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Determinants of exposure to volatile organic compounds in four Oklahoma cities

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To begin to develop generalized models for estimating personal exposure to ambient air pollutants within diverse populations, the design of the Oklahoma Urban Air Toxics Study incorporated eight dichotomous macroenvironmental and household factors that were hypothesized to be potential determinants of exposure. Personal, indoor, and outdoor samples of volatile organic compounds (VOCs) were collected over 24-h monitoring periods in 42 households, together with activity diaries and data on the participants' residences. The distributions of the VOC concentrations were moderately to highly left-censored, and were mostly bimodal. The ATSDR minimal risk level (MRL) was exceeded in a small number of the samples. Personal and indoor concentrations tended to be higher than outdoor concentrations, indicating that indoor exposures were dominated by indoor sources. However, indoor concentrations were not correlated with the permeability of the residence, suggesting that the observed indoor concentrations reflected mostly localized, short-term emissions. The influence of the eight dichotomous factors and of the presence of an attached garage was evaluated using the Wilcoxon rank-sum test and by comparison of "excursion fractions", that is, the fractions of each distributions exceeding 10% of the MRL. Dry weather and absence of children in the household were found to be associated with higher exposures in personal or indoor exposures. Given the small sample size, it is possible that these factors were confounded with unidentified household characteristics or activities that were the true determinants of exposure.
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Introduction

The total exposure assessment methodology (TEAM) (Wallace et al., 1987, 1991; Wallace, 1991) and similar studies have provided valuable information regarding which sources actually contribute significantly to personal exposure to ambient air toxics, and what factors affect the relative weighting of these sources. The ongoing National Exposure Assessment Survey (NHEXAS) (Callahan et al., 1995; Clayton et al., 1999; Gordon et al., 1999; Whitmore et al., 1999; Pellizzari et al., 2001) is expanding this knowledge base using a population-based multistage probability sampling design.

To develop generalized models for estimating personal exposure within diverse populations, it would also be advisable to incorporate determinants of exposure into study designs. The primary objective of the Oklahoma Urban Air Toxics Study was to investigate how external factors influenced the relationship between personal exposures and area measurements of air toxics. Using a multicity, multi-season, two-level factorial design, we proposed to study the distribution of personal exposures in relation to eight dichotomous macroenvironmental and household factors that were hypothesized to influence personal activity and exposure patterns. The results presented here suggest, however, that the determinants of exposure may arise primarily from microenvironmental and behavioral factors that are not so readily incorporated in study designs.

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Methods

Factorial Design

Fractional factorial designs are a highly efficient and systematic means of screening for the influence of potential factors (also referred to as "contrasts") and their interactions (Box et al., 1978). The factors in this study were city size, presence vs. absence of refineries in the urban area, presence vs. absence of precipitation, high vs. low ambient temperature, extreme vs. mild ambient temperature, working day vs. day off, "blue collar" vs. "white collar" occupation of study participant, and presence vs. absence of children in the household.

Among the four cities in this study, Oklahoma City (population 500,000) and Tulsa (population 390,000) are

large metropolitan areas traversed by major interstate highways, with similar levels of industrialization. Ponca City (population 26,000) and Stillwater (population 39,000) are small cities situated more than 10 miles away from any major highway, with some light manufacturing. Tulsa and Ponca City both have refineries located on the south side of the city, while Oklahoma City and Stillwater have no refineries. The prevailing wind direction in the region is from the south.

We dichotomized weather variables based on information reported by the National Weather Service (NWS) for each 24-h monitoring period at the weather station nearest to the study participant's residence. Monitoring events were classified as "wet" if measurable precipitation was reported. All other events were defined as "dry" days, including 5 days on which "trace" precipitation was reported by the NWS. "High-temperature" ("warm" or "hot") events were defined as days on which the daytime temperature exceeded 65°F (18°C). The daytime high temperature during the "low-temperature" ("cool" or "cold") monitoring events did not exceed 60°F (16°C). Monitoring events were classified as "extreme temperature" ("hot" or "cold") if the daytime high temperature was greater than 85°F (29°C) or lower than 40°F (4°C).

We classified monitoring events as "day off" if they occurred on a weekend, a national holiday, or a personal day off from the study participant's job. All other monitoring events were classified as "workdays". Only one study participant worked outside the home during a weekend monitoring event.

We classified study participants as "white collar" if they worked in professional, managerial, or clerical occupations, or were full-time students. The "blue collar" classification included such occupations as homemaker, cook, utility worker, construction worker, fire fighter, and security guard. One participant who was retired on disability was not classified. Households were classified as having children if one or more child under the age of 13 years lived in the house.

We initially planned a set of 64 monitoring events, representing a quarter-fraction of a full factorial design. Each planned monitoring event involved a different combination of the eight dichotomous factors. As in any two-level factorial design, both levels of each contrast were represented an equal number of times. Thus, for example, half the planned events were on "high-temperature" days and half were on "low-temperature" days. For a number of reasons to be discussed below, however, the planned factorial design was not completed. In all, 42 monitoring events were carried out, including 20 monitoring events with combinations of factors that did not appear in the planned matrix. Nevertheless, the design provided sufficient information to investigate the influence of most if not all of the contrasts. The combinations of factors in the actual monitoring events are listed in Table 1.

Study Population

Participation was limited to persons between 21 and 55 years of age living in nonsmoking households in detached, single-family homes in the four study cities. The restrictions were designed to (a) prevent indoor and outdoor pollutant exposures from being masked by tobacco combustion products, and (b) prevent the migration of indoor pollutants from nonparticipating households within the same building envelope. The age limitation was intended to ensure that subjects were in their prime working years with a similar capacity for physical activity. However, no volunteers were excluded on the basis of disability.

We recruited volunteers through notices and articles in the local news media, through flyers posted in public areas, and by personal contacts at shopping centers, schools, churches, and places of business. In total, 84 people were enrolled in the study; 39 people in 37 households actually took part in a monitoring event, including three people who participated twice.

Monitoring Procedure

Monitoring events were arranged 2 or 3 days in advance based on weather forecasts and the availability of a volunteer representing the desired combination of urban and personal factors. With one exception, the monitoring events began in the late afternoon or early evening. The study participant was fitted with a Pocket Pump® personal sampling pump (SKC, Eighty Four, PA, USA) equipped with Carbotrap 300 thermal desorption tubes (Gerstel, Mülheim an der Ruhr, Germany; or Supelco, Bellefonte, PA, USA) and a March II-E global positioning system (GPS) data recorder (Corvallis Microtechnology, Corvallis, OR, USA). As described elsewhere (Phillips et al., 2001), the GPS units were used in combination with activity diaries to track participants' locations throughout the monitoring period. The participants were instructed to remove the monitoring equipment only during sleep or bathing. Pumps with thermal desorption tubes (about 10% replicates) were also set up in the main living area of the house (usually the kitchen or living room) and in a sheltered location outside the house. Sampling flow rates were set at 35 ml/min and were stable to within 5% over the sampling period.

Sample tubes were changed after approximately 12 h. As participants typically preferred not to be visited by the investigators in the morning, the participants were instructed how to change from night-time to daytime samples. At the end of the 24-h monitoring event, the sample tubes were capped and placed in a cooler for transport. An investigator briefly reviewed the activities diary and worked with the participant to complete a questionnaire about the structural characteristics of the residence and potential emissions sources.

Table 1. Monitoring events actually conducted.

City	Temporal factors			Personal factors	
Oklahoma City	Cool	Work day	Dry	White collar	Children
Oklahoma City	Cool	Day off	Dry	White collar	No children
Oklahoma City	Cool	Work day	Wet	White collar	No children
Tulsa	Warm	Work day	Dry	Blue collar	No children
Tulsa	Warm	Work day	Dry	White collar	No children
Ponca City	Cool	Work day	Wet	White collar	Children
Ponca City	Cool	Work day	Wet	White collar	No children
Tulsa	Cool	Day off	Wet	Blue collar	No children
Tulsa	Cool	Day off	Wet	White collar	No children
Oklahoma City	Cool	Work day	Dry	Blue collar	Children
Stillwater	Warm	Work day	Dry	White collar	No children
Tulsa	Hot	Work day	Dry	Blue collar	No children
Tulsa	Warm	Work day	Dry	White collar	No children
Ponca City	Hot	Work day	Dry	Blue collar	Children
Oklahoma City	Warm	Day off	Dry	White collar	No children
Stillwater	Warm	Work day	Dry	Blue collar	No children
Oklahoma City	Warm	Day off	Wet	Blue collar	No children
Stillwater	Warm	Work day	Wet	Blue collar	No children
Tulsa	Hot	Day off	Dry	Blue collar	Children
Oklahoma City	Hot	Work day	Dry	White collar	Children
Tulsa	Hot	Work day	Dry	Blue collar	Children
Ponca City	Hot	Day off	Dry	White collar	No children
Ponca City	Hot	Work day	Dry	White collar	Children
Oklahoma City	Hot	Day off	Dry	White collar	No children
Stillwater	Warm	Work day	Dry	Blue collar	No children
Ponca City	Cool	Work day	Dry	White collar	Children
Oklahoma City	Warm	Day off	Dry	White collar	Children
Stillwater	Warm	Work day	Wet	White collar	No children
Tulsa	Cool	Work day	Wet	White collar	No children
Tulsa	Cool	Work day	Dry	White collar	No children
Oklahoma City	Cool	Work day	Dry	Blue collar	Children
Stillwater	Cold	Work day	Wet	Not classified	No children
Tulsa	Cool	Day off	Wet	White collar	Children
Ponca City	Warm	Day off	Dry	Blue collar	Children
Stillwater	Warm	Work day	Dry	White collar	No children
Stillwater	Warm	Work day	Dry	White collar	No children
Oklahoma City	Warm	Day off	Wet	Blue collar	No children
Oklahoma City	Warm	Work day	Wet	White collar	No children
Oklahoma City	Hot	Day off	Dry	Blue collar	No children
Tulsa	Hot	Day off	Dry	White collar	Children
Tulsa	Hot	Work day	Dry	White collar	No children
Tulsa	Hot	Work day	Dry	White collar	No children

Residence Permeability

After completion of each monitoring event, an investigator used a Model E-3 blower door with Model DM4 micromanometer (INFILTEC, Falls Church, VA, USA) to measure the air flow rate necessary to create a 50 Pa pressure difference between the inside and outside of the house. If a 50 Pa pressure difference could not be achieved, the flow rate Q was measured over a range of three to five pressure differences P and the results were extrapolated to 50 Pa based on a regression of $\log Q$ on $\log P$ using the relationship (Sherman, 1998):

$$Q = \kappa P^n$$

The flow rate at 50 Pa was divided by the volume of the house (floor area × ceiling height) to estimate ACH_{50}, the air change rate at 50 Pa. The normalized leakage NL for single-story houses was then estimated using the equation (Sherman, 1998)

$$NL = \frac{ACH_{50}}{20}$$

A correction factor of 0.8 (Meier, 1994) was applied for the six two-story houses in the study.

The blower door procedure initially used in this study lacked an explicit instruction to verify that all interior room doors are open during the test. After we recognized this

deficiency, we repeated blower door measurements in the affected residences. We omitted nine residences from the analysis of permeability data because two had undergone remodeling and seven households had moved or withdrawn from the study before the blower door measurements could be repeated.

Chemical Analysis

Desorption tubes were analyzed following EPA method T0-1 using an HP 6890/5973 gas chromatograph/mass spectrometer (Hewlett Packard/Agilent Technologies, Palo Alto, CA, USA) equipped with a TDS thermal desorption apparatus (Gerstel, Mülheim an der Ruhr, Germany). In addition to the field samples, 37 field blanks were analyzed. A spike sample was analyzed with approximately every 10 samples.

Calibration standards of known mass were generated using permeation tubes (Vici Metronics, Santa Clara, CA, USA). Six to nine point calibration lines were created for each VOC quantified: toluene, hexane, *o*-xylene, *p*-xylene, benzene, ethylbenzene, octane, dichlorobenzene, tetrachloroethylene, and trimethylbenzene. We set the limit of quantitation, which ranged from 1.2 ng for *p*-xylene to 16 ng for dichlorobenzene, at 50% of the lowest mass point in the calibration line for each compound.

Statistical Analysis

We performed statistical analysis only on those VOC results that were not more than 50% censored. For each dichotomous factor, we sorted the concentration data by level of the contrast to yield two data sets, for example, a ''large city'' distribution and a ''small city'' distribution. Each distribution was log-transformed and then a nonlinear least-squares-determined bimodal or unimodal normal probability curve was used to interpolate or extrapolate the empirical results. The excursion fraction — that is, the fraction of the fitted distribution that exceeded a predetermined cut-off value — was computed for each data set. Taking the excursion fraction as the binomial parameter, 95% confidence limits were calculated using interpolated values of the F distribution with noninteger degrees of freedom (Brownlee, 1965). We set the cut-off value at 10% of the minimal risk level (MRL) developed by the Agency for Toxic Substances and Disease Registry for inhalation of intermediate or chronic duration (ATSDR, 2001). We considered the excursion fraction to be significantly different ($P<0.05$) for a contrast if the point estimate of the excursion fraction for one level of the contrast fell outside the 95% confidence limits on the excursion fraction for the other level of the contrast. If the point estimates for both levels of the contrast fell outside the confidence limits for the other level, we considered the result significant at $P<0.025$.

Nonparametric statistical analyses of the experimental data were run using S-PLUS 2000 (MathSoft, Seattle, WA, USA). Wilcoxon signed-rank tests and Spearman's correlation tests were performed on sets of paired data. Wilcoxon rank-sum tests were run on unpaired data for each contrast.

Results

The distributions of concentrations of volatile organic compounds (VOCs) measured in the personal, indoor, and outdoor air samples are summarized in Table 2. The median and maximum values in Table 2 are from the actual measured data, not the fitted data. The total number of samples varied by type due to inclusion of replicate samples and loss of several samples through breakage, equipment malfunction, or failure to change sample tubes.

All contaminant concentrations were found to be less than 1 mg/m^3. Five of 82 personal samples and five of 90 indoor samples exceeded the ATSDR MRL of 12.8 $\mu g/m^3$ (4 ppb) for benzene inhalation of intermediate duration. Two of the personal samples exceeded the MRL of 301 $\mu g/m^3$ (80 ppb) for chronic inhalation exposure to toluene. Two personal and two area samples exceeded the MRL of 434 $\mu g/m^3$ (100 ppb) for chronic inhalation exposure to *p*-xylene.

The distributions were moderately to highly left-censored, and tended to be bimodal, with the lower mode falling below the censoring point. Lognormal probability plots of the benzene, toluene, *o*-xylene, and *p*-xylene data are provided in Figures 1–8, along with the fitted curves. Owing to censoring, we limited further statistical analysis mostly to the benzene, toluene, and *p*- and *o*-xylene results.

Exceptional Values

The concentration data for benzene, *o*-xylene, *p*-xylene, and ethylbenzene included a few exceptionally high values that could be described as outliers, in the sense that they lay well above the fitted log-normal probability curves and their inclusion in parametric analyses tended to bias the outcome. Exceptional values in the personal daytime, personal night-time, indoor daytime, and indoor night-time distributions for ethylbenzene, *o*-xylene, and *p*-xylene all occurred in one monitoring event. The participant in this case reported using a scented potpourri crock in the evening and hairspray in the morning. No other sources of exposure were identified in this monitoring event. An exceptional value for the indoor night-time benzene distribution occurred during another monitoring event for which no activity or product in the house was identified as a possible exposure source.

Indoor vs. Outdoor Concentrations

As illustrated in Figures 1–8, the distribution of outdoor concentrations fell well below the distribution of indoor concentrations. The differences between the indoor and outdoor distributions of benzene, toluene, *o*-xylene, and *p*-xylene as evaluated by the Wilcoxon signed-rank test were

Table 2. Distribution of concentrations of VOCs in ambient air samples ($\mu g/m^3$).

	Indoor day	Indoor night	Outdoor day	Outdoor night	Personal day	Personal night
Number of samples	46	44	38	39	40	42
Toluene						
Median	12	22	0.40	1.9	12	11
Max	120	140	73	26	380	140
LOQ	0.01	0.01	0.01	0.01	0.01	0.01
% Censored	13	9	34	38	5	10
Benzene						
Median	0.62	1.2	—	—	1.3	1.1
Max	14	110	2.5	4.4	30	23
LOQ	0.03	0.02	0.03	0.03	0.07	0.03
% Censored	33	25	50	51	25	21
o-Xylene						
Median	0.30	0.67	—	—	0.66	—
Max	240	280	2.3	6.5	230	200
LOQ	0.07	0.07	0.07	0.07	0.07	0.02
% Censored	46	39	87	69	35	50
p-Xylene						
Median	2.3	3.2	0.08	—	3.1	2.3
Max	490	570	7.5	4.5	520	460
LOQ	0.02	0.01	0.01	0.01	0.01	0.01
% Censored	26	14	45	51	13	21
Ethylbenzene						
Max	270	310	2.8	5.1	280	250
LOQ	0.07	0.03	0.04	0.07	0.11	0.06
% Censored	59	61	89	90	60	64
Trimethylbenzene						
Max	67	14	1.5	1.0	41	45
LOQ	0.04	0.03	0.03	0.06	0.07	0.04
% Censored	67	73	87	90	85	81
Octane						
Max	170	35	—	—	30	10
LOQ	0.12	0.12	0.07	0.16	0.18	0.10
% Censored	85	86	100	100	85	83
Hexane						
Max	20	100	9.3	11	37	42
LOQ	0.04	0.06	0.04	0.04	0.07	0.04
% Censored	76	80	74	85	70	81
Tetrachloroethylene						
Max	12	130	58	0.18	6.5	29
LOQ	0.06	0.06	0.04	0.07	0.09	0.05
% Censored	72	73	92	97	80	74
Dichlorobenzene						
Max	120	100	29	14	310	570
LOQ	0.33	0.53	0.33	0.70	0.36	0.32
% Censored	85	91	89	92	78	86

highly significant ($P<0.005$). No significant correlation was found between simultaneous indoor and outdoor concentrations for benzene, toluene, and *o*-xylene. The daytime indoor and outdoor concentrations of *p*-xylene were correlated ($r_s=0.51$, $P=0.0026$), but night-time *p*-xylene concentrations were not correlated. No correlation was found between simultaneous outdoor and personal concentrations.

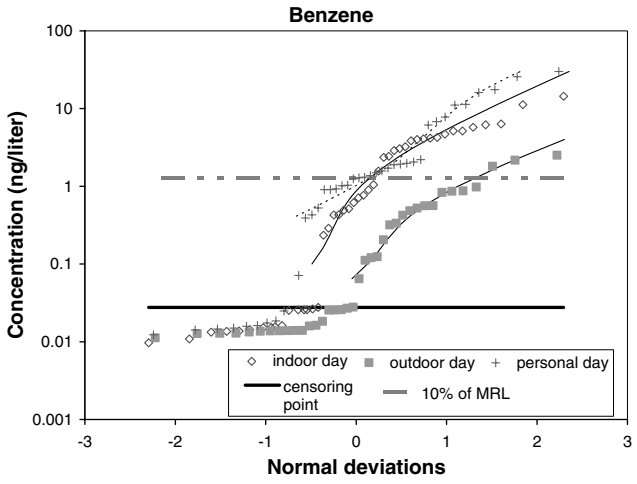

Figure 1. Benzene-monitoring results from daytime samples.

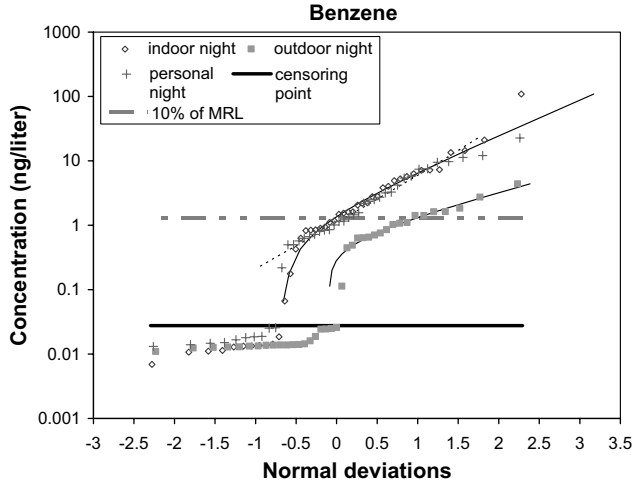

Figure 2. Benzene-monitoring results from night-time samples.

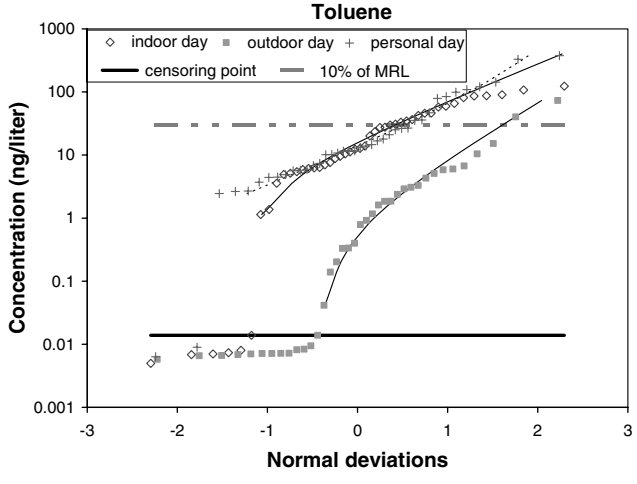

Figure 3. Toluene-monitoring results from daytime samples.

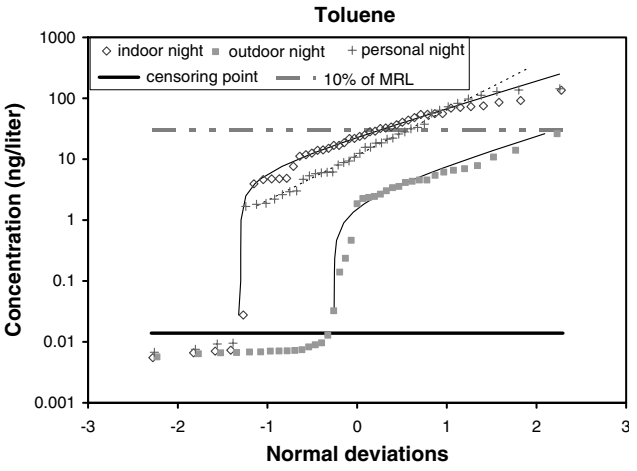

Figure 4. Toluene-monitoring results from night-time samples.

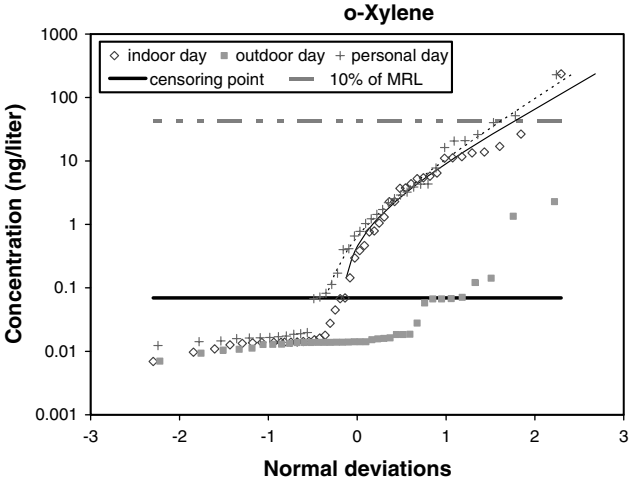

Figure 5. *o*-Xylene-monitoring results from daytime samples.

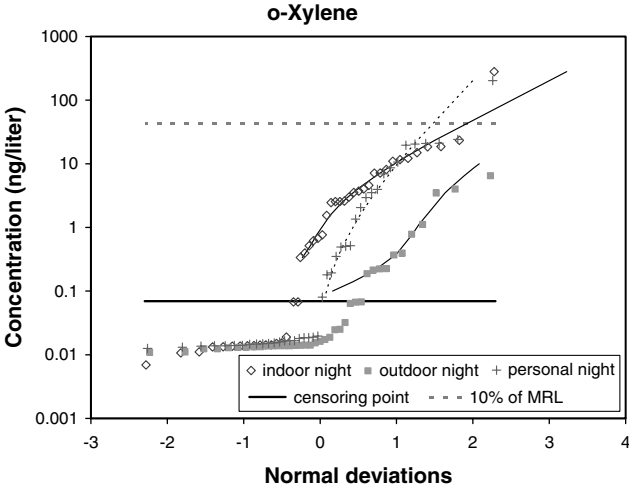

Figure 6. *o*-Xylene-monitoring results from night-time samples.

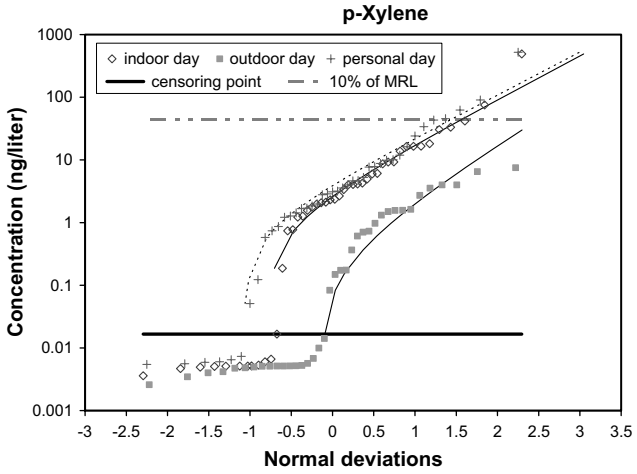

Figure 7. *p*-Xylene-monitoring results from daytime samples.

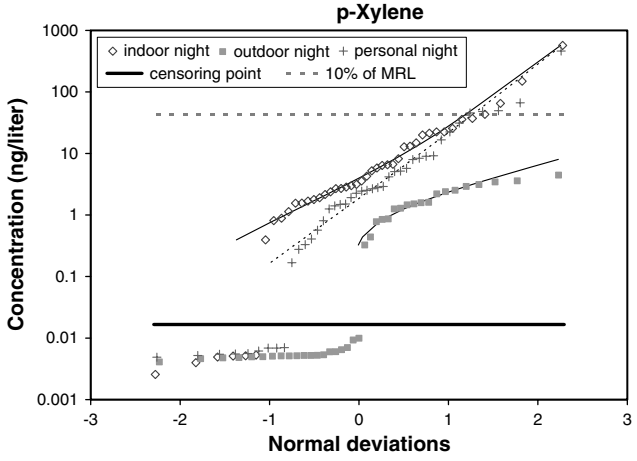

Figure 8. *p*-Xylene-monitoring results from night-time samples.

Indoor vs. Personal Concentrations

The distributions of personal and indoor concentrations appeared to be highly overlapped. No significant differences were found between the distributions of personal and indoor concentrations for benzene, toluene, *o*-xylene, and *p*-xylene by the Wilcoxon signed-rank test. Simultaneous daytime personal and indoor concentrations were correlated, with Spearman's correlation coefficients ranging from 0.55 to 0.62 ($P<0.001$) for the four VOCs. Among the night-time samples, personal and indoor concentrations were correlated for benzene ($r_s=0.34$, $P=0.033$), toluene ($r_s=0.45$, $P=0.0042$), and *o*-xylene ($r_s=0.39$, $P=0.013$).

Daytime vs. Night-time Concentrations

The distributions of daytime and night-time concentrations appeared to be fairly similar. The Wilcoxon signed-rank test showed a significant difference only for indoor *p*-xylene concentrations, where the night-time distribution was higher than the daytime distribution. The correlation between daytime and night-time concentrations was significant for benzene ($r_s=0.69$, $P<0.0001$), toluene ($r_s=0.65$, $P<0.0001$), *o*-xylene ($r_s=0.55$, $P=0.0006$), and *p*-xylene ($r_s=0.42$, $P=0.0084$) in personal samples; for *o*-xylene ($r_s=0.49$, $P=0.0021$) and *p*-xylene ($r_s=0.49$, $P=0.0018$) in indoor samples, and for benzene ($r_s=0.50$, $P=0.0039$), toluene ($r_s=0.37$, $P=0.040$), and *p*-xylene ($r_s=0.47$, $P=0.0092$) in outdoor samples.

Urban, Temporal, and Household Factors

Typically in a two-level factorial experimental design, contrasts are evaluated by subtracting the mean of the "negative" level from the mean of the "positive" level for each factor or interaction. The distribution of contrast differences is then examined for outliers. The factors or interactions corresponding to these outliers would be considered significant (Box et al., 1978). In the present study, the degree of censoring, the spread of the data, and the tendency to bimodality rendered such an analysis problematic. Although a median, if not a mean, could have been estimated from the fitted data for each level of a factor, it was not clear what practical meaning could be attached to such a number; it could not be considered representative of a central tendency, since the data points did not tend to cluster around a central value.

When the fitted distributions of the data for the two levels of a contrast were compared, in about 20% of contrasts the fitted distributions had similar medians but diverged above the median, as illustrated in Figure 9. In about 12% of contrasts, the medians differed by a factor of at least 3, but the upper parts of the fitted distributions converged or crossed, as illustrated in Figure 10. In only about 5% of contrasts were the two distributions clearly offset as illustrated in Figure 11. About 46% of contrasts had similar

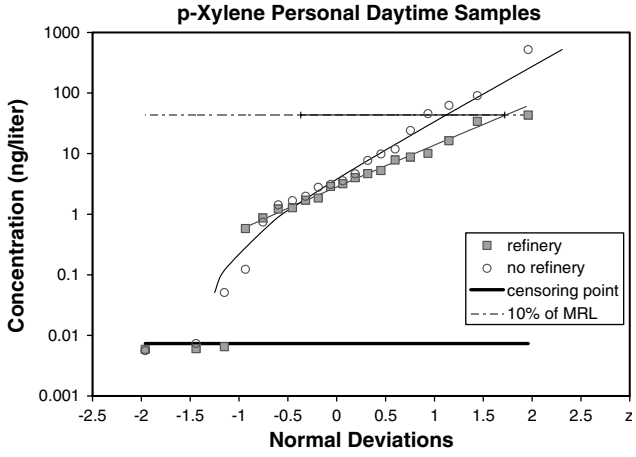

Figure 9. Example of contrasting distributions with similar medians and divergent upper quantiles. The differences between the excursion fractions were significant, as indicated by the error bars.

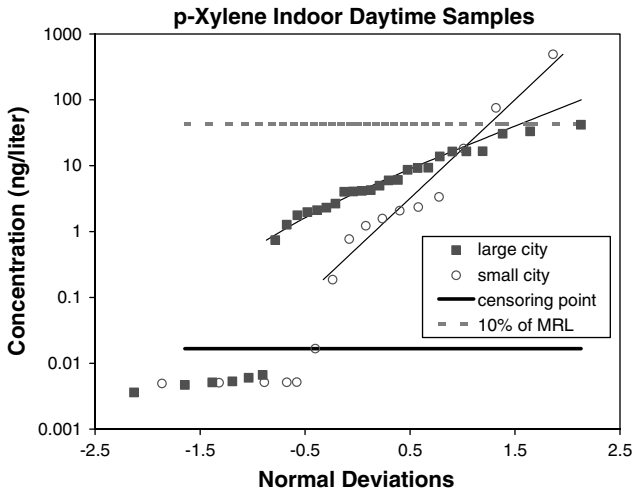

Figure 10. Example of contrasting distributions that crossed above the median.

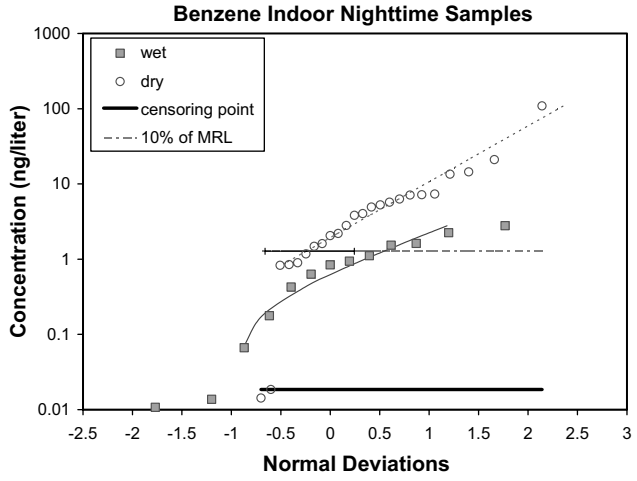

Figure 11. Example of contrasting distributions that were offset. The differences between the excursion fractions were significant, as indicated by the error bars.

distributions, and the remaining contrasts involved data sets that were too sparse to fit.

Statistically significant differences in the "excursion fractions" for the contrasts are presented in Table 3. The results for *o*-xylene should be viewed with caution because they are almost entirely based on extrapolations from the fitted data. Excluding the outdoor data, which were generally too highly censored and/or too sparse to produce a reasonable fit or to calculate useful binomial confidence intervals, 144 contrasts were analyzed, encompassing the eight design factors plus the presence/absence of an attached garage. In all, 50 of these contrasts showed significant differences in the excursion fraction, 73 showed no significant difference, and 21 had insufficient data to make a comparison.

Owing to the large number of comparisons that were made, some of the differences that met the $P < 0.05$ criterion for statistical significance were probably, in fact, merely due to random variation. If significant differences for a contrast were consistent across sample types (indoor day, indoor night, personal day, and personal night) for the same compound, or consistent across compounds for the same sample type, that consistency could be taken as evidence that the factor was influential. By this standard, the results suggest that among the nine contrasts evaluated, the factors most likely to be associated with significantly higher excursion fractions were the absence of children in the household and dry weather.

The contrasts were also evaluated using the Wilcoxon rank-sum statistic. Significant contrasts were identified as the possible outliers on a single normal probability plot of all of the rank-sum test statistics for benzene, toluene, *o*-xylene, and *p*-xylene. The indoor contrasts for residences reporting/not reporting likely VOC sources were included in this analysis. Outdoor contrasts for *o*-xylene were not determined, giving 194 contrasts in all. Seven contrasts appeared to be borderline significant: the distributions were higher for indoor daytime concentrations of toluene and *p*-xylene in households without children, for indoor night-time concentrations of toluene on dry days, for personal daytime concentrations of toluene on dry days, for personal night-time concentrations of benzene in cities without refineries, for outdoor night-time concentrations of benzene in residences without attached garages, and for outdoor daytime concentrations of benzene in small cities.

Residence Characteristics

The median construction date of the 37 houses in the study was 1969, with a fairly flat distribution by decade from the 1930s through the 1990s. The oldest house was built in 1906. All except one house had central heating and air conditioning. Some changes in construction styles were evident over time: (1) All houses dating from 1969 onward were built on a slab, whereas 72% of the houses built before 1969 had a crawlspace or basement. (2) A total of 90% of the houses built in or after 1969 included at least some brick, stone, or other masonry in the veneer, compared to only 56% of the houses built before 1969. (3) The average number of exterior openings — doors plus windows — decreased from 18.4 in the pre-1969 houses to 13.2 in the houses built in 1969 or later.

Of the 28 houses in which valid blower door measurements were taken, 20 had normalized leakage values in the range 0.4–1.13, which is considered sufficiently permeable to provide adequate natural ventilation in residential dwellings while also meeting energy use guidelines (Sherman, 1998). The remaining eight houses had normalized leakage values less than 0.4, indicating that these houses could be "too tight". Normalized leakage showed a moderately negative

Table 3. Fraction of fitted distribution exceeding 10% of MRL.

	Indoor day	Indoor night	Personal day	Personal night
Large city vs. small city				
Benzene		0.55 > 0.37*		0.57 > 0.28**
Toluene	0.40 > 0.22*	0.44 > 0.27*		0.37 > 0.13*
o-Xylene			0.08 > 0.01*	
p-Xylene			0.03 < 0.15*	
Refinery vs. no refinery				
Benzene	0.31 < 0.55**			0.35 < 0.61**
Toluene			0.19 < 0.40*	0.20 < 0.37*
p-Xylene			0.04 < 0.13*	0.03 < 0.17*
High vs. low temperature				
Benzene				
Toluene		0.47 > 0.29*		0.35 > 0.19*
o-Xylene		0.05 > 0.001*		
p-Xylene		0.14 > 0.002*	0.12 > 0.03*	0.11 > 0.01*
Extreme vs. mild temperature				
Benzene				0.35 < 0.52*
p-Xylene	0.004 < 0.10*		0.03 < 0.11*	0.01 < 0.14*
Wet vs. dry				
Benzene	0.31 < 0.51*	0.29 < 0.60*		
Toluene	0.17 < 0.40*	0.17 < 0.54**	0.10 < 0.40*	0.14 < 0.37*
p-Xylene			0.007 < 0.10*	
Workday vs. day off				
p-Xylene	0.11 > 0.006*	0.17 > 0.05*		
Attached garage vs. no attached garage				
Benzene		0.55 > 0.37*		0.52 > 0.36*
p-Xylene		0.14 > 0.02*	0.14 > 0.01*	0.14 > 0.03*
Blue collar vs. white collar				
Benzene	0.31 < 0.54*	0.39 < 0.57*		
o-Xylene			0.17 > 0.01**	
p-Xylene	0.13 > 0.03*		0.22 > 0.05**	
Children vs. no children				
Benzene	0.20 < 0.53*	0.40 < 0.56*		
Toluene	0.16 < 0.39*		0.14 < 0.38*	0.18 < 0.36*
o-Xylene				0.03 < 0.10*
p-Xylene			0.03 < 0.13*	0.04 < 0.12*

*$P < 0.05$.
**$P < 0.025$.
Only statistically significant differences between contrasts are shown.

correlation with year of construction ($r = -0.59$), masonry veneer ($r = -0.55$), and slab foundation ($r = -0.46$). Normalized leakage was very weakly correlated with the number of exterior openings ($r = 0.21$).

The relationship between permeability and concentrations of indoor air contaminants was examined for monitoring events in which the indoor VOC concentrations were quantifiable and exceeded the simultaneous outdoor concentrations. The net indoor concentration (indoor concentration minus outdoor concentration) was found not to be correlated with normalized leakage for any contaminants.

Activities and Sources

The frequency of reporting of various activities and sources potentially associated with exposure to air contaminants is summarized in Table 4. Chemical products used or stored in the home included toiletries, cleaning products, scented candles and potpourri, glues, paints, and sealants. We used

Table 4. Frequency of reported activities and sources during monitoring events.

	Daytime		Night-time	
	Fraction of events (%)	Minutes spent[a]	Fraction of events (%)	Minutes spent[a]
Pumped gas	0		7	
Used chemical at home	60		55	
Used chemical at work	17		2	
Used chemical elsewhere	7		2	
Drove/rode in car	74	50	24	27
Spent time in home	100	272	100	709
Spent time outdoors	83	30	50	27
Likely VOC sources in home	64		52	
Exposed to tobacco smoke	19		10	

[a]Average time spent by those engaging in activity.

professional judgment to identify products or activities that would be likely sources of VOC exposure. No correlation was found between the presence of likely VOC sources and any of the other dichotomous factors. Owing to the guesswork inherent in determining the presence or absence of VOC sources based on diary and questionnaire reports, we did not carry out a full analysis of the data for this contrast. Visual comparison of log-normal probability plots of the results of the indoor concentrations of benzene, toluene, *o*-xylene, and *p*-xylene for households with and without likely sources of VOCs suggested a possible relationship between the presence of likely VOC sources and increased concentrations of *o*-xylene, *p*-xylene, and toluene over part of the distribution. On the other hand, benzene concentrations tended to be higher in residences where VOC sources were not identified. None of these apparent differences were significant by the Wilcoxon rank-sum test.

When we planned the study, we anticipated that residents of the households might engage in extra housecleaning in advance of a scheduled monitoring event, and that such cleaning could result in higher than ordinary concentrations of VOCs in the indoor samples. To control for this possibility, on the monitoring day the study participants were asked how recently the house had been cleaned. In 18 of 42 monitoring events, the house had been cleaned within 1 day of the monitoring event; 20 of the houses had been cleaned 2–7 days before the monitoring event, and three houses had not been cleaned in 30 days or more. No response was recorded for one house. No correlation was found between indoor VOC concentrations and time since last cleaning.

Discussion

Of the 11 VOCs measured in this study, only benzene, toluene, *o*-xylene, and *p*-xylene were quantified in a sufficient number of samples to support statistical analysis. The MRL for benzene, toluene, and *p*-xylene was exceeded in a small number of the samples, indicating that ambient exposures to these air contaminants can reach levels at which adverse health effects may occur.

Effect of Observational Variables

The distributions of outdoor exposures were consistently and significantly lower than the indoor and personal exposure distributions. Except for daytime *p*-xylene concentrations, simultaneous indoor and outdoor exposures were largely uncorrelated. It may therefore be concluded that indoor exposures were dominated by indoor sources. This is consistent with the finding of the TEAM Study (Wallace et al., 1987, 1991).

If the sources of exposure are indoor, one might predict that exposures would tend to be higher in tight houses than in relatively leaky houses. However, the results of this study did not support this prediction; we found that normalized leakage, a measure of the permeability of a house, was not correlated with indoor-sourced exposure. The absence of any significant correlation between permeability and indoor exposure in the predicted negative direction suggests that the observed indoor concentrations might reflect mostly localized, short-term emissions inside the house, which would be largely independent of the leakage ventilation rate, as distinct from steady-state concentrations of contaminants, which would be inversely related to the leakage rate. On the other hand, it should be noted that the blower door method, which had been chosen over tracer gas methods because of its ease of use, provides only an indirect measure of air change rate. Furthermore, in our questionnaire on household characteristics and activities, we failed to note whether windows were open at any time during the sampling period, which could obviously affect the natural ventilation rate.

The distributions of personal exposures and indoor residential exposures were fairly similar, in contrast to the finding of the TEAM study that daytime personal exposures

tended to be higher than indoor exposures (Wallace et al., 1991). The exclusion of smokers from our study might be partially responsible for this difference. Simultaneous personal and indoor residential concentrations were correlated in daytime samples for all four VOCs and in night-time samples for benzene, toluene, and *o*-xylene. This suggests that personal activities that occurred mostly in the daytime simultaneously influenced personal and indoor exposures. Also, the large portion of the day spent away from home did not appear to contribute substantially to the overall distribution of personal daytime exposures, possibly because few of the participants reported exposure to chemical products or environmental tobacco smoke while away from home. The lack of correlation between night-time personal and indoor concentrations of *p*-xylene could reflect spatial variability in ambient exposure levels within the home, given that the night-time ''personal'' samples were collected mostly in the subject's bedroom and the indoor samples were collected in the main living area.

The influence of urban, temporal, and household factors on exposure distributions as evaluated by the Wilcoxon rank-sum test and the excursion fraction test were consistent. In both tests, dry weather and absence of children in the household were found to be associated with higher exposures in personal or indoor exposures. In view of the finding that indoor concentrations were driven primarily by indoor sources rather than by outdoor sources, it is somewhat surprising that the external factor precipitation had an apparent effect on indoor concentrations. The apparent association between absence of a refinery and elevated excursion fraction, shown in Table 3, is also surprising. This result is in contrast with the TEAM study, in which little difference in VOC concentrations was found in homes within 1.5 km of refineries and chemical plants compared to homes more distant from such facilities (Wallace et al., 1987). A reasonable explanation of these results is that the design factors, including refineries and precipitation, could have been confounded with household characteristics or activities that were the true determinants of exposure. Fortuitous confounding with unrecognized factors is a problem inherent in small sample sizes. The borderline significance of the findings of the Wilcoxon rank-sum tests also suggests that none of the contrasts represented strong determinants of exposure.

Assessment of the Factorial Approach

The combinations of factors represented in the actual experiments deviated considerably from the planned fractional factorial set for a number of reasons: (1) the weather during the monitoring event often did not conform to the forecast; (2) some combinations of temporal factors were relatively rare; (3) the representation of blue collar occupations, households with children, and residents of Ponca City fell short of our recruitment goals. Although the factorial design could not be adhered to, in practice, the attempt at a factorial approach was effective in preventing severe collinearity between factors. High temperature, extreme temperature, and dry weather were weakly correlated ($r = 0.31$–0.36), reflecting Oklahoma's climate with its hot, dry summers and mild winters. The factor ''attached garage'', which was not part of the original factorial design, was moderately correlated with the factors ''workday'' ($r = 0.51$) and ''blue collar'' ($r = -0.43$), providing an example of the type of happenstance collinearity that can occur in the absence of a factorial design.

The efficiency of the fractional factorial design can actually be a disadvantage in field-based studies because the small data sets generated are susceptible to high sampling variability and to confounding by unrecognized factors. This problem can be addressed — at considerably increased cost — by conducting replicate experiments. A 2^{-p} fractional factorial design in n factors would then become in essence a stratified random sampling design with 2^{n-p} strata.

Relevance of Distinction Between Daytime and Night-time Exposures

Dividing the 24-h monitoring period into night-time and daytime samples made the monitoring procedure more complicated because the study participant had to change sample tubes halfway through the monitoring event. Collection of a single 24-h sample would simplify the procedure while also increasing the total volume of air sampled, resulting in a lower limit of detection. It is therefore reasonable to question whether it was productive to collect separate daytime and night-time samples. In response, it should be noted that (1) activity patterns differed between day and night, (2) although sequential night-time and daytime exposures tended to be correlated, one was not strongly predictive of the other, and (3) some observed effects were not consistent across daytime and night-time data sets. Combining daytime and night-time samples into a single 24-h sample could thus reduce the resolution of a study. Therefore, although 24-h or longer-term samples are reasonable for surveillance studies, 12-h or shorter-term samples would be preferable for studies seeking to elucidate determinants of exposure.

An additional consideration is the evidence from this study that indoor concentrations might be dominated by short-term events within the house. This possibility warrants further investigation involving multiple short-term samples.

Use of Excursion Fraction

Estimates of central tendency are of limited usefulness for many environmental data sets that tend to be censored and to exhibit very broad multimodal log-normal distributions. The

95th percentile value of a distribution is sometimes used as an estimate of the ''worst case'' exposure because this measure takes account of the spread of the data. It is not clear, however, what degree of difference between two 95th percentile values would be considered statistically significant. Another potential problem with the use of the 95th percentile point is that it implicitly discounts the most exposed portion of the population.

In this study, we evaluated contrasts by determining the fraction of each fitted distribution that exceeded a biologically based ''operational exposure level'' (OEL), which was set at 10% of the ATSDR MRL. This measure explicitly recognizes the portion of the population that could be considered at risk of adverse effects from exposure. The OEL was set at 10% of the MRL rather than at the MRL to increase the power of the test; given the small fraction of concentrations that exceeded the MRL, the data set was too small to find significant differences using the higher limit. Significance testing of contrasts was straightforward using the binomial confidence limits on the excursion fraction. The excursion fraction approach proved to be consistent with, but more powerful than, the nonparametric Wilcoxon rank-sum test at evaluating contrasts, but its use was limited to data sets with at least seven uncensored values, and it had a relatively low power for detecting differences between small excursion fractions. Another important distinction between the tests was that the rank-sum test weighted all portions of a distribution equally, while determination of the excursion fraction was typically dominated by the upper portions of the distribution. Notwithstanding these limitations, the excursion fraction approach can be an intuitively simple and meaningful tool for describing and comparing environmental data distributions in a variety of applications.

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References

ATSDR. Minimal risk levels for hazardous substances. Agency for Toxic Substances and Disease Registry, Atlanta GA, 2001, (available at http://www.atsdr.cdc.gov/mrls.html. accessed November 1, 2002).

Box G.E.P., Hunter W.G., and Hunter J.S. *Statistics for Experimenters*. John Wiley Interscience, New York, 1978.

Brownlee K.A. *Statistical Theory and Methodology in Science and Engineering*. John Wiley, New York, 1965.

Callahan M.A., Clickner R.P., Whitmore R.W., Kalton G., and Sexton K. Overview of important design issues for a National Human Exposure Assessment Survey. *J Expos Anal Environ Epidemiol* 1995: 5: 257–282.

Clayton C.A., Pellizzari E.D., Whitmore R.W., Perritt R.L., and Quackenboss J.J. National Human Exposure Assessment Survey (NHEXAS): distributions and associations of lead, arsenic and volatile organic compounds in EPA Region 5. *J Expos Anal Environ Epidemiol* 1999: 9: 381–392.

Gordon S.M., Callahan P.J., Nishioka M.G., Brinkman M.C., O'Rourke M.K., Lebowitz M.D., and Moschandreas D.J. Residential environmental measurements in the National Human Exposure Assessment Survey (NHEXAS) pilot study in Arizona: preliminary results for pesticides and VOCs. *J Expos Anal Environ Epidemiol* 1999: 9: 456–470.

Meier A. Infiltration: just ACH_{50} divided by 20? *Home Energy* 1994, January/February: 35–37.

Pellizzari E.D., Smith D.J., Clayton C.A., Michael L.C., and Quackenboss J.J. An assessment of the data quality for NHEXAS-Part I: exposure to metals and volatile organic chemicals in Region 5. *J Expos Anal Environ Epidemiol* 2001: 11: 140–154.

Phillips M.L., Hall T.A., Esmen N.A., Lynch R., and Johnson D.L. Use of global positioning system technology to track subjects' location during environmental exposure sampling. *J Expos Anal Environ Epidemiol* 2001: 11: 207–215.

Sherman M. *The use of blower-door data. LBL Report #35173*. Lawrence Berkeley Laboratory, Berkeley, CA, 1998.

Wallace L.A. Comparison of risks from outdoor and indoor exposure to toxic chemicals. *Environ Health Perspect* 1991: 95: 7–13.

Wallace L., Nelson W., Ziegenfus R., Pellizzari E., Michael L., Whitmore R., Zelon H., Hartwell T., Perritt R., and Westerdahl D. The Los Angeles TEAM study: personal exposures, indoor–outdoor air concentrations, and breath concentrations of 25 volatile organic compounds. *J Expos Anal Environ Epidemiol* 1991: 1: 157–192.

Wallace L.A., Pellizzari E.D., Hartwell T.D., Sparacino C., Whitmore R., Sheldon L., Zelon H., and Perritt R. The TEAM study: personal exposures to toxic substances in air, drinking water, and breath of 400 residents of New Jersey, North Carolina, and North Dakota. *Environ Res* 1987: 43: 290–307.

Whitmore R.W., Byron M.Z., Clayton C.A., Thomas K.W., Zelon H.S., Pellizzari E.D., Lioy P.J., and Quackenboss J.J. Sampling design, response rates, and analysis weights for the National Human Exposure Assessment Survey (NHEXAS) in EPA Region 5. *J Expos Anal Environ Epidemiol* 1999: 9: 369–380.