Environment International, Vol. 12, pp. 369-387, 1986
Printed in the USA. All rights reserved.

0160-4120/86 $3.00 + .00

TOTAL EXPOSURE ASSESSMENT METHODOLOGY (TEAM) STUDY: PERSONAL EXPOSURES, INDOOR-OUTDOOR RELATIONSHIPS, AND BREATH LEVELS OF VOLATILE ORGANIC COMPOUNDS IN NEW JERSEY

Lance A. Wallace
U.S. Environmental Protection Agency 401 M Street, S.W. Washington, DC 20460
(Presently Visiting Scientist Harvard University School of Public Health Department of Environmental Sciences and Physiology 665 Huntington Avenue Boston, MA 02115 U.S.A.)

Edo D. Pellizzari, Tyler D. Hartwell, Roy Whitmore, Charles Sparacino,
Harvey Zelon
Research Triangle Institute Research Triangle Park, NC 27709

(Received 17 May 1985; Accepted 18 February 1986)

A total of 20 toxic, carcinogenic, or mutagenic organic compounds were measured in the air and drinking water of 355 residents of Bayonne and Elizabeth, New Jersey, in the fall of 1981. The participants were selected from over 10,000 residents screened by a probability sampling technique to represent 128,000 persons (over the age of seven) who live in the two neighboring cities. Over one hundred geographic areas throughout the two cities were selected for monitoring. Each participant carried a personal sampler with him during his normal daily activities for two consecutive 12-h periods. (One resident in each of the 108 sampling segments had an identical sampler operating in the backyard for the same two 12-h periods.) All participants also collected two drinking water samples. At the end of the 24-h sampling period, all participants gave a sample of exhaled breath, which was analyzed for the same compounds. All participants also completed a questionnaire on their age, sex, occupations and activities during the sampling period. An extensive quality assurance program was carried out on all sampling/analysis activities. Eleven of the 20 chemicals were prevalent in air and 3 in water. Air was the most important pathway of exposure for 10 compounds, and drinking water was most important for 3 trihalomethanes. Ranges of exposures were extremely large, with differences of 3–4 orders of magnitude common. Median personal exposures were 2–5 times larger than median outdoor concentrations; maximum personal exposures were as much as 100 times corresponding maximum outdoor concentrations. Residence near major point sources had no effect on exposure but many common activities (filling a gas tank, visiting a dry cleaner, smoking) had significant effect on exposures.

Introduction

Several studies have compared indoor and outdoor concentrations of volatile organic compounds. Mølhave (1979) found elevated levels of benzene and toluene in 39 Danish dwellings. Jarke (1981) found increased concentrations in 34 Chicago homes. Lebret (1984) found that all 35 target organic compounds displayed elevated concentrations inside 134 Netherlands homes. Seifert (1982) reported that 15 homes in Berlin displayed increased levels of toluene and xylenes from printed material. DeBortoli (1984) found that all 32 target organics in 15 Northern Italy homes exceeded outdoor levels. Wallace (1984) found slightly increased levels in a home for the elderly in Washington, D.C. Pellizzari (1984b) made repeated measurements in a new office building, finding that some chemicals declined from 100 times outdoor levels to 10 times outdoor levels in several months, while other chemicals showed little change or even increased their concentrations after workers occupied the building.

Another study has combined personal air, outdoor air, drinking water, and breath measurements of toxic organics in three cities: Baton Rouge, LA; Houston, TX; and Greensboro, NC (Pellizzari 1983). Findings included extreme variability of exposures, higher personal exposures than outdoor concentrations, and significant correlations between breath and personal air concentrations of several chemical compounds.

The U.S. Environmental Protection Agency sponsored a Total Exposure Assessment Methodology (TEAM) Study between 1980 and 1984. The long-term goal of this research program was to develop and test statistical and chemical methods for estimating total human exposure to selected toxic or hazardous substances. The approach was to select a sample of persons with known probability to represent the entire population of an urban area. Direct measurements of 20–30 organic compounds were made on the air each person breathes, outdoor air in his neighborhood and the water he drinks. Concurrently, the same chemicals in each person's breath were measured. The study attempts to establish for each chemical (1) the frequency distributions of exposures for the target population, (2) the relative importance of certain routes of exposure (air, water), (3) whether a predictable correlation exists between exposure and body burden (breath), and (4) whether personal activities or household characteristics affected exposure.

Pilot studies to evaluate equipment and methodology were carried out at Lamar University in Beaumont, TX, and the University of North Carolina in Chapel Hill, NC (Wallace, 1982; Zweidinger, 1982), and also at Bayonne and Elizabeth, NJ, and Research Triangle Park, NC (Wallace, 1984a). The full-scale study of 355 persons in Bayonne and Elizabeth was carried out between September and November 1981 and will be summarized here. A more detailed report is available as an EPA Report (Pellizzari, 1984a).

Methods and Materials

Study Design

The sample design for the first season of the TEAM study was a stratified three-stage design. A stratified sample of 110 geographic area segments was selected at the first stage with probabilities proportional to estimated housing unit counts based upon preliminary 1980 Census data. Stratification variables included proximity to major point sources (chemical plants, petroleum refineries) and socioeconomic status (based on real estate values). At the second stage, a compact cluster of 51 housing units within each area segment was screened; data on age, sex, smoking status, and potential occupational exposure to hazardous chemicals were collected for each member of the housing unit. The screening data then were used to select a third stage stratified sample of individuals for personal exposure monitoring.

The target population for the first season TEAM study contains individuals who satisfy all the following conditions:

1. Primary residence was in Bayonne or Elizabeth, New Jersey (using 1980 Census Place boundaries) when the household screening was conducted in July and August 1981.
2. Age 7 years or older at the time of the household screening.
3. Did not change status with regard to a key stratification variables between screening and personal exposure monitoring, i.e.,
   (a) Did not change smoking status, and
   (b) Did not change occupational exposure status, and
   (c) Did not move outside the economic-proximity stratum where they resided at the time of screening.
4. Physically and mentally capable of participating, i.e., able to complete the consent form and the study questionnaire, when personal exposure monitoring was conducted, from September 3, 1981, through November 23, 1981.
5. Not living in group quarters (as defined for the 1980 Census) or on a military reservation.

A sample of 110 geographical areas was then selected with probability proportional to size, as measured by the estimated number of housing units. These areas were ordered to achieve implicit stratification based upon the following characteristics:

1. city (Bayonne or Elizabeth)
2. proximity to major point sources
3. percentage white population; and
4. expected number of housing units.

The first stratification variable proportionally allocated the sample of 110 first stage units to the two cities. The second dimension of stratification was based upon proximity to a suspected point source and socioeconomic status. The purpose of this stratification was to insure representation of residents from areas near suspected point sources and low socioeconomic areas in case their exposure to hazardous chemicals might be significantly different from that of the overall target population. The purpose of stratification by race was to insure proportional representation of non-whites in the sample in case their exposure to hazardous chemicals was significantly different from that of whites. Finally, the fourth dimension of stratification by size of the first stage unit helped to insure that both large and small first stage units would be selected using sequential selection from the ordered frame with probabilities proportional to size. In summary, the major purpose of stratification of the first stage sample was to control the distribution of the sample with regard to variables that could be confounded with exposure to hazardous chemicals.

The sample of 110 first stage units was selected from the ordered sampling frame discussed in the previous paragraph using a sequential minimum probability replacement technique. Of the 110 sample segments that were selected by this procedure, two segments were ultimately discarded. One of these segments was a group quarters on board a ship that was docked in Elizabeth at the time of the 1980 Decennial Census. The other discarded segment was a military base. Thus, the final data base for the first phase sample contained 108 sample segments—43 in Bayonne and 65 in Elizabeth.

Second stage sampling of housing units. The second stage in selection of the first season TEAM sample was selection of sample housing units within the 108 eligible sample segments. First, the number of housing units in each sample segment was counted. Whenever the counted number of housing units was 51 or less, all housing units in the segment were selected into the sample. Otherwise, a compact cluster of 51 housing units was selected.

Third stage sample of persons. In the third stage, population subgroups defined by age, occupational exposure to hazardous chemicals, proximity to a suspected point source, socioeconomic status, and smoking status were all of special analytical interest. Hence, the main study sample was stratified by these variables. Eighteen occupations were classified as resulting in potential occupational exposure, including painters; chemical, textiles, plastics, and petroleum workers; service station attendants; photographers; taxi, bus, and truck drivers; custodians; dry cleaning workers; and others.

Three age groups were considered: 7–16, 17–64, and 65 or older. Persons residing within 1.5 kilometers of major point sources (chemical plants, petroleum refineries, etc.) were classed as potentially highly exposed; those living farther than 1.5 km were classified as moderately exposed if near heavy traffic arteries, and not exposed if residing away from such highways.

Questionnaire design. A set of three questionnaires was designed and submitted for clearance to the Office of Management and Budget. The Household Screening Questionnaire contained 13 questions determining key stratification variables (address, occupation, smoking status) for the 5500 target households (~12,000 household members) of Stage II. The Household Characteristics Questionnaire contained 51 questions on occupation, indoor-outdoor time budgets, smoking habits, hobbies, physical condition, diet, heating/ventilation, and sources of drinking water for the target households of Stage III. The third instrument was a 24-h activity recall questionnaire containing questions on exposure to smokers, occupations or businesses, cleaning agents, chemical mixtures, pesticides, food groups and auto or bus trips during the two 12-h monitoring periods.

Chemical Sampling and Analysis

Personal air, breath and water samples were collected from each of 355 participants. Two consecutive 12-h integrated personal air samples were collected on Tenax GC cartridges: an ''overnight'' sample (6 p.m.–6 a.m.) collected in people's homes for the most part; and a ''daytime'' (6 a.m.–6 p.m.) sample. The breath sample was collected immediately following the daytime air sample. Drinking water samples from home and work were also collected. Two consecutive 12-h outdoor air samples were collected from identical Tenax cartridges located in the backyards of 86 participants concurrently with their personal monitoring samples. This allowed comparison of simultaneous indoor and outdoor concentrations.

A sampling team (two field technicians) could sample up to three participants in a day. With three sampling teams in the field, a maximum of nine people per day could be sampled. However, due to scheduling difficulties usually six to seven participants were sampled per day.

Air

Volatile organic compounds were collected by pulling air through a glass cartridge containing Tenax GC using a constant flow pump. Approximately 20 L of air was sampled over a 12-h period using flow rates of about 30 mL/min. For fixed-site sampling the pump and Tenax cartridge were placed inside a metal box with only the inlet end of the sample cartridge protruding. For personal air sampling, the pump and cartridge were carried by the study participant with the inlet of the sample cartridge placed in the subject's breathing zone. A sampling vest was designed to minimize participant inconvenience during sampling. Cartridges were stored in sealed cans until analysis.

Recovery of volatile organic compounds from Tenax GC cartridges was accomplished by thermal desorption and purging with helium into a liquid nitrogen cooled nickel capillary trap. The vapors were then introduced into a high resolution fused silica chromatographic column where the constituents were separated from each other. Characterization and quantification of the constituents in the sample were accomplished by electron impact mass spectrometry by measuring the intensity of the extracted ion current profile. Instrumental limits of detection were calculated for each of the mass spectrometers (a Finnigan 3300 quadrupole and an LKB 2091 magnetic sector system) used during analysis. Representative values for most target compounds ranged from 2–10 ng/cartridge.

Initial and final flow checks differed by less than 5% for virtually all samples. Fixed-site samples showed slightly greater flow differences (up to 10%) than the personal air samples; the greater temperature range experienced by the pumps at the fixed site is a likely reason for the difference. Although sampling pump flow rates were constant (±5%), different flow rates were used, covering a range of 25 mL/min to 35 mL/min. Total air volume collected was also dependent on sampling times, which, depending on each participant's availability, varied from 7 h to 15 h.

The main problem (although detected in far fewer than 1% of the samples) was accidental disconnection of the sampling cartridge tubing from the pump. In some instances the tubing was immediately re-attached by the respondent, and the sample collection was not invalidated. In other instances the sample was lost. Loss of sample due to pump failure was negligible.

Recoveries of the target chemicals from the Tenax GC cartridges were evaluated by analyzing field and laboratory controls. These controls were Tenax GC cartridges spiked with ~200 ng of the target compounds prior to sampling. Some control cartridges were stored in the laboratory (lab controls); others were taken to the field and returned with the field samples (field controls). Control cartridges (N = 201) were analyzed simultaneously with field samples drawn from the same batch of Tenax.

Because of the large number of samples collected, and the time required to analyze all samples, some of the samples were stored for about 10 weeks before analysis. Control samples run with the field samples showed expected losses on the order of 10–20% of target compounds resulting from storage. Final calculated concentrations were corrected by dividing by the observed recovery efficiencies.

Field and laboratory blank cartridges (N = 155) were also analyzed along with field samples to assess potential background contamination. Average blank backgrounds for each batch were subtracted from observed cartridge loadings in calculating concentrations.

Precautions have been instituted to minimize contamination of sample cartridges in the field-based operations. All cartridges are kept in a large Tedlar bag under a constant flow of charcoal-cleaned helium. This practice has been used during several field trips and appears effective in controlling contamination.

Water

Water samples were collected in the morning and evening from the kitchen tap in each respondent's home. Water was turned on and allowed to run for 20 sec prior to sampling. Grab samples were collected without headspace in 40 mL amber glass vials with sodium thiosulfate added to quench residual chlorine, if present. The samples were refrigerated and remained sealed until analysis. The small sample size collected allowed for convenience in transport and storage.

For analysis, a 5 mL water sample was introduced into the purging apparatus, and purgeable organics were purged from the aqueous phase with helium. The organic constituents were swept from the purging device onto a short Tenax GC column. The trapped components were thermally desorbed and backflushed with carrier gas onto the head of the GC column packed with SP-1000 and separated under programmed conditions. At the end of the column the effluent was split; half for flame ionization detection of aromatic hydrocarbons and half for electrolytic conductivity detection of organohalides. The extraction/concentration technique increases the quantity of compounds by a factor of 1000 over what may be directly injected onto a GC column. Splitting the effluent for benzene analyses halves the sensitivity of the method.

Samples were analyzed on a 1.8 m × 2 mm I.D. glass column packed with 1% SP-1000 on Carbopak B using temperature program conditions. All target analytes were resolved under these conditions except *m*- and *p*-dichlorobenzene and *m*- and *p*-xylene.

Control samples were analyzed to obtain recovery factors for each analyte. Each target was spiked into prepurged, distilled/deionized water samples (25 mL). Laboratory controls remained in storage at Research Triangle Institute (RTI) during the field operations while field controls were transported to the field work station and returned with the field samples.

Breath

Human breath samples were collected by having the subject inhale humidified ultrapure air and exhale through a Douglas 2-way valved mouthpiece of a specially-designed spirometer into a 40 L Tedlar holding bag. The 40 L breath sample was then pumped through two parallel Tenax cartridges to concentrate the organic compounds for later analysis. Tenax cartridges were analyzed by thermal desorption/glass capillary gas chromatography/mass spectrometry using methods identical to those described for volatiles in air.

Several problems occurred with sample collection during the study. Many breath samples were highly contaminated with aromatic hydrocarbons (i.e., benzene, toluene). Apparently, van air was being pulled into the sampling bags and through the Tenax cartridge during pumping. To remedy this problem, the spirometer design was modified and new spirometers constructed. These spirometers did not leak and problems with sample contamination were eliminated.

The analysis procedure is identical to that used for ambient air samples. Spirometer controls were spiked cartridges sent to the field or stored in the laboratory and then analyzed with the field samples. No data were available on analyte recovery from the spirometer itself. However, during spirometer validation, recovery experiments were performed with acceptable results.

Table 1. Blank and control data for air and breath volatiles.

	Blanks (ng/cartridge ± S.D.)		Controls (% Recovery ± S.D.)	
Compound	Field (N = 76)	Lab (N = 79)	Field (N = 110)	Lab (N = 91)
Vinylidene chloride	1 ± 2	< 1	85 ± 23	110 ± 33
Chloroform	22 ± 20	8 ± 6	89 ± 22	90 ± 39
1,2-Dichloroethane	< 1	< 1	100 ± 15	106 ± 31
1,1,1-Trichloroethane	33 ± 21	14 ± 17	87 ± 19	94 ± 28
Benzene	97 ± 64	41 ± 26	86 ± 22	90 ± 26
Carbon tetrachloride	2 ± 3	15 ± 42	80 ± 20	93 ± 24
Trichloroethylene	3 ± 5	1 ± 2	95 ± 12	99 ± 24
Bromodichloromethane	ND	ND	96 ± 19	98 ± 11
Dibromochloromethane	ND	ND	95 ± 17	93 ± 13
Tetrachloroethylene	11 ± 10	2 ± 4	108 ± 18	109 ± 29
Chlorobenzene	1 ± 3	2 ± 3	110 ± 24	109 ± 32
Bromoform	ND	ND	96 ± 19	92 ± 15
Dibromochloropropane	< 1	ND	96 ± 17	77 ± 24
Styrene	2 ± 3	2 ± 3	104 ± 14	92 ± 15
p-Dichlorobenzene	3 ± 7	1 ± 1	101 ± 11	87 ± 13
Ethylbenzene	12 ± 13	5 ± 10	95 ± 14	95 ± 18
o-Xylene	8 ± 9	3 ± 5	100 ± 13	88 ± 19
p-Xylene	22 ± 21	7 ± 11	100 ± 14	91 ± 18
o-Dichlorobenzene	1 ± 2	1 ± 1	96 ± 13	85 ± 15

Quality of the Data

An extensive quality assurance (QA) program was carried out. About 30% of all samples were either blanks, spikes, or duplicates. Every type of analysis was repeated for 10% of samples in an external QA laboratory (IIT Research Institute). Audits of laboratory activities were undertaken by EPA's Environmental Monitoring Systems Laboratory at Research Triangle Park, NC (EMSL-RTP). A separate QA report (McSorley 1982) was written by an independent laboratory (Northrop Services, Inc.) concluding that no significant analytical differences could be found among the three laboratories (Research Triangle Institute, IIT Research Institute, and EMSL-RTP).

Results

Quality Control

Recovery efficiencies for 201 laboratory and field control cartridges used for air and breath samples ranged from 85–110% for nearly all target compounds (Table 1). Background levels of field blanks were usually less than 20 ng/cartridge (the equivalent of 1 μg/m^3) with the exception of benzene (97 ± 64 ng/cartridge) and 1,1,1-trichloroethane (33 ± 21). Field blanks (N = 76) often doubled or tripled the laboratory blank levels (N = 79), indicating some contamination during transport or storage (Table 1).

Recoveries for the water control samples were lower (typically 60–80%) but also had smaller variation than for air and breath samples (Table 2). Two targets—bromoform and dibromochloropropane—were poorly recovered and difficult to analyze.

Results of 202 duplicate analyses for 11 target compounds in air and breath samples indicate that median coefficients of variance range from 20–40% in most cases (Table 3).

Table 2. Control data[a] for volatiles in water from Elizabeth.

	% Recovery ± S.D.	
Compound	Field[b]	Lab[c]
Vinylidene chloride	67 ± 17	50 ± 25
Chloroform	63 ± 4[d]	60 ± 8[e]
1,2-Dichloroethane	68 ± 74	68 ± 9
1,1,1-Trichloroethane	56 ± 16	44 ± 15
Carbon tetrachloride	62 ± 15	46 ± 16
Trichloroethylene	64 ± 6	57 ± 10
Bromodichloromethane	57 ± 6	54 ± 6
Chlorodibromomethane	48 ± 7	45 ± 6
Tetrachloroethylene	69 ± 5	62 ± 12
Chlorobenzene	70 ± 12	63 ± 7
Bromoform	45 ± 5	45 ± 6
Dibromochloropropane	27 ± 5	27 ± 5
Dichlorobenzene	62 ± 4	62 ± 6
Benzene	83 ± 7	78 ± 9
Toluene	83 ± 8	83 ± 11
Ethylbenzene	79 ± 9	75 ± 12
Styrene	65 ± 9	64 ± 11
Xylene	75 ± 9	71 ± 11

[a] Spiked at 20 ppb
[b] N = 14
[c] N = 12
[d] N = 10
[e] N = 6

Table 3. Coefficients of variance (%) for duplicate air and breath samples in New Jersey—Season I.

	Median			75th Percentile		
	Personal[a]	Outdoor[b]	Breath[c]	Personal	Outdoor	Breath
Chloroform	20	24	36	35	70	63
1,1,1-Trichloroethane	27	23	46	45	67	56
Benzene	36	47	41	69	67	72
Carbon tetrachloride	24	15	42	37	32	59
Trichloroethylene	14	25	28	31	37	48
Tetrachloroethylene	21	20	18	37	31	41
Styrene	18	18	22	38	37	41
p-Dichlorobenzene	23	22	16	40	27	43
Ethylbenzene	20	27	30	42	35	66
o-Xylene	19	21	15	41	43	56
m,p-Xylene	24	24	23	50	48	58

[a]N = 134
[b]N = 34
[c]N = 35

Results and Discussion

Response Rate

Overall, 4,426 homes completed the screening questionnaire out of a target number of 5,208. Information was collected on ~11,414 persons in these households. From these, 618 persons were classified as eligible for monitoring and 355 (57%) completed all scheduled sampling (Table 4). Some individuals were classified as ineligible for exposure monitoring because they had moved outside the target area, were physically or mentally unable to participate, or were not correctly identified with regard to a key stratification variable. Combining these results with the screening response rate of 85% produces an overall response rate of 49%. Low response rates are typical of personal monitoring studies due to the unusually high respondent burden. Nonresponse produces bias to the extent that exposure levels are related to whether or not a person is willing to participate in the study. Hence, the effect of high nonresponse may not be as severe for a personal monitoring study as it would be for a study that measures personal attitudes and opinions. Moreover, nonresponse weight adjustments reduce this bias to the extent that respondents and nonrespondents are more alike within weighting classes than in the overall population. Finally, a small study of sample nonrespondents revealed no significant differences between respondents and nonrespondents for the New Jersey TEAM sample. Thus, the findings of the study should be applicable to the target population of 128,000 New Jersey residents.

Samples Collected

About 1900 air, breath, and water samples were collected and chemically analyzed (Table 5). An additional 900 quality control samples (duplicates, spikes, and blanks) were analyzed.

Percent Detected of Target Chemicals

For each of the 20 target chemicals in each of the sample types (breath, water, personal, and outdoor air) the weighted percent of quantifiable concentrations is shown in Table 6. Eleven chemicals were found in 50–100% of the air and breath samples; only the three chlorinated trihalomethanes were ubiquitous in drinking water samples.

Concentrations

Summary statistics for breath, air, and water samples are given in Tables 7 to 12. These values are weighted estimates for the combined target population of 128,000. Only the eleven most prevalent chemicals are included in the air and breath tables.

Table 4. Final status of eligible housing units and known[a] eligible respondents.

Eligible Housing Units	5208	(100.0%)
Completed screening	4426	(85.0%)
Refused screening	260	(5.0%)
Never at home	343	(6.6%)
Other	179	(3.4%)
Eligible Respondents[a]	618	(100.0%)
Completed study	355	(57.4%)
Partial completion	40	(6.5%)
Refused	216	(35.0%)
Other	7	(1.1%)

[a]Not included are 77 persons who could not be located after repeated attempts.

Table 5. Sample collection results of main study samples.

	Personal Air	Fixed Air	Breath	Drinking Water
Field Samples (scheduled/collected)	724/705	196/183	364/358	724/718
Field Duplicates (scheduled/collected)	144/131	36/32	38/37	72/70
Field Blanks (scheduled/utilized)	81/74	22/19	56/50	53/47
Field Controls (scheduled/utilized)	81/73	22/19	56/51	53/49
Lab Blanks (scheduled/utilized)	70/70	26/26	42/42	33/33
Lab Controls (scheduled/utilized)	66/66	26/26	42/42	33/33
Total	1166/1119 (96%)	328/305 (93%)	598/580 (97%)	968/850 (98%)

Table 6. Percentage of population[a] with compound concentrations measurable[b] for Bayonne and Elizabeth:

	Breath	Overnight Personal Air	Daytime Personal Air	Overnight Outdoor Air	Daytime Outdoor Air	Drinking Water
Minimum Sample Size:	295	346	339	81	86	265
Maximum Sample Size:	339	348	341	86	90	354
Vinylidene chloride	12	3	6	1	0.4	45
Chloroform	60	59	44	47	37	100
1,2-Dichloroethane	2	3	3	3	4	0.4
1,1,1-Trichloroethane	80	80	73	85	81	50
Benzene	89	95	90	89	75	0.1
Carbon tetrachloride	18	30	23	53	50	6
Trichloroethylene	29	52	46	54	46	57
Bromodichloromethane	0.1	2	2	0.1	0	100
Dibromochloromethane	0	0	0	0	0	100
Tetrachloroethylene	93	92	88	80	80	58
Chlorobenzene	3	9	4	2	4	0.9
Bromoform	0	0	0	0	0	3
Dibromochloropropane	0	1	0	0	0	–
Styrene	48	83	77	34	18	0
m,p-Dichlorobenzene	62	82	76	44	23	2
o-Dichlorobenzene	2	7	9	0.6	1	–
Ethylbenzene	93	93	89	87	82	0
o-Xylene	85	87	83	82	74	–
m,p-Xylene	95	99	98	98	90	0

[a] Estimated population: 128,000

[b] Median quantifiable limits range between 1 and 5 $\mu g\ m^{-3}$ for most compounds (0.1 ng/mL for water)

Table 7. Weighted summary statistics for breath ($\mu g/m^3$) with Bayonne and Elizabeth combined—TEAM first season.

Estimated Population Size:	128,603
Minimum Sample Size:	295
Maximum Sample Size:	339

							Percentiles				
Compound	Mid Q.L.[a]	Arith. Mean	S.E.[b]	Geo. Mean[c]	S.E.[d]	Median	75	90	95	99	Range
Chloroform	2.08	3.12	0.34	1.30	1.13	1.80	3.70	8.20	11.5	26.0	.05–29.0
1,1,1-Trichloroethane	2.30	15.0	2.57	4.79	1.13	6.60	13.0	30.0	42.0	185	.06–520
Benzene	0.44	18.7	1.40	8.19	1.22	12.0	24.0	42.0	56.0	120	.02–200
Carbon Tetrachloride	1.70	1.31	0.26	0.60	1.15	0.69	1.06	2.25	2.7	20.0	.05–250
Trichloroethylene	1.50	1.77	0.21	0.93	1.13	0.88	1.80	3.94	5.9	14.0	.08–30.0
Tetrachloroethylene	4.10	13.3	1.83	7.33	1.08	6.80	12.9	31.0	44.0	190	.12–280
Styrene	0.97	1.15	0.13	0.72	1.11	0.79	1.25	2.40	3.0	7.2	.06–31.0
m,p-Dichlorobenzene	1.32	8.10	1.54	1.72	1.12	1.30	3.50	21.0	44.0	110	.11–158
Ethylbenzene	0.40	4.58	0.55	2.45	1.14	2.90	5.30	8.90	12.0	29.0	.02–290
o-Xylene	1.10	3.35	0.36	1.99	1.12	2.20	3.70	6.30	9.2	17.0	.05–220
m,p-Xylene	0.52	8.95	0.93	5.34	1.12	6.35	11.0	19.0	21.0	53.0	.05–350

[a] Mid Q.L. = Median Quantifiable Limit

[b] S.E. = Standard Error of Arith. Mean

[c] Geo. Mean = Geometric Mean

[d] S.F. = Geometric Standard Error = exp(s) where s is the standard error of the weighted mean of log(x).

Table 8. Weighted summary statistics for overnight personal air ($\mu g/m^3$) with Bayonne and Elizabeth combined—TEAM first season.

Estimated Population Size: 128,603
Minimum Sample Size: 346
Maximum Sample Size: 348

Compound	Mid Q.L.[a]	Arith. Mean	S.E.[b]	Geo. Mean[c]	S.E.[d]	Median	Percentiles 75	90	95	99	Range
Chloroform	2.96	8.73	2.62	3.32	1.08	3.30	7.90	16.0	24.0	215	.06–215
1,1,1-Trichloroethane	24.0	113	71.7	18.6	1.11	16.9	38.0	78.0	180	880	.36–8300
Benzene	0.60	29.7	4.98	12.5	1.09	15.0	32.0	54.0	73.0	320	.02–510
Carbon Tetrachloride	2.50	13.9	10.3	1.79	1.12	1.50	2.44	5.75	18.0	200	.14–1100
Trichloroethylene	2.45	7.27	2.28	2.60	1.11	2.25	4.80	12.0	22.5	140	.16–350
Tetrachloroethylene	2.16	11.3	0.95	6.30	1.10	6.35	12.0	26.0	35.0	70	.08–250
Styrene	0.76	2.68	0.33	1.52	1.08	1.75	3.00	4.60	6.20	16	.05–76
m,p-Dichlorobenzene	1.20	56.0	14.6	5.12	1.20	3.80	13.0	82.0	260	1200	.08–1500
Ethylbenzene	4.50	12.6	2.11	6.43	1.06	6.30	12.0	22.0	35.0	110	.17–380
o-Xylene	4.90	15.7	6.56	5.26	1.07	4.90	8.70	15.0	27.0	250	.16–750
m,p-Xylene	4.80	54.6	26.8	16.0	1.08	14.0	25.0	47.0	87.0	605	.20–3100

[a] Mid Q.L. = Median Quantifiable Limit
[b] S.E. = Standard Error of Arith. Mean
[c] Geo. Mean = Geometric Mean
[d] S.E. = Geometric Standard Error = exp(s) where s is the standard error of the weighted mean of log(x).

Frequency distributions for the combined Bayonne-Elizabeth target population of 128,000 persons are shown for all personal air, outdoor air, and breath samples of benzene and 1,1,1-trichloroethane (Figs. 1–2). Notable are the great range of exposures (<1 $\mu g/m^3$ to >100 $\mu g/m^3$); the greater personal exposures than outdoor concentrations; and the greater breath concentrations than outdoor concentrations.

The arithmetic and geometric means were calculated by assigning a value of ½ the limit of detection (LOD) to all undetected quantities, and a value midway between the LOD and the quantifiable limit (QL) to all trace quantities (detectable but not quantifiable). Since the QL varied depending on the volume of the sample, a median QL is listed for each chemical in each environmental medium. Thus, values below the median QL are somewhat arbitrary. Also, the geometric mean may be sensitive to this arbitrary choice; the arithmetic mean is barely affected.

All distributions were analyzed for normality and for log-normality using the Kolmogorov-Smirnov test statistic. Only a few appeared to be log-normal, but nearly all were closer to log-normal than to normal. On closer examination, most distributions were ap-

Table 9. Weighted summary statistics for daytime personal air ($\mu g/m^3$) with Bayonne and Elizabeth combined—TEAM first season.

Estimated Population Size: 128,603
Minimum Sample Size: 339
Maximum Sample Size: 341

Compound	Mid Q.L.[a]	Arith. Mean	S.E.[b]	Geo. Mean[c]	S.E.[d]	Median	Percentiles 75	90	95	99	Range
Chloroform	4.10	7.43	1.27	3.01	1.09	3.10	7.70	19.0	28.5	89	.08–89
1,1,1-Trichloroethane	8.20	816	667	19.1	1.13	16.9	47.0	130	380	4400	.16–333,000
Benzene	1.10	26.2	1.40	11.2	1.12	17.0	32.0	65.0	81.0	160	.02–270
Carbon Tetrachloride	3.08	4.62	1.50	1.29	1.09	1.50	2.63	4.9	9.9	37	.06–900
Trichloroethylene	3.44	18.9	6.20	3.05	1.10	2.63	6.65	15.5	46.0	405	.19–1,400
Tetrachloroethylene	3.25	78.0	44.6	9.15	1.06	8.40	20.0	50.0	69.0	1100	.12–12,000
Styrene	1.06	15.2	7.17	1.84	1.10	2.00	3.50	6.0	10.0	140	.07–6,500
m,p-Dichlorobenzene	1.40	35.1	7.88	4.73	1.15	3.50	8.70	85.0	210	490	.11–790
Ethylbenzene	1.80	24.8	7.62	7.33	1.10	7.90	14.0	32.0	52.0	420	.08–1,500
o-Xylene	5.50	17.2	4.49	6.12	1.08	5.81	11.0	22.0	35.0	300	.08–830
m,p-Xylene	2.10	48.6	10.5	18.5	1.09	18.0	32.0	59.0	100	860	.12–1,800

[a] Mid Q.L. = Median Quantifiable Limit
[b] S.E. = Standard Error of Arith. Mean
[c] Geo. Mean = Geometric Mean
[d] S.E. = Geometric Standard Error = exp(s) where s is the standard error of the weighted mean of log(x).

Table 10. Weighted summary statistics for overnight outdoor air (μg/m^3) with Bayonne and Elizabeth combined—TEAM first season.

Estimated Population Size: 128,603
Minimum Sample Size: 81
Maximum Sample Size: 86

Compound	Mid Q.L.[a]	Arith. Mean	S.E.[b]	Geo. Mean[c]	S.E.[d]	Median	Percentiles				Range
							75	90	95	99[e]	
Chloroform	1.20	1.22	0.22	0.55	1.26	0.66	1.40	2.90	4.80	6.60	.04–21.5
1,1,1-Trichloroethane	3.00	5.41	0.80	3.66	1.21	4.50	8.40	11.0	12.0	19.0	.05–40.0
Benzene	0.36	8.61	1.17	4.10	1.23	6.70	11.0	15.0	24.0	49.0	.04–91.0
Carbon Tetrachloride	1.20	1.16	0.13	0.79	1.12	0.81	1.30	2.06	2.50	5.90	.04–14.0
Trichloroethylene	4.80	2.13	0.37	1.37	1.20	1.30	3.00	3.88	7.50	11.0	.08–15.0
Tetrachloroethylene	0.99	3.72	0.31	2.08	1.17	2.60	4.10	6.90	15.0	23.0	.06–27.0
Styrene	1.30	0.90	0.10	0.55	1.11	0.61	0.94	1.70	2.80	5.40	.06–11.0
m,p-Dichlorobenzene	1.30	1.54	0.23	1.00	1.12	1.20	1.69	2.50	4.00	13.0	.07–13.0
Ethylbenzene	1.20	3.76	0.47	2.49	1.24	2.90	5.30	6.90	11.0	13.0	.04–20.0
o-Xylene	1.70	3.97	0.54	2.80	1.23	2.90	5.50	7.90	11.0	11.6	.18–27.0
m,p-Xylene	1.04	11.0	1.40	8.34	1.20	9.90	16.0	21.0	26.0	29.0	.13–70.0

[a] Mid Q.L. = Median Quantifiable Limit
[b] S.E. = Standard Error of Arith. Mean
[c] Geo. Mean = Geometric Mean
[d] S.E. = Geometric Standard Error = exp(s) where s is the standard error of the weighted mean of log(x).
[e] Because of limited sample sizes, the estimated percentages should be viewed with caution.

proximately log-normal up to the 90th percentile but then departed from log-normality in the direction of more extremely high values than expected (heavy-tailed distributions). A set of 29 very high values (>1000 μg/m^3) were closely investigated for possible transcription errors; none were found. In 27 of the cases, an activity was listed (often occupation-related) that appeared to account for the extremely high value.

Therefore, both the arithmetic mean and the geometric mean (with their associated standard errors) have been calculated. Assuming log-normality, the geometric standard error may be interpreted as the square root of the factor that the geometric mean must be multiplied and divided by to provide the 95% confidence limits for the estimation of the true geometric mean of the target population. Since the geometric standard error is generally close to 1.1 for the personal air and breath measurements, the 95% confidence limits around the estimated geometric means are within a factor of 1.1^2, or roughly within ±20% of the true

Table 11. Weighted summary statistics for daytime outdoor air (μg/m^3) with Bayonne and Elizabeth combined—TEAM first season.

Estimated Population Size: 128,603
Minimum Sample Size: 86
Maximum Sample Size: 90

Compound	Mid Q.L.[a]	Arith. Mean	S.E.[b]	Geo. Mean[c]	S.E.[d]	Median	Percentiles				Range
							75	90	95	99[e]	
Chloroform	1.32	1.50	0.31	0.58	1.21	0.60	2.00	3.50	6.90	8.70	.04–8.80
1,1,1-Trichloroethane	1.44	8.60	1.98	3.32	1.28	4.80	10.0	14.3	20.0	41.0	.05–470
Benzene	1.08	9.54	0.83	3.77	1.20	7.80	16.0	20.0	27.0	33.0	.05–44.0
Carbon Tetrachloride	1.50	1.00	0.15	0.70	1.23	0.94	1.20	1.80	1.95	3.80	.04–7.10
Trichloroethylene	1.86	2.37	0.30	1.43	1.13	1.60	2.60	6.30	6.88	8.10	.08–11.0
Tetrachloroethylene	1.20	8.32	1.58	3.76	1.27	3.60	13.0	23.0	26.0	57.0	.11–57.0
Styrene	1.40	0.82	0.07	0.61	1.10	0.71	1.13	1.50	1.90	3.44	.07–5.12
m,p-Dichlorobenzene	1.60	1.94	0.51	0.78	1.11	0.81	1.30	2.88	5.40	57.0	.10–57.0
Ethylbenzene	2.10	4.26	0.64	2.61	1.24	3.20	5.90	8.70	14.0	15.0	.06–16.0
o-Xylene	2.30	3.99	0.61	2.65	1.21	3.10	5.30	8.30	11.0	12.0	.08–19.0
m,p-Xylene	2.90	11.6	1.89	6.96	1.27	8.20	18.0	24.5	31.0	35.0	.08–37.0

[a] Mid Q.L. = Median Quantifiable Limit
[b] S.E. = Standard Error of Arith. Mean
[c] Geo. Mean = Geometric Mean
[d] S.E. = Geometric Standard Error = exp(s) where s is the standard error of the weighted mean of log(x).
[e] Because of limited sample sizes, the estimated percentages should be viewed with caution.

Table 12. Weighted summary statistics for water (ng/mL) with Bayonne and Elizabeth combined—TEAM first season.

Estimated Population Size: 128,603
Minimum Sample Size: 265
Maximum Sample Size: 354

Compound	Mid Q.L.[a]	Arith. Mean	S.E.[b]	Geo. Mean	S.E.[d]	Median	Percentiles 75	90	95	99	Range
Vinylidene Chloride	.05	0.25	0.03	0.08	1.13	0.03	0.24	0.82	1.19	1.85	.03–2.36
Chloroform	.05	69.9	1.49	65.9	1.03	66.8	83.4	94.8	102	128	.03–168
1,1,1-Trichloroethane	.05	0.59	0.08	0.13	1.18	0.03	0.75	2.14	3.03	4.43	.03–5.34
Trichloroethylene	.05	0.56	0.08	0.15	1.19	0.06	0.73	1.87	2.50	3.93	.03–4.18
Bromodichloromethane	.10	13.6	0.16	13.2	1.02	13.4	14.6	16.5	17.5	19.9	.06–23.4
Dibromochloromethane	.10	2.45	0.06	2.34	1.02	2.38	2.71	3.21	3.38	4.17	.06–8.39
Toluene	.50	0.42	0.04	0.36	1.04	0.31	0.31	0.64	0.97	2.63	.31–2.73
Tetrachloroethylene	.05	0.44	0.06	0.13	1.18	0.06	0.60	1.45	1.82	2.65	.03–3.32

[a] Mid Q.L. = Median Quantifiable Limit
[b] S.E. = Standard Error of Arith. Mean
[c] Geo. Mean = Geometric Mean
[d] S.E. = Geometric Standard Error = exp(s) where s is the standard error of the weighted mean of log(x).

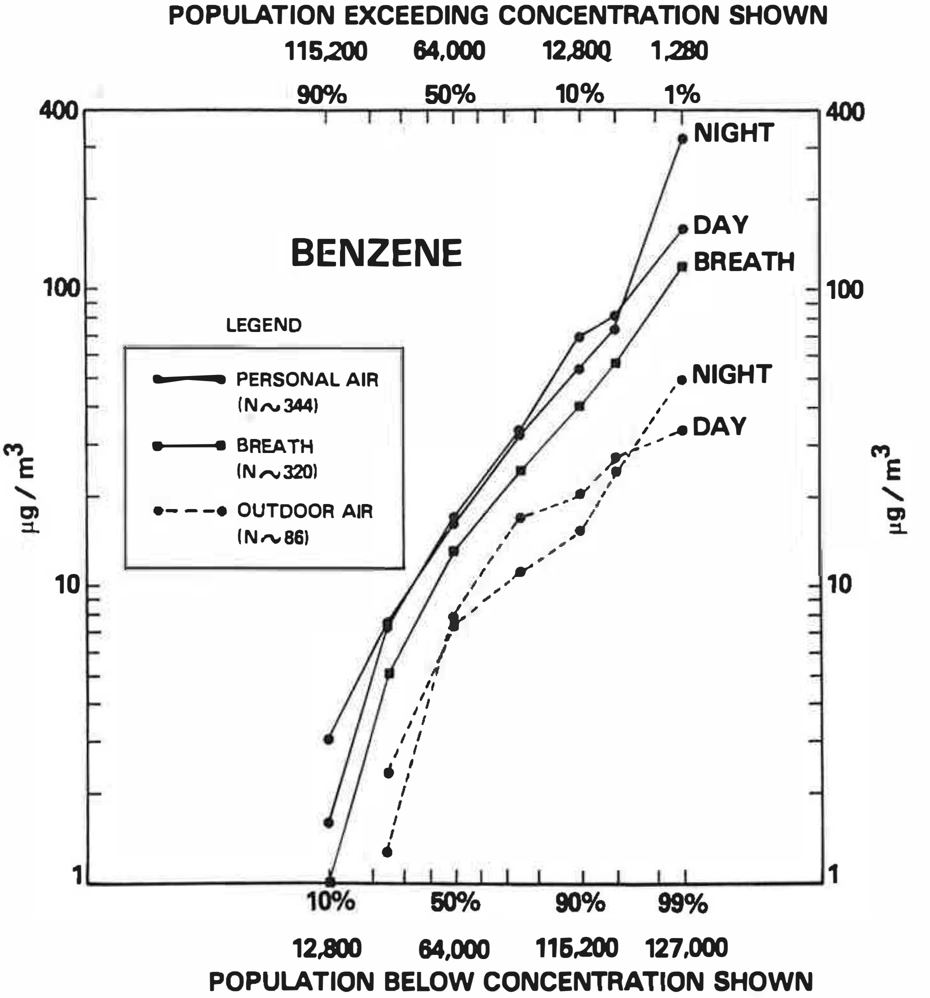

Fig. 1. Benzene: Estimated frequency distributions of personal air exposures, outdoor air concentrations, and exhaled breath values for the combined Elizabeth-Bayonne target population (128,000). All air values are 12-hour integrated samples. The breath value was taken following the daytime air sample (6:00 a.m.-6:00 p.m.). All outdoor air samples were taken in the vicinity of the participants' homes.

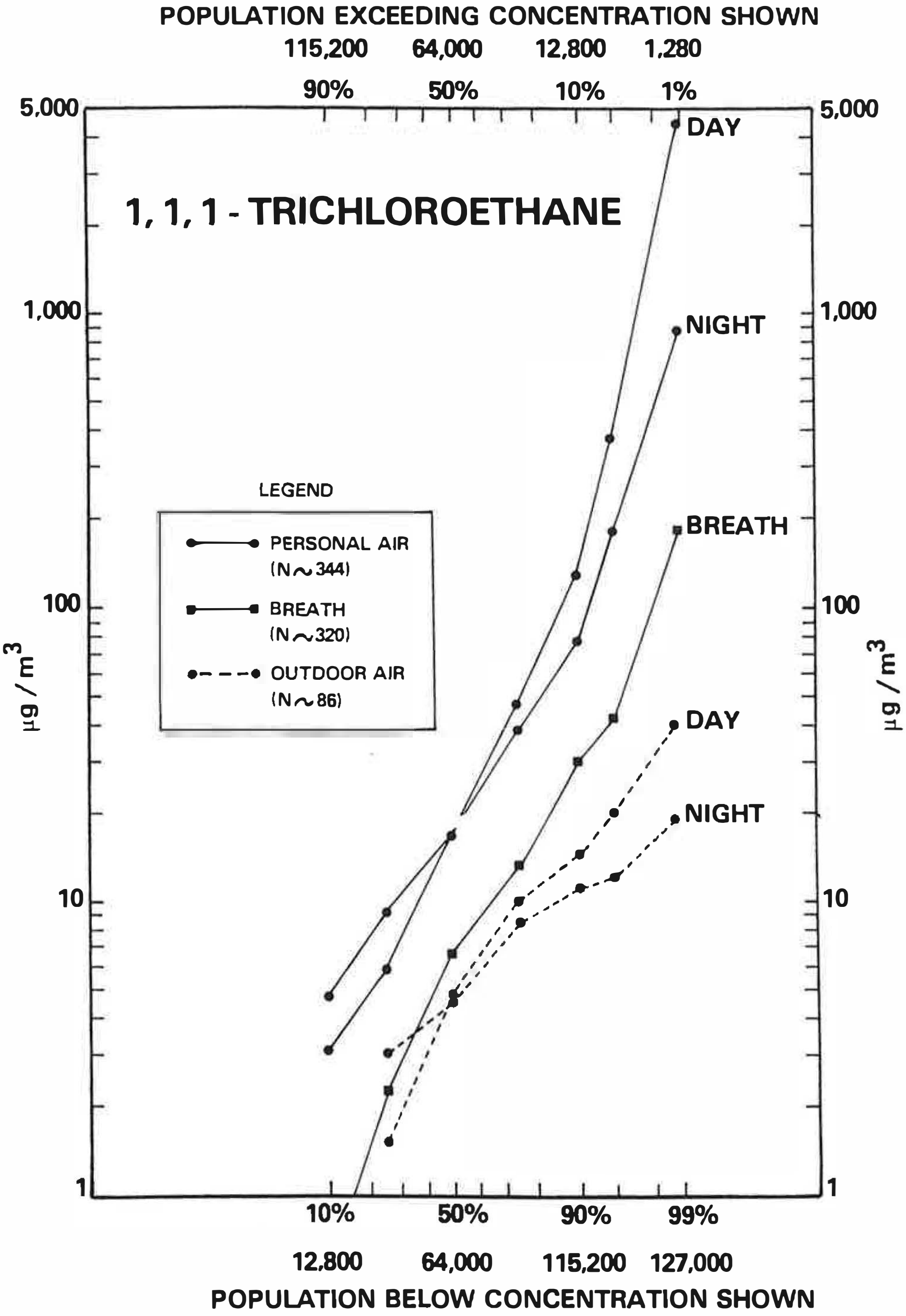

Fig. 2. 1,1,1-Trichloroethane: Estimated frequency distributions of personal air exposures, outdoor air concentrations, and exhaled breath values for the combined Elizabeth-Bayonne target population (128,000). All air values are 12-hour integrated samples. The breath value was taken following the daytime air sample (6:00 a.m.-6:00 p.m.). All outdoor air samples were taken in the vicinity of the participants' homes.

value. Owing to the smaller number of outdoor air samples, the 95% confidence limits around the estimated geometric mean are relatively larger, averaging about ±40%.

Nearly all overnight personal air exposures were essentially indoor air measurements, since the monitor was on the bedside table while the participant slept. Thus, these indoor measurements may be compared with outdoor measurements for those homes with an outdoor monitor on their property (Table 13). The indoor/outdoor ratios for the eleven prevalent compounds ranged between 1.5 and 4.0 for median values, and between 1.0 and 70 for maximum concentrations.

Table 13. Summary of medians, maximum concentrations, and their ratios for matched overnight personal air and overnight outdoor air—TEAM first season.

Compound	Overnight Outdoor Air	Overnight Personal Air	Personal/ Outdoor Ratio
Chloroform	0.74 (21.5)*	2.94 (215)	3.97 (10.0)
1,1,1-Trichloroethane	4.20 (40.0)	15.6 (880)	3.71 (22.0)
Benzene	7.00 (91.0)	13.0 (120)	1.86 (1.32)
Carbon Tetrachloride	0.81 (14.0)	1.38 (14.0)	1.70 (1.00)
Trichloroethylene	1.34 (15.0)	2.00 (47.0)	1.49 (3.13)
Tetrachloroethylene	2.60 (27.0)	5.60 (250)	2.15 (9.26)
Styrene	0.67 (11.0)	1.80 (53.5)	2.69 (4.86)
m,p-Dichlorobenzene (isomers)	0.80 (13.0)	2.80 (915)	3.50 (70.4)
Ethylbenzene	3.20 (20.0)	6.10 (320)	1.91 (16.0)
o-Xylene	3.00 (27.0)	4.98 (46.0)	1.66 (1.70)
m,p-Xylene (isomers)	9.90 (70.0)	15.5 (120)	1.57 (1.71)

*Median (maximum).

It is evident from these tables that personal exposures, whether day or night, are consistently greater than outdoor concentrations. Even breath levels usually exceed ambient concentrations.

Correlations

Spearman correlations for measurable concentrations of the prevalent chemicals in selected media are listed in Table 14. Breath levels for nine of the eleven

Table 14. Spearman correlations of measurable amounts for breath, daytime personal air, daytime outdoor air and water for selected chemicals—TEAM first season.[a]

	Breath and Daytime Personal Air		Breath and Daytime Outdoor Air		Daytime Personal Air and Daytime Outdoor Air	
Compound	Spearman Correlation	Sample Size	Spearman Correlation	Sample Size	Spearman Correlation	Sample Size
Chloroform	.07	84	.51*	18	.39	19
1,1,1-Trichloroethane	.28*	203	.39*	56	.31*	52
Benzene	.17*	275	.31*	58	.03	59
Carbon Tetrachloride	.01	24	.83*	6	.23	11
Trichloroethylene	.54*	58	−.03	12	.35	24
Tetrachloroethylene	.44*	286	.41*	63	.51*	66
Styrene	.23*	129	−.27	8	−.19	11
m,p-Dichlorobenzene	.63*	140	.53	14	.29	18
Ethylbenzene	.37*	282	.08	69	.27*	63
o-Xylene	.28*	233	.15	62	.16	59
m,p-Xylene	.30*	309	.20	77	.15	80

	Breath and Water		Overnight Personal Air and Overnight Outdoor Air		Daytime Outdoor Air and Water	
Compound	Spearman Correlation	Sample Size	Spearman Correlation	Sample Size	Spearman Correlation	Sample Size
Chloroform	.25*	188	.27	28	.28	35
1,1,1-Trichloroethane	−.04	125	.20	57	−.27	41
Benzene	—	0	.19	69	—	0
Carbon Tetrachloride	−.20	4	.57*	17	—	0
Trichloroethylene	−.13	39	.49*	31	.36	27
Tetrachloroethylene	−.04	167	.26	56	−.03	40
Styrene	—	0	.09	33	—	0
m,p-Dichlorobenzene	−.60	6	.63*	32	—	0
Ethylbenzene	—	0	.26*	68	—	0
o-Xylene	—	0	.23	63	—	0
m,p-Xylene	—	0	.22	77	—	0

*Significantly different from zero at .05 level.

compounds are significantly related to the previous 12-h exposures in air. Of the two compounds showing no relationship, chloroform in breath is significantly related to levels in drinking water. Only carbon tetrachloride showed no relationship of body burden to exposure.

Frequency of Appearance of Target Compounds

Air and breath. The target chemical compounds may be sorted into four classes based on their presence in air and breath samples (Table 15). The first class, ubiquitous compounds that were present in 60–98% of all air and breath samples, includes two common solvents (1,1,1-trichloroethane and tetrachloroethylene) and four aromatic components of gasoline, paints, and other petrochemical products (benzene, two xylene isomers, and ethylbenzene).

The second class, compounds often but not always present in all sample types, includes two additional solvents (carbon tetrachloride and trichloroethylene); a compound (chloroform) whose main source is chlorinated water, and which may be released into indoor air during hot showers; and two components of common consumer products (styrene, used in insulation and plastics; and *p*-dichlorobenzene, used in moth crystals and room deodorizers). An indication that the source of the last two compounds was in the home can be discerned in the much greater frequencies of detection in overnight personal air samples (70–80%) compared to outdoor air samples (20–40%).

Table 15. Target compounds sorted by percent measurable in air and breath samples.

	Range of % Measurable
Ubiquitous	
m,p-xylene	86–98
tetrachloroethylene	70–91
ethylbenzene	75–89
benzene	72–89
1,1,1-trichloroethane	67–79
o-xylene	60–78
Often present	
chloroform	23–59
carbon tetrachloride	10–38
trichloroethylene	26–36
styrene	4–79
m,p-Dichlorobenzene	10–71
Occasionally found	
ethylene dichloride	0–2
chlorobenzene	2–3
o-dichlorobenzene	0–7
bromodichloromethane	0–2
Never found in air or breath	
bromoform	0
dibromochloromethane	0
dibromochloropropane	0

The third class of substances were only occasionally found (< 10% detected in most sample types). This class includes ethylene dichloride; bromodichloromethane; chlorobenzene; and o-dichlorobenzene.

Finally, three brominated substances not found in air or breath included two trihalomethanes and dibromochloropropane. (Previous work—Phase I of the TEAM Study—showed that levels of bromodichloromethane increased sufficiently in summer to become readily detectable in the personal air and exhaled breath of selected Elizabeth and Bayonne residents. Thus, the present negative findings for bromodichloromethane in air and breath should not be extrapolated to apply to all seasons.)

Drinking water. In drinking water, a different set of the target compounds were present. All of the four trihalomethanes except bromoform were present in >99% of all samples from both Bayonne and Elizabeth. In Bayonne, no other target compounds appeared with much frequency. In Elizabeth, four target compounds (carbon tetrachloride, tetrachloroethylene, trichloroethylene, 1,1,1-trichloroethane) often appeared, but their levels were generally so low that they made no appreciable contributions to the exposures of the participants. (Bayonne and Elizabeth have separate surface water supplies). Thus, although several of the target compounds were often present in drinking water, only chloroform and perhaps bromodichloromethane appeared to make important contributions to the exposures of the 350 participants.

Concentrations of Target Compounds

Air and breath. Examination of the concentrations in Tables 7–12 and Figs. 1–2 suggest the following observations:

1. Exposures were highly variable. For many compounds, the range in personal air exposures exceeded a factor of 1000 or even 10,000. This was far greater than for typical criteria pollutants such as carbon monoxide and suspended particulates. The range in breath concentrations was almost equally variable, strongly indicating that the higher exposures were resulting in a higher body burden.
2. All eleven prevalent chemicals had higher personal air concentrations than outdoor air concentrations. This is the case even for nighttime exposures, when participants were normally at home for the entire 12 h.
3. Breath levels were also often higher than outdoor levels. Since levels in exhaled breath are often only 20–40% of total intake, the remainder being metabolized or excreted through other pathways, this is further indication that exposures are higher than would be expected from observed outdoor concentrations.

4. The ratio of personal exposures to outdoor levels increased with higher exposures. The decreasing importance of outdoor levels in contributing to the higher exposures was illustrated by comparing indoor nighttime exposures (when persons were almost invariably inside their homes) to outdoor nighttime concentrations for the 75th percentile and the 99th percentile of each distribution. The ratios increased from 2–5 at the 75th percentile up to 10–20 at the 99th percentile for most of the target compounds. The daytime personal air exposures were usually the highest, as expected since this time period included the commuting and occupational activities. However, the nighttime personal air exposures, when people were normally sleeping, were nearly as high. In fact, all eleven prevalent chemicals had much higher nighttime indoor concentrations than nighttime outdoor concentrations, often 10 times higher.

5. The higher overnight personal exposures appear to implicate the home or personal activities within the home as the major source of exposure to most or all of the 11 compounds.

Drinking Water

6. Only chloroform and possibly bromodichloromethane are important contributions to total exposure from drinking water in the study area. The median value for chloroform in drinking water was 67 μg/L; in air, 3.2 μg/m^3. Assuming 2L of water intake per day and 20 cubic meters of air intake per day, the median intake of chloroform in water (134 μg) is about twice that in air (64 μg). Drinking water also accounts for most exposures to bromodichloromethane, since it was detected in less than 3% of the personal air samples.

Effects of residence, occupation, and activities on exposure city. Student *t*-tests on the geometric mean indicated that few chemicals showed significant ($p < 0.05$) differences between cities, with Elizabeth generally slightly higher on personal air and breath samples.

Proximity to Point Sources. Census tracts were classified as high and low proximity strata by whether they were within 1.5 km of any major point sources or not. In general, no differences in percent measurable or mean concentrations in any medium were seen between the high and low proximity strata.

Activities and Potential Sources. All participants were asked if they had been exposed to potential sources of the target chemicals on the day they were monitored or within the previous week. Sources included indus-

Table 16. Median concentrations ($\mu g/m^3$) of chemicals significantly ($p < .05$) higher in breath of persons exposed to potential sources during the day (week) they were monitored.

	N	a	b	c	d	e	f	g	h	i	j	k
All subjects	350	16	3.2	2.6	6.6	1.3	7.8	1.9	9.1	1.1	3.2	2.3
Chemical plants	21	25*	7.4*		12	2.3		6		12		
	37	(25)*				(2.3)*		(5.7)		(3.3)		
Plastics	11	27*	8.4*	7.1*	16*	2.4				12*		
	15	(22)	(7.2)	(5.8)*	(12)	(2)		(9.8)				
Paint	28	30*	5.7*	3.6	11*	3.3*	13*					
	58		(5.1)*	(3.4)	(10)*	(2)	(10)					
Petroleum plants	9	32				2.3						
	19	(27)				(2.4)						
Printing	9	28	5.5									
	17											
Auto exhaust	62	22										
	93											
Gas station	67	28*										
	154	(20)*										
Science lab	14	32										
	18											
Metal working	17								17			
	22						(10)					
Dry cleaners	13						22					
	37						(15)*					
Hospitals	13											
	38											
Furniture refinishing	7											
	10											
None of the above	124											

*$p < 0.005$

a = Benzene, b = Ethylbenzene, c = *o*-Xylene, d = m,p-Xylene, e = Styrene, f = Tetrachloroethylene, g = Trichloroethylene, h = 1,1,1-Trichloroethane, i = Carbon Tetrachloride, j = Chloroform, k = m,p-Dichlorobenzene trial plants, auto exhaust, and paint. For 10 of the 12 sources, at least one (and as many as 6) of the 11 most prevalent chemicals appeared at significantly higher levels in the breath of persons exposed during the day or week compared to those not exposed to the source. In most cases, the chemicals that were elevated were those expected to be associated with a given source, such as tetrachloroethylene with dry cleaners and benzene with service stations (Table 16).

A second series of questions concerned exposure to chemical groups or mixtures directly, without regard to source. These chemical mixtures included solvents, odorous compounds, and tobacco smoke. Again, certain compounds appeared at significantly higher levels in the breath of exposed persons compared to those not exposed (Table 17).

Thus breath analysis alone was effective in detecting increased exposures due to specific sources or chemical mixtures. The magnitude of the increases ranged from 30–40% up to factors of 3 or higher.

Personal air exposures to certain chemicals were also significantly elevated for persons recently exposed to potential sources compared to persons not exposed to that source. chemicals for which both personal air *and* breath levels of exposed persons were significantly elevated over persons unexposed to that source are listed in Table 18.

Since many chemicals have multiple sources, some members of the so called ''unexposed'' groups in the above analyses may have been exposed to the same chemical through a different source, thus blurring the distinction between exposed and unexposed groups. Therefore the breath and personal air levels of groups exposed to each source were compared to the group of persons who responded that they were not exposed to any source. The number of chemicals showing significant differences increased considerably. The number showing simultaneously elevated air and breath values doubled (Table 19).

Other personal or household characteristics appeared to be significantly associated with exposure, as determined by analysis of variance and stepwise regression (Table 20). Exposure to certain chemicals appeared to be strongly associated with smoking, employment, and other characteristics and activities.

By far the strongest association—stronger than any occupational category—was found between benzene and styrene levels in the breath of smokers. In both cases, levels in exhaled breath actually exceeded the previous exposures in air as measured by the personal

Table 17. Median concentrations (μg/m³) of chemicals significantly ($p < .05$) higher in breath of persons exposed to chemical groups or chemical mixtures during the day (week) they were monitored.

	N	Benzene	m,p-Xylene	o-Xylene	Ethylbenzene	Styrene	1,1,1-Trichloroethane	Tetrachloroethylene
Solvents	37	27	13.5*	4.4*	6*	2.2*		11
	79	(22)	(7.8)			(1.8)		
Odorous chemicals	82	23	8.3	3.2	3.9*			
	162							8
Degreasing compounds	19						20*	
	39							
Dust	63		7.7					
	87							
Tobacco smokers	161	23*	7.6		4	1.4		11
Tobacco smoke (including smokers)	209	20.5*	7.2		3.7			8.3
Tobacco smoke (nonsmokers only)	99							
Toxic chemicals	27							
	49							
Cleaning solutions	94							
	165							
"Other"	19							
	28							

*$p < .005$

Four chemicals (carbon tetrachloride, trichloroethylene, m,p-dichlorobenzene, and chloroform) showed no significant increases in breath of exposed persons.

Table 18. Chemicals showing significantly ($p < .05$) higher concentrations in air and breath of persons recently exposed to potential sources compared to persons not exposed to that source.

Potential Source	No. of Persons Exposed	Ratio of Median Concentrations: Breath	Ratio of Median Concentrations: Air
Dry Cleaners	37		
Tetrachloroethylene		2.8	2.0 (.01)[a]
Paint	28		
Styrene		2.6	1.6 (.002)
Ethylbenzene		1.8	1.8 (.0009)
o-Xylene		1.4	1.9 (.006)
m,p-Xylene		1.8	2.1 (.0003)
Auto Exhaust	62		
None			
Tobacco Smokers	161		
Styrene		1.4	1.4 (.0002)
Chemical Plant	21		
Styrene		1.8	1.8 (.02)
Ethylbenzene		2.3	1.5 (.008)
m,p-Xylene		1.8	1.6 (.01)
Pesticides	20		
None			
Furniture Refinishing	7		
None			
Printing Shop	9		
None			
Petroleum Plant	19		
None			
Science Laboratory	14		
None			
Service Station	67		
Benzene		1.9	1.2 (.03)
Plastics Manufacturing	11		
Styrene		2.0	2.4 (.04)
Metal Working	17		
1,1,1-Trichloroethane			4.3 (.02)
Tetrachloroethylene			1.6 (.05)
Hospital	13		
None			
Solvents	37		
Styrene		1.7	1.5 (.03)
Ethylbenzene		1.9	1.5 (.01)
o-Xylene		1.7	2.2 (.002)
m,p-Xylene		2.0	1.5 (.005)
Odorous Chemicals	83		
Tetrachloroethylene		1.1	1.2 (.02)
Benzene		1.3	1.5 (.003)
Ethylbenzene		1.2	1.6 (.0001)
o-Xylene		1.2	1.8 (.0000)
m,p-Xylene		1.2	1.5 (.0001)
Degreasing Compounds	19		
None			
Dust	63		
m,p-Xylene		1.1	1.2 (.002)
Tobacco Smoke			
(non-smokers only)	99		
None			
Cleaning Solutions	94		
None			
Toxic Chemicals	27		
None			

Table 19. Chemicals with significantly ($p < .05$) higher concentrations in air and breath of persons recently exposed to potential sources compared to persons not exposed to any source.

Potential Source	No. of Persons Exposed	Ratio of Median Concentrations: Exposed vs. Unexposed Groups: Breath	Ratio of Median Concentrations: Exposed vs. Unexposed Groups: Air
Paint	28		
Benzene		2.3 (.0002)[a]	1.3 (.03)
Tetrachloroethylene		2.0 (.0000)	2.7 (.02)
Styrene		2.8 (.0004)	1.8 (.0005)
Ethylbenzene		1.9 (.0004)	2.1 (.0001)
o-Xylene		1.4 (.009)	2.5 (.0003)
m,p-Xylene		1.7 (.002)	2.5 (.0000)
Chemical Plant	21		
Styrene		1.9 (.02)	2.0 (.004)
Ethylbenzene		2.5 (.0008)	1.8 (.0006)
o-Xylene		1.4 (.05)	2.3 (.0003)
m,p-Xylene		1.9 (.004)	1.9 (.0006)
Plastics Manufacturing	11		
Styrene		2.0 (.01)	2.6 (.02)
Ethylbenzene		2.8 (.003)	1.8 (.03)
o-Xylene		3.4 (.0006)	2.3 (.02)
m,p-Xylene		2.5 (.001)	2.1 (.02)
Dry Cleaning	37		
Tetrachloroethylene		2.3 (.0000)	2.2 (.003)
Benzene		2.2 (.02)	1.7 (.03)
Petroleum Plant	19		
None			
Service Station	67		
Benzene		2.2 (.0000)	1.3 (.02)
Printing	9		
Ethylbenzene		1.8 (.02)	1.6 (.03)
o-Xylene		1.3 (.03)	2.2 (.02)
Metal Working	17		
Tetrachloroethylene		1.4 (.01)	1.8 (.03)
Ethylbenzene		1.8 (.05)	3.7 (.0000)
o-Xylene		1.8 (.05)	4.4 (.0000)
Science Laboratory	14		
Ethylbenzene		1.7 (.03)	2.2 (.002)
o-Xylene		1.4 (.05)	2.7 (.001)
Furniture Refinishing	7		
Ethylbenzene		2.8 (.03)	2.2 (.02)
o-Xylene		2.5 (.04)	2.4 (.006)
Hospital	13		
None			

[a] Probability of no difference between exposed and unexposed groups —Wilcoxon Rank-Sum Test

monitors. Breath levels in smokers were about twice those of non-smokers, while personal exposures were about 50% higher. Similarly, air levels of benzene in homes containing smokers were about 30–50%

Table 20. Questionnaire variables related to concentrations of volatile organics in personal air, indoor air, and exhaled breath of 350 residents of New Jersey: Significant relationships on both analysis of variance and stepwise regression analyses.

Chemical	Variable ($p < .01$)
Benzene	Smoking Smoker in home Employment
Styrene	Smoking Smoker in home Employment Chemical plant Paint Degreasing compounds Scale model building Race (Black/Hispanic)
Xylenes and Ethylbenzene	Smoking Smoker in home Employment Paint Chemical plant Garage Solvents Odorous chemicals Metal working Degreasing compounds
p-Dichlorobenzene	Central air conditioning Hospital work
Chloroform	None
Carbon Tetrachloride	Employment Outdoor concentration
1,1,1-Trichloroethane	Employment
Trichloroethylene	Scientific lab Outdoor concentration
Tetrachloroethylene	Employment Dry cleaners Pesticide Use Age (Adults)

greater than air levels in non-smokers' homes. Since about 60% of U.S. children live in homes with smokers, their increased exposure to a known leukemogen may be a matter of public health concern. (Note that this speculation has recently been supported by a study [Sandler, 1985] showing greater risks of leukemia mortality among persons exposed to parental smoking.)

Caution in interpreting these results is indicated because of the small numbers of persons in some of the exposed groups. Nonetheless, the number and strength of many of the associations leads us to the following conclusion: Breath levels and personal air exposures to certain toxic and carcinogenic chemicals are significantly elevated in persons exposed to common household sources (consumer products and building materials) and normal daily activities (visiting service stations and dry cleaners, engaging in hobbies, smoking, painting, refinishing, etc.).

Summary and Conclusions

The major finding of this study has been the observation that personal exposures are nearly always greater—often much greater—than outdoor concentrations of several toxic and carcinogenic compounds. We are led to the conclusion that indoor air in the home and at work far outweighs outdoor air as a route of exposure to these chemicals. A natural next step would be to investigate the sources of these exposures more systematically than was possible in the TEAM Study. Such studies have begun with chamber investigations of adhesives (Girman 1984) and paint, carpets, wallpaper, cleansers, and cigarettes (Wallace 1985). The relative contribution of building materials, furnishings, personal activities, and consumer products to personal exposures should be determined by intensive studies in a number of homes, office buildings, schools, and other structures where people spend much of their time.

A second major finding has been the great utility of breath sampling to estimate levels in the body due to normal daily exposure to toxic chemicals. Breath sampling is noninvasive and is much more sensitive and less costly and difficult than blood sampling. In this study, breath sampling alone was effective in distinguishing between populations exposed to specific sources and those not so exposed. The technique should be investigated for possible use in the following situations:

a. To estimate dosages of persons exposed to chemical spills or releases;

b. To survey healthy persons to establish normal baselines and ranges of biological variability;

c. To study diseased persons to establish possible early diagnostic procedures.

A third finding has been the demonstration of the utility of this personal monitoring approach not only in estimating the exposure of entire urban area populations, but also in gaining an understanding of the sources of exposure. The general methodology appears applicable to determining exposures to pesticides and metals provided adequate sampling and analysis protocols for individually cooked meals can be developed to determine exposure through food intake. With the development of better instruments, it should also be possible to carry out large-scale studies of exposure to inhalable particulates and NO_2 in the near future.

Acknowledgments—Local and state officials in New Jersey gave essential support to this study. Special efforts were made by Dr. John Sakowski and Mr. David Roach of the Bayonne Department of Health, Mr. John Surmay and Mr. Robert Travisano of the Elizabeth Health, Welfare and Housing Department, and Dr. Thomas Burke of the New Jersey Department of Environmental Protection. We thank Shirley Barbour for deciphering the author's hieroglyphics and creating readable typescripts throughout many revisions. One of us (LAW, presently a visiting scientist at Harvard University of Public Health) is grateful to Dr. John D. Spengler and his colleagues at the Harvard University School of Public Health for providing an atmosphere conducive to study. We are most indebted to the hundreds of citizens who conscientiously wore monitors, kept diaries, and answered questions about their activities.

Disclaimer

Opinions expressed are those of the authors and do not reflect official positions of the U.S. Environmental Protection Agency. Mention of products or brand names does not imply endorsement by the government.

References

De Bortoli, M., Knoppel, H., Pecchio, E., Peil, A., Rogora, L., Schauenberg, H., Schlitt, H. and Vissers, H. (1984) Integrating 'Real Life' Measurements of Organic Pollution in Indoor and Outdoor Air of Homes in Northern Italy, *Indoor Air*, **4**, 21–26. Swedish Council for Building Research. Stockholm, Sweden.

Evans, J. S., Cooper, D. W. and Kinney, P. (1984) On the Propagation of Error in Air Pollution Measurements, *Env. Mon. and Assess.* 4:139–153.

Girman, J. R., Hodgson, A. T. and Newton, A. S. (1984) Volatile organic emissions from adhesives with indoor applications, *Indoor Air* **4**, 271–276. Swedish Council for Building Research, Stockholm, Sweden.

Jarke, F. H. (1981) Organic contaminants in indoor air and their relation to outdoor contaminants. ASHRAE Rp. 183, IITRI, Chicago.

Lebret, E., Van de Wiel, H. J., Bos, H. P., Noij, D. and Boleij, J. S. M. (1984) Volatile Hydrocarbons in Dutch Homes, *Indoor Air* **4**, 169–174. Swedish Council for Building Research, Stockholm, Sweden.

Mølhave, L. and Moller, J. (1979) The Atmospheric Environment in Modern Danish Dwellings: Measurements in 39 Flats, *Indoor Climate* 171–186. Danish Building Research Institute, Copenhagen.

Pellizzari, E. D., Hartwell, T., Zelon, H., Williams, S., Breen, J., Cooper, S. and Wallace, L. (1983) Human Exposure to Vapor-Phase Halogenated Hydrocarbons: Fixed-Site vs. Personal Exposure, *Proceedings: National Symposium on Recent Advances in Pollutant Monitoring of Ambient Air and Stationary Sources*, Environmental Monitoring Systems Lab., Research Triangle Park, N. C., EPA-600/9-83-007, pp. 264–288.

Pellizzari, E., Hartwell, T., Sparacino, C., Sheldon, L., Whitmore, R., Leininger, C. and Zelon, H. (1984a), *Total Exposure Assessment Methodology (TEAM) Study: First Season, Northern New Jersey—Interim Report*, contract No. 68-02-3679, U.S. Environmental Protection Agency, Washington, D.C.

Pellizzari, E., Sheldon, L., Sparacino, C., Bursey, J., Wallace, L. and Bromberg, S. (1984b) Volatile Organic Levels in Indoor Air, *Indoor Air 4, Chemical Characterization and Personal Exposure*, pp. 303–308. Swedish Council for Building Research, Stockholm, Sweden.

Sandler, D. P., Everson, R. B., Wilcox, A. J., and Browder, J. P. (1985) Cancer risk in adulthood from early life exposure to parent's smoking, *Am. J. Public Health* May 1985.

Seifert, B. and Abraham, H. J. Indoor Air Concentrations of Benzene and Some Other Aromatic Hydrocarbons *Ecotoxicol. Environ. Safety*, **6**: 190–192 (1982).

Suggs, J. C. and Curran, T. C. (1983) An Empirical Bayes Method for Comparing Air Pollution Data to Air Quality Standards, *Atmos. Env.* **17**, 837–841.

Wallace, L. A., Zweidinger, R., Erickson, M., Cooper, S., Whitaker, D. and Pellizzari, E. D. (1982) Monitoring Individual Exposure: Measurements of Volatile Organic Compounds in Breathing-Zone Air, Drinking Water, and Exhaled Breath, *Environment International* **8**, 269–282.

Wallace, L. A., Pellizari, E., Hartwell, T., Rosenzweig, M., Erickson, M., Sparacino, C., and Zelon, H. (1984a) Personal Exposure to Volatile Organic Compounds: I. Direct Measurement in Breathing-Zone Air, Drinking Water, Food, and Exhaled Breath, *Environmental Research* **35**, 293–319.

Wallace, L., Bromberg, S., Pellizzari, E., Hartwell, T., Zelon, H. and Sheldon, L. (1984b) Plan and Preliminary Results of the U.S. Environmental Protection Agency's Indoor Air Monitoring Program, *Indoor Air* **1**, *Recent Advances in the Health Sciences and Technology*, 173–178. Swedish Council for Building Research, Stockholm, Sweden.

Wallace, L., Pellizzari, E., Hartwell, T., Sparacino, C., Sheldon, L. and Zelon, H. (1985) Personal Exposures, Indoor-Outdoor Relationships and Breath Levels of Toxic Air Pollutants Measured for 355 Persons in New Jersey, *Atmos. Env.* **19**, 1651–1661.

Wallace, L., Pellizzari, E., Leaderer, B., Hartwell, T., Perritt, K., Zelon, H. and Sheldon, L. (1985) Assessing Sources of Volatile Organic Compounds in Homes, Buildings, Building Materials, and Consumer Products, paper presented at Conference on Characteristics of Contaminant Emissions from Indoor Sources, May 13–15, 1985, Raleigh, N.C. *Atm. Environ.* (in press)

Zweidinger, R., Erickson, M., Cooper, S., Whittaker, D., Pellizzari, E. and Wallace, L. (1982) *Direct Measurement of Volatile Organic Compounds in Breathing-Zone Air, Drinking Water, Breath, Blood, and Urine*, U.S. Environmental Protection Agency, Washington, D.C., NTIS No. PB-82-186-545.

Appendix

A Method for Correcting Observed Frequency Distributions for Multiplicative Measurement Errors

Measurement errors increase the apparent variance of a distribution. If the true distribution is normal, and the measurement errors are additive and are also normally distributed, the observed variance is the sum of the true variance and the measurement error variance:

$$\sigma^2_{\mathrm{OBS}} = \sigma^2_{\mathrm{TRUE}} + \sigma^2_{\mathrm{ERROR}} \tag{1}$$

It has been shown (Suggs, 1983; Evans, 1984) that a similar relationship holds for the variances of the logarithms of log-normal distributions with *multiplicative* measurement errors distributed log-normally. (An error in calculating flow rate is an example of a multiplicative error, since the amount collected is the product of the volume and the concentration.)

In the present case, for all compounds except possibly benzene, which had high background levels (an additive error), the errors as determined from duplicate measurements appear to be multiplicative, and are distributed nearly log-normally between the 10th and 90th percentiles. These errors are defined as the ratio of one member (say x_1) of a duplicate pair (x_1, x_2) to their geometric mean $\sqrt{x_1 x_2}$:

$$\text{Error} = {}^{x_1}\!\big/\!_{\sqrt{x_1\, x_2}} = \sqrt{\frac{x_1}{x^2}} \tag{2}$$

Table A-1. Correction factors due to measurement errors—Fall 1981.

		Personal Air		Outdoor Air	
	Breath[a]	Night[a]	Day[a]	Night[b]	Day[b]
Chloroform	0.70	0.96	0.92	—[c]	0.87
1,1,1-Trichloroethane	0.60	0.93	0.81	0.82	0.91
Benzene	—	0.75	0.62	—	0.66
Carbon tetrachloride	0.97	0.92	0.63	0.95	—
Trichloroethylene	0.84	0.96	0.84	0.98	0.86
Tetrachloroethylene	0.85	0.93	0.96	0.92	0.97
Styrene	0.90	0.89	0.68	—	0.77
m,p-Dichlorobenzene	0.96	0.96	0.92	0.98	0.97
Ethylbenzene	—	0.89	0.92	0.98	0.92
o-Xylene	0.55	0.74	0.92	0.95	0.92
m,p-Xylene	0.50	0.81	0.84	0.93	0.75

[a] "True" 90th percentile value/Observed 90th percentile
[b] "True" 75th percentile value/Observed 75th percentile
[c] "True" value cannot be calculated—measurement errors too large

For each chemical and each environmental medium, the variance σ^2_{ERROR} associated with the duplicate measurements can be calculated and subtracted from the observed variance to give the true variance. (For the error as defined above, $\sigma^2_{ERROR} = 2S^2$ where S^2 is the variance of the duplicates). Then the variance σ^2_{TRUE} of the true log-normal distribution can be calculated from equation (1). This distribution passes through the observed median but is "rotated clockwise" (as seen when plotted on log-normal probability paper) such that the slope (the geometric standard deviation) is reduced. Thus all percentiles higher than the median are likely to be overestimates of the true values. The "correction factors" by which these overestimates should be multiplied to obtain the true values are tabulated in Table A-1. Although in most cases these correction factors are between 0.8 and 1, indicating overestimates at the 75th or 90th percentile of less than 25%, there are some cases (6 out of 55) in which the measurement error was too large to allow any estimate of the frequency distribution other than the observed geometric mean.

Since both the observed field samples and the errors associated with the duplicates depart from log-normality at the tails of the distribution, the validity of the above method should be tested by simulation methods. That is, the postulated "true" distribution can be convoluted with the observed distribution of errors by sampling with replacement from the set of duplicates to obtain a resultant distribution that should agree with the observed distribution. One such set of simulations has been run on one chemical in this study with satisfactory results.