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Measurements of Indoor Toxic VOC Concentra- tions Attributed to the Residential Storage of Household Products

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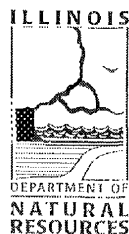
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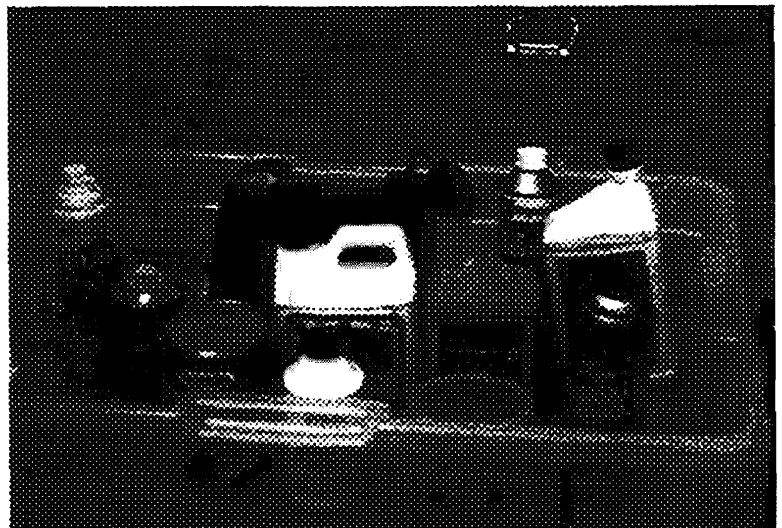


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ABSTRACT

Breathing indoor air is an important exposure route and human health risk factor for many toxic chemicals. In this study, the concentrations of toxic volatile organic compounds (VOCs) generated by the storage of hazardous household products (HP) were measured in an experimental test house and in three actual residences. Experiments in the test house indicate that high concentrations (>100 ng/L) can develop in areas where HP are stored and in attached living areas. The concentrations of individual VOCs in the residences were influenced by stored HP but were also affected by differences in air infiltration rates, temperature, volatilization from building materials, and the presence of other sources such as freshly dry-cleaned clothes or emissions from showers. When infiltration rates in the test house were matched to those measured in the actual residences, the range of measured toxic VOC concentrations was similar. It was concluded that storage and use of HP in homes and in attached storage areas can cause a significant elevation in the levels of toxic VOCs in indoor air and that the test house reflects conditions found in homes in Illinois. It is recommended that the amount of HP stored in the home be minimized by purchasing only the amount of product needed for a particular job. If HP materials must be stored, it should be in an area separate from the living area with high infiltration rates.

EXECUTIVE SUMMARY

Introduction

Research has shown that residential indoor concentrations of toxic volatile organic compounds (VOCs) are up to an order of magnitude higher than outdoor concentrations. It is estimated that Americans normally spend 90% of their time indoors so that most of the health risk from VOCs is from exposure to indoor air. The storage of hazardous household products (HP) in a residence may be a substantial source of indoor air pollution. According to curbside refuse surveys, up to 1% of household waste is household hazardous waste (HHW). For many households the preferred "disposal" option is long-term storage. The objective of this project was to investigate stored HP as a source of VOCs in indoor air.

The sources of indoor air pollution are numerous and complex; but, in general, can be categorized as: (1) building materials and furnishings; (2) use of household products; and (3) automobiles and related products. This study concentrates on automotive and household maintenance products and VOCs generated from these products. Indoor VOC concentrations have been evaluated in test houses with infiltration rates representative of those in houses in Illinois. For comparison, measurements were also made in 3 actual residences.

Test House

The test house used for this work is an experimental structure operated by the Building Research Council at the University of Illinois. The work was carried out in two 8' x 20' x 8' rooms that were individually climate controlled. One room was an unheated exterior room that simulated an attached garage for our study while the second interior room simulated a living area. Air exchange between the rooms and with the outside was regulated using adjustable ports. During the experiments, temperature and air flow were monitored in the house along with outside conditions.

A standard blower door test method was used to determine infiltration in units of air changes per hour (ACH) in the test house and in actual residences. For the first test period, the test house infiltration rate was 0.2 ACH, equivalent to the best currently used energy efficient construction technology. This represents a "worst case" situation for indoor air pollution because a minimum of outside air passes through the house to dilute emissions of VOCs. The actual residences had infiltration rates between 0.3 and 1.4 ACH. These values are typical of the range of infiltration rates found in homes in Illinois. In a second round of experiments in the test house, infiltration rates were set between 0.6 and 1.2 ACH to better reflect average "real world" conditions.

Methods

The HP were introduced into the test house in the "garage". The HP consisted of standard containers of automotive products (gasoline, carburetor cleaner, brake fluid, etc.) or household maintenance products (cleaning products, paint, paint stripper, caulking, adhesives, etc.). The containers were opened then resealed and placed in a plastic tub for the experiments. In the first phase of the work, both HP groups were evaluated. Only automotive products were tested in the later phases because the household maintenance products were found to emit lower amounts of mostly the same VOCs.

Sampling was carried out using 6L Summa-polished stainless steel canisters. Time-averaged, 2-hour samples were collected using a 30 gauge needle as a critical orifice. VOCs in the air samples were cryogenically concentrated in a liquid nitrogen trap and analyzed by gas chromatography using simultaneous electron capture and flame ionization detection. The target compounds were 15 toxic VOCs including several polychlorinated solvents, benzene, and freon.

Test House - Phase I

During the first part of this work, the test house was in a "tight" configuration with minimum infiltration rates (0.2 ACH). VOCs reached maximal concentrations much above background levels in the garage within 5 hours. Concentrations of VOCs in the living area were only slightly above those in outdoor air. When small ports between the garage and living area were opened to allow air exchange between the rooms, levels of VOCs in the living area increased well above those in outdoor air.

In the next part of the work, the infiltration rate was left at 0.2 ACH and the ports between the rooms in the test house were left open. Both automotive and household maintenance HP were stored in the garage for 16-day periods. Under these conditions, VOC concentrations exhibited one of three patterns. Some VOCs reached peak concentrations of up to 1000 ng/L or more in the garage section of the test house within 24 hours, and then slowly declined over a 16-day period. Concentrations of these VOCs in the living area followed the same trend but were initially about half as much as those in the garage. Other VOCs including carbon tetrachloride, chlorobenzene, trichloroethylene, and chloroform varied with outside air levels indicating the products tested were not significant sources for these chemicals. Finally, concentrations of freon were consistently higher in the living area of the test house even before the introduction of the products. This pattern indicates a source in the living area of the test house (air conditioner). Concentrations of the VOCs emitted by the stored HP usually reached a plateau within the first 4 days of the storage period and subsequent fluctuations were attributed to changes in temperature.

Test House - Phases II and III

During the next part of this work, the infiltration rate through the test house rooms was increased to 0.56 and 1.2 ACH by opening additional ports to the outside. The opening between the garage and living area remained the same as in the previous experiments. The intent was to determine the influence of an increased infiltration rate on the movement of VOCs from the garage to the living area and the relationship of

infiltration rate to the overall concentration of VOCs that builds up in the test house. As before, the automotive product mix was placed in the garage simulating the storage of partially used automotive products. The maximum VOC concentrations measured in the garage reached approximately half of comparable levels measured with the tighter configuration (0.2 ACH). VOCs in the living area were decreased by up to 50% at 0.56 ACH and by 90% at 1.2 ACH. It is apparent from the data that a larger infiltration rate limits the buildup of VOCs in both the garage and living area.

Tests in Actual Residences

VOC concentrations in 3 actual residences were measured. These houses had infiltration rates that reflected the range found in Illinois housing. A new house was tested before and after the introduction of the household maintenance product mixture. Although the home with the highest infiltration rate generally had lower concentrations of VOCs, there was not a consistent difference. The pattern of VOC concentrations indicates that there were sources other than stored HP including building materials, tap water emissions (showers), and freshly dry-cleaned clothes. In some cases, HP clearly contributed to the VOC levels measured in the residences especially in the rooms where the HP was stored and used.

Conclusions and Recommendations

The test house used in this work allowed for the evaluation of the impact of stored automotive and household maintenance hazardous products (HP) on concentrations of volatile organic compounds (VOCs) in indoor air without the confounding variables encountered when making measurements in actual homes. Infiltration rates from the outside and exchange between the rooms in the test house could be adjusted to values representative of typical homes in Illinois. To the extent that indoor air quality is dependent on dilution of toxic materials through natural ventilation, then the concentration of these materials must be maintained low enough to ensure indoor air quality at infiltration rates of 0.5 air changes per hour or less.

It is evident from this work that the storage of partially used HP contributes to elevated concentrations of many VOCs that may pose a human health risk. Automotive products are associated with emissions to indoor air of benzene, styrene, m,p,o-xylenes, methyl ethyl ketone, toluene, and ethylbenzene. Household maintenance products are associated with elevated levels of methylene chloride, methyl chloroform, toluene, ethylbenzene, and m,p-xylenes. In both the test house and in actual residences, storage of partially used HP can result in a rapid buildup of VOCs (up to 1000 ng/L) in the rooms used for storage.

In the test house, increases of VOCs in the "living area" were associated with product sources in the "garage" when a connection between the rooms was made. VOC levels were inversely related to infiltration rate with lowest VOC concentrations corresponding to the highest infiltration rate. VOC concentrations peaked one or two days after the product introductions in the garage. VOC concentrations in the living area rose more gradually, approaching the level in the garage after about 4 days. These concentrations were often 100 times greater than initial background levels. Increases in the VOC levels in the living area could be minimized by limiting the exchange of air between the garage and living area.

In actual homes, the concentrations of some VOCs in indoor air were related to the storage of partially used containers of automotive and household maintenance products. For other VOCs, alternate sources were probably responsible for the elevated levels found in indoor air. The highest levels of VOCs were found in the areas where the products were actually stored or used. Where air exchange between the storage area and living areas was limited, increases in VOC concentrations attributable to stored HP were minimized.

Recapping a container of many automotive and household maintenance products does not prevent the escape of many toxic VOCs to the air. To minimize the buildup of toxic VOCs in residential indoor air, the storage of used (i.e. seal broken)

products in a garage, basement, or other storage area within a residence is not advisable. Ideally only the amount of a product needed to do the job should be purchased. If extra product does need to be stored, it should be in a separate building from living areas. If products must be stored in the same building with the living areas, either low air exchange between the living and storage areas or high overall infiltration rates are needed to prevent VOCs from building up in the living area. In any case, elevated concentrations of VOCs can be expected in the storage area especially when ambient temperatures are high.

INTRODUCTION

Indoor air quality is receiving increased attention from researchers and public health officials as a critical component of the total exposure of humans to a variety of air pollutants. Indoor air quality is not a reflection of outdoor conditions. Research has shown that residential indoor concentrations of toxic volatile organic compounds (VOCs) are up to an order of magnitude higher than outdoor concentrations (Wallace *et al.*, 1985; Parkhurst *et al.*, 1988; Lewis *et al.*, 1988). The impact of these concentrations on human health are compounded by the large proportion of time spent indoors, evidenced by the ability of Lewis *et al.* (1988) to show good agreement between residential indoor air quality and personal air exposure monitoring. It is estimated that Americans normally spend 90% of their time indoors (Dockery and Spengler, 1981) and, for most individuals, the largest fraction of that time is spent within a residence.

The 1984 Hazardous and Solid Waste Amendments to the Federal Resource Conservation and Recovery Act (RCRA) and section 22.19 of the Illinois Environmental Protection Act recognize homes as sources of hazardous materials to the waste stream from the disposal of hazardous household products (HP). These materials are referred to as household hazardous waste (HHW) when they are thrown away. The amounts of hazardous HP used, stored and thrown away are difficult to determine. Estimates of 0.5 to 1% by weight of curbside refuse (Tufts University, 1988), and annual disposal of 10 containers per household (Liebert, 1988) and 35 pounds per household (Albuquerque, 1986) have been reported. While actual quantities are difficult to pin down and definitions of HHW vary, even the most conservative estimates of HHW are considered "substantial" (Tufts University, 1988). The interest in HHW generally has been in its proper disposal to prevent environmental contamination (IEPA, 1989). Furthermore, the dangers of fire, explosion and poisoning have been recognized. Efforts have been made to educate the public in the safe handling of these materials and to suggest the use of safer alternatives (i.e. HWRIC brochures TN88-008a and TN88-008b). A number of

communities across the United States and in Illinois have participated in HHW collection and disposal programs. These programs have only been able to attract HHW from about 1% of households (USEPA, 1986; IEPA, 1989). Only 0.7% participation has been reported for Champaign County (Liebert, 1988).

In addition to a threat of improper disposal, the storage of HP in a residence may be a substantial source of indoor air pollution. A public information document from the U.S. Environmental Protection Agency (USEPA, 1986) advises homeowners to throw away any partially full containers of hazardous HP to lower concentrations of organic chemicals in the home. Storage is a management option for many people. A survey in Champaign county found that the majority of households did not dispose of hazardous HP in any way and that up to two thirds of containers collected in HHW programs were taken from storage rather than being diverted from improper disposal (Liebert, 1988). The degree to which the household storage of HP may affect the health of the occupant has not been well characterized (USEPA, 1986). A first step is to identify and quantify the contributions of stored HP to indoor air pollution. The objective of this research was to determine the influence of stored HP on indoor air quality. We hope this information will help define HHW and will be useful in promoting and increasing participation in HHW programs.

The sources of indoor air pollution are numerous and complex; but, in general, can be categorized as: (1) building materials and furnishings; (2) use of household products; and, (3) automobiles and related products. If these products are not properly disposed of after use, they can continue to contribute to indoor air pollution. Emission factors for some of these sources have been evaluated in test chambers (Tichenor and Mason, 1988) allowing model calculations of indoor VOC concentrations. We have concentrated on household and auto-related products and VOCs generated from these products. In this work, indoor VOC concentrations generated by HP have been evaluated in test houses with conditions representative of those in actual residences. The rationale for using a test house rather than an actual

residence is that the construction materials, household product use, and lifestyles in lived-in residences are highly variable and difficult to control. The use of a test house allows us to reproduce precise conditions of air exchange that can be related to conditions in actual residences.

ENGINEERING STUDIES

Simulation in Test Cells under Natural Conditions

The Building Research Laboratory building was constructed for field testing of building assemblies. This "test house" contains ten study bays, which were constructed for uniformity of workmanship. Each bay measures 8' (2.3m) by 20' (6.0m) in surface area. The ceiling height is 8' (2.3 m) and the interior volume is 1280 ft³ (31.8 m³). The building is on a crawl space foundation. Materials and workmanship are similar in all of the bays.

The aim in using the test house was to have a simple test system with air exchange similar to that in actual houses. In this work, bay 9 was considered the "garage" and bay 8 was considered the "living area" (figure 1). Each bay had openings to the outside, which were 3" (7.6 cm) diameter PVC tubing. To reduce excessive air flows through the tubing, fiberglass insulation buffers were added. There was also 1.5" (3.8 cm) diameter PVC tubing between bays 8 and 9 with fittings that permitted the size of the opening to be reduced in 0.5" steps down to 0.5" (1.3 cm) in diameter.

The bays were individually heated, cooled, and humidified. Bay 8 was maintained at 70°F (21°C) during the winter and 75°F (24°C). Bay 9 was not heated or cooled. Temperature was monitored in the 2 bays, the crawl space, and outdoors. Airflow measurements through the port openings and air pressure difference measurements between the bays were also taken. Weather data (outdoor temperature, wind speed, wind direction, and solar insolation) were collected throughout the study from a weather station located at the Building Research Laboratory. Figure 2 provides an example of a typical day's weather data.

Infiltration is the uncontrolled flow of air through unintentional openings driven by wind, temperature difference, and/or appliance-induced pressures across the building shell. Air leakage is a measure of the air tightness of the building shell, and

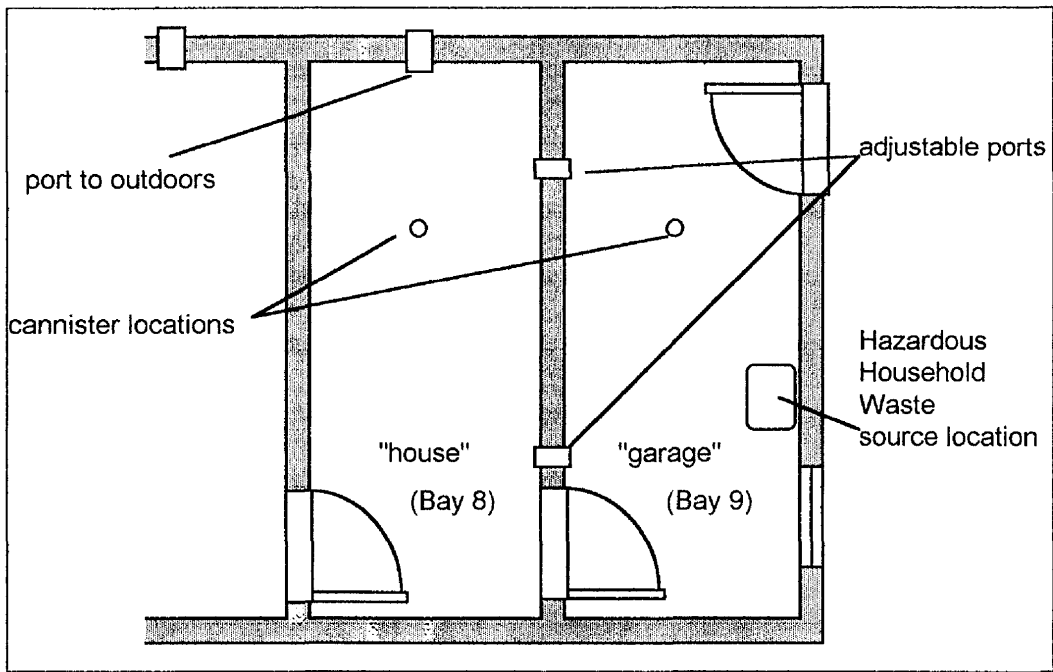


Figure 1. Schematic of Bays 8 and 9, Building Research Council Test House

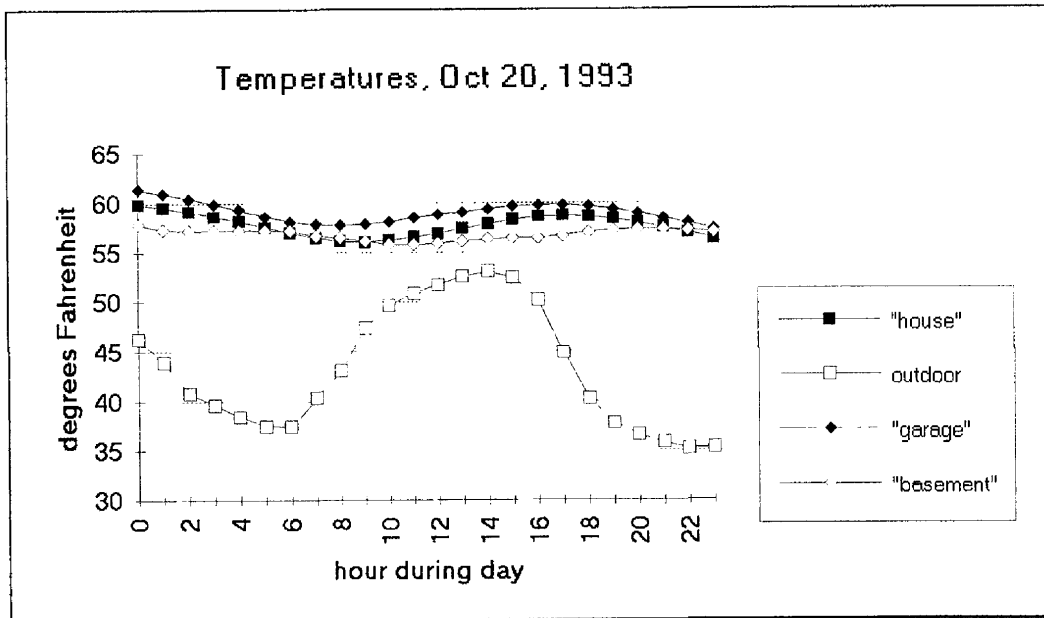


Figure 2. Test House Temperatures for Oct. 20, 1993

is determined by the total area and placement of the unintentional openings. Greater air leakage area causes a higher infiltration rate. Infiltration, the air entering the structure, is always equal to the exfiltration, the air leaving the structure. A useful measure of infiltration is air changes per hour (ACH).

Bays 8 and 9 of the Research laboratory were tested for air infiltration using a blower door test and calculating an equivalent leakage area (ELA) and infiltration rate (ACH). With ports to the outside closed, both bays had measured air exchange rates of 0.2 ACH. The only evident air leakage paths were those used for routing instrument cables. This configuration was used for the initial tests.

Later in the project, additional holes were installed between the outdoors and Bay 8. The purpose of these holes was to allow for configuring the test house to approximately replicate the air change rates found in actual homes in Illinois. An additional 3" PVC pipe was installed in the north wall, and two 3" PVC pipes were installed in the south wall. Plastic inserts, with holes of varying diameter in the center, were fabricated to fit within the 3" PVC pipes. Bay 8 was then tested for ACH under different hole configurations using a blower door test. The selected hole configurations for various phases of the work are shown in Table 1.

Table 1. 1994 Test Building Configuration

Test Number	Bay 8 Configuration	Infiltration Rate	Corresponding Residence
Phases I	no holes	0.20 ACH	Residence #1
Phases III	3" hole, north and south	0.56 ACH	Residence #3
Phases II	two 3" holes, north and south	1.20 ACH	Residence #2

Although the test house can be calibrated and tested to provide an estimated infiltration rate, there exists a qualitative difference between the test house and typical field conditions. The total air leakage area in a residence is comprised of many small

openings and cracks associated with doors, windows, wall penetrations, etc. Air change rate is dependent on wind conditions and direction, and the placement and balance of the leakage sites. Clearly, the grouping of leakage sites into two or four larger opposing openings, as compared to numerous small, randomly placed openings, presents a qualitatively different condition for interaction with wind conditions.

Blower Door Method of Analysis of Air Leakage

Tests were performed to determine the infiltration rates of the residences and various configurations of the test house. The test method was a blower door analysis. Blower doors have been in use since the 1980's. The equipment has been steadily refined but continues to exhibit certain limitations. Accuracy may vary as much as 8% between doors and windy conditions will cause fluctuations in the readings. Testing was conducted using the same blower door in all instances, thus eliminating the potential discrepancy between fans. Blower door tests on the test house bays were done during periods with low wind speed (2 km/hr) to avoid the effects of wind gusts on the readings.

When testing a residential structure for air tightness the following protocols are used. All exterior windows (including storm windows) and doors are closed. The home should be in the same state as it normally would be for the winter/heating season. The heating system and water heating system is shut off or set so that they will not cycle during the infiltration test. Interior doors to all rooms, floor levels, spaces and closets are opened. This allows for the identification of air leakage sites throughout the building. Exhaust fans, clothes dryers, and other devices designed to exhaust air from the building were shut off. Fireplace dampers and/or glass doors are placed in a closed position.

The device used in these tests was a Minneapolis Blower Door Model 3 (The Energy Conservatory Inc., Minneapolis, MN) with three concentric low-flow rings. Flow is adjusted by adding or removing these rings. Air flow from 100 to 6000

$\text{ft}^3\text{min}^{-1}$ could be measured. The blower door was mounted in an exterior door opening. A nylon door panel covered an expandable aluminum frame which fitted with and sealed the existing door opening thus providing a tight seal at the test location. A large variable speed fan was mounted in a opening provided in the nylon door panel.

The equipment and associated diagnostic software give results in a variety of formats. Testing may be done as a single point test or as a multi-point test to improve accuracy. The equipment is set to depressurize the building, thus accentuating the infiltration points. A multi-point depressurization test was used to measure the pressure of the house and fan at five to six different pressures. Readings begin when the house or bay space has been depressurized to between 50 and 60 pascals below the exterior pressure. The pressure difference is changed by 5 to 10 pascals by slowing the fan motor (thereby pulling less air through the holes, cracks and crevices) and allowing the house to stabilize at the new pressure. Readings were taken of the new house pressure and the corresponding fan pressure at 50, 45, 40, 35, 30 and 25 pascals.

The infiltration rate (ACH) of the space being evaluated corresponds to the air flow in cubic feet per hour divided by the volume of the space. This value is easily calculated from the blower door test and is commonly used for the sizing of mechanical equipment, air-to-air heat exchangers, fans, and ducts. ACH can be easily compared from building to building.

An estimate of the natural infiltration was made using protocols (ASHRAE, 1993) which were developed by Lawrence Berkeley Lab (LBL). The Effective Leakage Area (ELA) estimates the natural infiltration area in square inches when the difference between the interior and exterior pressures is 4 pascals. The 4 pascal value is a realistic approximation of what might occur in a residential structure due to exterior wind loads, stack effect, and duct leakage (mechanical system interference).

This value is related to the area of the building envelope (area of the shell of the building) and generally can not be readily compared between buildings. For example, 100 square inches of leakage area for a large home may be too much while 100 square inches in a small home may be too little. ELA is also a useful number in evaluating the success rates for air sealing operations relative to before and after blower door testing. ACH can be calculated from the ELA and the volume of the building. Table 2 gives physical dimensions, ELA and ACH for the test bays and the 3 residences evaluated in this work.

Table 2. Configurations and Characteristics of Residences and Test House Bay

House or Bay	Area ft ²	Shell ft ²	ACH	ELA cm ²	ELA % of Shell
Residence 1	1219	3961	0.336	48.64	1.23%
Residence 2	4074	7293	1.400	454.81	6.24%
Residence 3	1600	4588	0.621	97.45	2.12%
Bay 8, Phase I	160	860	0.200	4.92	0.58%
Bay 8, Phase III	160	860	0.560	8.29	0.96%
Bay 8, Phase II	160	860	1.189	29.65	3.45%

The three residences had air change per hour readings of 0.34, 0.62, and 1.4. There was no home in this study equal to or less than 0.2 ACH, which is typical of new construction which has been designed and built by contractors with a primary interest in energy efficiency. ASHRAE (1993) recommends mechanical ventilation for homes which have air change rates of less than 0.35 ACH. The range of infiltration rates produced in the test house approximates the range of ACH values found in the Illinois housing stock (see next section) and are similar to the ACH values measured at the 3 residences evaluated.

Infiltration Rates for Single Family Housing in Illinois

To date, North American single-family housing design has depended on

uncontrolled infiltration to provide the necessary ventilation to ensure indoor air quality for the occupants. The average house today, however, is tighter than houses built in the past. Energy saving products such as sill seals, house wraps, and weather-tight windows and doors have become standard. Some changes in materials such as the use of plywood instead of board sheathing have tightened the typical building shell. The trend to year-round climate control in houses with central air conditioning has increased the proportion of the year that the typical house is closed up and the occupants are dependent on infiltration of fresh air through the shell. Finally, the national focus on energy efficiency in the last two decades has resulted in tighter building shells in many older homes through retrofit procedures such as weather stripping and caulking.

Infiltration rates of individual buildings depend on weather conditions, topography, construction quality, equipment operation, and occupational activities. The rate can vary by a factor of five from weather effects alone (Malik, 1978). The location, size, and balance of air leakage sites (wall penetrations, windows, doors, etc.) also plays a major role in determining infiltration rate.

Typical infiltration rates in housing in North America vary by at least a factor of ten, with "tight" houses at around 0.2 ACH, and "leaky" houses at 2.0 ACH or more. ASHRAE (1993) cites data from several studies, including one giving the average seasonal infiltration of 312 houses located in different areas of North America (Grimsrud et al., 1982). These houses had a median infiltration rate of 0.5 ACH. Other data on infiltration rates in 266 houses located in 16 cities in the United States (Grot and Clark, 1979) showed a median infiltration of 0.90 ACH. The Northwest Residential Infiltration Survey found a heating season average of 0.40 ACH (range 0.13 to 1.11 ACH) in 134 houses in the Pacific Northwest (Palmiter and Brown 1989, and Parker et al., 1990). In a nationwide comparison of 292 houses incorporating energy efficient features to 331 control houses with typical construction features, Parker et al. (1990) found an average infiltration rate of about 0.25 ACH (range: 0.02

to 1.63) for the energy efficient houses versus a rate of 0.49 ACH (range: 0.05 to 1.63) for typical houses.

The Building Research Council at the University of Illinois operates the EnergyWise Homes program, a residential energy rating program sponsored by the Illinois Department of Energy and Natural Resources. Through this program, the infiltration rates of 160 houses in Illinois have been measured. Measurements are made using blower-door leakage tests, and combined with the LBL infiltration model discussed earlier. Infiltration rates are expressed as equivalent leakage area as well as air changes per hour.

A breakdown of the infiltration rates of the 160 house sample is shown in Figure 3. The average infiltration value is 0.54 ACH, and the median value is 0.45 ACH. The distribution pattern is similar to the other studies with one exception: there are considerably more houses in this sample with very low ACH (< 0.25). This is likely the result of a natural bias toward energy efficiency in the sample. The EnergyWise Home program is a service to Illinois residents, and the tested houses do not comprise a random sample. Rather, the energy ratings are performed upon the request of homeowners who have heard of the program and request an energy audit. They may represent a more energy conscious subset of all homeowners with houses that have received significant attention to the tightness of the building envelope prior to testing. Additionally, because EnergyWise Homes is a rating program, the sample contains houses that were tested at the request of contractors who consider a good rating a selling point.

A comparison of houses built before 1970 to those built post-1970 divides the sample exactly in half (80 houses in each category). The newer houses are only slightly tighter than the older houses, with an average ACH of 0.53 as compared to 0.55 ACH. The median value of the older houses is 0.49 ACH and the median of the newer houses is 0.45. The difference is primarily in those houses that tested very

tight, (< 0.25 ACH). Again, the probable bias towards energy efficiency likely effects these results.

The results of these surveys indicate that a large percentage of houses have fairly low infiltration rates. Infiltration rates between 0.25 and 0.5 ACH are common in both new and old residential construction. To the extent that indoor air quality is dependent on dilution of toxic materials through natural ventilation, then the concentration of these materials must be maintained at a level low enough to ensure indoor air quality at infiltration rates of 0.5 ACH or less.

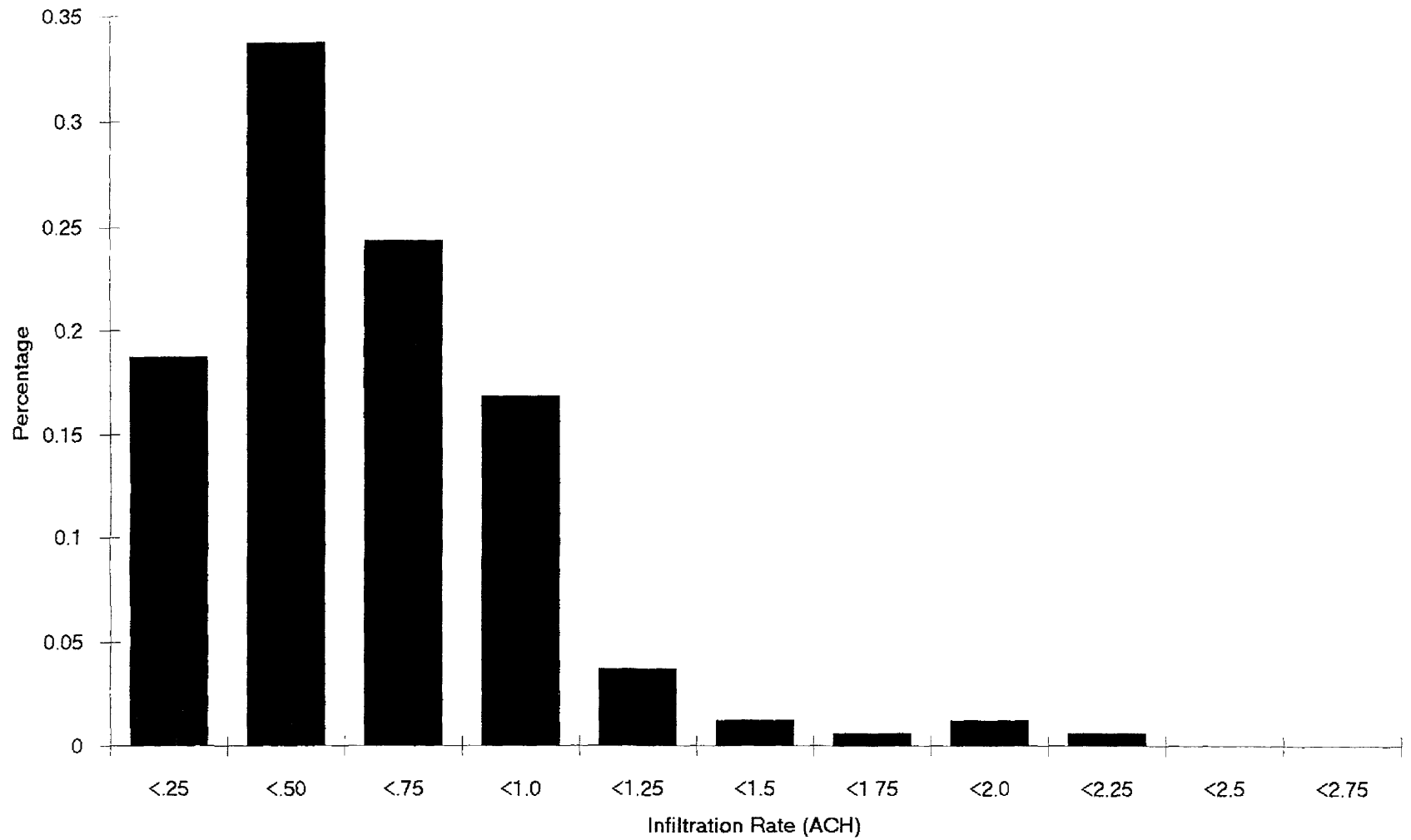


Figure 3. Infiltration Rates in Illinois Houses - BRC 160 House Database

METHODS

Hazardous Household Products and Test House

Hazardous household products (HP) typically stored in residences were purchased from a local automotive and department store in standard size commercial packaging. Prior to their introduction into the test house or residence, the HP was placed in a plastic tub. This was done to prevent accidental spillage and possible contamination of the test house or residence. Two type of product mixes were introduced: an automotive mix and a household maintenance product mix (Table 3). In addition to choosing HP typically stored in a residence, consideration was also given to the presence of VOCs in the product. Generally these were not identified by the product labels (see Table 3) and we relied on the previous work of others to characterize the range of VOCs found in these products (Calabrese and Kenyon, 1991; Sack *et al.* , 1992). VOCs are defined as compounds with a vapor pressure greater than 10^{-4} mm Hg, and thus can be found in the gas phase under ambient conditions. A list of targeted VOCs is given in Table 4. The choice of target compounds was also influenced by the capabilities of our analytical system.

The test house was set up for the desired infiltration rate (see table 2) and ventilated until all of the measured VOCs were at the concentrations measured in the outside air before placement of the HP in the test house "garage" (bay 9) and beginning the sampling.

Sampling Methods

Sampling was performed using standard 6.0 liter Summa-polished stainless steel canisters (SIS Inc., Moscow ID). These canisters are capable of holding air samples for up to 30 days with no significant change in the concentrations of target VOCs (Oliver and Pleil, 1986) Prior to sampling, each canister was thoroughly cleaned using repeated flushing with humidified zero grade air in a sealed system. Once cleaned, the cans were analyzed for contamination before release for sampling and recleaned if contamination was present.

Table 3. Hazardous Product Identification, Amounts, and Identified VOCs

Automotive Products	Size	Target VOCs Identified on Labels	
octane booster	8 oz.	xylene, ethylbenzene	
rust remover	12 oz.		
bug and tar remover	16 oz.	styrene	
body filler	32 oz.		
gasket remover	12 oz.		
transmission fluid	32 oz.	methylene chloride	
carburetor cleaner	16 oz.		
automotive grease	16 oz.	toluene, methyl ethyl ketone	
chrome cleaner	12 oz.		
unleaded gasoline	128 oz.		
brake fluid	12 oz.		
radiator protector	128 oz.		
motor oil	32 oz.		
Household Maintenance Products			
latex paint	128 oz.		methylene chloride, toluene
paint stripper	32 oz.		
caulking	11 oz.		xylene, methylene chloride methyl chloroform
adhesive remover	32 oz.		
formica cement	32 oz.		
metal primer	32 oz.		
polyurethane varnish	32 oz.		
mineral spirits	128 oz.		
spackling compound	16 oz.		
water sealant	128 oz.		
water putty	16 oz.		
roof cement	32 oz.		
primer/sealer	32 oz.		

Table 4. Targeted VOCs and Associated Toxicity Profiles

Selected VOC	Toxicity Profile^a
benzene	carcinogen, developmental toxicity
toluene	central nervous system dysfunction <i>fatigue and headaches</i>
methylene chloride	carcinogen and developmental toxicity <i>fatigue and light headedness</i>
trichloroethylene	carcinogen and liver damage <i>headaches and fatigue</i>
chloroform	carcinogen, developmental toxicity
carbon tetrachloride	carcinogen
tetrachloroethylene	carcinogen <i>dizziness, headaches, and eye irritation</i>
chlorobenzene	carcinogen, liver and kidney effects <i>headaches</i>
methyl chloroform (1,1,1-trichloroethane)	no known effects <i>light headedness</i>
styrene	carcinogen <i>irritation to eyes, nose, and skin</i>
o,m, and p-xylene	developmental toxicity <i>irritation to eyes, nose, and throat</i>
ethylbenzene	liver and kidney effects <i>irritation to eyes and skin</i>
methyl ethyl ketone (2-butanone)	developmental toxicity <i>irritation to eyes and respiratory tract</i>
freon 12	none reported

a) It is yet to be determined whether low levels of VOCs found in residences cause health problems. The first line refers to effects from higher doses than likely from residential indoor air. The second line (italics) refers to symptoms that might be experienced from VOCs emitted by HP.

Shortly before the sample was taken, the canisters were evacuated to between -24 and -25 inches of mercury via a sealed, cleaned, vacuum line. The valves were then closed. The valves were fitted with a sampling inlet consisting of a GC septum pierced by a 30-gauge needle. The needle acted as a critical orifice and provided a constant flow of about 30 mL/min for up to 2 hours when the valve was opened.

Outside air samples were taken upwind of the house being sampled, and when possible, out of the sun or rain. Indoor samples were taken in both test house bays and at selected locations in the residences. Once the canisters were in place, the sampling valves were fully opened, then backed off about 1/4 turn. The sampling needle function was checked by placing a finger over the collection orifice. Plugged needles were replaced and rechecked. The canisters were then left for approximately 2 hours to collect a time-integrated sample.

At the end of the sampling period, the valves were closed and the canisters were returned to the laboratory for processing. The canister was allowed to come to room temperature (25°C) before analysis and measurement of sample volume. The transfer lines were flushed with zero air, and the canister was hooked up to the system. The remaining vacuum in the canister was measured for calculation of the actual volume of air that had been sampled. The canister was then pressurized to 15 psi using zero grade air. The sampling valve was closed, and the sample was ready for analysis.

Analytical Methods

The analytical method was cryogenic preconcentration (USEPA TO-14) as described by McClenny *et al.*, 1991. Analysis of the samples was performed on a Hewlett-Packard 5890 gas chromatograph (GC) equipped with a cryogenic trap, subambient cooling, a gas sampling valve and flame ionization (FID) and electron capture (ECD) detectors. The FID is a general purpose detector with moderate sensitivity for all organic compounds. The ECD is a selective detector with high sensitivity to polyhalogenated compounds and low sensitivity to most other organics.

A 30-m, 0.325-mm-i.d., fused silica capillary column with a 1.0- μ m-thick film of DB-5 bonded liquid phase (J&W Scientific, Rancho Cordova, CA) was used. The cryogenic trap was a 20 cm length of nickel tubing bent into a loop that could be immersed in a Dewar containing liquid nitrogen. Data from the GC was collected electronically through a Nelson Analytical analog to digital (A to D) converter to a two pen strip chart recorder and a computerized software package with analytical capabilities. At a later time the A to D converter was replaced by a commercial software package, FrontRunner2, which performed both functions more efficiently.

At the start of an analytical cycle, the entire system of transport, sampling and trap lines was flushed with zero grade air for several minutes at the purge temperature of approximately 80°C. The sample canister was attached to the sampling loop while pressurized zero grade air was still on, this prevents contamination from laboratory air. Once attached, the pressurized air was shut off, and the line pressure was allowed to drop to the level of the canister pressure. At this point, the sample valve on the canister was opened; and the vacuum source was allowed to pull sample air through the sampling loop and the heated trap lines for approximately 1 minute.

After the one minute sample flush, the sampling valve was closed, and the sealed sampling lines were evacuated. The hot water bath heating the trap was removed, and the trap was allowed to cool to near room temperature. Then the trap was placed in a Dewar of liquid nitrogen. The temperature of the trap dropped to nearly -200°C. When the nitrogen stopped boiling, both ends of the vacuum canister were closed; and the vacuum pressure was recorded to the nearest 0.1 inch of mercury.

Once the system had stabilized, a valve on the vacuum canister was opened to the sampling loop allowing sample air to pass through the cold trap which caused the VOCs to condense. The amount of air sample analyzed was monitored by a gauge on the vacuum canister. When an appropriate amount of sample was collected (0.2 to 1.0 L), the valve to the cryogenic trap was closed off; and the sampling lines were

flushed with zero air. At this point, the liquid nitrogen was removed from the trap for approximately 15-20 seconds. This allowed any condensed oxygen to boil off but left the VOC sample intact in the trap. At the end of the 15-20 seconds, the trap was again immersed in liquid nitrogen. When cooling was complete, a final reading was taken on the vacuum gauge. The difference between the initial vacuum and the final vacuum in the canister was used to calculate the sample size.

To start the analysis, the liquid nitrogen was removed from the trap and replaced with water in a hot water bath (80°C). At the same time, a gas sampling valve was activated which back-flushed carrier gas and volatilized VOCs through the trap and onto the cold GC column (-35°C). Hot water was poured over the inlet and outlet of the trap line to complete the heating cycle. VOCs volatilized from the heated trap were cryofocused at the head of the GC column. The GC run proceeded for just over 30 minutes. The column effluent was split (9:1) to the FID and ECD detectors so that both detectors analyze the same sample simultaneously. Data were transferred in near real time to the data collection system and saved for later analysis.

The temperature was initially held at -35°C for one minute. This allowed separation of the freon peak from residual oxygen. After 1 minute, the temperature was ramped at 8 degrees per minute from -35 to 200°C. The temperature was then held at 200°C for one minute and the oven allowed to cool to -35°C with liquid nitrogen. The oven was allowed to equilibrate for a minimum of 1.5 minutes at -35°C before the start of the next run.

The injection port of the GC was set at 200°C, and the gas sampling valve was set at 150°C. Both the ECD and FID detectors were set at 275°C. The FID was run with zero grade air, and ultra high purity (UHP) hydrogen, with UHP nitrogen used as the makeup gas. UHP nitrogen was also the makeup gas for the ECD. The carrier gas was chromatographic grade helium. All gases were passed over moisture and charcoal traps before use.

Analytical Quality Assurance

The analytes of interest are found in very low levels in the indoor air samples and are even lower in outside air. It is important that cleaning, sampling, and analysis all be meticulously performed to insure data integrity. A Quality Assurance Plan was prepared for this work. The data quality objective for precision was specified as $\pm 25\%$ for all analytes. Detection limits were the same as those reported earlier (Sweet and Vermette, 1991) and were generally $< 1\%$ of the sample concentrations measured in this work.

The entire sampling system was kept under positive pressure with zero grade air when not in use, and whenever possible when in use. A complete flush of the system with zero air was performed at the beginning of each day of analysis, and between each sample run. All analytical samples are run in triplicate, non-consecutively, and when possible on 3 separate days. In general, samples were run in the morning and afternoon of one day (to help compare drift during the day) and the third sample on another day. Samples were rerun if unusually high concentrations of any target compound were found, if carry over was suspected, or if other difficulties were encountered. Samples were generally analyzed within one week on collection. Other workers have shown that all target compounds are stable in the canisters for up to 30 days storage time (Oliver *et al.*, 1986).

The calibration standard was a custom mixture of 17 target VOCs that was commercially prepared and certified (Scott Specialty Gases, Plumsteadville, PA). Concentrations were 10 ppb for the polychlorinated VOCs (ECD compounds) and 100 ppb for the non-chlorinated VOCs (FID compounds). This standard mix was diluted by half with humidified zero air to make a working standard with VOC concentrations similar to those encountered in the samples. Response factors from the working standard are compared with past values for consistency. The working standard was also used to check the recovery of VOCs from the canisters and the analytical system. Recoveries of VOC compounds in the working standard mix sampled from a canister were 80-120% of the results obtained by direct injection of the same working

standard. Serial dilutions of the working standard were used to verify linear behavior in the response of the detectors over the concentration ranges analyzed (Sweet and Vermette, 1991).

After the zero grade air and calibration samples were analyzed and approved, unknown samples are run. Levels of carbon tetrachloride (CCl₄) are tracked between each sample as a surrogate internal standard. CCl₄ concentrations are relatively constant and ubiquitous throughout the atmosphere, so ambient air samples should have a similar amount if no source is present.

Duplicate samples were generally run during each sampling day. This includes the studies in private homes and the Building Research Council test house. Sampling canisters were placed side by side and the sampling valves opened and closed simultaneously. It was determined for this series of experiments that the sampling duplicate variations were approximately equal to the replicate variations in the lab ($\pm 20\%$).

During analysis of the chromatograms and the data, any peaks that could not be positively identified and quantified were not included in the data sets. Causes of this included retention time drift with the lighter gases, coelution, baseline drift, and peak values near the noise levels. Duplicates and replicates were compared with each other and across days. Outside air samples were compared across sampling days.

RESULTS AND DISCUSSION

Test House - Phase I

During the first phase of this work, the test house was in a "tight" configuration with minimum infiltration rates (0.2 ACH). Automotive products (see Table 3), with their seals broken and caps removed, were placed in the exterior room simulating a garage. An adjacent room in the test house represented a "living area". This configuration simulated active use of the products and represented a worst case emission scenario. Air samples were taken in the "garage" and "living areas" 5 hours after the product introduction. These samples were analyzed for benzene, the most toxic of the target compounds. The concentration of benzene reached 280 ng/L (over 200 times outdoor values) in the "garage"; however, levels in the "living area" (4.2 ng/L) were measured at levels only slightly higher than those found in outdoor air (1 to 2 ng/L).

The air movement between the garage and living area was increased by removing the caps from two adjustable ports connecting the two rooms creating a leakage area of 10 cm². Air samples were again taken in the garage and living areas 5 hours after opening the air exchange ports and 29 hours after the original introduction of the products into the garage. Benzene levels in the garage were too high to quantify accurately without considerable dilution. They were estimated at over 900 ng/L (about 600 times over blank values). The benzene concentration measured in the living area was 39 ng/L. We concluded from these experiments that short-term storage of HP in a room with low infiltration can result in a rapid build-up of VOCs. The low infiltration rate also seems to be effective in preventing high levels of VOCs from building up in rooms adjacent to the storage area, but with air exchange between rooms, elevated levels of VOCs occur in this room as well.

During the second phase of the work, the test house remained in a "tight" configuration with infiltration rate at 0.2 ACH. Automotive and household maintenance HP were placed in the garage with their caps in place but having their original seals broken. This configuration simulated the storage of partially used HP.

Air samples were collected outside, upwind of the test house, in the garage, and in the living area. The first test was prior to the introduction of the automotive and household maintenance products. Later tests were 6 hours, 24 hours, 2 days, 4 days, 8 days, 12 days, and 16 days after the introduction of HP. A summary of VOCs emitted by the HP is given in table 5.

Under these conditions, VOC concentrations exhibited one of three patterns. Some VOCs reached peak concentrations of up to 1000 ng/L or more in the garage section of the test house within 24 hours, and then slowly declined over a 16-day period (figures 4 through 14). Concentrations of these VOCs in the living area followed the same trend but were initially about half as much as those in the garage. Styrene/o-xylene seemed to behave somewhat differently under these conditions showing continued increases during the sampling period. Overall concentrations in the living area approached those in the garage but remained lower throughout the 16-day period. The data suggest that an equilibrium concentration between 10 and 100 times the background concentration is established in both the garage and the adjacent living area after long-term storage of these products. This equilibrium may be perturbed by changes in the ambient temperature. During experiments with the household maintenance products (figures 10 to 14), the outside temperature increased from a high of 23°C on day 4 to a high of about 30°C on days 8 through 12. VOC levels in both the garage and living area increased during this period. The automotive products were sources of benzene, toluene, and several other aromatic hydrocarbons; and the household maintenance products were sources of methyl chloroform, methylene chloride, and, to a lesser degree, aromatic hydrocarbons.

Other VOCs including carbon tetrachloride, chlorobenzene, trichloroethylene, and chloroform (figures 15 through 19) varied with the outside air indicating the products tested were not significant sources for these chemicals. Finally, concentrations of freon were consistently higher in the living area of the test house even before the introduction of the products (figure 20). This pattern indicates a source in the living area of the test house (air conditioner).

Table 5. Sources of VOCs in the Test House

Automotive	Household Maintenance	Other Sources
<i>benzene</i>	<i>methylene chloride</i>	freon
toluene*	toluene	chlorobenzene
ethylbenzene*	ethylbenzene	trichloroethylene
m,p-xylene*	m,p-xylene	chloroform
<i>styrene/o-xylene</i>	<i>methyl chloroform</i>	carbon tetrachloride
<i>methyl ethyl ketone</i>		tetrachloroethylene

* indicates the dominant source. Compounds in *italics* are associated with only one of the product mixes.

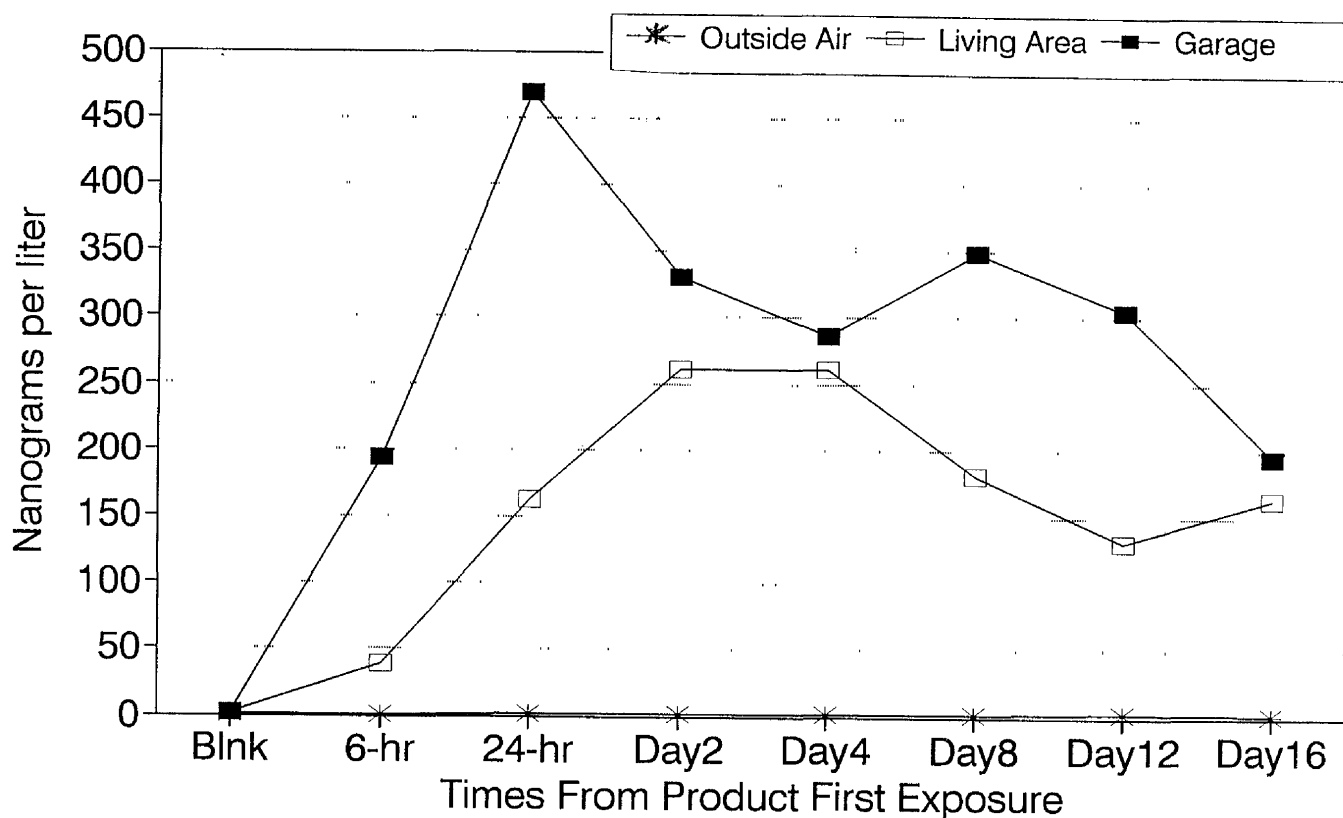


Figure 4. Concentrations of Benzene from Automotive Products in the Test House "Garage" (0.2 ACH)

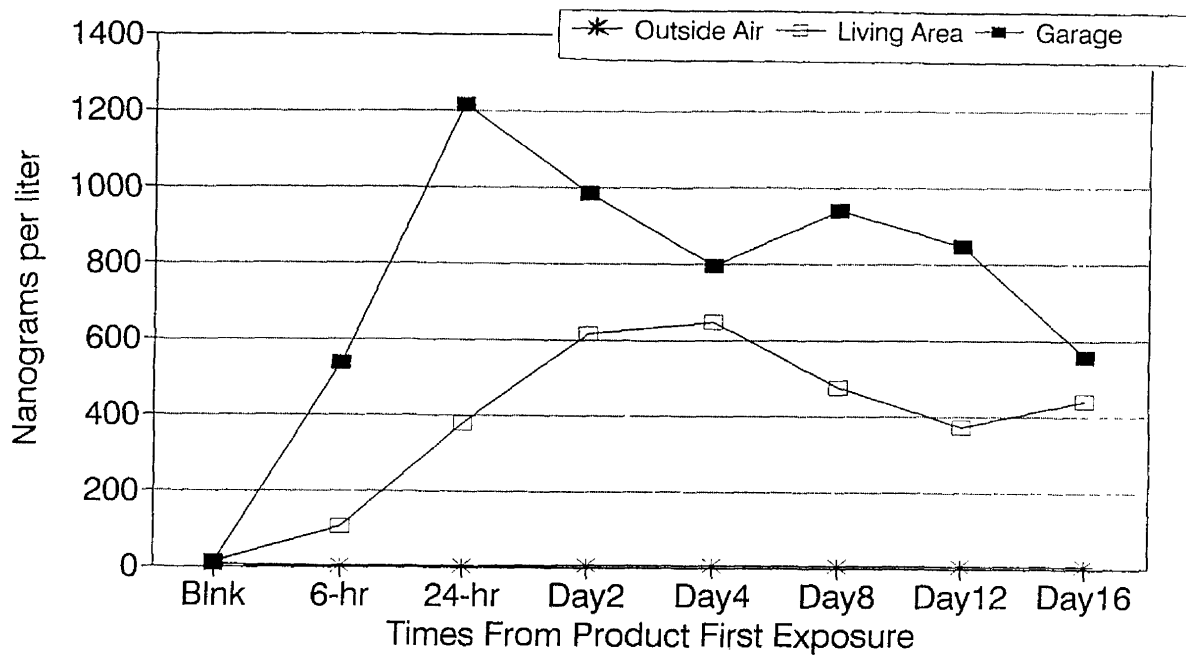


Figure 5. Concentrations of Toluene from Automotive Products in the Test House "Garage" (0.2 ACH)

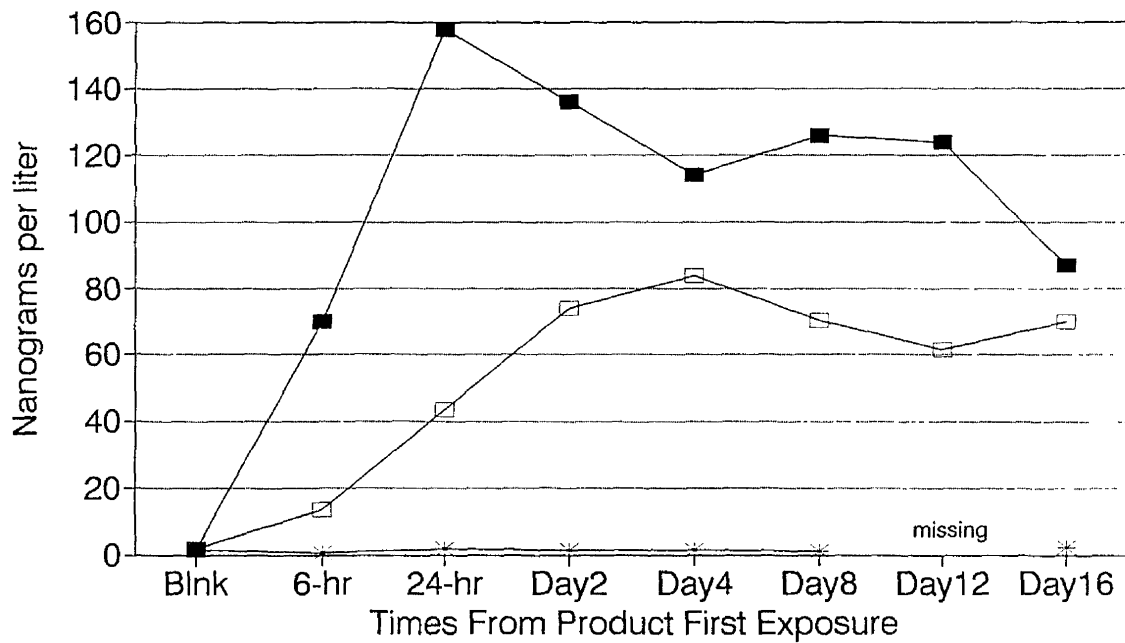


Figure 6. Concentrations of Ethylbenzene from Automotive Products in the Test House "Garage" (0.2 ACH)

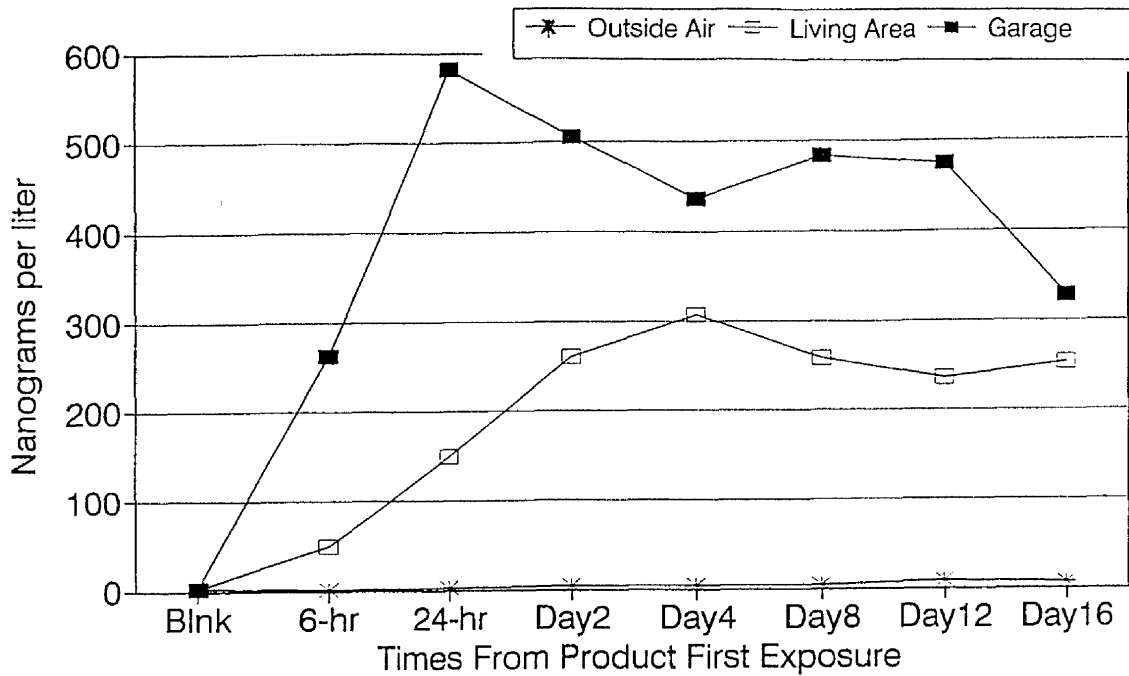


Figure 7. Concentrations of m,p-Xylene from Automotive Products in the Test House "Garage" (0.2 ACH)

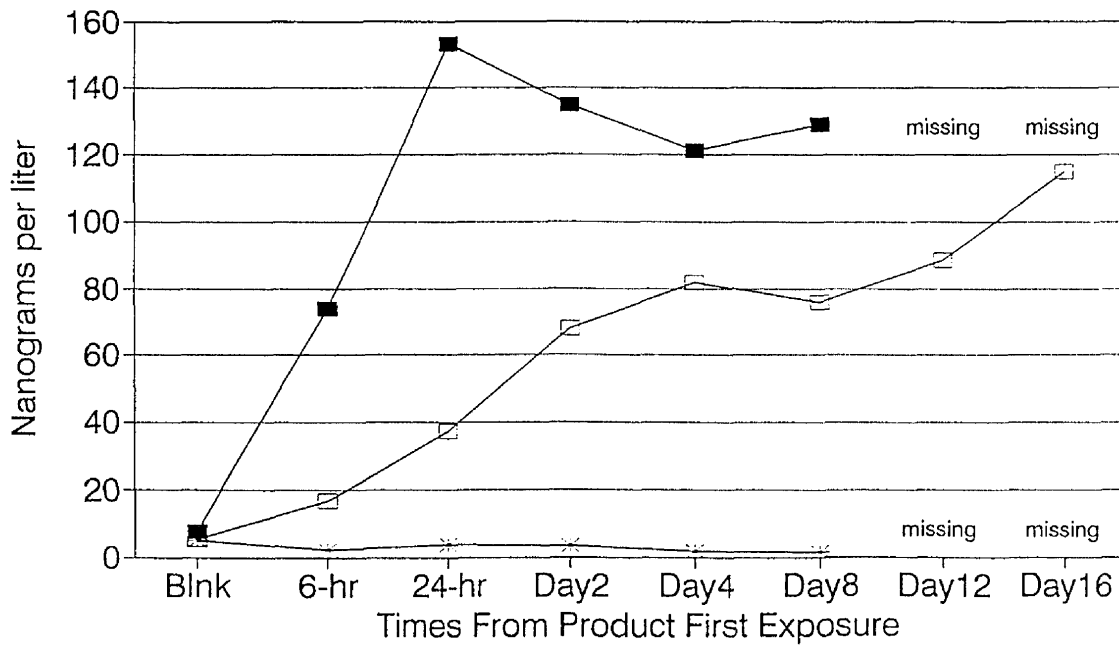


Figure 8. Concentrations of Styrene/o-Xylene from Automotive Products in the Test House "Garage" (0.2 ACH)

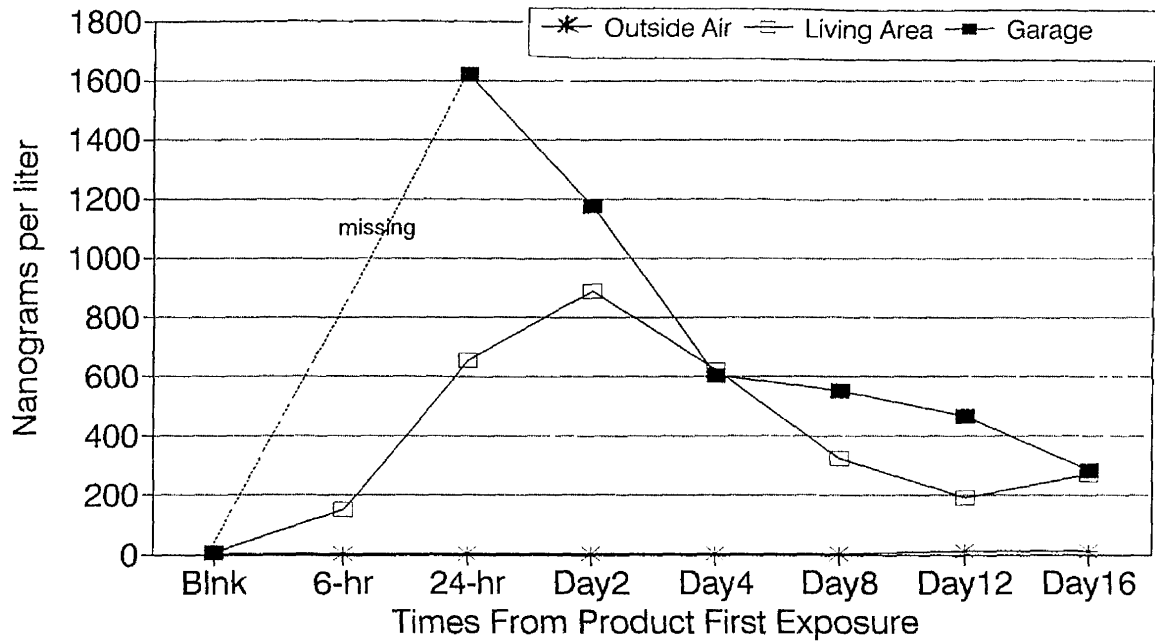


Figure 9. Concentrations of Methyl Ethyl Ketone from Automotive Products in the Test House "Garage" (0.2 ACH)

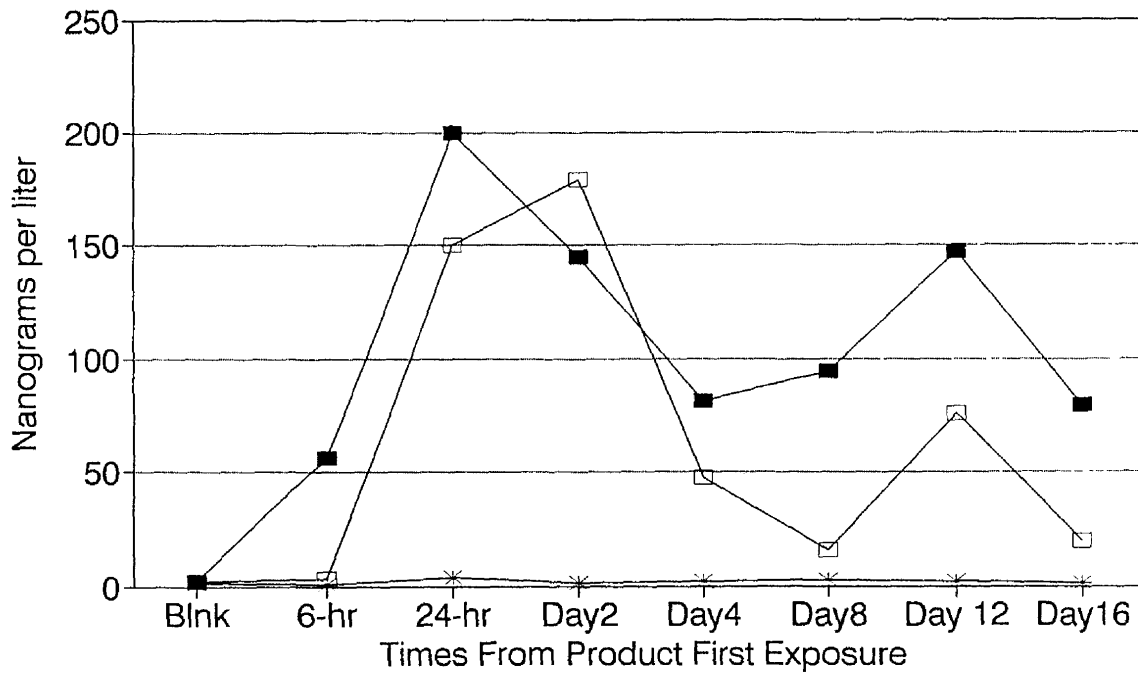


Figure 10. Concentrations of Methyl Chloroform from Household Maintenance Products in the Test House "Garage" (0.2 ACH)

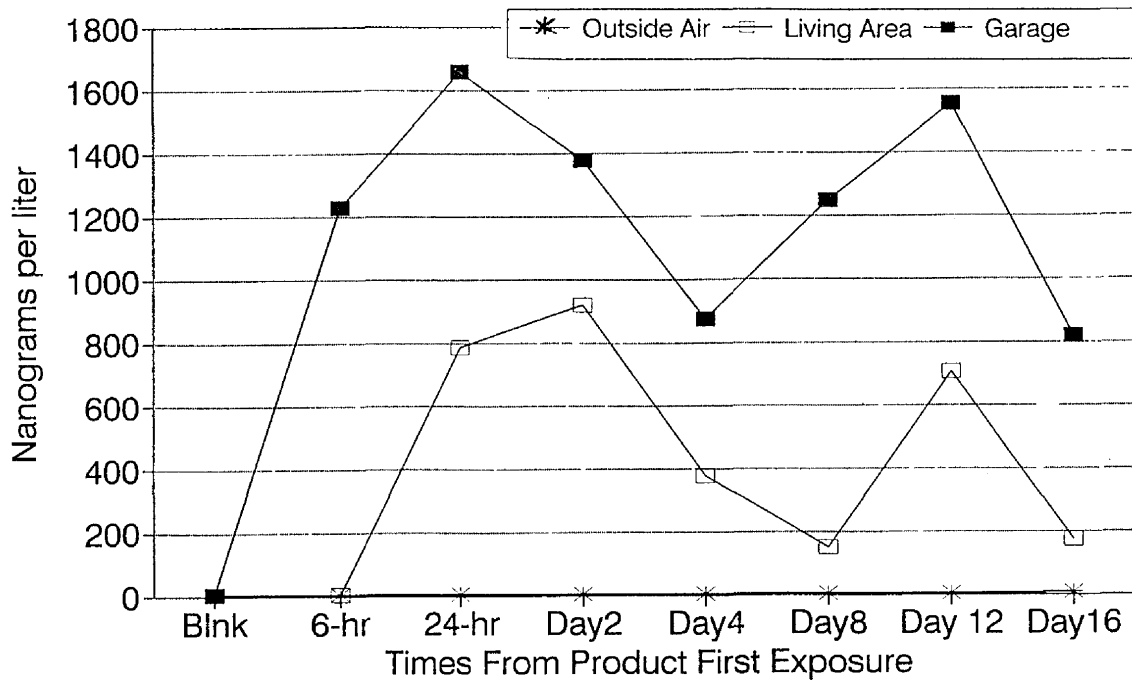


Figure 11. Concentrations of Methylene Chloride from Household Maintenance Products in the Test House "Garage" (0.2 ACH)

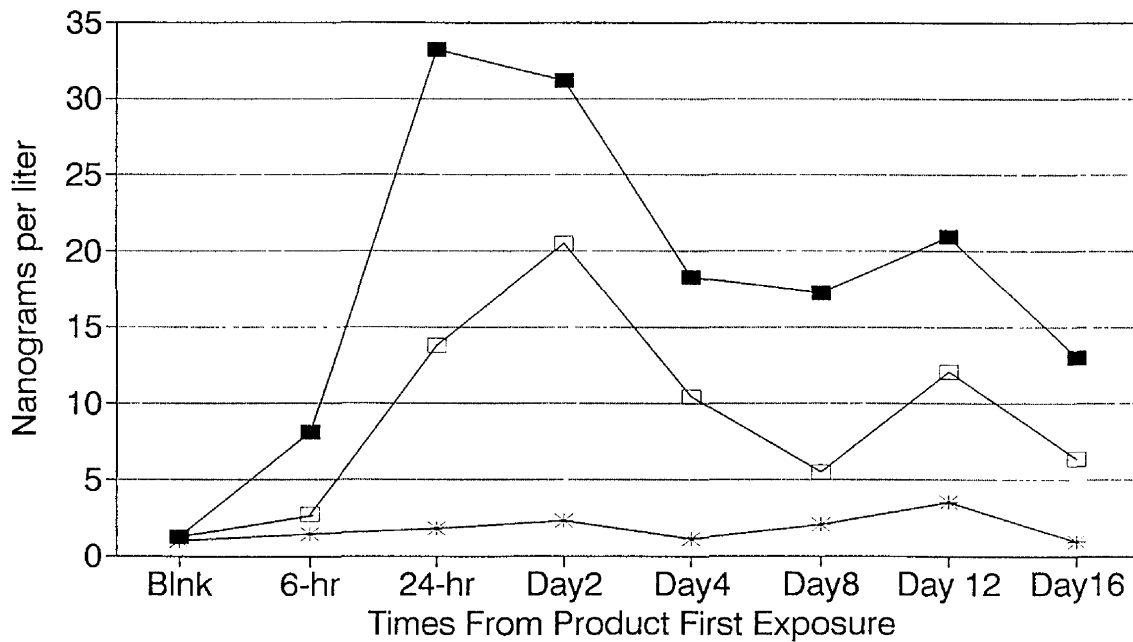


Figure 12. Concentrations of m,p-Xylene from Household Maintenance Products in the Test House "Garage" (0.2 ACH)

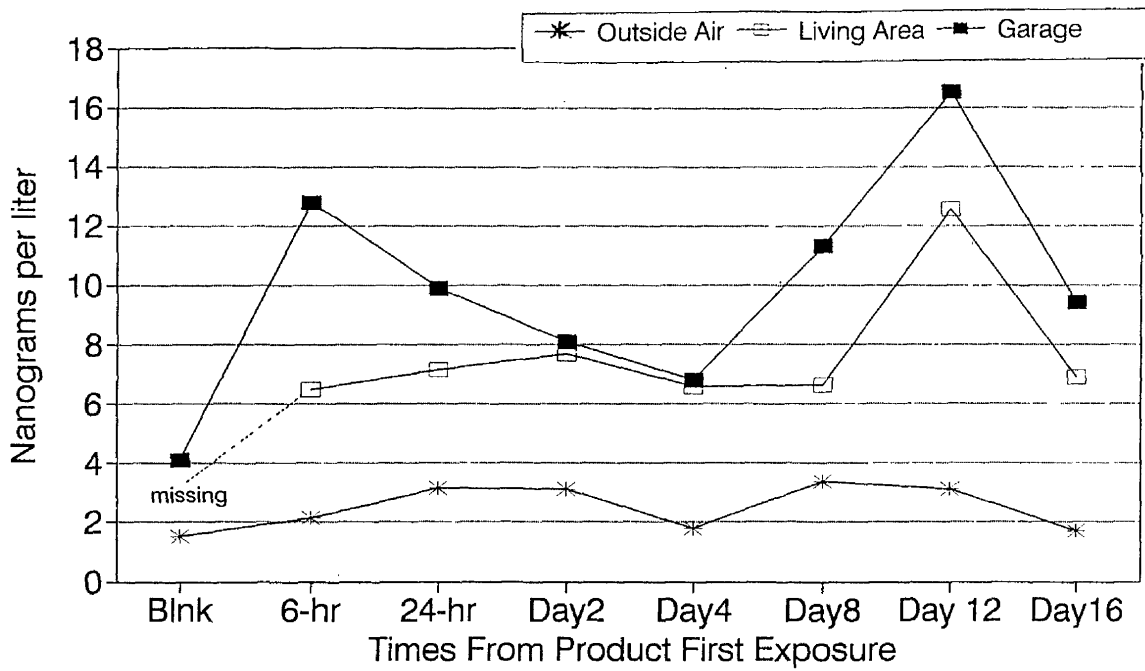


Figure 13. Concentrations of Toluene from Household Maintenance Products in the Test House "Garage" (0.2 ACH)

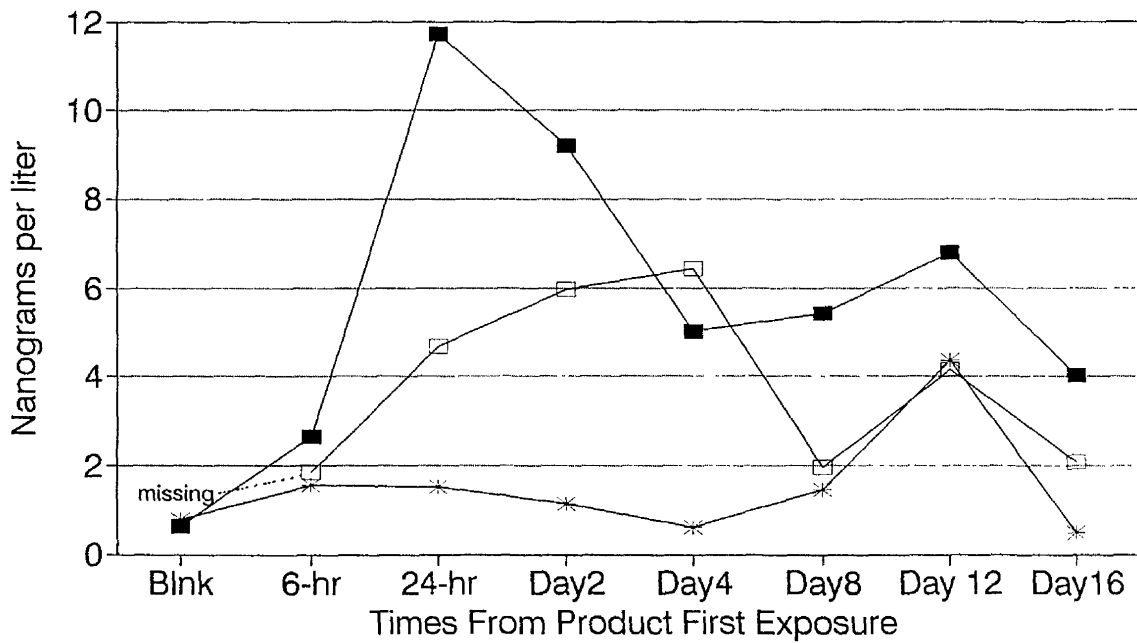


Figure 14. Concentrations of Ethylbenzene from Household Maintenance Products in the Test House "Garage" (0.2 ACH)

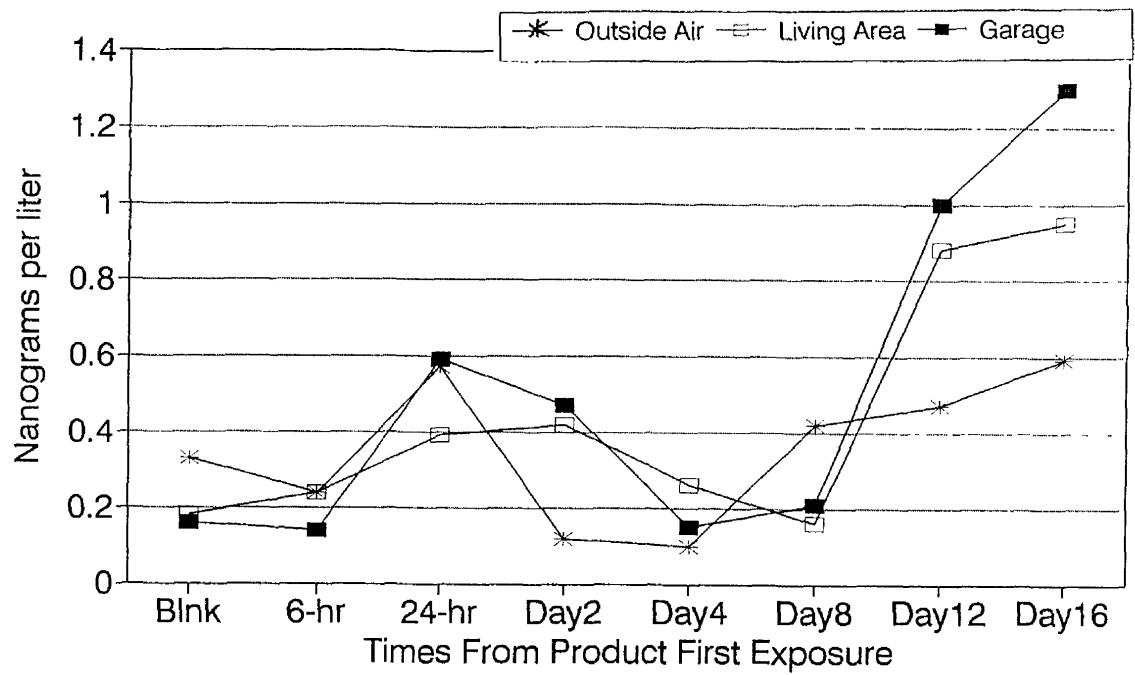


Figure 15. Concentrations of Tetrachloroethylene from Ambient Air Exchange in the Test House (0.2 ACH)

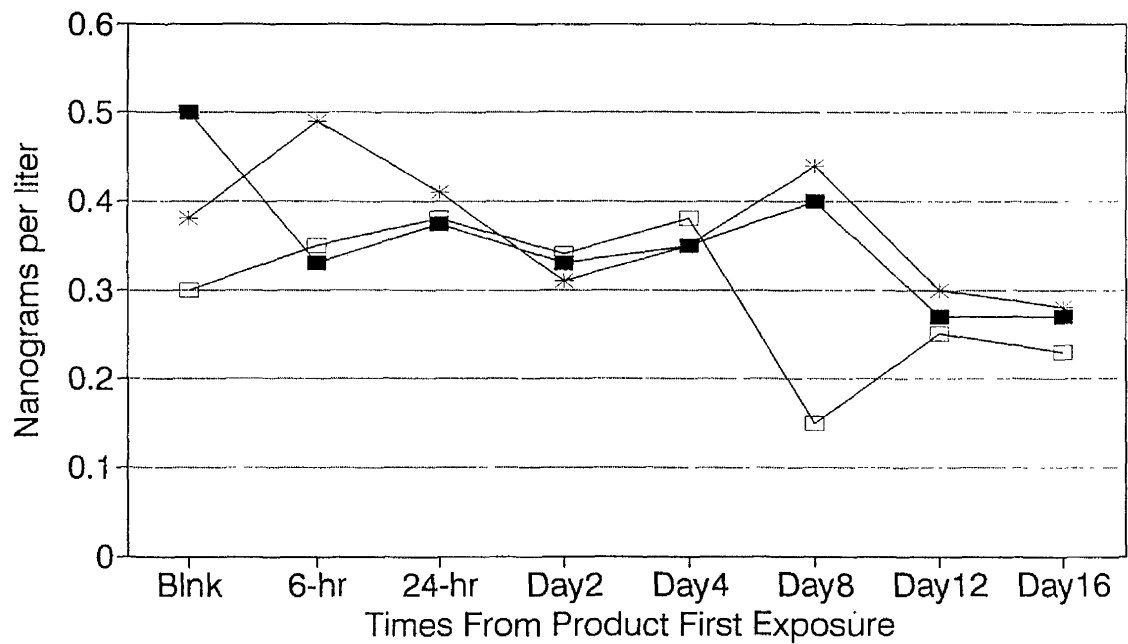


Figure 16. Concentrations of Carbon Tetrachloride from Ambient Air Exchange in the Test House (0.2 ACH)

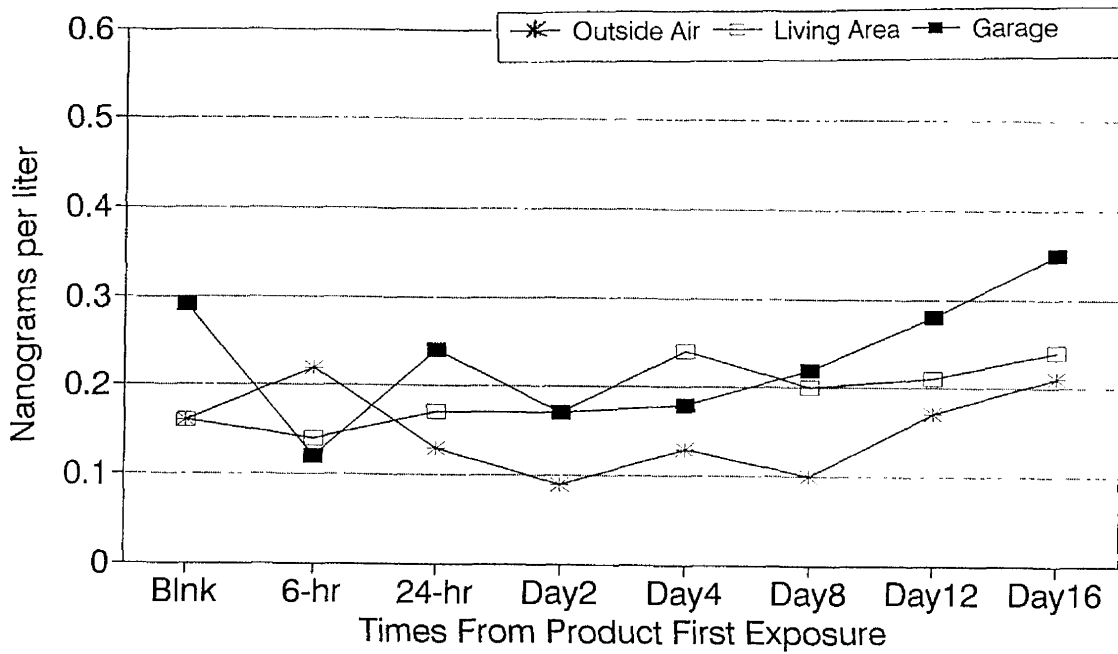


Figure 17. Concentrations of Trichloroethylene from Ambient Air Exchange in the Test House (0.2 ACH)

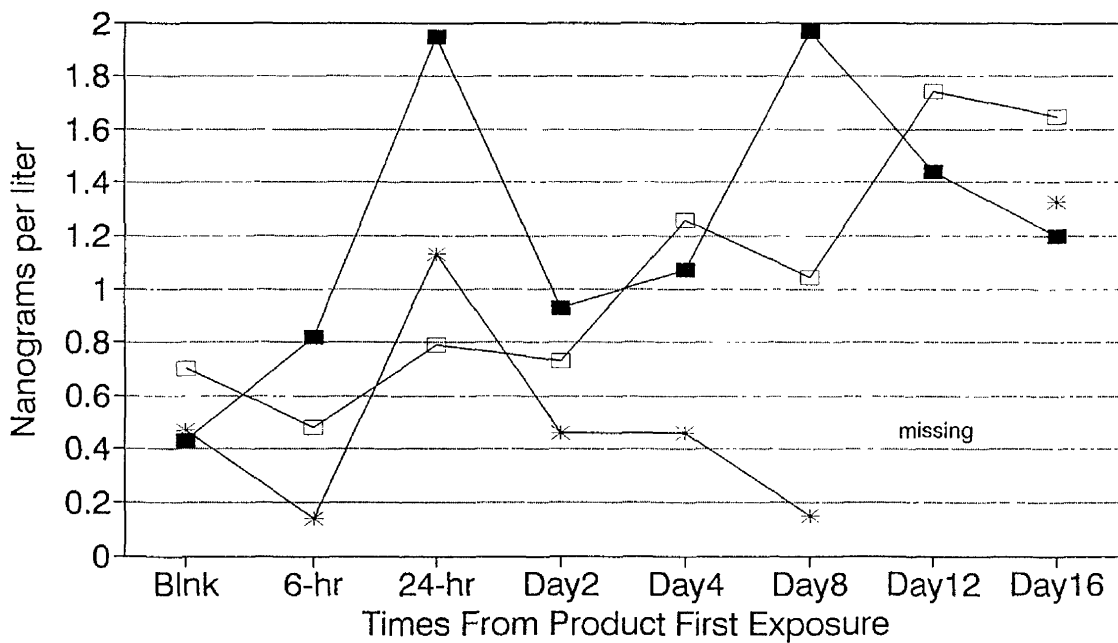


Figure 18. Concentrations of Chlorobenzene from Ambient Air Exchange in the Test House (0.2 ACH)

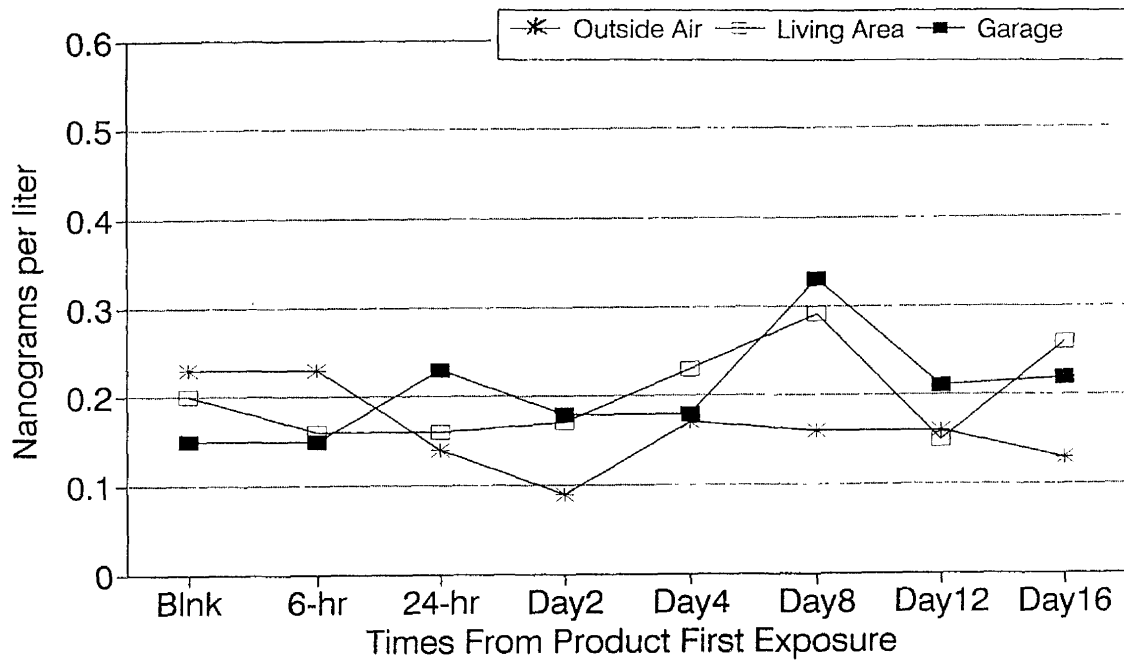


Figure 19. Concentrations of Chloroform from Ambient Air Exchange in the Test House (0.2 ACH)

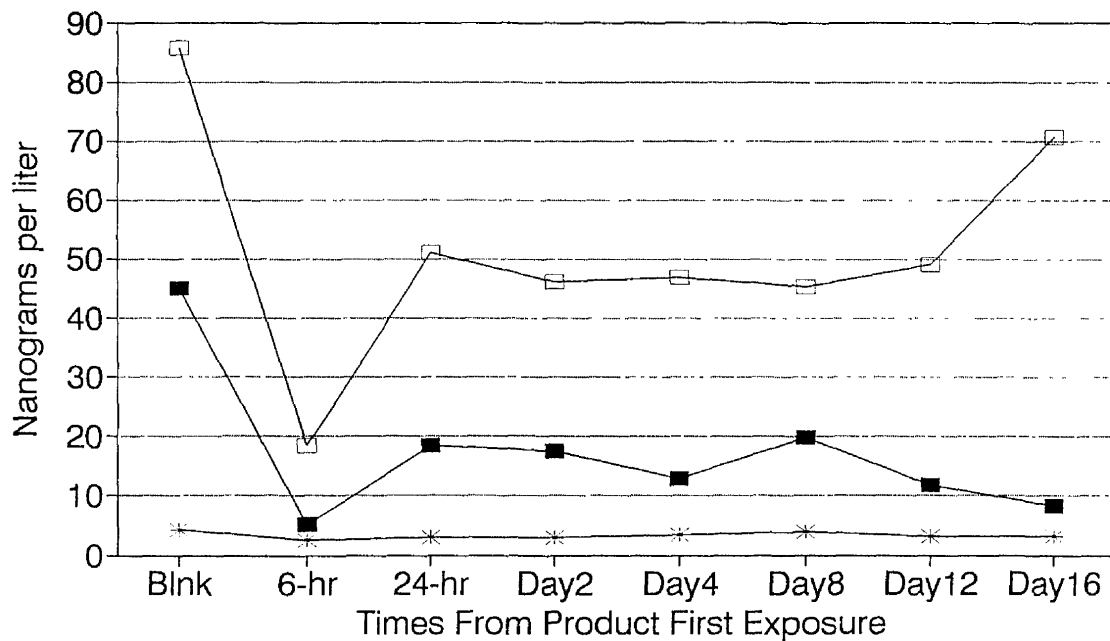


Figure 20. Concentrations of Freon from Ambient Air Exchange and Sources in the Test House (0.2 ACH)

Test House - Phase II

During the next phase of this work, the test house infiltration rate was increased to 1.2 ACH by opening additional ports to the outside. The opening between the garage and living area remained the same as in the previous experiment. The intent was to determine the influence of an increased infiltration rate on the movement of VOCs from the garage to the living area and its effect on the overall concentrations of VOCs that build up in the test house. As before, the automotive product mix was placed in the garage simulating the storage of partially used automotive products. Since the household maintenance products emitted lower amounts of many of the same VOCs, they were not tested in this part of the study.

Measurement of targeted VOCs identified the same five compounds previously associated with the automotive products (see table 3). The VOCs measured in the garage reached about half of comparable levels measured with the tighter configuration (0.2 ACH). VOCs in the living area were reduced compared to the earlier study (figures 21 through 26). It is apparent from the data that a larger infiltration rate limits the buildup of VOCs in both the garage and living area.

The automotive products were removed after day 8, and subsequent samples were taken after 24 hours (day 9), 3 days (day 11), and 6 days (day 14). The removal of the automotive products dramatically reduced the VOC concentrations within 2 days. It is apparent that under conditions of high infiltration the removal of the HP results in a rapid decrease in the VOC concentrations in indoor air to levels consistent with those present prior to the introduction of the products.

The increase in VOC concentrations on day 8 (figures 21 to 26) again corresponded to an increase in outside air temperatures. This suggests that a continuing decrease or a stable equilibrium in VOC concentrations can not be expected over the long term. Rather, indoor VOC concentrations attributable to the storage of HP may be subject to considerable fluctuations due to changes in the temperature of unheated storage areas.

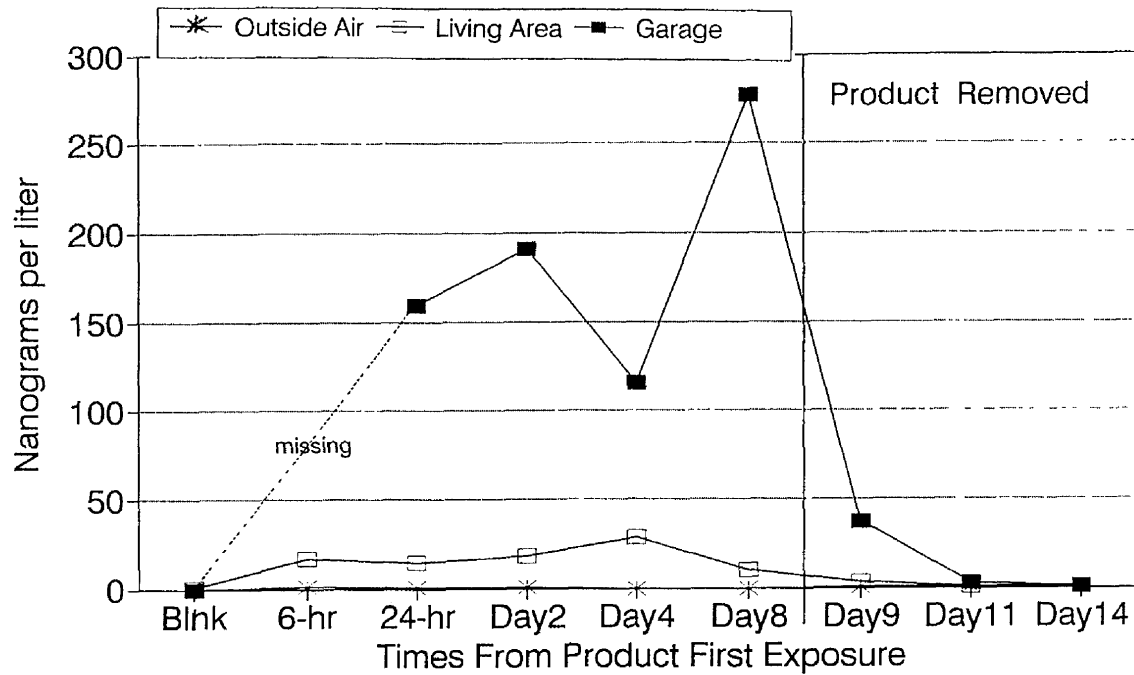


Figure 21. Concentrations of Benzene from Automotive Products in the Test House "Garage" (1.2 ACH)

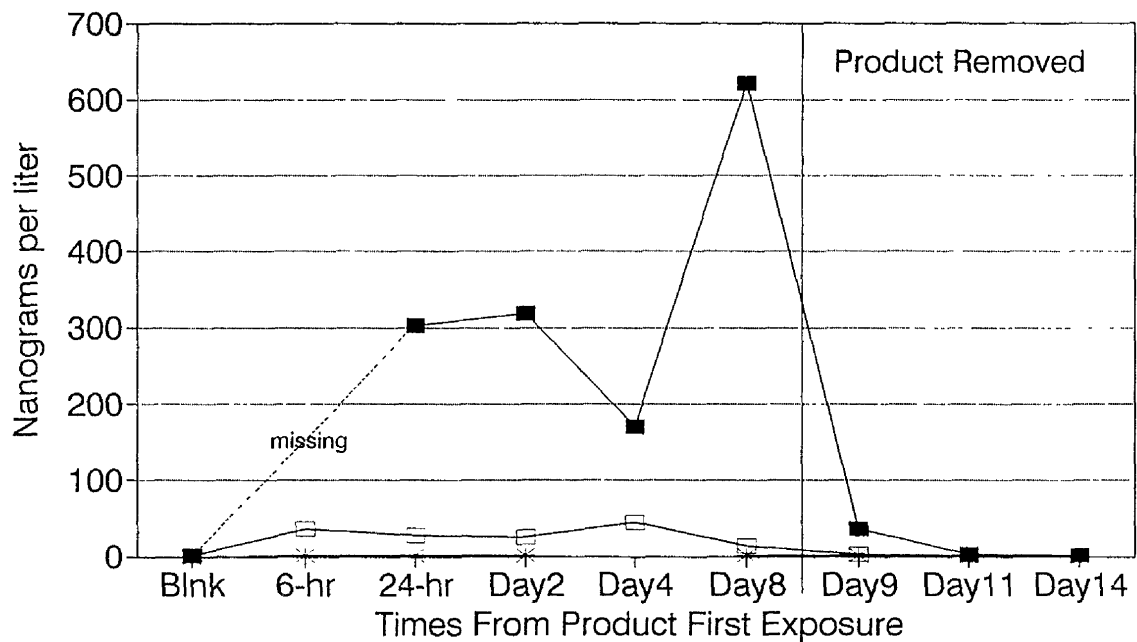


Figure 22. Concentrations of Toluene from Automotive Products in the Test House "Garage" (1.2 ACH)

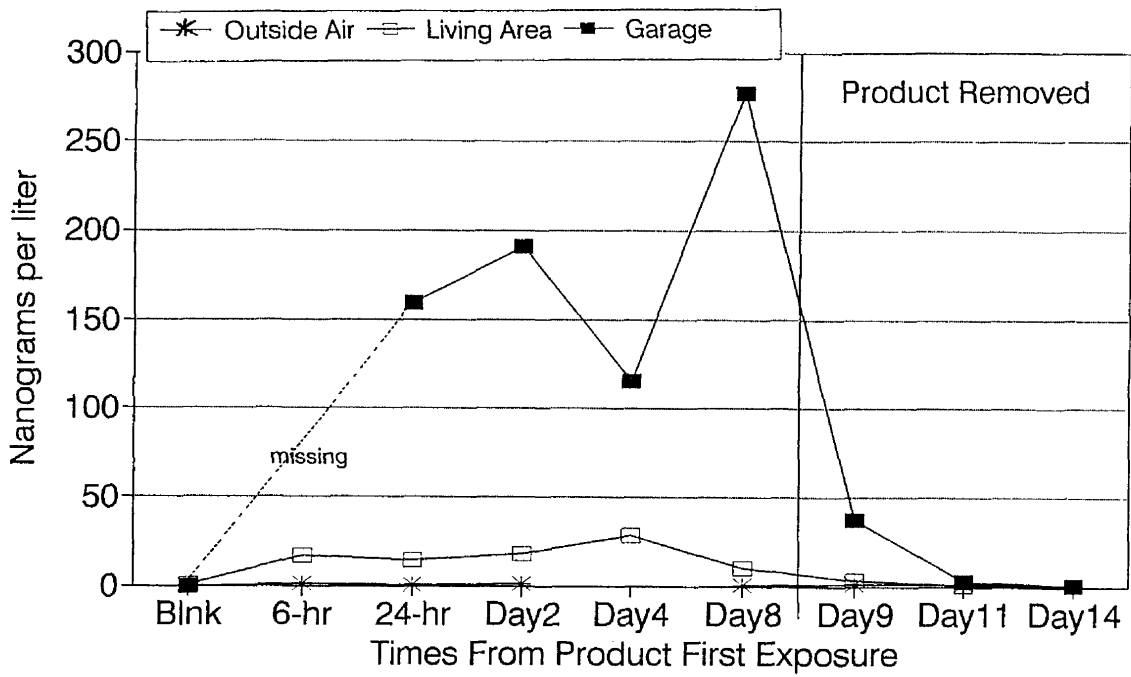


Figure 23. Concentrations of m,p-Xylene from Automotive Products in the Test House "Garage" (1.2 ACH)

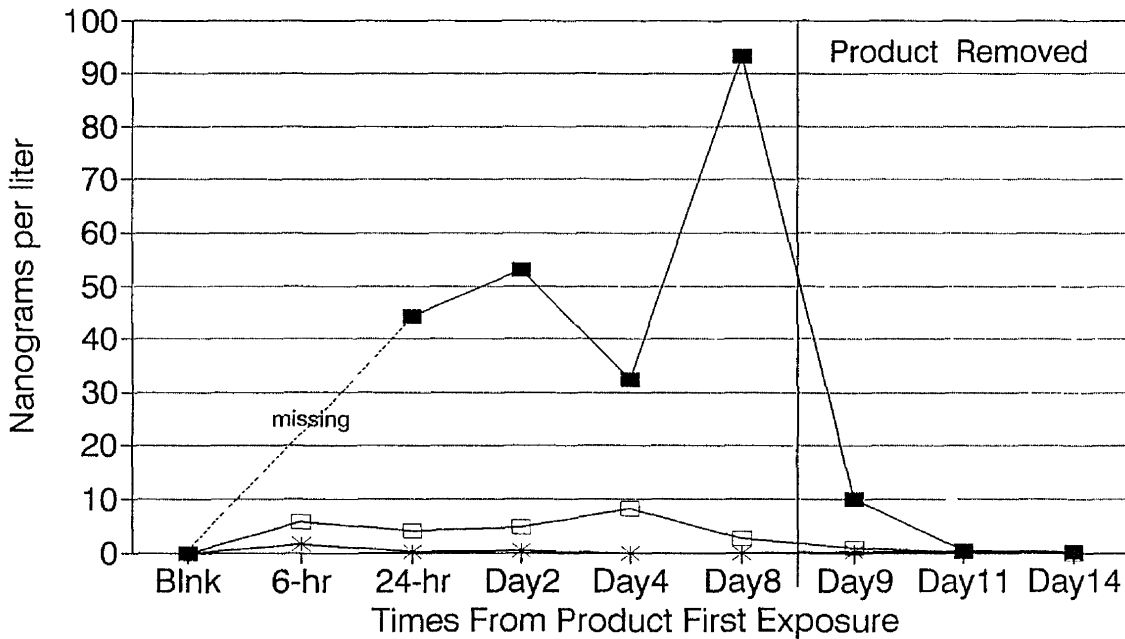


Figure 24. Concentrations of Ethylbenzene from Automotive Products in the Test House "Garage" (1.2 ACH)

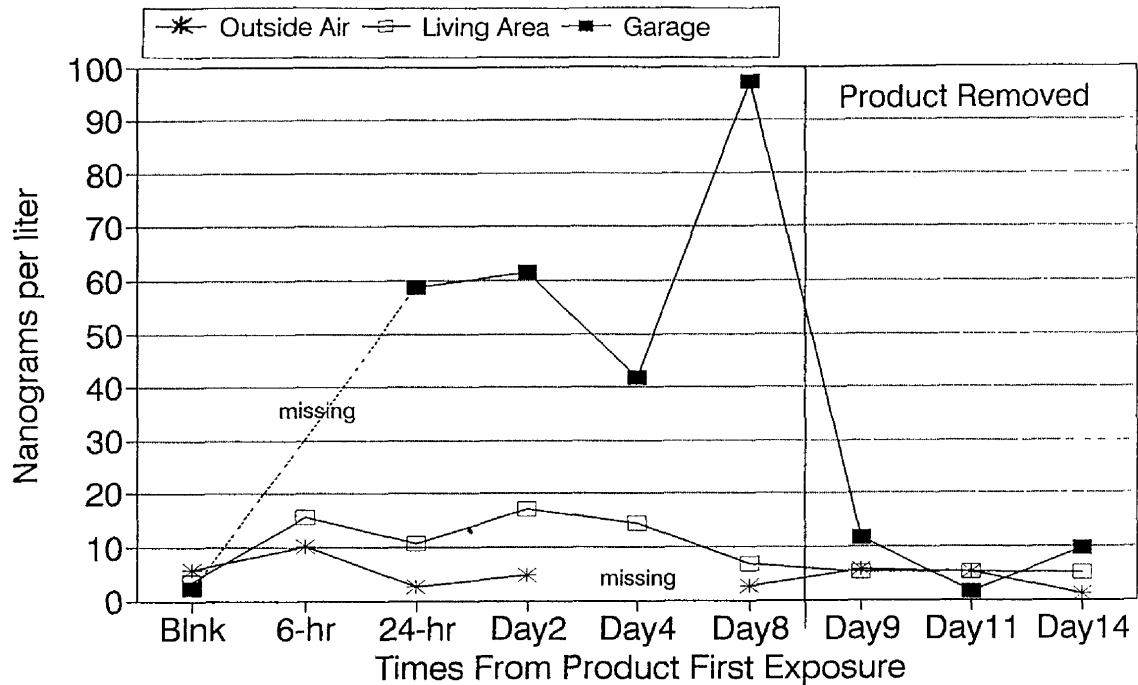


Figure 25. Concentrations of Styrene/o-Xylene from Automotive Products in the Test House "Garage" (1.2 ACH)

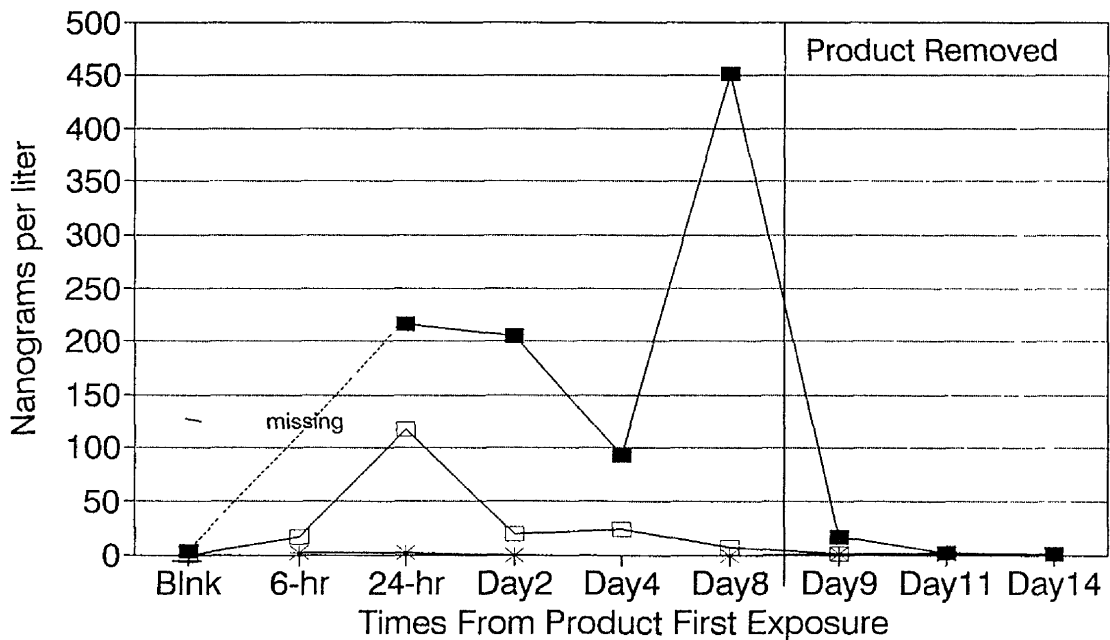


Figure 26. Concentrations of Methyl Ethyl Ketone from Automotive Products in the Test House "Garage" (1.2 ACH)

Test House - Phase III

A final set of experiments was carried out in the test house with the infiltration rate set at an intermediate value (0.56 ACH). The automotive product mix was also used for this test. The VOC concentrations followed the same pattern as in the two previous experiments reaching a plateau after about 4 days. Table 6 gives the concentration in the living area of 3 representative automotive product VOCs and one VOC not emitted by the automotive products (methyl chloroform, MeCHCl₃) after 4 days storage of the HP.

Table 6. Equilibrium Concentrations of VOCs at Various Infiltration Rates^a

Infiltration Rate (ACH)	Benzene	Toluene	Xylenes	MeCHCl ₃
0.20	260	625	300	3.0
0.56	154	570	365	2.0
1.20	25	50	30	0.1

a) concentrations in ng/L.

Tests in Actual Residences

Indoor air samples were taken at three actual residences during periods when the houses were closed up to compare VOC concentrations in indoor air to those measured at the test house and to identify the sources of the VOCs. Air samples were taken outside (upwind of the houses), in the garages (attached and detached), and at several locations throughout the living areas. The results of the blower door tests on these houses are given in Table 2.

Residence 1. The first residence tested was a single-story, 15-year-old, frame house with a 2-car attached garage and storage area. The blower door test measured 0.34 ACH. The garage contained numerous containers of automotive products (gasoline, cleaners) and household maintenance products (paints and solvents). In the living area, there were some household cleaning products in the utility area just off the garage and in the bathrooms (Figure 27).

Figures 28 and 29 show concentrations of selected VOCs in this house. For non-chlorinated species (benzene, toluene, etc.), concentrations in the garage were 100-200 ng/L, almost as high as the worst case concentrations in the test house "garage" at the end of the 16-day storage period. These VOCs were also from the same group of chemicals previously associated with the automotive product mix. The elevated levels of automotive VOCs in the garage suggest that they may be a potential source to the living area of residence 1. The concentrations of benzene measured in the garage and living area of residence 1 (100 ng/L and 4 ng/L) were similar to corresponding values measured in the test house (280 ng/L and 4 ng/L) at similar infiltration rates before the connection between the garage and living area was opened. Freon levels were also elevated in the garage (50 ng/L) presumably from an auto air conditioner. The concentrations of m,p-xylene, ethylbenzene, and toluene also show some increase in the living area (up to 10 times outside air), but they were not as high as those recorded in the living area of the test house when the connection between the bays was opened. The elevated levels of VOCs in the living areas of residence #1 could be associated with movement of VOCs from the garage or with storage of household maintenance products in the living area of the house.

Several chlorinated VOCs (trichloroethylene, chloroform, and methyl chloroform, chlorobenzene) were present in the living area of the house at levels higher (5 to 50 times) than those found in outside air (figure 29). This indicates that sources were present in the living areas of residence 1. The concentrations of chloroform and methylene chloride are evenly distributed in the residence, but they drop off dramatically in the garage. The highest levels of methyl chloroform, trichloroethylene, chlorobenzene, and tetrachloroethylene were measured in the bathroom of residence 1, with concentrations decreasing in rooms further from the bathroom (figure 29). Sources may be stored household maintenance products or emission from a shower that was used during the sampling period. McKone (1987, 1991) determined that household tap water can be a source of chlorinated VOCs in indoor air. Of the target compounds measured, only carbon tetrachloride showed no difference between inside and outside concentrations.

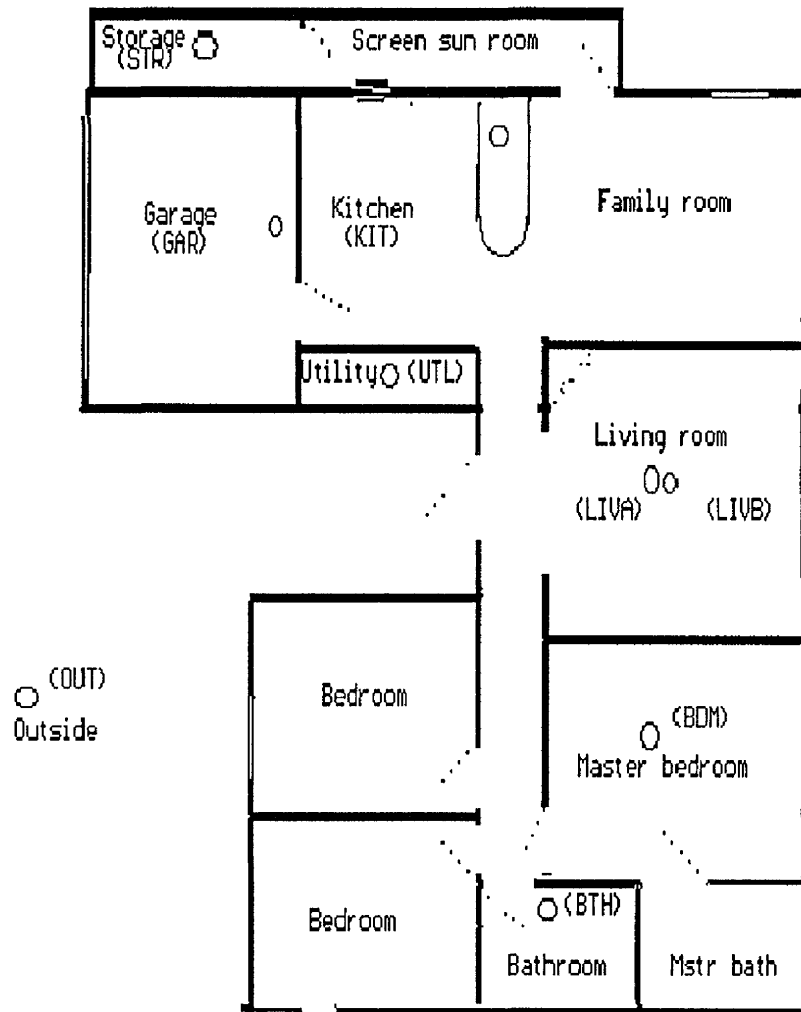


Figure 27. Floorplan and Sampling Locations for Residence 1

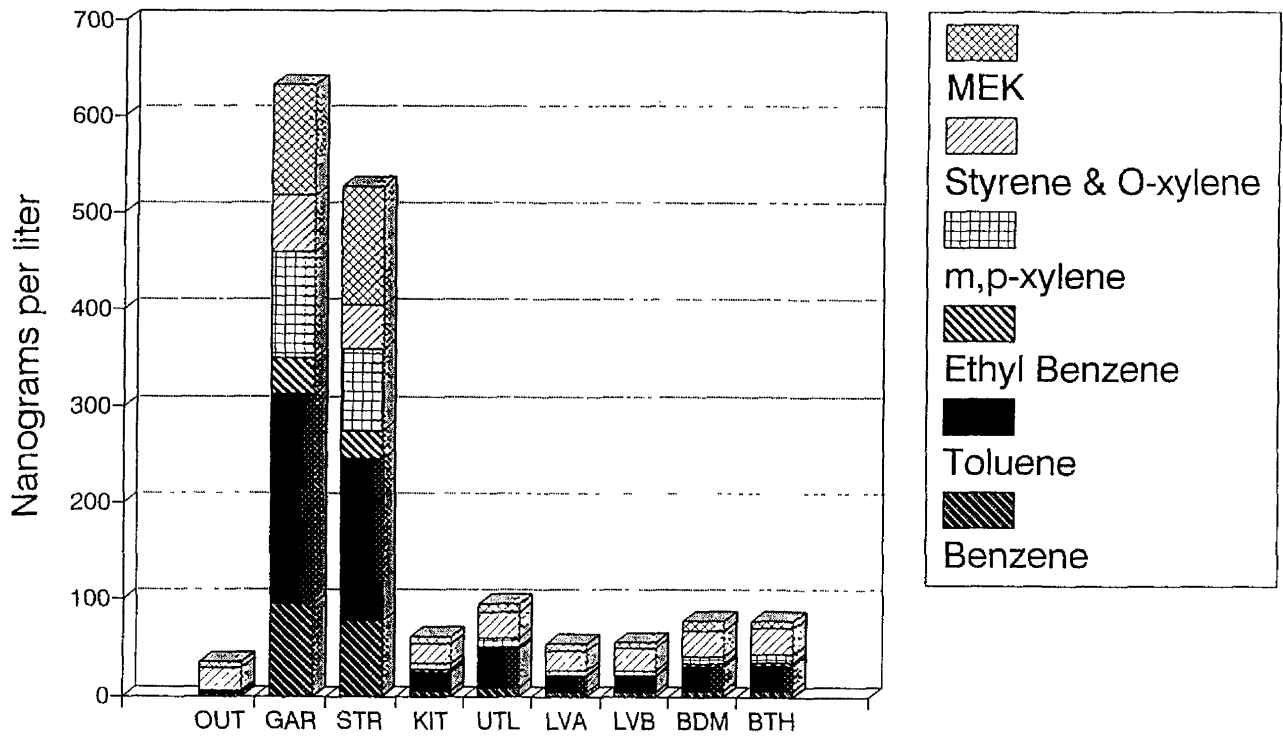


Figure 28. Concentrations of Non-chlorinated VOCs in Residence 1 (0.34 ACH)

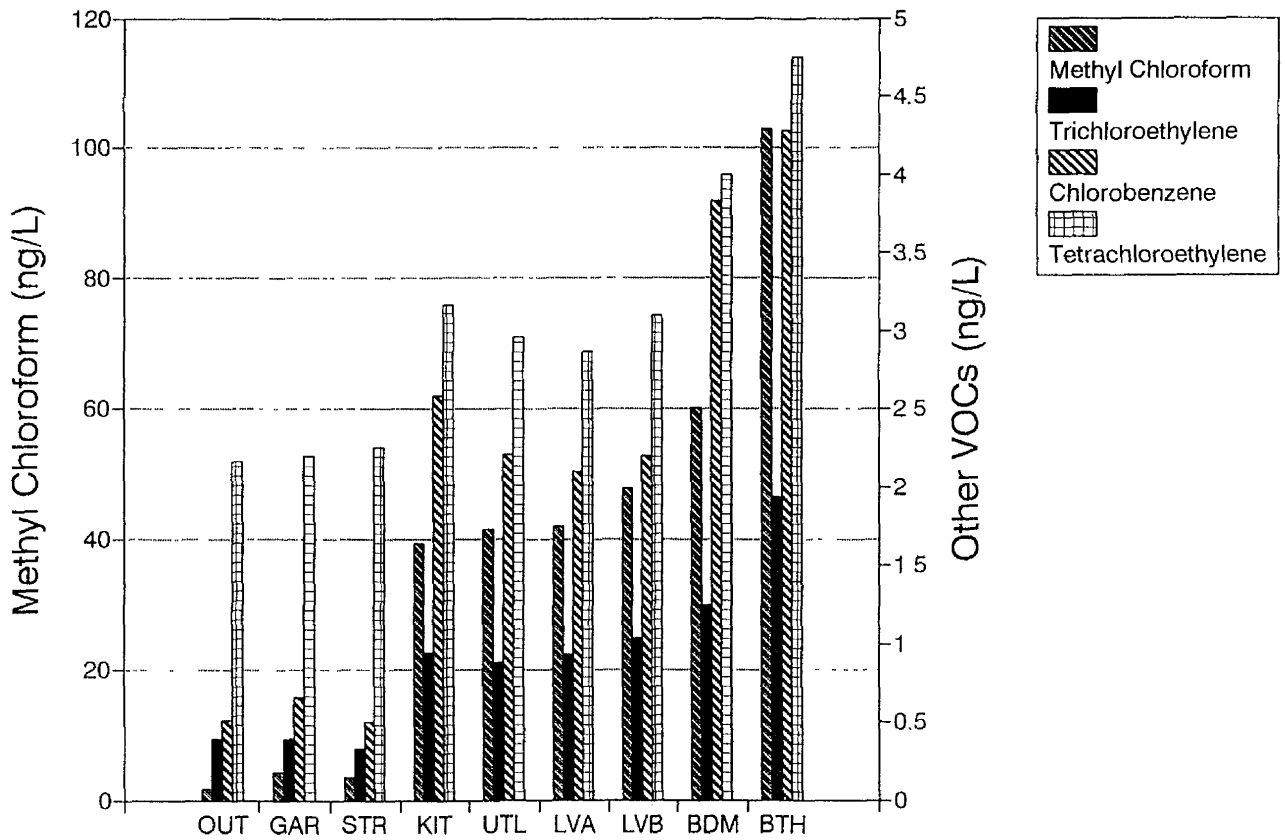


Figure 29. Concentrations of Chlorinated VOCs in Residence 1 (0.34 ACH)

Residence 2. The second residence (figure 30) was a 100-year-old, 3-story, frame house with a basement and a separate garage building. Household maintenance products were stored in the basement (paints and solvents) and in the living area of the home (household cleaners). This house had the highest infiltration rate (1.4 ACH).

As indicated in figure 31, VOCs previously associated with the automotive product mix (benzene, toluene, xylene, etc.) exhibited the highest indoor air concentrations in the detached garage. The levels of these VOCs closely resembled those measured in the test house garage at the end of the 16-day storage period (figures 6 to 11). The levels of methylene chloride and methyl chloroform (previously attributed to the household maintenance product mix) were also elevated in the detached garage.

The concentrations of VOCs (figure 32) in the living areas were, with 3 exceptions, similar to those found in the living area of the first house. The concentration of methyl chloroform was lower than in the first house (5 to 20 ng/L versus 40 to 100 ng/L). The concentration of methyl ethyl ketone found in the kitchen was 50 ng/L, about 10 times higher than that found in the living areas of the first residence. Tetrachloroethylene was found at elevated concentrations (5 to 20 ng/L) in the living areas of the second residence compared to the first residence (<5 ng/L). This compound is the principal solvent used in dry cleaning and is emitted from freshly dry-cleaned clothes (Thomas *et al.*, 1989). Tetrachloroethylene was not found above ambient levels in either the test house or in the first home which had no freshly dry-cleaned clothes. There is no indication that HP storage in the basement has an effect on VOC concentrations in the living area. Rather, VOC sources seem to be confined to the living areas of the second residence, especially the living room (figure 34). The elevated levels of VOCs in the living room suggest that the source is volatilization of product components from household maintenance products after their intended use rather than volatilization from stored HP.

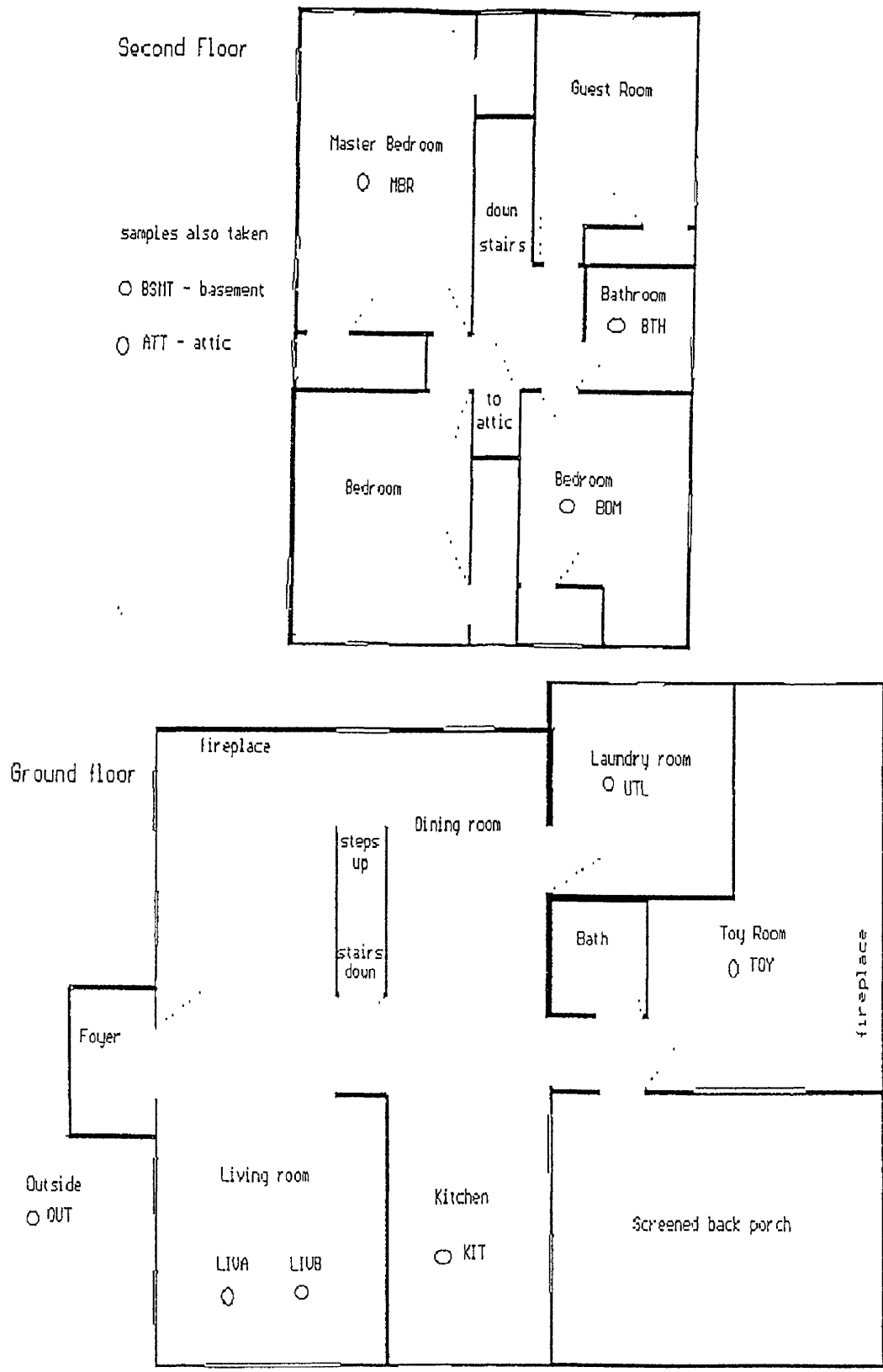


Figure 30. Floorplan and Sampling Locations for Residence 2

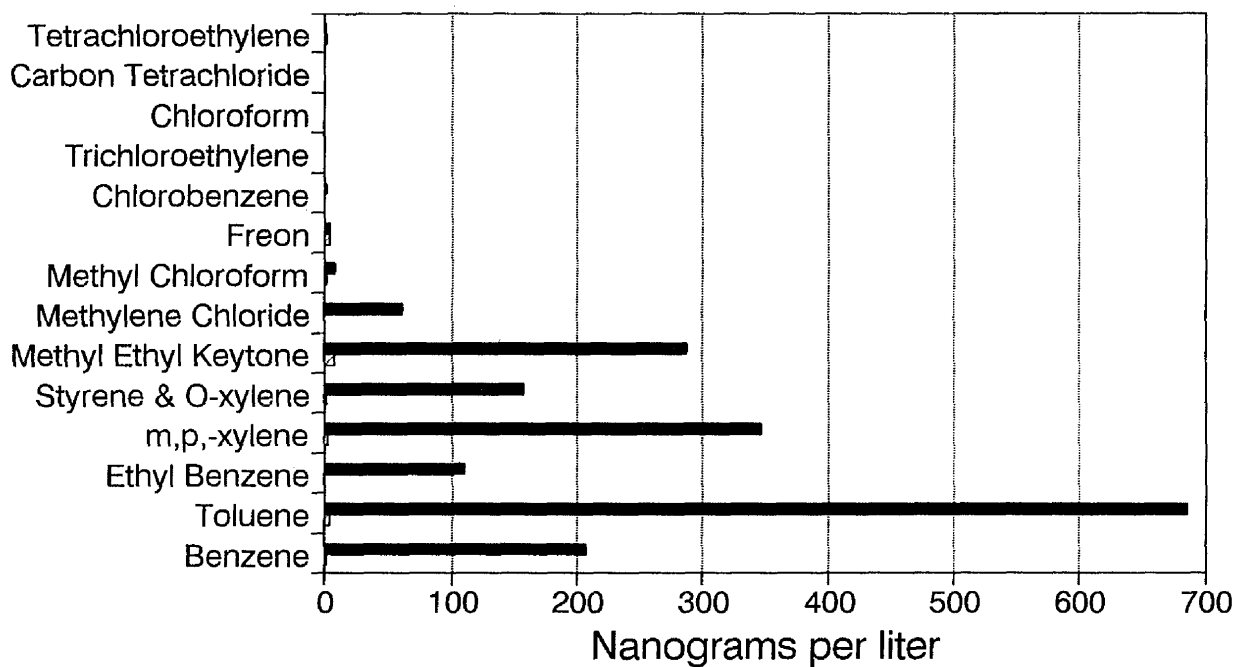
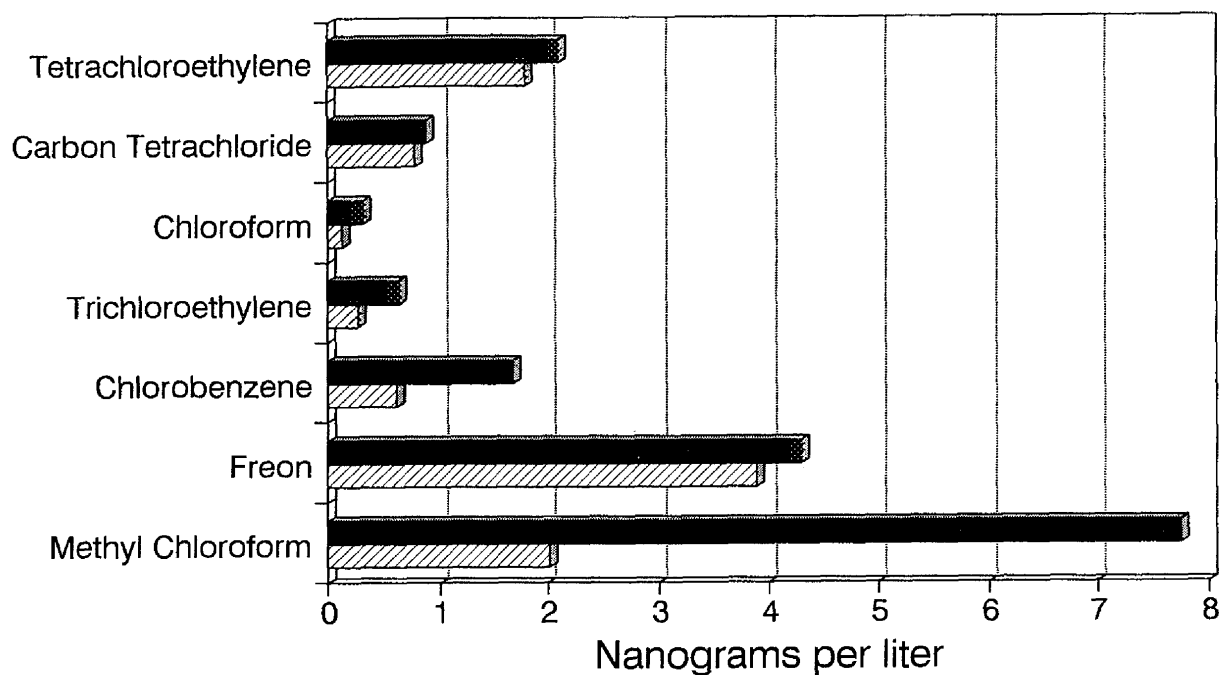


Figure 31. Concentrations of VOCs in the Garage of Residence 2 (1.4 ACH)

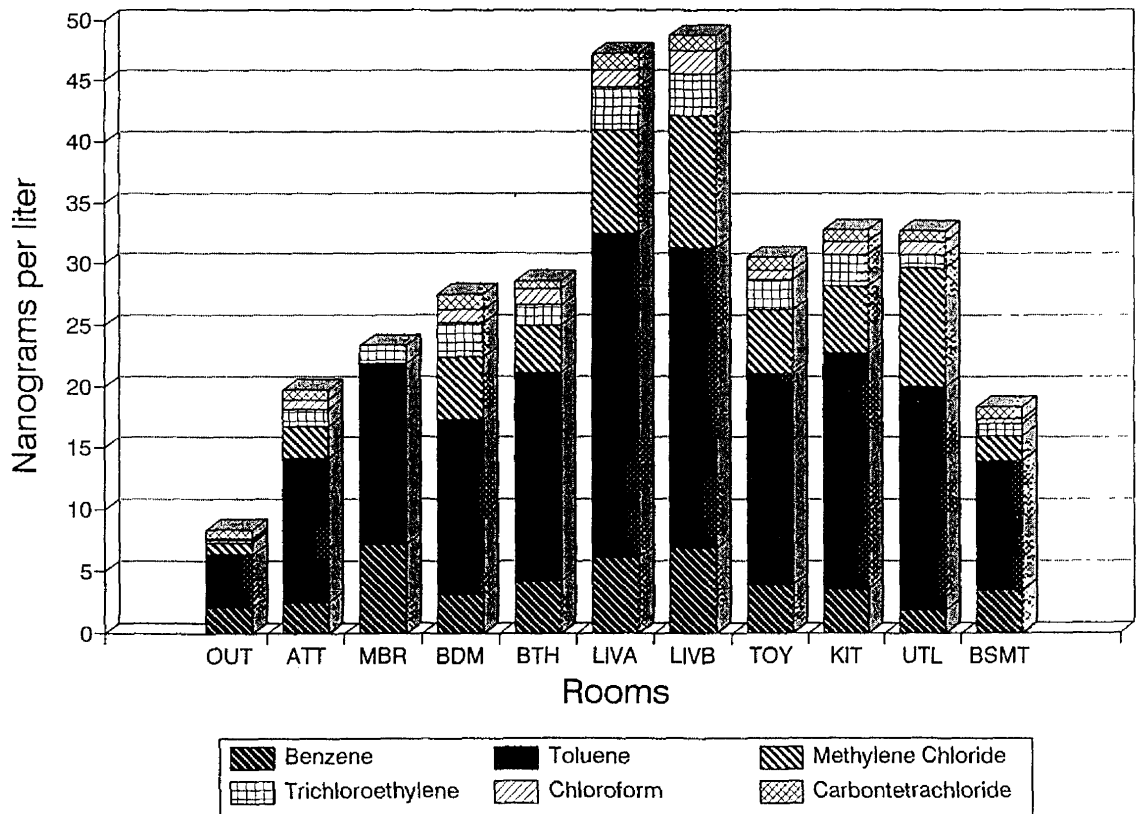
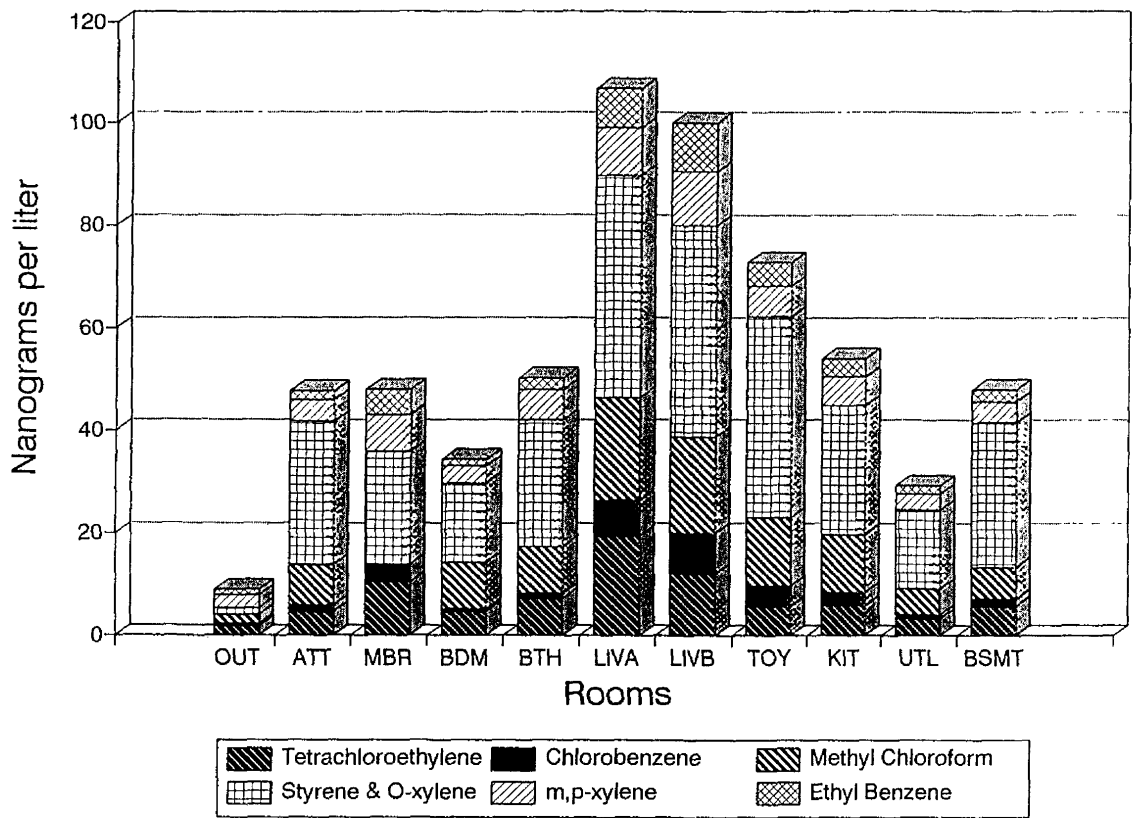


Figure 32. Concentrations of VOCs in the Living Areas of Residence 2 (1.4 ACH)

Residence 3. The third residence (figure 33) that was tested was a new modular home with a separate garage building. The home had been built 10 months before the tests and was never occupied. These tests were conducted during the summer, however the house was closed and air circulation systems were not operating. Outdoor temperatures were about 30°C during the tests. The infiltration rate at this house was intermediate between the first two houses tested (0.62 ACH).

As there were no products in the home, the standard mix of household maintenance products was introduced in the utility area and VOC concentrations were measured at several locations in the house before and after HP introduction. Indoor levels were higher than outdoor levels for most of the compounds. In some cases (benzene, methylene chloride, freon, trichloroethylene, chlorobenzene) chemicals were present at significant concentrations before the introduction of HP indicating that other sources within the home were contributing most of these VOCs to indoor air. This is probably due to volatilization from new building materials, a known source in VOCs in indoor air (Berglund *et al.*, 1987). Figure 34 illustrates the distribution of toluene in the home. Toluene in the indoor air was substantially elevated over the level in outside air before the placement of the HP. After introduction of the HP, indoor toluene concentrations increased by a factor of 2. In the case of methyl chloroform (figure 35), the indoor concentrations measured were essentially the same as those in outside air prior to product introduction. There was a substantial increment after the placement of the HP mixture. These results suggest that both the new building materials and the added HP were sources of VOCs in indoor air in this residence.

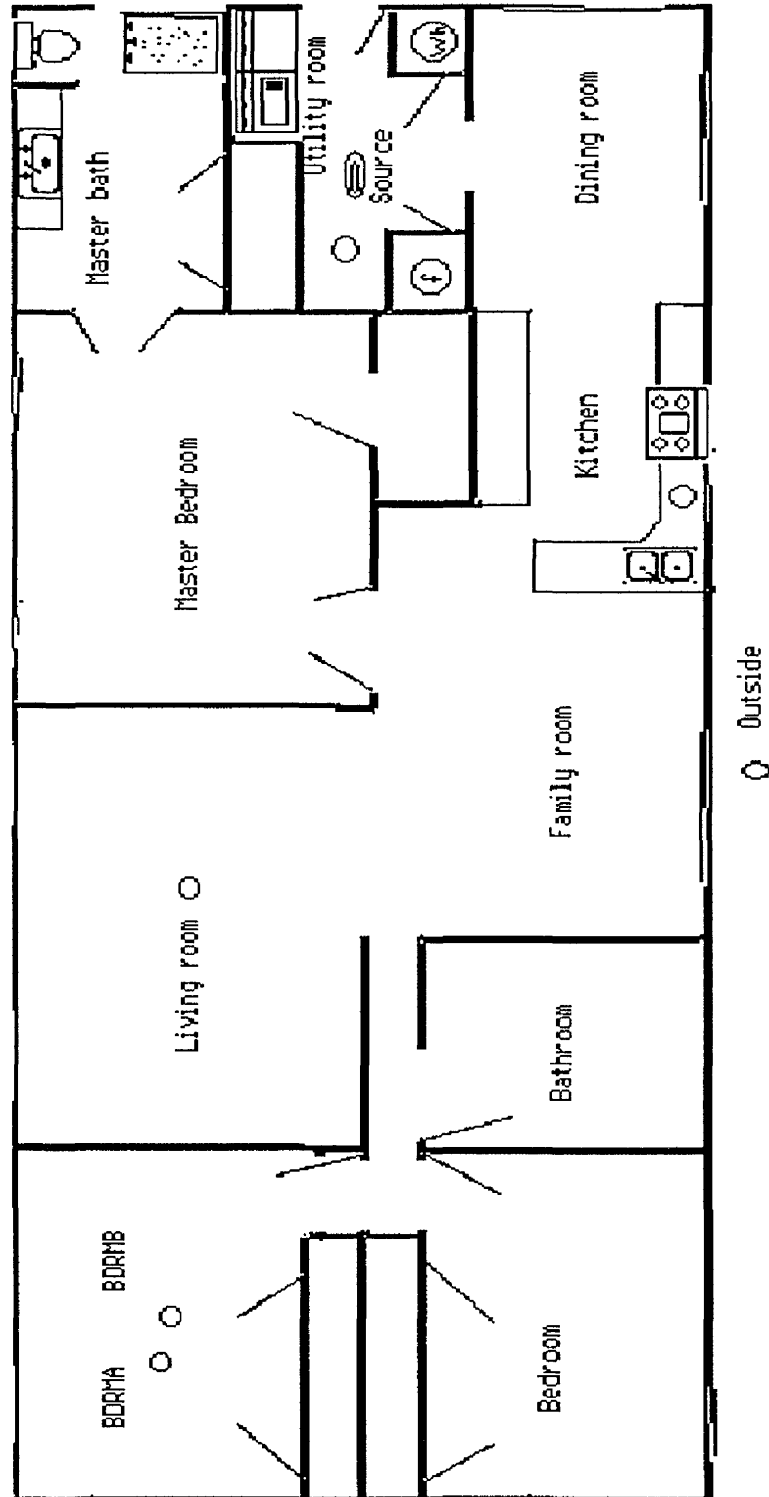


Figure 33. Floorplan and Sampling Locations for Residence 3

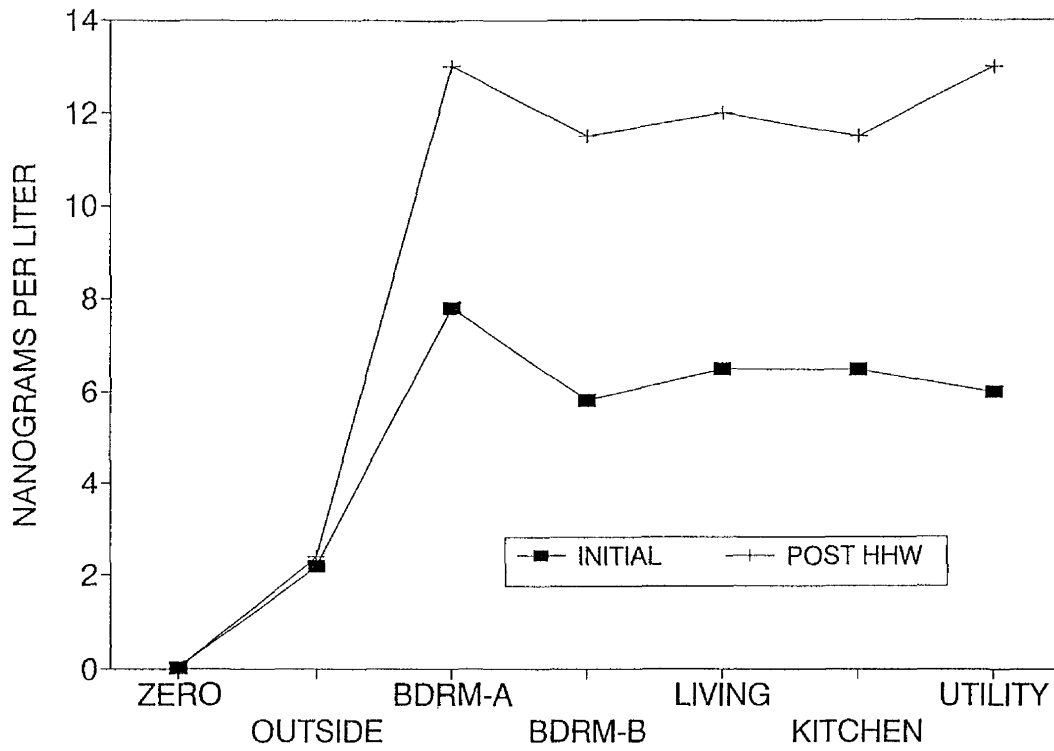


Figure 34. Toluene Concentrations in Residence 3 (0.62 ACH)

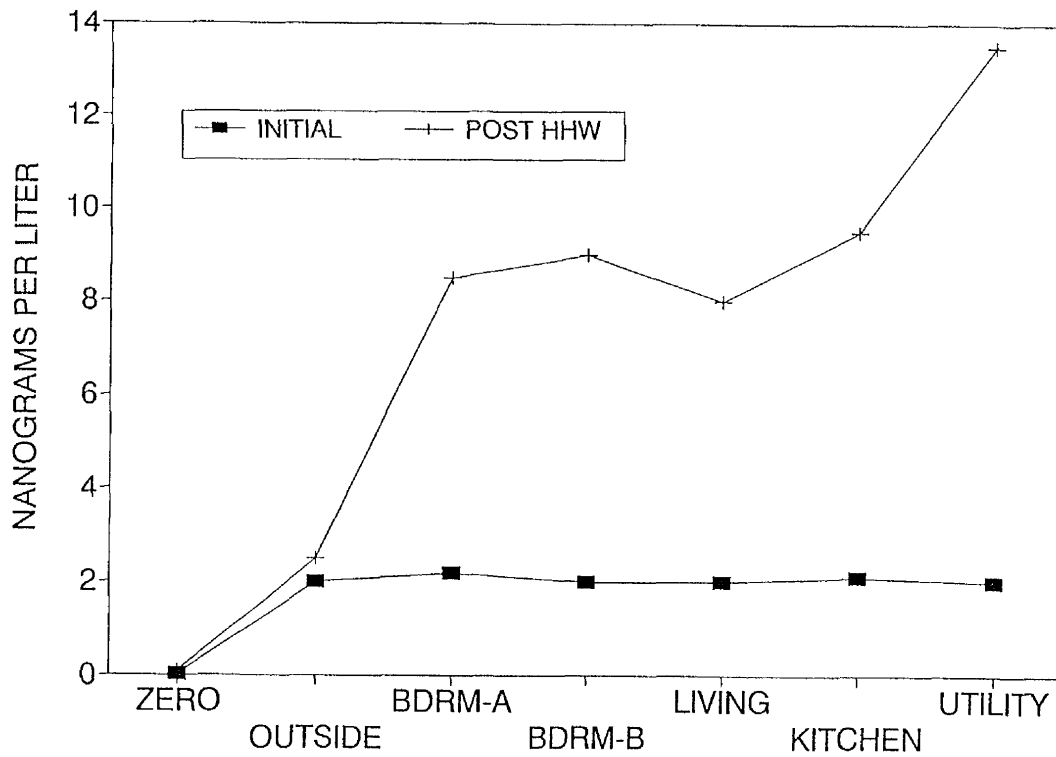


Figure 35. Concentrations of Methyl Chloroform in Residence 3 (0.62 ACH)

CONCLUSIONS AND RECOMMENDATIONS

The test house used in this work allowed for the evaluation of the impact of stored automotive and household maintenance hazardous products (HP) on the concentrations of volatile organic compounds (VOCs) in indoor air without the confounding variables encountered when making measurements in actual homes. Infiltration rates from the outside and exchange between the rooms in the test house could be adjusted to values representative of typical homes in Illinois. To the extent that indoor air quality is dependent on dilution of toxic materials through natural ventilation, then the concentration of these materials must be maintained low enough to ensure indoor air quality at infiltration rates of 0.5 air changes per hour or less.

It is evident from this work that the storage of partially used HP contributes to elevated concentrations of many VOCs that may pose a human health risk. Automotive products are associated with emissions to indoor air of benzene, styrene, m,p,o-xylenes, methyl ethyl ketone, toluene, and ethylbenzene. Household maintenance products are associated with elevated levels of methylene chloride, methyl chloroform, toluene, ethylbenzene, and m,p-xylenes. In both the test house and in actual residences, storage of partially used HP can result in a rapid buildup of VOCs (up to 1000 ng/L) in the rooms used for storage.

In the test house, increases of VOCs in the "living area" (the room adjacent to the "garage" where the products were stored) were associated with product sources in the garage when a connection between the rooms was made. VOC levels were inversely related to infiltration rate with lowest VOC concentrations corresponding to the highest infiltration rate. VOC concentrations peaked one or two days after the product introductions in the garage. VOC concentrations in the living area rose more gradually, approaching the level in the garage after about 4 days. These concentrations were often 100 times greater than initial background levels. Increases in the VOC levels in the living area could be minimized by limiting the exchange of air between the garage and living area.

In actual homes, the concentrations of some VOCs in indoor air were related to the storage of partially used containers of automotive and household maintenance products. For other VOCs, alternate sources were probably responsible for the elevated levels found in indoor air. The highest levels of VOCs were found in the areas where the products were actually stored or used. Where air exchange between the storage area and living areas was limited (detached garage, basement, attached garage in a "tight" house), increases in VOC concentrations attributable to stored HP were minimized.

Recapping a container of many automotive and household maintenance products does not prevent the escape of many toxic VOCs to the air. To minimize the buildup of toxic VOCs in residential indoor air, the storage of used (i.e. seal broken) products in a garage, basement, or other storage area within a residence is not advisable. Ideally only the amount of a product needed to do the job should be purchased. If extra product does need to be stored, it should be in a separate building from living areas. If products must be stored in the same building with the living areas, either low air exchange between the living and storage areas or high overall infiltration rates are needed to prevent VOCs from building up in the living area. In any case, elevated concentrations of VOCs can be expected in the storage area especially when ambient temperatures are high.

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ABSTRACT

Breathing indoor air is an important exposure route and human health risk factor for many toxic chemicals. In this study, the concentrations of toxic volatile organic compounds (VOCs) generated by the storage of hazardous household products (HP) were measured in an experimental test house and in three actual residences. Experiments in the test house indicate that high concentrations (> 100 ng/L) can develop in areas where HP are stored and in attached living areas. The concentrations of individual VOCs in the residences were influenced by stored HP but were also affected by differences in air infiltration rates, temperature, volatilization from building materials, and the presence of other sources such as freshly dry-cleaned clothes or emissions from showers. When infiltration rates in the test house were matched to those measured in the actual residences, the range of measured toxic VOC concentrations was similar. It was concluded that storage and use of HP in homes and in attached storage areas can cause a significant elevation in the levels of toxic VOCs in indoor air and that the test house reflects conditions found in homes in Illinois. It is recommended that the amount of HP stored in the home be minimized by purchasing only the amount of product needed for a particular job. If HP materials must be stored, it should be in an area separate from the living area with high infiltration rates.

EXECUTIVE SUMMARY

Introduction

Research has shown that residential indoor concentrations of toxic volatile organic compounds (VOCs) are up to an order of magnitude higher than outdoor concentrations. It is estimated that Americans normally spend 90% of their time indoors so that most of the health risk from VOCs is from exposure to indoor air. The storage of hazardous household products (HP) in a residence may be a substantial source of indoor air pollution. According to curbside refuse surveys, up to 1% of household waste is household hazardous waste (HHW). For many households the preferred "disposal" option is long-term storage. The objective of this project was to investigate stored HP as a source of VOCs in indoor air.

The sources of indoor air pollution are numerous and complex; but, in general, can be categorized as: (1) building materials and furnishings; (2) use of household products; and (3) automobiles and related products. This study concentrates on automotive and household maintenance products and VOCs generated from these products. Indoor VOC concentrations have been evaluated in test houses with infiltration rates representative of those in houses in Illinois. For comparison, measurements were also made in 3 actual residences.

Test House

The test house used for this work is an experimental structure operated by the Building Research Council at the University of Illinois. The work was carried out in two 8' x 20' x 8' rooms that were individually climate controlled. One room was an unheated exterior room that simulated an attached garage for our study while the second interior room simulated a living area. Air exchange between the rooms and with the outside was regulated using adjustable ports. During the experiments, temperature and air flow were monitored in the house along with outside conditions.

A standard blower door test method was used to determine infiltration in units

of air changes per hour (ACH) in the test house and in actual residences. For the first test period, the test house infiltration rate was 0.2 ACH, equivalent to the best currently used energy efficient construction technology. This represents a "worst case" situation for indoor air pollution because a minimum of outside air passes through the house to dilute emissions of VOCs. The actual residences had infiltration rates between 0.3 and 1.4 ACH. These values are typical of the range of infiltration rates found in homes in Illinois. In a second round of experiments in the test house, infiltration rates were set between 0.6 and 1.2 ACH to better reflect average "real world" conditions.

Methods

The HP were introduced into the test house in the "garage". The HP consisted of standard containers of automotive products (gasoline, carburetor cleaner, brake fluid, etc.) or household maintenance products (cleaning products, paint, paint stripper, caulking, adhesives, etc.). The containers were opened then resealed and placed in a plastic tub for the experiments. In the first phase of the work, both HP groups were evaluated. Only automotive products were tested in the later phases because the household maintenance products were found to emit lower amounts of mostly the same VOCs.

Sampling was carried out using 6L Summa-polished stainless steel canisters. Time-averaged, 2-hour samples were collected using a 30 gauge needle as a critical orifice. VOCs in the air samples were cryogenically concentrated in a liquid nitrogen trap and analyzed by gas chromatography using simultaneous electron capture and flame ionization detection. The target compounds were 15 toxic VOCs including several polychlorinated solvents, benzene, and freon.

Test House - Phase I

During the first part of this work, the test house was in a "tight" configuration with minimum infiltration rates (0.2 ACH). VOCs reached maximal concentrations

much above background levels in the garage within 5 hours. Concentrations of VOCs in the living area were only slightly above those in outdoor air. When small ports between the garage and living area were opened to allow air exchange between the rooms, levels of VOCs in the living area increased well above those in outdoor air.

In the next part of the work, the infiltration rate was left at 0.2 ACH and the ports between the rooms in the test house were left open. Both automotive and household maintenance HP were stored in the garage for 16-day periods. Under these conditions, VOC concentrations exhibited one of three patterns. Some VOCs reached peak concentrations of up to 1000 ng/L or more in the garage section of the test house within 24 hours, and then slowly declined over a 16-day period. Concentrations of these VOCs in the living area followed the same trend but were initially about half as much as those in the garage. Other VOCs including carbon tetrachloride, chlorobenzene, trichloroethylene, and chloroform varied with outside air levels indicating the products tested were not significant sources for these chemicals. Finally, concentrations of freon were consistently higher in the living area of the test house even before the introduction of the products. This pattern indicates a source in the living area of the test house (air conditioner). Concentrations of the VOCs emitted by the stored HP usually reached a plateau within the first 4 days of the storage period and subsequent fluctuations were attributed to changes in temperature.

Test House - Phases II and III

During the next part of this work, the infiltration rate through the test house rooms was increased to 0.56 and 1.2 ACH by opening additional ports to the outside. The opening between the garage and living area remained the same as in the previous experiments. The intent was to determine the influence of an increased infiltration rate on the movement of VOCs from the garage to the living area and the relationship of infiltration rate to the overall concentration of VOCs that builds up in the test house. As before, the automotive product mix was placed in the garage simulating the storage of partially used automotive products. The maximum VOC concentrations

measured in the garage reached approximately half of comparable levels measured with the tighter configuration (0.2 ACH). VOCs in the living area were decreased by up to 50% at 0.56 ACH and by 90% at 1.2 ACH. It is apparent from the data that a larger infiltration rate limits the buildup of VOCs in both the garage and living area.

Tests in Actual Residences

VOC concentrations in 3 actual residences were measured. These houses had infiltration rates that reflected the range found in Illinois housing. A new house was tested before and after the introduction of the household maintenance product mixture. Although the home with the highest infiltration rate generally had lower concentrations of VOCs, there was not a consistent difference. The pattern of VOC concentrations indicates that there were sources other than stored HP including building materials, tap water emissions (showers), and freshly dry-cleaned clothes. In some cases, HP clearly contributed to the VOC levels measured in the residences especially in the rooms where the HP was stored and used.

Conclusions and Recommendations

The test house used in this work allowed for the evaluation of the impact of stored automotive and household maintenance hazardous products (HP) on concentrations of volatile organic compounds (VOCs) in indoor air without the confounding variables encountered when making measurements in actual homes. Infiltration rates from the outside and exchange between the rooms in the test house could be adjusted to values representative of typical homes in Illinois. To the extent that indoor air quality is dependent on dilution of toxic materials through natural ventilation, then the concentration of these materials must be maintained low enough to ensure indoor air quality at infiltration rates of 0.5 air changes per hour or less.

It is evident from this work that the storage of partially used HP contributes to elevated concentrations of many VOCs that may pose a human health risk. Automotive products are associated with emissions to indoor air of benzene, styrene,

m,p,o-xylenes, methyl ethyl ketone, toluene, and ethylbenzene. Household maintenance products are associated with elevated levels of methylene chloride, methyl chloroform, toluene, ethylbenzene, and m,p-xylenes. In both the test house and in actual residences, storage of partially used HP can result in a rapid buildup of VOCs (up to 1000 ng/L) in the rooms used for storage.

In the test house, increases of VOCs in the "living area" were associated with product sources in the "garage" when a connection between the rooms was made. VOC levels were inversely related to infiltration rate with lowest VOC concentrations corresponding to the highest infiltration rate. VOC concentrations peaked one or two days after the product introductions in the garage. VOC concentrations in the living area rose more gradually, approaching the level in the garage after about 4 days. These concentrations were often 100 times greater than initial background levels. Increases in the VOC levels in the living area could be minimized by limiting the exchange of air between the garage and living area.

In actual homes, the concentrations of some VOCs in indoor air were related to the storage of partially used containers of automotive and household maintenance products. For other VOCs, alternate sources were probably responsible for the elevated levels found in indoor air. The highest levels of VOCs were found in the areas where the products were actually stored or used. Where air exchange between the storage area and living areas was limited, increases in VOC concentrations attributable to stored HP were minimized.

Recapping a container of many automotive and household maintenance products does not prevent the escape of many toxic VOCs to the air. To minimize the buildup of toxic VOCs in residential indoor air, the storage of used (i.e. seal broken) products in a garage, basement, or other storage area within a residence is not advisable. Ideally only the amount of a product needed to do the job should be purchased. If extra product does need to be stored, it should be in a separate building

from living areas. If products must be stored in the same building with the living areas, either low air exchange between the living and storage areas or high overall infiltration rates are needed to prevent VOCs from building up in the living area. In any case, elevated concentrations of VOCs can be expected in the storage area especially when ambient temperatures are high.

ENGINEERING STUDIES

Simulation in Test Cells under Natural Conditions

The Building Research Laboratory Building was constructed for field testing of building assemblies. This "test house" contains ten study bays, which were constructed for uniformity of workmanship. Each bay measures 8' (2.3m) by 20' (6.0m) in surface area. The ceiling height is 8' (2.3 m) and the interior volume is 1280 ft³ (31.8 m³). The building is on a crawl space foundation. Materials and workmanship a similar in all of the bays.

The aim in using the test house was to have a simple test system with air exchange similar to that in actual houses. In this work, bay 9 was considered the "garage" and bay 8 was considered the "living area" (figure 1). Each bay had openings to the outside, which were 3" (7.6 cm) diameter PVC tubing. To reduce excessive air flows through the tubing, fiberglass insulation buffers were added. There was also 1.5" (3.8 cm) diameter PVC tubing between bays 8 and 9 with fittings that permitted the size of the opening to be reduced in 0.5" steps down to 0.5" (1.3 cm) in diameter.

The bays were individually heated, cooled, and humidified. Bay 8 was maintained at 70°F (21°C) during the winter and 75°F (24°C). Bay 9 was not heated or cooled. Temperature was monitored in the 2 bays, the crawl space, and outdoors. Airflow measurements through the port openings and air pressure difference measurements between the bays were also taken. Weather data (outdoor temperature, wind speed, wind direction, and solar insolation) were collected throughout the study from a weather station located at the Building Research Laboratory. Figure 2 provides an example of a typical day's weather data.

Infiltration is the uncontrolled flow of air through unintentional openings driven by wind, temperature difference, and/or appliance-induced pressures across the building shell. Air leakage is a measure of the air tightness of the building shell, and

is determined by the total area and placement of the unintentional openings. Greater air leakage area causes higher a infiltration rate. Infiltration, the air entering the structure, is always equal to the exfiltration, the air leaving the structure. A useful measure of infiltration is air changes per hour (ACH).

Bays 8 and 9 of the Research laboratory were tested for air infiltration using a blower door test and calculating an equivalent leakage area (ELA) and infiltration rate (ACH). With ports to the outside closed, both bays had measured air exchange rates of 0.2 ACH. The only evident air leakage paths were those used for routing instrument cables. This configuration was used for the initial tests.

Later in the project, additional holes were installed between the outdoors and Bay 8. The purpose of these holes were to allow for configuring the test house to approximately replicate the air change rates found in actual homes in Illinois. An additional 3" PVC pipe was installed in the north wall, and two 3" PVC pipes were installed in the south wall. Plastic inserts, with holes of varying diameter in the center, were fabricated to fit within the 3" PVC pipes. Bay 8 was then tested for ACH under different hole configurations using a blower door test. The selected hole configurations for various phases of the work are shown in Table 1.

Table 1. 1994 Test Building Configuration

Test Number	Bay 8 Configuration	Infiltration Rate	Corresponding Residence
Phases I	no holes	0.20 ACH	Residence #1
Phases III	3" hole, north and south	0.56 ACH	Residence #3
Phases II	two 3" holes, north and south	1.20 ACH	Residence #2

Although the test house can be calibrated and tested to provide an estimated infiltration rate, there exists a qualitative difference between the test house and typical field conditions. The total air leakage area in a residence is comprised of many small

CONCLUSIONS AND RECOMMENDATIONS

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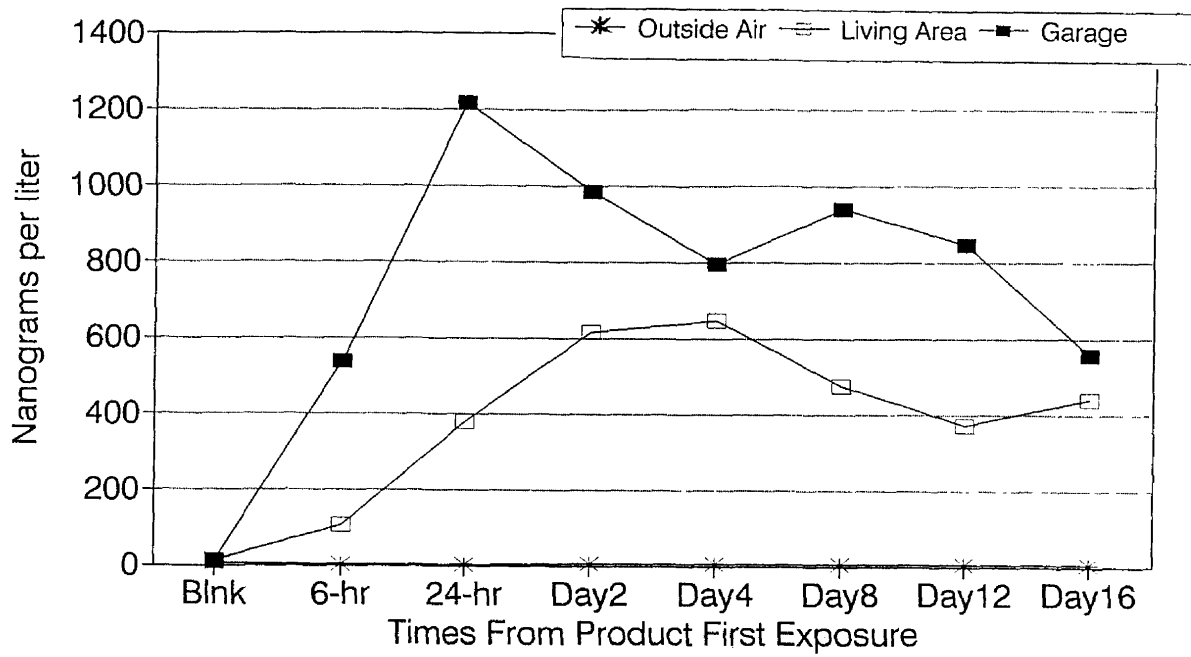


Figure 5. Concentrations of Toluene from Automotive Products in the Test House "Garage" (0.2 ACH)

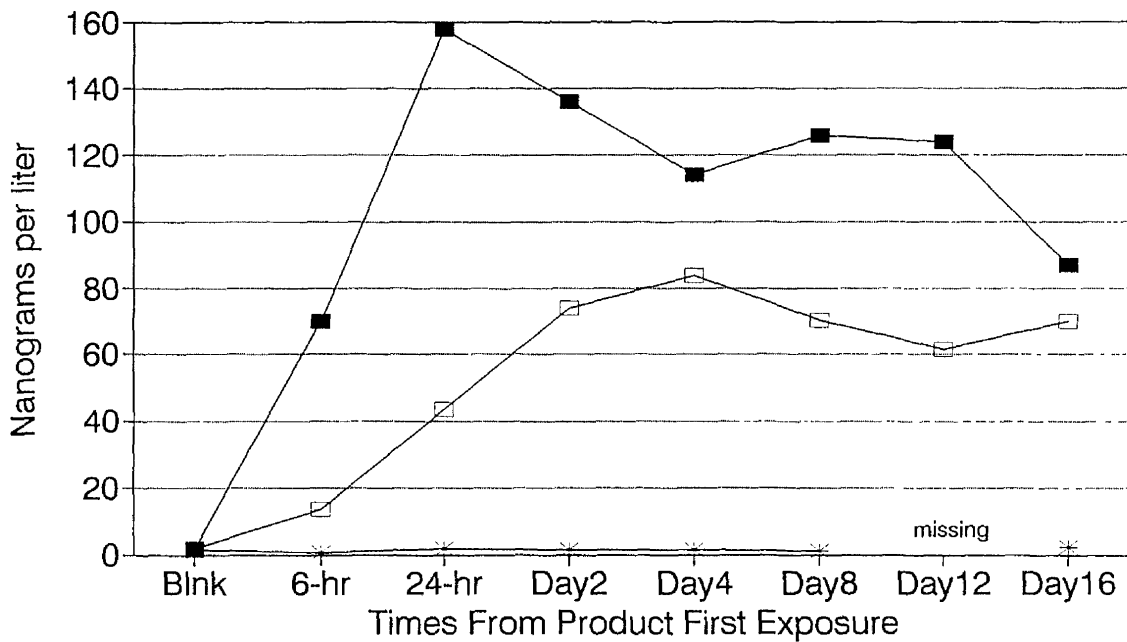


Figure 6. Concentrations of Ethylbenzene from Automotive Products in the Test House "Garage" (0.2 ACH)

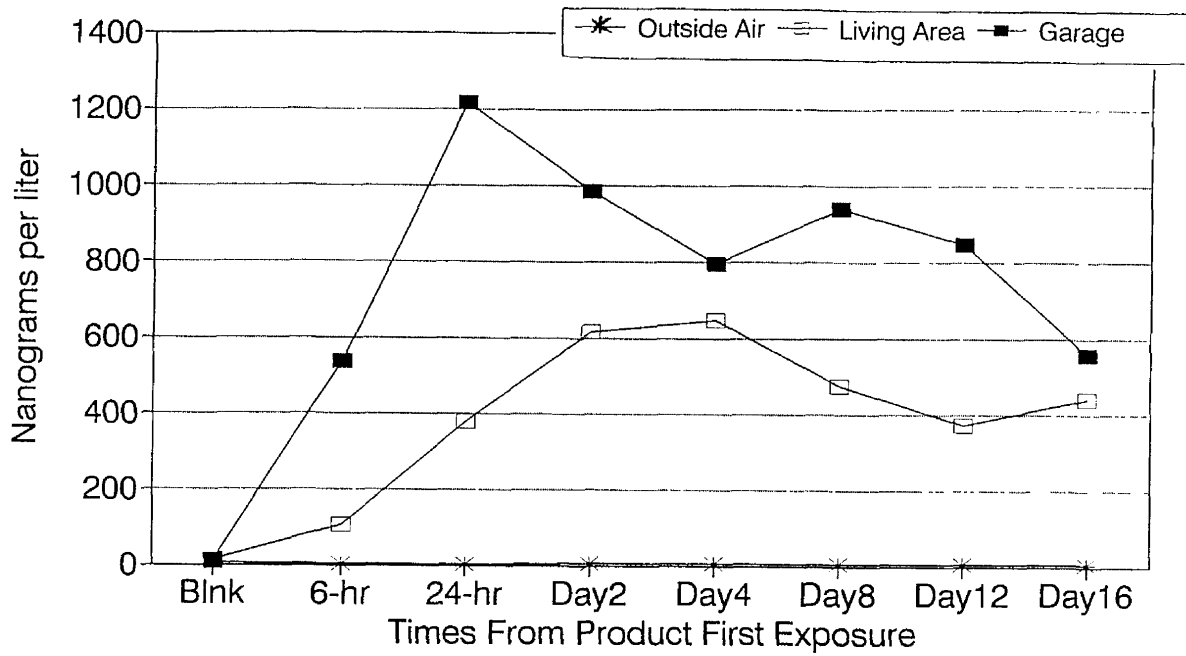


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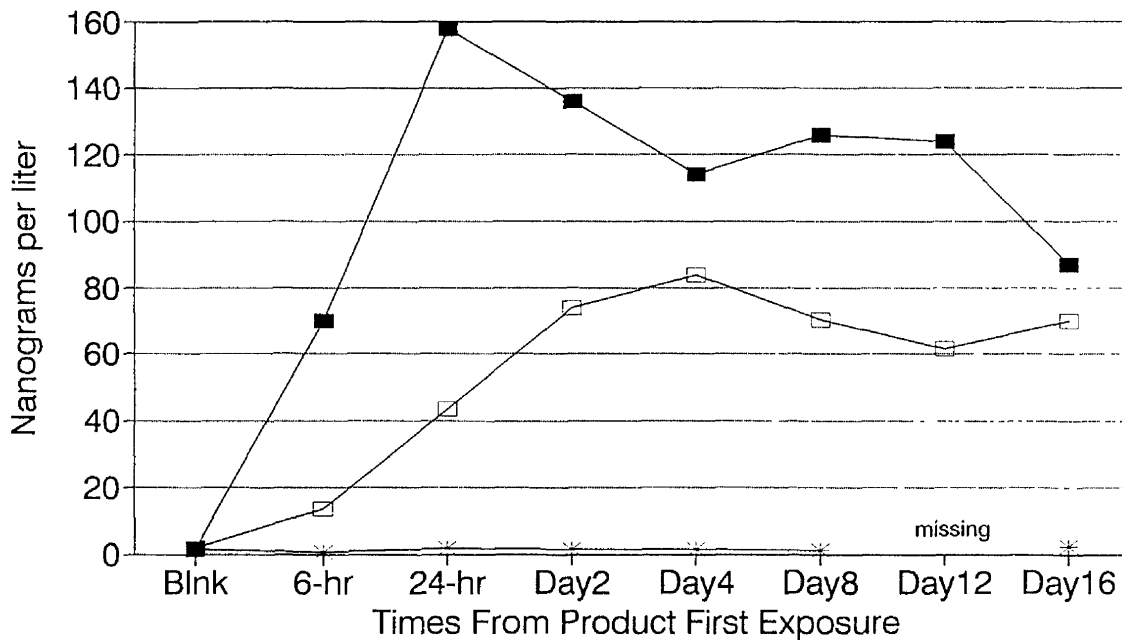


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