Paneled ☐ Yes ☐ No Age __________

Particleboard ________________ sq. ft.

Windows (number) ______________

HOMEOWNER'S AGREEMENT

In this Agreement, made as of ______________________, 1981, between ______________________ and wife ______________________ residing at ______________________, County of ______________________, State of ______________________, and Union Carbide Corporation, Nuclear Division, with an office at Oak Ridge, Tennessee 37830, acting under its Prime Contract W-7405-Eng-26 with the United States of America as represented by the Department of Energy;

The recent trend toward making homes more energy efficient by using tighter construction techniques has resulted in the deterioration of indoor air quality. Indoor pollutants may be generated in the home from structural materials, furnishings, combustion sources, consumer products, and human activity. Residential air has been found to contain pollutants identified as formaldehyde, other organic chemicals used in the manufacture of plastics, inorganic chemicals, and particulates.

Union Carbide has been directed to conduct a study of the residential indoor air quality.

The objectives of this study are:

a) Identify various organic pollutants in the residential indoor air.

b) Determine the amount of selected organic pollutants along with formaldehyde, CO, NO_x, and particulates in older as well as newer built homes.

NOW, THEREFORE, the parties agree as follows:

Union Carbide will

(1) provide data collection equipment and install said equipment on participants' premises;

(2) maintain and service such equipment at its expense during the term of this agreement;

(3) periodically monitor the air on the premises;

(4) make available to the Participants the results of the data collected from the Participant's premises;

(5) assume responsiblity for any damage to Participants' property directly caused by its representatives monitoring the air or by the installation or removal of the data collection equipment, or for claims brought by third persons for personal injury or damage to property directly related to the installation of the equipment or monitoring of the air during the term of this agreement; and

(6) notify Participants at least 24 hours in advance of the date and date access to premises is desired.

Participants will

(1) permit representatives of Union Carbide reasonable access to Participants' premises for the purpose of sampling air indoors and outdoors;

(2) allow the installation and servicing of data collection equipment;

(3) allow Union Carbide to take photographs at Participants' premises; and

(4) allow Union Carbide to disseminate information collected to the public.

PARTICIPANTS

By ______________________________

Title ____________________________

Date _____________________________

By ______________________________

Title ____________________________

Date _____________________________

UNION CARBIDE CORPORATION
NUCLEAR DIVISION

By ______________________________

Title ____________________________

Date _____________________________

DATA SHEET FOR PASSIVE DOSIMETERS — H_2O

Participant Name ____________________

Date ______________________________

Investigator _______________________

Kitchen

Sampler #'s	Time Left:
__________	__________
__________	
__________	Time Picked Up:
__________	____________

Living Room

Sampler #'s	Time Left:
__________	__________
__________	
__________	Time Picked Up:
__________	____________

Bedroom or Study

Sampler #'s	Time Left:
__________	__________
__________	
__________	
__________	Time Picked Up:
__________	____________

Outdoors

Sampler #'s	Time Left:
__________	__________
__________	
__________	
__________	Time Picked Up:
__________	____________

Weather Conditions

At time samplers left: ☐ Sunny ☐ Rain ☐ Snow ☐ Cloudy

At time samplers picked up: ☐ Sunny ☐ Rain ☐ Snow ☐ Cloudy

Indoor temperature: Min. ____________ Max. ____________

Outdoor temperature: Min. ____________ Max. ____________

APPENDIX A

METHODOLOGY

Details of Routine Procedures

ONE-DAY SURVEYS

The function of this 2-person monitoring team is to conduct intensive measurements in one house at a time for one day. Each of the 40 houses will be visited four times to conduct these measurements. Both pollutant concentration and method validation measurements will be taken. Visits will be scheduled so that each house is sampled once during the spring, summer, fall, and winter seasons.

Initial Conditioning of House

Initially the house will be closed for two hours. This is to allow build-up of pollutant concentrations and the conduct of operations connected with measurements of air-exchange. Previous work indicates that build-up requires about 1 hour (see October-November, 1981, Monthly Report).

During the two-hour period, two one-hour NIOSH double impinger measurement and four (1/4-hr each) pumped molecular sieve measurements will be made to measure maximum formaldehyde build-up.

Air Infiltration

Concurrent with the battery of other pollutant measurements, the overall air infiltration rate for the house will be measured. The house will be closed up for two hours, all room-to-room doors opened, the air stirred thoroughly, and a tracer gas injected and thoroughly mixed. Freon-12, initially at a whole-house concentration of 20 ppm, will be used and the Freon concentration will be followed over time to measure air exchange. At the conclusion of the air-exchange measurement, door blower fans will be used to measure ventilation rates. This will also help to remove Freon-12 from the indoor air, as it could interfere with the use of the portable gas chromatograph.

Formaldehyde

On four occasions at each home, formaldehyde measurements will be made in two ways for the purpose of quality control. The standard NIOSH double impinger method with chromatropic acid and colorimetric analysis will be made to validate the readings obtained with pumped molecular sieve dosimeters. This process would be aimed at measurements taken in one particular room.

NO_x, SO_2, Particulates, CO_x

On four different occasions (spring, summer, fall, and winter) each home will be monitored for each of these pollutants during the course of the standard working day.

NO,NO_2. The chemiluminescence monitor will measure the outdoors, upwind NO_x concentration early in the morning when atmospheric NO_x concentrations are likely to be maximum. The indoors concentration will then be monitored in the living room (and/or kitchen) to obtain, with unattended operation, peak and integrated concentrations over a fraction or, if necessary, the total daily cycle of human activities. The instrument, with a 2 ppb low LOD, will be calibrated dynamically for NO_2 with NBS traceable permeation tubes and by producing known concentrations of NO_2 from NO and ozone using gas-phase titration. Calibration for NO will be done similarly.

SO_2. Those residences that use space heaters fueled by coal or kerosene will also be monitored for SO_2. The most prevalent form of fuel for these heaters is grade-2 kerosene, which contains a significant proportion of sulfur (0.3%), as does coal. DUVAS (Derivative UV-Absorption Spectrometer) will be used in these measurements. Some measurements will also be done in homes that do not use kerosene space heaters as a background check. Estimated LOD is 10 ppb.

Particulates. In conjunction with the NO_2 measurements, an early morning outdoor and 8-hour peak and average measurements of indoor particulate concentrations in one or more rooms will be made. The sensing stack of the analyzer has been fitted to a tripod for more stability and height adjustment, therefore, both standing and sitting measurements can be made. A ten-stage cascade impactor, quartz crystal microbalance will be used to separate and measure particles down to 0.05 μm. A microporcessor and printer will provide size distribution and mass concentration data automatically. If crystal loading of 300 Hz or greater is encountered, there will be reduced measurement accuracy. Therefore, a limit of 8 hours for measurement time is considered necessary.

CO, CO_2. Four small personnel CO (electrochemical) samplers will be used to provide a direct reading of ambient levels of CO down to 1 ppm concentration levels (1-3 ppm levels are typical for urban environments) which will be recorded intermittently every 30 minutes over a period of 7-8 hours. An output terminal is available for strip chart recording of fluctuations in CO concentrations. One meter will be placed in each of three rooms (kitchen, living, and bedroom), and one outside, upwind of the house. In the room where the strongest source of CO is present, the fluctuations in CO concentration will be monitored with a strip chart recorder or data logger. The CO monitor will be calibrated using a calibration system obtained from MDA Scientific, Inc. The calibration range will be set at 0 ppm and 60 ppm CO. Once the air-exchange measurements have been completed, the infrared analyzer will be free to measure CO_2 concentrations.

Radon

Two modified Wrenn chambers will be used to monitor ^{222}Rn. A selected number of homes will be monitored over an extended 4-day period, primarily because of the ingrowth of radon daughters. This extended period limits the number of homes that can be surveyed. The preferred location for the 2 chambers is the basement (or crawl space) and the main living room. The chambers have been calibrated by a radium source. Variations with humidity will be taken into account since weather conditions will be recorded. The lower limit of detection is 0.24 pCi/liter.

Organics

An attempt will be made, with a portable gas chromatograph, to locate in different areas of the house the major sources of some of the more prevalent organics identified by the Tenax sorbent. Sampling will be conducted in and around specific consumer products. These tests will be conducted as a special project in a time frame and with a frequency dictated by availability of manpower and time.

Meteorological Data

At the same time air-infiltration measurements are being taken, a number of other climatic parameters will be measured. These will include indoor and outdoor maximum and minimum temperatures and relative humidities, and wind direction and speed. Measurement will be continued throughout the day and checked against official data from NOAA.

ROUTINE FORMALDEHYDE AND ORGANICS SURVEYS

This measurement team (2 persons) has the primary task of conducting biweekly CH_2O monitoring and organics collection at each of the 40 homes. This task comprises driving to and from, and between, ORNL and 10 homes, on a given morning. Colorimetric analyses then have to be conducted on 61 samples (15 per house, plus blanks). Additionally, samplers have to be cleaned and prepared for the following day, results have to be logged and processed by computer, and homeowners have to be telephoned about the following day's visit.

Formaldehyde

Passive dosimeters will be used for integrated measurement over 24 hours. These will be of the semi-permeable, membrane-water type. Subsequent analysis is by the pararosaniline colorimetric method selective for formaldehyde.

Twenty homes per working week will be sampled using the following procedure:

Day 1: Samplers prepared at ORNL and placed in homes in Area A (a.m.). Measure sampling rates of 10 randomly-selected passive dosimeters using an environmental chamber. This test is to check the constancy of sampling rates for purposes of quality control.

Day 2: Area A — samplers picked up, the contents analyzed, and samplers cleaned (a.m.).
Area B — samplers prepared and placed (p.m.).

Day 3: Area C — samplers prepared and placed (a.m.).
Area B — samplers picked up, etc (p.m.).

Day 4: Area C — samplers picked up, etc (a.m.).
Area D — samplers prepared and placed (p.m.).

Day 5: Area D — samplers picked up, etc (p.m.). Typically, Day 5 will be a Friday, no samplers will be placed in the houses.

Each area will contain approximately five houses. Four passive dosimeters will be placed in each of three rooms plus three outside (15 dosimeters per house). Dosimeters will be placed on non-formaldehyde-emitting surfaces (such as metal or glass). In addition, six blanks will be prepared for each area of five homes. A total of 81 dosimeters is needed for each area of 5 houses. If the outdoor concentrations of formaldehyde in the cluster area prove to be uniform, then the number of outdoor measurements on a given day would be reduced to one at each of the five houses. In addition, if formaldehyde levels are uniform in the three different rooms, the number of rooms tested will be reduced to one at a later time.

To meet the above schedule of dosimeter preparation, cleaning, placement, and analysis, 3 sets of 81 devices have to be on hand for a total of 243 dosimeters. One set (81 samplers) will be used twice in one week. The set used twice per week will be rotated, so that all sets are used an equal number of times.

In addition to the house monitoring, spot checks will be made to determine long-term variations in the sampling rates of individual dosimeters (to be conducted the first day of each week).

For additional quality assurance, there will be multiple instrument monitoring of two homes, one old and one new, conducted monthly. The monitoring protocol will include 24-hour passive samplers and one Lawrence Berkeley

Laboratory (LBL) Reference Method run over a 24-hour period. Colorimetric analysis will be conducted with pararosaniline for the first technique. During this time the 3M 3750 Formaldehyde Monitor and the DuPont Formaldehyde Badge, Series II, Type C-60, will be tested. Four badges of each sort will be left in the vicinity of the LBL sampler. Since the LBL sampler is a modified small refrigerator, transportation problems will require the help of another staff member or senior staff member on those days when the measurements are performed. For quality assurance, four of each type of badge will be tested in an environmental chamber. It should be noted that the LOD sensitivity of the 3M monitor is 0.8 ppm-hours, or over 24 hours equivalent to 33.3 ppb formaldehyde concentration. For the DuPont Monitor, LOD is 2 ppm-hours or over 24 hours equivalent to 83.3 ppb formaldehyde concentration. Our passive samplers are sensitive down to 25 ppb formaldehyde concentrations.

Over the twelve-month period of house monitoring, there will be 16,000 samples taken using the passive dosimeters with a standard 24-hour sampling time. For quality assurance, there will be twenty-four 24-hour LBL Reference Method tests and 520 additional passive dosimeter tests. In addition, 96 tests for each of the 3M 3750 Formaldehyde Monitors and DuPont Formaldehyde Badge, Series II, Type C-60 will be performed during the course of the 24-hour LBL Reference Method.

Meteorology Data

On every visit, the indoor and outdoor maximum and minimum temperatures will be measured over the course of 24 hours.

Other weather conditions, such as whether it is raining, snowing, sunny, or cloudy and passage of frontal systems, will also be noted. Precipitation and its accumulation over the total monitoring period will be measured at one particular house for each of the three main clusters of homes, using portable rain guages. This will be done by the most cooperative homeowners, most probably ORNL staff members. Official weather data from local NOAA weather stations will also be obtained.

Organics

The objectives are three-fold: identify (qualitatively) various organic pollutants; quantitate the levels of organic pollutants (10 or more selected on the basis of prevalence, adequately measurable concentrations, and toxicity); and make correlations, based on variations between building products used in the construction of each house. These tasks shall be accomplished in the following ways.

Tenax Collection

Three Tenax samplers and pumps (two currently in use and a third is planned) have been supplied on loan by the Analytical Chemistry Division (ORNL). Nine homes per week maximum can be sampled until all 40 homes and outside measurements have been performed in any quarter of the year. This will coincide with the routine formaldehyde surveys. Although this may be modified for fewer homes in any given week, the following is a tentative schedule for maximum speed of collection.

Day 1: Area A* - Tenax samplers prepared and placed (a.m.).

Day 2: Area A - Tenax samplers picked up (a.m.)
Area B - Tenax samplers prepared and placed (p.m.).

Day 3: Area B - Tenax samplers picked up (p.m.).

Day 4: Area D - Tenax samplers prepared and placed (p.m.).

Day 5: Area D - Tenax samplers picked up (p.m.).

*Areas correspond to area A through D in the formaldehyde surveys. Because of logistics area C cannot be covered in the time available.

Tenax Analysis

Quantitative Analysis. Tenax collection and gas chromatography (GC) analysis will be used. Each house will be monitored once indoors (40 samples) during each season. One outdoor sample will be collected for each of the eight (5 house) clusters (8 samples). The 48 analyses will be performed by the Analytical Chemistry Division.

Qualitative Identification. Each house will be monitored once via Tenax collection and gas chromatography/mass spectral (GC/MS) analysis. One sample will be taken from indoors in each home (40 samples). One sample from each of the eight (5-house) clusters (8 samples) will be collected from outside. In all, 48 samples will be collected.

A total of 96 GC or GC/MS analyses will have to be performed per season. The sample time will be 24 hours with the sampler operating at 0.3 ℓ/min. Standard procedure for mass spectral analysis involves using the UCON-660 column. The samples are run using electron impact ionization with the gas chromatograph oven programmed from 40-160°C at 2°C/min with a two-minute initial hold.

Correlation with Consumer Products. Collection and measurement of organics is made preferably in a time period when the weather is hot and the houses are closed up and air conditioned. A June-August time period would be the most desirable for the summer quarter measurements. Vapor pressures and concentrations are likely to be a maximum at this time.

APPENDIX B

MAJOR INSTRUMENTS

The primary instruments used routinely in the indoor air survey are given in the following two tables.

Table I.

One-day Measurements	
Measurement	Instrument
Air Exchange	
Freon-12	1 Wilkes Model M80 Infrared Analyzer
Formaldehyde	
Molecular Sieve	3 DuPont P-2500 pumps
NIOSH	1 Turner Model 350 Spectrophotometer
NO_2,NO	1 Monitor Laboratories Model 8840 NO_x Analyzer 1 Permacal NO_x Calibration Unit 1 Linear Strip Recorder Model 142
SO_2	1 ORNL Prototype Derivative UV-Absorption Spectrometer (DUVAS)
Particulates	1 PC-2 Quartz Crystal Microbalance Cascade Impactor by California Measurements, Inc.
CO_2	1 Wilkes Model M80 Infrared Analyzer
CO	4 General Electric CO Detectors (EPA)*
Radon	2 Modified Wrenn Chambers (DOE)*
Organics	1 Photovac 10A10 Portable Gas Chromatograph 1 Hewlett Packard X-Y Recorder
Meteorological Data	5 Fans 1 Weather station (NOAA)* 1 Heath (Zenith) Strip Chart Recorder (NOAA)* 1 3-Cup Anomometer Model 12102D Photochopper Type, R. M. Young Co., Gill Design (NOAA)* 1 R. M. Young Co., Gill Design Microvane Model 12302, wind speed and direction (NOAA)* 1 Sylron/Taylor Rain Gauge (5") No. 2702

*Equipment supplied on loan from this agency.

Table II.

Routine Formaldehyde and Organics	
Measurement	**Instrument**
Passive Sampler formaldehyde	1 Turner Model 350 Spectrophotometer
Lawrence Berkeley Laboratory Method formaldehyde	1 LBL refrigerated air-sampling unit on loan from Lawrence Berkeley Laboratory
Meteorological Data	12 Stortz MMT-15 Max-Min Thermometer 3 Sybron/Taylor Rain Gauge No. 2702
Organics gas chromatograph mass spectral analysis	3 Tenax Samplers (2,6-diphenyl-p-phenylene oxide resin) 3 DuPont P-4000 pumps 1 UCON 50HB660 GC/column qualitative 1 Hewlett-Packard 5895 GC/MS 1 Perkin-Elmer Model 900 GC quantitative

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