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Indoor Air VOC Concentrations in Suburban and Rural New Jersey

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Abstract

Indoor VOC air concentrations of many compounds are higher than outdoor concentrations due to indoor sources. However, most studies have measured residential indoor air in urban centers so the typical indoor air levels in suburban and rural regions have not been well characterized. Indoor VOC air concentrations were measured in 100 homes in suburban and rural areas in NJ to provide background levels for investigations of the impact from subsurface contamination sources. Of the 57 target compounds, 23 were not detected in any of the homes, and 14 compounds were detected in at least 50% of the homes with detection limits of \sim 1 μ g/m³. The common compounds identified included aromatic and aliphatic hydrocarbons from mobile sources, halogenated hydrocarbons commonly used in consumer products or from chlorinated drinking water, acetone and 2-butanone emitted from cosmetic products, and Freons. Typical concentrations were in the low *μ*g/m³ range, though values of tens, hundreds or even thousands of *μ*g/m³ were measured in individual homes in which activities related to specific sources of VOCs were reported. Compounds with known similar sources were highly correlated. The levels observed are consistent with concentrations found in the air of urban homes.

Introduction

Indoor air concentrations have been reported for homes in the U.S., Canada, and Europe in a number of large studies such as TEAM, NHEXAS, TEACH, EXPOLIS, and RIOPA, where the majority of the homes sampled, though not all, have been in urban settings (1-14). There have also been smaller studies examining indoor air, again focusing on homes in cities (15-20). Two studies that have reported indoor VOC air concentration in rural regions were the TEAM study done in North Dakota (1, 21) and part of the Minnesota Children's Pesticide Exposure Study which included the measurement of 10 VOCs (22). When paired indoor–outdoor measurements exist, it has been documented that ambient air levels of VOCs surrounding the home provides a baseline for the indoor air concentration that is augmented by contributions from indoor sources. In some cases indoor sources of VOCs are the

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Supporting Information Available

The correlation matrix. This material is available free of charge via the Internet at<http://pubs.acs.org>.

dominant sources contributing to their indoor concentrations. Thus, while the ambient air levels of VOCs in suburban and rural settings are expected to be lower than in urban settings, the concentration of many VOCs in homes within those settings may not be. An additional source of VOCs in indoor air is vapor intrusion of volatile compounds from contaminated groundwater (23, 24). The most frequently measured VOCs in indoor air are chlorinated hydrocarbons and aromatic hydrocarbons. However, there are additional VOCs that are often regulated as groundwater contaminants but have been rarely measured in indoor air. The current study measured indoor air concentrations of VOCs in suburban and rural homes where no known groundwater or soil contamination was present. These measured background concentrations should be taken into account when assessing potential impacts of vapor intrusion from contaminated groundwater on indoor air quality.

Experimental Section

One hundred homes were selected from 13 counties across the state of New Jersey using a convenience sampling approach. The selection criteria were (1) single family or semiattached homes in municipalities with population densities of less than 4000 people/ square mile (suburban and rural areas); and (2) not located within 0.8 km of known or reported contaminated groundwater plumes or soil or within 330mof gasoline stations with leaking storage tanks. Samples were collected for 24 h using a canister sampler placed on the ground floor of the home in an actively used living space other than a kitchen. The kitchen was excluded to minimize collection of compounds emitted during cooking. A questionnaire was administered before sampling to identify potential indoor sources of the target compounds. Each house location was compared against geographic information systems (GIS) databases containing the population density and existing NJ Department of Environmental Protection GIS databases containing locations of contaminated groundwater and soil sites in New Jersey to verify that the selection criteria were met.

Subjects were asked to keep the windows and doors closed (except when entering or leaving the home), and not to use an exhaust fan or air conditioner (except on a recirculation mode), fireplaces, kerosene heaters, or gasoline powered equipment within the house during the sampling in accordance with procedures used during New Jersey vapor intrusion investigations.

Subject Recruitment

Contact for potential participants was made using three approaches: direct contact with members and staff at a science institute (EOHSI), through solicitation by Agricultural Cooperative Extension Agents of Rutgers University at their facilities and local meetings, classes or workshops, and via word of mouth from the existing participants in the study. In all cases informed consent was obtained from each subject and all protocols were approved and reviewed annually by the UMDNJ/RWJMS Institutional Review Board for Human Subjects.

Sample Collection and Analysis

Indoor air samples were collected and analyzed for 58 volatile organic compounds (Table 1) using U.S. Environmental Protection Agency (USEPA) Method TO-15 (25), a canisterbased collection method coupled with GC/MS analysis. The canisters were analyzed by STL Burlington, a NJ Department of Environmental Protection (NJDEP) certified contract laboratory. A 6 L Summa canister, with a flow controller, was used. The inlet of the sampler was placed at breathing zone height (3–5 ft). A nominal 24 hour sample was collected, with the exact duration of the sample recorded. A pressure gauge was used to ascertain the integrity of the vacuum prior to sampling and when the canister was retrieved. Samples were collected in most months between December 2003 and April 2006.

Six pairs of samples were collected in duplicate, approximately after every 20 samples, to assess the overall precision. A significant number of compounds in all of the duplicate pairs, and samples throughout the study, were below the detection limit, typically between 0.5 and $3 \mu g/m^3$. The agreement of the measurements above the MDL for paired samples was within 30%. In addition, most pairs with one value below detection had the second value also below or only slightly above detection (within 30% of the detection limit). The exceptions to the ±30% agreement were a single sample pair for acetone and a single pair for Freon. Additional quality control procedures that are part of the NJDEP certification on the TO-15 method include the use of certified standards, daily performance checks, calibration checks, analysis of method blanks, instrument blanks and quality control samples.

Questionnaire

A short questionnaire was developed to ascertain the presence of possible indoor sources of compounds being measured. The questions focused on general information about the home (e.g., number of floors, age, type of heating, when renovations were last done), and typical products located or used in home (e.g., smoking, presence of pets, cleaning or purchase of furniture, painting) that could release the target VOCs.

Source Analysis

Correlation analyses, factor analysis and general linear model (GLM) univariate analysis were used to evaluate sources of compounds measured in the homes (SPSS Version 13.01 for Windows). The concentration data were consistent with log-normal distributions. Therefore, a Spearman correlation matrix, which does not assume a normal distribution, was calculated (Supporting Information). The options selected for the factor analysis were principal component method with varimax rotation, to optimize the factor selection, and a maximum of 25 iterations for convergence. A dummy variable indicating when the detection limit changed in the study was also included in the factor analysis. The program's default values were used in selecting which factors to retain in the final analysis combined with a visual analysis of the scree plot to select the number of factors where the eigenvector value dropped below unity and the values began to plateau. Seven factors were selected based on a cutoff of the eigenvalues declining to below unity, though the declines in the eigenvalues were not sharp. While the inclusion of compounds with large numbers of MDLs could result in factor(s) solely because most samples concentrations were below the MDL, it could also generate unique factors that had distinct indoor sources in a small number of homes. For the

GLM univariate analysis each compound's concentration was used as the dependent variable and selected questionnaire responses were used as fixed factors independent variables. A full factorial model was run that included factor by factor interactions; significance level determined for the F-statistic test; type III sum of squares; and no contrasts specified. All questionnaire responses were solely binary (yes/no), so no posthoc tests were run.

Results and Discussion

Concentrations

Summary statistics from the one hundred homes are given in Table 1. Thirty two homes are classified as rural (<1 dwelling unit per acre) (26) and 68 homes suburban. Many of the compounds had the vast majority of the concentrations below the detection limit. The detection limit of some compounds was improved approximately halfway through the project resulting in two different detection limits reported. Compounds routinely detected above the detection limit in >50% of the homes were acetone, benzene, 2-butanone, chloromethane, dichlorofluoromethane, ethyl benzene, *n*-heptane, *n*-hexane, methyl *tert* butyl ether (MTBE), toluene, trichlorofluoromethane (Freon 11), 1,2,4 trimethylbenzene, m/p xylenes and *o* xylene. Compounds above the detection limit in >10% but <50% of the homes were: chloroform, cyclohexane, 1,4 dichlorobenzene, ethyl benzene, *p* ethyltoluene, methylene chloride, 4 methyl 1,2 pentanone (MIBK), styrene, tetrachloroethene (PERC), 111 trichloroethane, 1,3,5 trimethylbenzene, and 2,2,4 trimethylpentane. Many of the measurable compounds have been reported previously to have indoor sources (e.g., emissions from automobiles parked in a garage attached to a house, tobacco use, cleaning products, dry cleaning agents, air fresheners, paints, home furnishing, and volatilization from chlorinated tap water) (1, 2, 12, 27) or are at quantifiable levels in outdoor air due to emissions from mobile, industrial, and commercial sources (14).

Source Evaluation

Potential similarities in composition and sources of the compounds to the indoor air were evaluated using correlation coefficients, factor analysis, and GLM univariate analysis. These complementary approaches provide information on which compound concentrations vary together and what personnel activities or housing properties contribute to the concentration's variability. Compounds which had measurable levels in at least 10% of the homes were included, with the MDL level used when the concentration was below detection. This approach can result in false correlations when the percent of values below detection is large. Scatter plots of correlated compounds were visually examined to evaluate if the correlation might be an artifact of a large number of samples being below the MDL.

Correlation Analysis

As other studies have reported (14, 28, 29), aromatic compounds, alkanes and MTBE, compounds with mobile sources, were significantly correlated $(p < .001)$ with each other, with correlation coefficients (R^2) for MTBE, BTEX, and more highly substituted aromatic compounds generally exceeding 0.7 and >0.9 for o-xylene with ethyl benzene and for 4 ethyltoluene with 1,2,4 trimethylbenzene. Correlation coefficients among alkanes, and between alkanes and aromatic compounds were statistically significant with values of 0.3–

0.6. Other compound pairs with common sources that were highly correlated were acetone and 2-butanone, both components in nail polish remover (0.536), and Freon 11 and Freon 12 from refrigeration and air conditioning units (0.407). However, correlation coefficients exceeding 0.4 were also calculated for compounds expected to come from different indoor sources. One possible explanation for the highly statistically significant correlation coefficients for the indoor air concentrations of compounds with different indoor sources is their concentrations are affected by the air exchange rate in a similar fashion. Thus their indoor air concentrations vary together, i.e., are correlated.

Factor Analysis

Factor analysis groups highly intercorrelated variables together in representative factors. It is possible to suggest sources for different factors if the compounds assigned to those factors are known to have common sources. Factor analyses of VOC data collected in the EXPOLIS study identified mobile sources, transport of outdoor air indoors, environmental tobacco smoke, emissions from plastic, paint, building material biologics, emissions from carpets, rubber and adhesives, and cleaning products as potential sources (30). Seven factors were identified for the VOC indoor air concentrations measured in this study (Table 2), several having similar compounds groupings as found for the EXPOLIS data. The first factor includes aromatic compounds and MTBE while the second factor includes benzene, MTBE, and alkanes. The most likely source of these compounds is emission from gasoline engines. These compounds also come from environmental tobacco smoke (28) but there were few smokers in the study homes. The first factor could be associated with transport of ambient air that contains mobile sources emissions or emissions from automobiles or other gasoline powered engines in attached garages. The inclusion of more volatile gasoline components in the second factor suggests a stronger association with evaporative emissions rather than exhaust emissions. Evaporative emissions are important for homes with attached garages. The third factor contains compounds present in cleaning products or added as fragrances to consumer products. The fourth factor includes the two Freon compounds indicative of a refrigeration or air-conditioning source. The fifth factor contains chloroform and styrene which have different indoor sources. The combination of these two compounds is likely an artifact of both having the majority of the samples below their detection limits. The sixth factor includes acetone and 2-butanone, compounds commonly present in nail polish and nail polish remover. The last factor includes only tetrachloroethene, the common drycleaning compound.

GLM Univariate Analysis

A GLM Univariate Analysis of Variance (ANOVA) was run to evaluate the effect of housing characteristics or participant's activity as reported in the questionnaire on the indoor air concentrations of individual VOCs. The questions included in each compound's analysis were based on a-priori assumptions about what indoor sources and housing conditions would alter that compound's indoor air concentration. The *p* values from the F-test indicate whether there are statistical differences in the mean compound concentrations with the presence or absence of the housing characteristic based on the questionnaire response (Table 3). The aromatic compounds, alkanes, and MTBE, which have emissions from gasoline considered presences/absence of an attached garage; a doorway between the garage and

WEISEL et al. Page 6

house; a car or other gasoline powered engines in the garage during sampling; the location of the room with sampler relative to the garage and the age of the house. For these compounds, the location of the garage relative to the room sampled, presence of a doorway between the garage and house or a car being in the garage, and the age of the home were generally included $(p < 0.05)$ in the GLM univariate models. There was a significant interactions term between the age of the home and having an attached garage consistent with older homes (pre 1950s) generally not built with attached garages. Elevated levels of aromatic compounds have been reported in homes with attached garages (22, 31, 32).

For the compounds methylene chloride, 1,1,1 trichloroethane, and 1,4 dichlorobenzene, which are in cleaning agents and/or deodorizers, questions about use of deodorizers, mothballs, room fresheners, paint thinner/stripper were examined. For 1,4 dichlorobenzene, the use mothballs, (but not deodorizers nor scented candles), age of home, and factor interaction effects were statistically significant. For 1,1,1, trichloroethane, only the age of home was significant, with concentrations increasing with home age, the reverse of what would be expected if older homes are less airtight and have higher air exchange rates. Source apportionment models have suggested that indoor use of cleaning products can contribute to personal exposure for 1,1,1, trichloroethane (33, 34). For methylene chloride no variables reached statistical significance.

For chloroform when the MDL changed in the analyses was the only significant variable, but use of a public water supply (which are typically chlorinated) had a *p*-value of 0.094 just below significance. Previous studies have found use of chlorinated water and bleach contribute to indoor air chloroform concentrations (35).

For acetone, age of home was significant and the use of nail polish had a *p*-value of 0.051, with an interaction between the two being significant. For 2-butanone age of home and use of nail polish were just below significance with *p*-values of 0.085 and 0.072, respectively. The lack of statistical significant may be because not all nail polish and nail polish removers contain acetone and 2-butanone and these products are used for a relatively short time period (minutes) compared to the sampling duration of 24 h. Additionally, it is not known if these products were used in the same room where the sample was collected.

For tetrachloroethene, bringing dry cleaning into a home and the sample MDL were statistically significant, while use of paint thinner was just below significance, *p*-value of 0.084. The interactive terms between dry cleaning and paint thinner use and for paint thinner use and age of home were statistically significant. Paint thinner was used in a limited number of homes including the home with the maximum tetrachloroethene concentration. Bringing dry cleaning into homes has been shown to increase the average indoor air concentration of tetrachloroethene (36).

The univariate analysis confirmed the likely source assignments made from the factor analysis, indicative of the activities or housing conditions that lead to potential elevated indoor air concentrations in homes in suburban and rural areas.

Comparison to Other Data Bases

The median and 90th percentile indoor air concentrations measured in this study are compared to values reported in the literature from urban studies done in NJ (TEAM in the 1980s and RIOPA in 1999–2001), and other indoor air studies done within the past decade (Table 4). Among the three studies in NJ, the concentrations measured during the TEAM study are generally much higher. This reflects the higher ambient levels and greater presence of these compounds in consumer products and fewer emission controls on automobiles in the 1980s than currently exist. The RIOPA indoor air concentrations from an urban center in Elizabeth, NJ during 1999–2001, are comparable or slightly lower than the concentrations measured in the suburban and rural NJ homes of this study. Since the median and mean indoor air concentrations measured during the RIOPA study were slightly elevated relative to the outdoor levels for most of the compounds measured, the similarities in the indoor air concentration suggest similar indoor sources of compounds across the state, in urban and the suburban/rural settings. Overall, the values reported from various studies are very similar suggesting that indoor emissions from consumer products are a critical source for the air concentration indoors for VOCs.

Implications, limitations, and future needs

Indoor air concentrations in rural and suburban residences are highly affected by indoor sources so while outdoor levels in these locations may be lower for many VOCs than in urban settings, indoor levels are not. Identifying the sources of VOCs in indoor air and the resulting exposures becomes particularly important for contaminants whose air concentrations exceed health-based indoor screening levels (39, 40). One limitation in this study was that detection limits were above health-based indoor air screening levels for many carcinogens so additional contaminants not detected in this study may be present above screening levels. As detection limits improve and the list of contaminants of concern continues to grow more data on background indoor air concentrations and exposure may be needed to implement effective remediation strategies to remediate the sources of contaminants that affect indoor air quality such as intrusion of contaminants from subsurface water and soil contamination below residences in suburban and rural settings.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

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TABLE 1

a

Summary Statistics of VOC Concentrations (*μ*g/m3) Measured in New Jersey Suburban and Rural Homes

t-1,2-dichloroethene, 1,2-dichloropropene, 1,3-dichloropropene, 1,2-dichloroetrafluoroethane, hexachlorobutadiene, tertiary butyl alcohol, 1,1,2,2-tetrachloroethane, 1,2,4-trichlorobenzene, 1,1,2-trichloroethane, vinyl chl r-1,2-dichloropropane, cis-1,3-dichloropropene, 1-1,3-dichloropropene, 1-1,3-dichlorovertafluorochane, hexachlorochane, hexachlorochane, hexachlorochane, 1,2,2-terachlorochane, 1,2,2-terichlorochane, vinyl chloride. Compou a Compounds never detected: bromodichloromethane, bromoform, bromomethane, carbon tetrachloride, chlorobenzene, chloromethane, 3-chloropropene, dibromochloromethane, 1,2-dibromoethane, 1,3-dichlorobenzene, 1,1-dichloroe $a_{\rm compounds}$ never detected: bromodichloromethane, bromothene, bromothame, carbon tetrachloride, chlorobenzene, athoromethane, 3-chloropropene, dibromochloromethane, 1,2-dibromoethane, 1,3-dichlorobenzene, 1,1-dichloroethane, only 1, 2, or 3 samples: 1,2 dichlorobenzene, 1,2 dichloroethane, 1,2 dichloroethene (cis), 1,2-dichloroethane (Freon 114), carbon disulfide, 1,2,3-Trichloro-1,2,2-trifluoromethane, 2-chlorooluene. only 1, 2, or 3 samples: 1,2 dichlorobenzene, 1,2 dichloroethane, 1,2 dichloroethene (cis), 1,2-dichlorotetrafluoroethane (Freon114), carbon disulfide, 1,2,3-Trichloro-1,2,2-trifluoromethane, 2-chlorotoluene.

 b Mean values calculated using the MDL value. $b_{\text{Mean values calculated using the MDL value.}}$

TABLE 2

Principal Component Analysis of Target VOCs Indoor Air Concentrations Showing the Compounds Grouped Together in Seven Factors, Suggesting Principal Component Analysis of Target VOCs Indoor Air Concentrations Showing the Compounds Grouped Together in Seven Factors, Suggesting *a* Common Sources Rotated Component Matrix

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*a*Extraction Method: Principal Component Analysis. Rotation Method: Varimax with Kaiser Normalizationa. Rotation converged in six iterations.

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TABLE 3

p-Values and Overall model p-Values and Overall model R^2 from the GLM Univariate Analysis for Each Compound with the Presence/Absence of Different Housing Characteristics 2 from the GLM Univariate Analysis for Each Compound with the Presence/Absence of Different Housing Characteristics Selected to Contribute the Compound's Variability *a*

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Comparison of Indoor Air Concentrations (µg/m³) Measured in this Study with Reported Literature Values Values are Median and 90th Percentiles Reported Comparison of Indoor Air Concentrations (*μ*g/m3) Measured in this Study with Reported Literature Values Values are Median and 90th Percentiles Reported

