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Major Sources of Benzene Exposure

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Data from EPA's TEAM Study allow us to identify the major sources of exposure to benzene for much of the U.S. population. These sources turn out to be quite different from what had previously been considered the important sources. The most important source of exposure for 50 million smokers is the mainstream smoke from their cigarettes, which accounts for about half of the total population burden of exposure to benzene. Another 20% of nationwide exposure is contributed by various personal activities, such as driving and using attached garages. (Emissions from consumer products, building materials, paints, and adhesives may also be important, although data are largely lacking.) The traditional sources of atmospheric emissions (auto exhaust and industrial emissions) account for only about 20% of total exposure. Environmental tobacco smoke is an important source, accounting for about 5% of total nationwide exposure. A number of sources sometimes considered important, such as petroleum refining operations, petrochemical manufacturing, oil storage tanks, urban-industrial areas, service stations, certain foods, groundwater contamination, and underground gasoline leaks, appear to be unimportant on a nationwide basis.

Introduction

Benzene is recognized as a human leukemogen (1). It is regulated in the workplace in most countries. In the U.S., it is one of only a few chemicals that are regulated under Section 112 of the Clean Air Act as a Hazardous Air Pollutant.

Nonetheless, until recently the main sources of exposure to the general population have remained obscure. Chemical plants, petroleum refining operations, oil storage tanks, major urban-industrial areas, and gasoline service stations have been suspected major sources of exposure. Food, water supplies, and landfills have also been mentioned as possible major sources.

Now, a large study of human exposure to benzene (EPA's TEAM Study) has been completed, with the surprising result that the main sources of human exposure are associated with personal activities, not with the so-called "major point sources" mentioned above. This paper will attempt to create a nationwide exposure budget, identifying the main sources of benzene exposure for the U.S. population, by drawing on the TEAM Study findings.

Review of TEAM Study Findings

The TEAM Study is described in detail in a four-volume EPA publication (2-5) and in several journal articles (6-10). Following is a brief summary of the study and its findings with respect to benzene. The study measured

24-hr personal exposures in air and drinking water to 20 to 25 target volatile organic compounds for a probabilistically selected group of subjects in Elizabeth-Bayonne, NJ; Los Angeles, CA; Antioch-Pittsburg, CA; Greensboro, NC; and Devils Lake, ND. (In 1987, another city, Baltimore, MD, was sampled. Preliminary results are included in this paper.)

Subjects were selected according to a three-stage stratified survey design. In each city, a target population was selected using census information. Blocks of homes were stratified according to socioeconomic factors and proximity to potential industrial and mobile sources. In the second stage, a large number of homes (about 5500 in New Jersey and 2000 in California) were visited, and trained interviewers collected information on age, sex, occupation, smoking status, and other factors for each person in the household. This information was used to determine the prevalence of potential exposure factors in the target population and to allow selection of those persons more likely to be exposed. Since the probability of selection was known for the entire target population, the measured concentrations, when weighted by the inverse of the probability of selection, apply to the entire population.

A total of about 700 subjects representing more than 800,000 residents of the various cities collected two 12-hr air samples and 1 to 2 tap water samples during a 24-hr period. Concurrent outdoor air samples were collected from the backyards of a subset (about 200) of the subjects' homes. At the end of 24 hr, each subject provided a sample of exhaled breath to a van-mounted spirometer. Both air and breath samples were collected on Tenax cartridges and analyzed by GC/MS.

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Results

Population-weighted personal exposures to benzene (Table 1) exceed the outdoor air concentrations (Table 2) in every city. The overall mean personal exposure is about $15 \mu\text{g}/\text{m}^3$, compared to an overall mean outdoor concentration of only $6 \mu\text{g}/\text{m}^3$. When maximum exposures are compared to maximum outdoor concentrations, the difference is even more striking: $500 \mu\text{g}/\text{m}^3$ for the personal exposure maximum compared to $90 \mu\text{g}/\text{m}^3$ for the outdoor maximum. These results imply that personal activities or sources in the home far outweigh the contribution of outdoor air to human exposure to benzene. Since most of the traditional sources exert their effect through outdoor air, we must find new sources to explain the increased personal exposures observed.

Smoking

Examination of exhaled breath concentrations against personal activities identified one activity as paramount: smoking tobacco. Smokers typically have breath concentrations of benzene around $14 \mu\text{g}/\text{m}^3$, while nonsmokers range around $2 \mu\text{g}/\text{m}^3$ (Fig. 1). From measurements of

benzene content in mainstream smoke [$57 \mu\text{g}$ benzene in the average sales-weighted tar and nicotine cigarette (11)] we can calculate that the average smoker (32 cigarettes/day) takes in about 1.8 mg of benzene per day. This is nearly 10 times the average daily intake of nonsmokers (12,13).

Passive Smoking

Passive smoking was also an important source of benzene exposure. Median levels of benzene in 200 homes without smokers were $7 \mu\text{g}/\text{m}^3$; in 300 homes with one or more smokers, median levels were $10.5 \mu\text{g}/\text{m}^3$. This represents a 50% increase in benzene exposures of spouses and children in homes of smokers. A recent study of 500 homes in West Germany (14) replicated this result, with median values of $6.5 \mu\text{g}/\text{m}^3$ in nonsmoking homes and $11 \mu\text{g}/\text{m}^3$ in smoking homes (Fig. 2). Work exposures were also increased; nonsmokers not exposed at home who stated they were exposed to tobacco smoke more than 50% of the time they were at work showed significantly higher breath concentrations (Mann-Whitney nonparametric test) than those exposed to tobacco smoke at work less than 50% of the time.

Table 1. Population-weighted personal exposures to benzene in five U.S. cities.

TEAM site ^a	n	Arithmetic	Arithmetic	Geometric	Geometric	Median	Percentile			Maximum
		mean	SE	mean	SD		75	90	95	
NJ night	347	29.7	5.22	12.5	2.6	15	32	64	73	610
NJ day	340	26.2	1.68	11.2	2.6	17	32	65	81	270
NC night	24	10.2	1.87	2.23	1.9	12	16	30	41	43
NC day	24	7.93	1.55	1.55	2.1	7.6	13	20	32	36
LA1 night	112	16.5	1.30	13.6	2.6	15	21	30	34	43
LA1 day	112	19.1	1.53	15.1	2.1	15	23	35	61	86
LA2 night	50	7.78	1.31	4.69	2.9	4.4	9	25	29	35
LA2 day	50	10.5	1.62	6.88	2.4	7.2	12	25	34	54
AP night	69	6.47	1.13	4.63	2.5	4.4	7.5	16	18	32
AP day	67	8.47	0.87	6.83	2.1	6.3	11	17	21	25
MD night	70	20.7	1.42	12.3	2.5	13	26	42	66	104
MD day	70	16.4	1.24	8.38	2.3	11	22	32	45	129

^aNJ: Bayonne-Elizabeth, NJ; fall 1981; population, 130,000. NC: Greensboro, NC; May 1982; population, 130,000. LA1: Los Angeles, CA; February 1984; population, 360,000. LA2: Los Angeles, CA; May 1984; population, 330,000. A P Antioch-Pittsburg, CA; June 1984; population, 90,000. MD: Baltimore, MD; March 1987; population, Not yet weighted.

Table 2. Outdoor concentrations of benzene in three U.S. cities.

TEAM site ^a	n	Arithmetic	Arithmetic	Geometric	Geometric	Median	Percentile			Maximum
		mean	SE	mean	SD		75	90	95	
NJ night	84	8.6	1.04	4.1	2.0	6.7	11	15	24	91
NJ day	88	9.5	0.95	3.8	2.1	7.8	16	20	27	44
LA1 night	24	18.9	1.86	16.5	1.4	19	25	32	33	33
LA1 day	24	13.2	1.34	11.2	1.3	14	18	21	22	35
LA2 night	23	3.1	0.45	2.6	2.0	2.5	4.4	5.8	6.7	8.6
LA2 day	24	4.2	0.82	3.2	2.2	3.1	4.8	8.7	12	15
AP night	10	1.8	0.32	1.6	1.7	1.7	1.9	3.2	—	3.6
AP day	10	2.0	0.63	1.5	2.5	1.3	1.6	6.3	—	6.3

^aNJ: Bayonne-Elizabeth, NJ; fall 1981; population, 130,000. LA1: Los Angeles, CA; February 1984; population, 360,000. LA2: Los Angeles, CA; May 1984; population, 330,000. AP: Antioch-Pittsburg, CA; June 1984; population, 90,000.

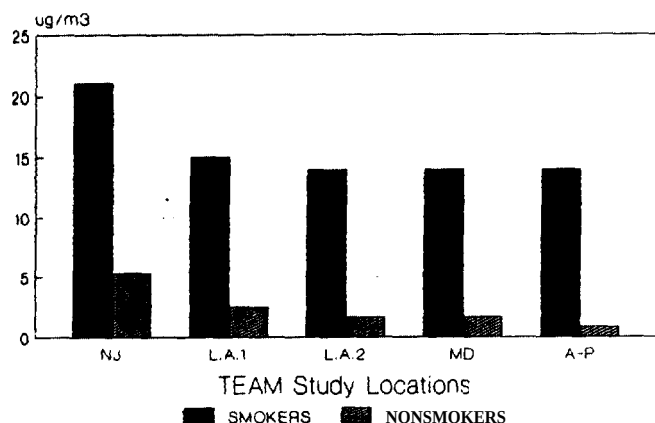


FIGURE 1. Geometric mean benzene concentrations in the breath of smokers exceeded breath concentrations of nonsmokers at all TEAM Study sites: Bayonne-Elizabeth, NJ (smokers, $n = 150$; nonsmokers, $n = 188$); Los Angeles, CA in February 1984 (smokers, $n = 29$; nonsmokers, $n = 85$); Los Angeles, CA, in May 1984 (smokers, $n = 11$; nonsmokers, $n = 40$); Baltimore, MD (smokers, $n = 30$; nonsmokers, $n = 45$); and Antioch-Pittsburg, CA (smokers, $n = 19$; nonsmokers, $n = 49$).

Auto-Related Activities

Stepwise regressions of breath concentrations and personal air exposures identified several auto-related activities as sources of benzene exposure. Exposure to auto exhaust, time spent in an auto, or pumping gas all resulted in increased personal exposure to benzene. Since benzene forms 1 to 2% of most gasoline blends, it is given

off as a vapor by hot engines and by fuel tanks and is also a constituent of auto exhaust. Therefore we may identify three potential major sources of auto-related exposure to benzene: auto travel, filling gas tanks, and parking hot cars in attached garages.

Auto Travel. All four studies in New Jersey and California showed increases in the amount of benzene exposure in proportion to time spent in the car. Benzene concentrations in the car could not be reliably determined because exposures were averaged over 12 hr; however, concentrations of 3 to 4 times normal exposures (i.e., 40 to 60 $\mu\text{g}/\text{m}^3$) were calculated.

Pumping Gasoline. Several stepwise regressions identified pumping gasoline as a significant source of benzene exposure. Concentrations were estimated to be on the order of 1 ppm (3000 $\mu\text{g}/\text{m}^3$). Since that calculation, a study found about 1 ppm exposure at breathing level while pumping gasoline (15).

Attached Garages. Gasoline vapors from attached garages have been observed in homes in several studies (16,17). No quantitative estimates of benzene concentrations due to these emissions have yet been made.

Occupational Exposures

According to the National Institute of Occupational Safety and Health, about 240,000 workers are exposed to benzene (18). The occupational limit is presently 1 ppm. Thus, a realistic estimate of exposure might be 100 ppb/worker, or 2.4×10^4 person-ppm.

Consumer Products

About 400 of 5000 materials and products tested by the National Air and Space Administration emitted benzene vapors, in amounts ranging from 0.01 $\mu\text{g}/\text{g}$ up to 140 $\mu\text{g}/\text{g}$ (19). Paints, adhesives, marking pens, rubber products, tapes, and other common categories of materials emitted benzene. Other studies have also shown that latex paints emit benzene (20,21). Insufficient data exist to estimate exposures from any one category, but it seems likely that a substantial portion of the indoor excess of benzene (once contributions from tobacco smoke and auto emissions in attached garages are subtracted) can be attributed to the category of emissions from materials, surface coatings, or Consumer products.

Other Sources of Benzene Exposure

We have mentioned other possible sources of benzene exposure. Several of these sources have been investigated in the TEAM Study and have been found to be relatively unimportant. For example, persons living close to the heavy petrochemical and refining operations at New Jersey and Los Angeles had no greater exposures than those living farther away. Although outdoor levels were higher in New Jersey and Los Angeles than in Greensboro, NC, this seems to be due to a higher intensity of automobile traffic, since the petrochemical manufacturing areas of Antioch-Pittsburg, CA, also had low outdoor levels of benzene.

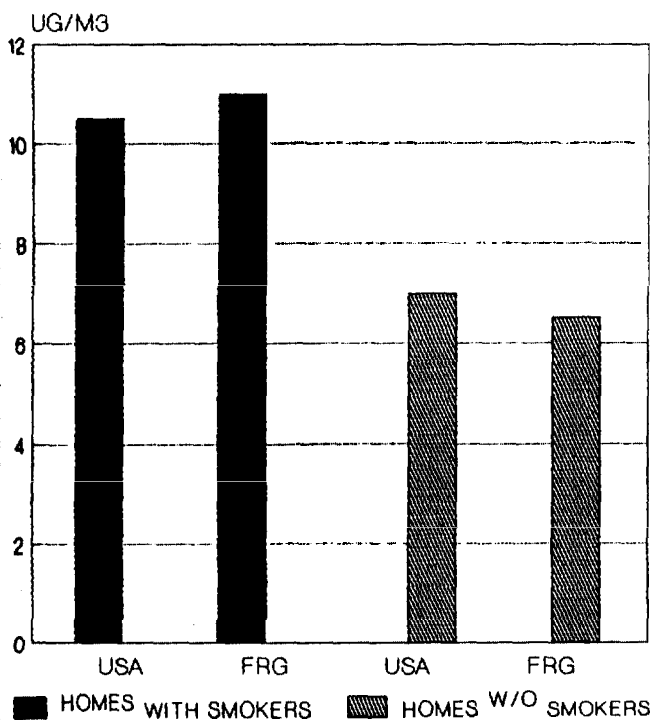


FIGURE 2. Benzene concentrations in the homes of smokers were about 50% higher than in the homes of nonsmokers, both in the U.S. and in West Germany. U.S. values are geometric means based on 528 homes (343 with smokers, 185 without) in New Jersey and California; West German values are medians based on 488 homes.

Since the TEAM Study measured breath levels of benzene, exposure to any important unmeasured sources (such as food and beverages) should have resulted in increased breath concentrations. In fact, this was the case for cigarette smokers, whose breath levels exceeded their apparent exposures through air as measured by the personal monitors. However, no other noticeable discrepancies between apparent exposure and measured breath concentrations have been found. Therefore, exposure through food, beverages, and drinking water is believed to be unimportant for most persons.

As the TEAM subjects were drawn from areas where little use of wood stoves or kerosene heaters was made, it remains possible that these combustion sources will prove to be important sources of exposure to benzene.

Calculation of Exposure Budget

Having identified the main sources of benzene exposure and the concentrations associated with each, it remains to estimate the number of people exposed to each source.

About 50 million persons smoke cigarettes in the U.S.⁽²²⁾; Perhaps 100 million persons pump gasoline 70 min per year, and the entire population (240 million) is exposed to indoor air, outdoor air, and air in autos. We can assume that two-thirds are passive smokers at home and at work (23) with workplace exposures to benzene from tobacco smoke equaling home exposures ($3 \mu\text{g}/\text{m}^3$).

These assumptions lead to the conclusion that more than half of the entire nationwide exposure to benzene results from smoking tobacco or being exposed to tobacco smoke. The remainder is split nearly evenly between personal and outdoor sources. The main personal sources are driving or riding in automobiles and using products that emit benzene. The main outdoor source is likely to be automobile exhaust, based on the lack of evidence for increased exposure in areas near petroleum refining and petrochemical operations.

Based on the TEAM Study findings, it appears that the following are not important sources of exposure to ben-

zene on a nationwide basis: chemical plants, petroleum refining operations, oil storage tanks, drinking water, food, and beverages.

Risk

An adequate calculation of benzene-related risk may be impossible with present knowledge. However, if risk is proportional to exposure, then the relative risks associated with major sources of exposure will be in the same proportion as the exposures themselves. The excess risk of leukemia associated with 70 years of exposure to $1 \mu\text{m}/\text{m}^3$ benzene has been estimated by EPA to be 8×10^{-6} (24) and by a group at Harvard University to be 4×10^{-6} (25). Using the EPA potency estimate and the measured TEAM Study mean exposure of $15 \mu\text{g}/\text{m}^3$ extrapolated to the U.S. population, one can calculate roughly 400 benzene-related leukemia cases/year due to the major indoor and outdoor sources: auto exhaust, driving, passive smoking and consumer product emissions (Table 3). An additional 500 cases can be calculated to occur among cigarette smokers inhaling benzene in mainstream smoke. Cigarette smokers have in fact been observed to be at about 50% higher risk of leukemia mortality (27), which would result in about 1000 excess cases of leukemia in smokers annually. Thus, benzene in cigarette smoke may account for a significant portion of the observed excess leukemia mortality among smokers.

Conclusion

On a nationwide basis, the most important single source of benzene exposure is active smoking of tobacco. Smoking accounts for about half of the total population exposure to benzene. Personal exposures due to riding in automobiles, passive smoking, and exposure to consumer products account for roughly one-quarter of the total exposure, with outdoor concentrations of benzene, due mainly to vehicle exhaust, accounting for the remaining

Table 3. Benzene exposures and risks.

Activity	Intake, $\mu\text{g}/\text{day}$	Population at risk	Cases/year ^a
Smoking	1800 ^b	53×10^6	500
Passive smoking	50 ^c	200×10^6	50
Outdoor levels	120 ^c	240×10^6	150
Driving/riding auto	40 ^d	200×10^6	40
Filling gas tank	10 ^e	100×10^6	5
Occupational	10000 ^b	240×10^{3i}	10
Other personal	150	240×10^6	200 ^j
Total			960

^aUsing a unit risk of $8 \times 10^{-6} (\mu\text{g}/\text{m}^3)^{-1}$.

^b57 $\mu\text{g}/\text{cigarette}$ (Higgins) $\times$ 32 cigarettes/day.

^c $3 \mu\text{g}/\text{m}^3 \times 17 \text{ hr}/\text{day indoors} \times 1 \text{ m}^3/\text{hr respiration}$.

^dApproximately 80% of persons exposed to environmental tobacco smoke.

^eTEAM outdoor average in eight locations = $6 \mu\text{g}/\text{m}^3 \times 20 \text{ m}^3/\text{day}$.

^fFew data available, assumed $40 \mu\text{g}/\text{m}^3$ in vehicle $\times$ 1 hr/day.

^g1 ppni $\times$ 70 min/year.

^hAssumed $1000 \mu\text{g}/\text{m}^3 \times 10 \text{ m}^3/8 \text{ hr workday}$.

ⁱNIOSH estimate of number of workers exposed to benzene.

^jObtained by subtraction from published estimate of 460 nonsmoking cases/year (26). Includes emissions from surface coatings, consumer products, evaporative emissions from autos in attached garages, etc.

portion. Occupational exposures, pumping gasoline, living near chemical plants or petroleum refining operations, food, water, and beverages appear to account for no more than a few percent of total nationwide exposure to benzene.

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