Geographical Distribution of Benzene in Air in Northwestern Italy and Personal Exposure

Giorgio Gilli, Enzo Scursatone, and Roberto Bono

Department of Hygiene and Community Medicine, University of Turin, Turin, Italy

Benzene is a solvent strictly related to some industrial activities and to automotive emissions. After the reduction in lead content of fuel gasoline, and the consequent decrease in octane number, an increase in benzene and other aromatic hydrocarbons in gasoline occurred. Therefore, an increase in the concentration of these chemicals in the air as primary pollutants and as precursors of photochemical smog could occur in the future. The objectives of this study were to describe the benzene air pollution at three sites in northwestern Italy throughout 1991 and 1994; to examine the relationship between benzene air pollution in indoor, outdoor, and personal air as measured by a group of nonsmoking university students; and to determine the influence of environmental tobacco smoke on the level of benzene exposure in indoor air environments. The results indicate a direct relationship between population density and levels of contamination; an indoor/outdoor ratio of benzene air pollution higher than 1 during day and night; a similar level of personal and indoor air contamination; and a direct relationship between levels of personal exposure to benzene and intensity of exposure to tobacco smoke. Human exposure to airborne benzene has been found to depend principally on indoor air contamination not only in the home but also in many other confined environments. — Environ Health Perspect 104(Suppl 6):1137-1140 (1996)

Key words: benzene, air pollution, personal exposure, gasoline, tobacco smoke, indoor air pollution

Introduction

Benzene is a solvent strictly related to some industrial activities and to automotive emissions (1). About 2 million U.S. workers are exposed to benzene each year (2,3) and an increase in benzene and other aromatic hydrocarbons in gasoline occurred after the reduction of lead content and the consequent decrease in octane number (4). Therefore, the concentration of these chemicals in air as primary pollutants and as precursors of photochemical smog could increase in the future (5,6).

The main sources of human exposure to benzene are associated with personal or professional activities (7–11), active or passive inhalation of cigarette smoke, and the use of certain consumer products in indoor environments (12).

For example, smoking can cause a 50% increase in benzene exposure of cohabitant nonsmokers; 400 of 5000 materials and products tested by the U.S. National Air and Space Administration emit benzene vapors in amounts ranging from 0.01 μg/g to 140 μg/g (13). The presence of benzene in the air is of extreme importance for public health because air is its primary route of entry into the body and because benzene has been proved to be a human leukemogen (14–16).

The purposes of this work are summarized as follows:

First, we described the benzene outdoor air pollution in three fixed sites in the Piedmont (northwestern) region of Italy, in 1991, prior to the anticipated atmospheric changes due to the introduction of new gasoline and again during 1994; the sites were chosen to reflect different degrees of urbanization. Such a comparison can show changes in benzene air pollution caused by the gradual reformulation of fuel from leaded to unleaded commercialized in Italy during those years. Furthermore, we compared the benzene data and some meteorological parameters to study the latter as potential favoring factors. Finally, a comparison between benzene and other airborne chemicals was investigated to determine whether they have similar atmospheric behavior.

Second, we observed the relationships between benzene pollution in indoor, outdoor, and personal air of volunteers living in Turin, the capital city of the Piedmont region.

Third, we evaluated the influence of environmental tobacco smoke on the level of exposure to benzene in indoor environments.

Experimental Methods

Outdoor Air, Fixed Sites

During all of 1991, the measurement of benzene air pollution was carried out in the center of an urban site (Turin), in a suburban site (Cuorgnè), and in a rural site (Mount Banchetta, 2000 m above sea level). Details concerning the three sites have been published (6). Urban sampling was done in two 1-year periods from January to December 1991 and 1994, testing for 10 consecutive days per month, 24 hr each day in an "urban canyon" in the center of Turin. Due to logistic difficulties, sampling at Cuorgnè in 1991 was limited to the months of March, May, July, October, and December; samples were taken in those months during the same days and hours as at the urban site. At the rural site, samples were collected in January and in July 1991 for a consecutive week in each month (7 samples of 24 hr each).

A private company, owner of the meteorological station in Turin city, measured four meteorological parameters: relative humidity, wind speed, solar irradiation, and ground temperature, and two airborne chemicals: nitrogen monoxide (NO) and carbon monoxide (CO). These measurements were obtained every hour during the same days as the benzene measurements and were mediated for the same 24 hr as benzene. Air measurements in urban, suburban, and rural sites were carried out using a sampling line (air flow = 1 liter/min) consisting of a membrane pump, a gas meter, and a granular activated carbon (GAC) cartridge.

Indoor, Outdoor, and Personal Air

Indoor, outdoor, and personal air were measured using a passive sampler equipped with a sorbent capsule containing GAC. All measurements were made simultaneously.
(from 8 AM to 8 PM and from midnight to 8 AM each day) by 10 nonsmoking university students who collected the samples for 10 consecutive days (December 1991). Indoor air was collected inside an apartment with a habitable density ranging from 25 to 35 m² per person. Outdoor air was collected outside the same apartment (balcony or window), and personal air was collected during normal daily activities by attaching the personal sampler to the subject’s collar. Each student recorded all the confined environments visited during sampling.

An additional 88 young students (14 years of age) were sampled for benzene using the same passive samplers. These measurements lasted 24 hr and concerned only personal air in relation to the intensity of smoking habits of the students and their parents and cohabitants; these data were obtained by means of a questionnaire validated by comparison to urinary cotinine. Methodology and further details of this epidemiological sample have been published elsewhere (17,18).

**Analytical Techniques**

Passive and active samples were analyzed with a gas chromatograph (GC) equipped with an F.I.D. Detector and a capillary column DB-624 30 m × 0.318 mm I.D., film 1.8 μm. A titration curve, suitable for the range of concentrations expected, was prepared by adding known concentrations of benzene. The GC oven temperature program: 45°C × 4 min – from 45 to 145°C, 10°C/min – 145°C × 2 min.

Precision of the analysis was determined by analyzing an environmental sample 20 times; the sample was collected by the active method. The mean results referred to toluene were: 14.4 μg/m³, SD = 0.6 μg/m, and coefficient of variation = 4.9%. Recovery (95%) has been calculated using techniques previously described (6,19,20).

The passive method was compared to the active one used in the three fixed sites. Twenty parallel measurements (active vs passive) were done and results show good overlapping (r = 0.97089).

**Statistical Analysis**

Statistical analyses were carried out using nonparametric tests performed by Statistical Analysis System Packages for the IBM PC (21).

**Results**

Findings obtained for the three sites are shown in Figure 1. Each monthly bar represents the mean of 10 daily results. The direct relationship between level of urbanization and benzene pollution is evident, as is the higher level of contamination in all the sites during the cold months. A comparison between 1991 and 1994 in Turin is reported in Figure 2. The two annual averages, 6.85 and 6.62 ppb, respectively, do not indicate differences in benzene pollution between the 2 years; furthermore, the interseasonal difference in 1991 is high and benzene pollution in winter 1994 is lower than in winter 1991 (Table 1).

![Figure 1. Benzene in air contamination in northwestern Italy (1991).](image1)

![Figure 2. Benzene air contamination in Turin (urban site).](image2)

| Year | Mean  |
|------|-------|
| 1991 | 6.85  |
| 1994 | 6.62  |

**Table 1.** Seasonal comparison between benzene pollution (ppb) in 1991 and 1994 (Turin).

![Image](image1)

**Table 2.** Correlations to some meteochemical parameters-Turin.

| Meteochemical parameters | Benzene, ppb |
|--------------------------|--------------|
| NO                       | +0.558       |
| CO                       | +0.511       |
| Relative humidity        | -0.299       |
| Wind speed               | -0.466       |
| Solar irradiation        | -0.329       |
| Ground temperature       | -0.253       |
and solar irradiation; no significant relationships are found for relative humidity and ground temperature. Instead, correlations among benzene, CO, and NO are significantly positive.

Table 3 shows the mean of indoor/outdoor ratios collected separately during day and night by 10 nonsmoking university students who collected the samples for 10 consecutive days in December 1991. In all cases the ratio is higher than 1 (higher level of contamination in the indoor environments), especially during the day, when personal activities are probably more numerous.

Table 4 shows mean and standard deviation (ppb) for indoor, outdoor, and personal benzene contamination during day and night. Comparative analysis of day and night means does not offer statistical differences between indoor or outdoor air. During the day, personal benzene contamination is higher than indoor and outdoor air. All types of air pollution measured by passive samplers are much higher than in outdoor air measured in fixed sites (Table 1).

A detailed analysis of single levels of contamination measured by personal samplers and the corresponding environments visited by the university students during sampling shows some cases of interest:

- 204.9 ppb of benzene in indoor air during the day, when the student was doing housework
- 254.7 ppb of benzene in personal air at a shoe-repair shop
- 70.8 ppb of benzene in personal air at a hairdresser and a supermarket

Results relative to the 88 young students are reported in Table 5. Exposure to benzene was proportional to the intensity of exposure to tobacco smoke. These levels of exposure to tobacco smoke were established by questionnaires validated by urinary cotinine (17,18).

### Discussion

Results obtained at the three fixed sites in the Piedmont region highlight a direct relationship between population density and level of contamination, confirming the effects of automotive traffic and industrial activities on benzene outdoor pollution. In each site, the solvent shows a higher level of concentration during winter, proving its primary pollutant nature. This fact depends on the minor mixing ratio during the coldest months which allows an accumulation of pollutants in the lowest layers of the atmosphere. Furthermore, the negative correlation with wind speed and solar irradiation confirms that the atmospheric behavior of benzene also depends on the meteorological events, many of which have not been considered in the present work (for example, thermal inversion, cloudiness, etc.).

An interesting finding shows significant correlations between benzene, carbon monoxide, and nitrogen monoxide, confirming a similar origin and atmospheric fate.

Table 6 compares benzene air pollution measured in this study and that reported in other urban and suburban sites; these results demonstrate similar ranges of contamination.

Even if a higher amount of aromatics were present in gasoline during the years of the two surveys in Turin, data reported in Table 1 show that in 4 years no significant differences in benzene air pollution have been recognized. This evidence can be explained by considering some restrictions in automobile traffic in the city center in recent years: a) the enforcement of parking meters, b) the enforcement of some limited traffic areas, c) the enforcement of no-parking areas, d) changes of one-way streets.

These innovations may induce some optimism about the usefulness of such actions to lessen human exposure to outdoor air pollution in the cities.

Indoor/outdoor ratios above 1 confirm a higher level of pollution in indoor environments during day and night, whereas when considering the results from indoor, outdoor, and personal air, no differences may be demonstrated between day and night contamination. However, considering the diurnal data, personal exposure measurements seem to demonstrate a higher level of contamination.

In general, the behavior of benzene is not very clear. Probably it is involved in several, and extremely unsteady, indoor and outdoor environmental events, some of which have been verified in the present work: a) The levels of emission in indoor environments can be very high considering the episodes mentioned above. Considering the use of or exposure to products containing benzene, housework and visits to certain kinds of shops can in fact represent behaviors that lead to exposure to this solvent. b) The seasonal trend measured in fixed sites is less remarkable in comparison to CO and NO, well-known markers of automobile traffic. c) Standard deviation of indoor, outdoor, and personal air measurements are, in general, very large, confirming the variability of benzene air pollution.

Considering data published elsewhere (22), toluene and xylene air pollution seem to demonstrate more linear environmental behavior and considering the similar origin, chemical characteristics, and environmental fate, could be used as indirect markers of benzene pollution.

Benzene is not a chemical specific for tobacco smoke pollution, and its presence in air can depend on many other indoor sources. Nevertheless, data obtained from the 88 young students show that the three degrees of exposure to tobacco smoke induce a corresponding trend in benzene personal air contamination. Thus, smoking induces an increase of benzene contamination in indoor environments and in personal human exposure (23), but the measurement of benzene personal air contamination is not the best and most unique technique to
distinguish between active and passive exposure to tobacco smoke.

Both groups of volunteers considered in the present study are nonprofessionally exposed subjects. Considering the temporal trend of occupational limit of benzene (TLV–TWA 1990–1991 = 10,000 ppb; actual = 100 ppb) and considering that it is a confirmed human carcinogen (A1), the measured environmental pollution of this chemical (indoor and outdoor) can be considered a human health hazard even if the levels of benzene found in the present study are much lower than levels showing toxicological effects on humans (16).

Human exposure to benzene, as well as to other chemicals (aldehydes, volatile halogenated hydrocarbons, etc.), is due principally to indoor air contamination, not only in the home but probably also in many other confined environments (office, car, shops, etc.). Furthermore, benzene exposure depends on lifestyle, smoking, and home habits (number of daily air exchanges, the use of air filter, etc.). We conclude that, considering the low level of exposure to aromatic hydrocarbons in the air, only biological markers such as benzene–DNA adducts (24,25) will permit epidemiological studies concerning this environmental and public health problem with correct sensitivity and specificity.

REFERENCES

1. Wallace L.A. Major sources of benzene exposure. Environ Health Perspect 82:165–169 (1989).
2. NIOSH. National Occupational Hazard Survey (NOHS). Cincinnati, OH: National Institute for Occupational Safety and Health, 1976.
3. NIOSH. Revised Recommendation for an Occupational Exposure Standard for Benzene. Cincinnati, OH: National Institute for Occupational Safety and Health, 1977.
4. Warhol JE. Measurements of benzene, toluene, and xylene in urban air. Atmos Environ 17(9):1713–1722 (1983).
5. Nate RJ. Proceedings of the VII International Symposium on Alcohol Fuel, 20–23 October 1986, Paris: Ed Tech, 1986.
6. Gilli G, Bono R, Scarsatone E, Guerrini E. Formaldehyde and acetaldehyde air contamination. A two year study before the introduction of new gasoline in Italy. Toxicol Environ Chem 33:219–229 (1991).
7. Wallace LA, Zweidinger R, Erickson M, Cooper S, Whittaker D, Pellizzi E. Monitoring individual exposure: measurement of volatile organic compounds in breathing-zone air, drinking water and exhaled breath. Environ Int 8:269–282 (1982).
8. Wallace LA, Pellizzi E, Harwell T, Rosenzweig R, Erickson M, Sparacino C, Zelton H. Personal exposure to volatile organic compounds. I: Direct measurement in breathing-zone air, drinking water, food, and exhaled breath. Environ Res 35:293–319 (1984).
9. Wallace LA, Pellizzi E, Harwell TD, Sparacino C, Sheldon L, Zelton H. Personal exposures, indoor/outdoor relationship and breath levels of toxic air pollutants measured for 355 persons in New Jersey. Atmos Environ 19:1651–1661 (1985).
10. Wallace LA. Personal exposure. Indoor and outdoor concentrations and exhaled breath concentrations of selected volatile compounds measured for 350 residents of New Jersey, North Dakota, North Carolina, and California. Toxicol Environ Chem 12:213–236 (1986).
11. Wallace LA, Pellizzi E, Harwell TD, Sparacino C, Whitmore R, Sheldon L, Zelton H, Perritt R. The TEAM Study personal exposure to toxic substances in air, drinking water, and breath of 400 residents of New Jersey, North Carolina, and North Dakota. Environ Res 43:290–307 (1987).
12. Wallace LA. The exposure of the general population to benzene. Cell Biol Toxicol 5(3):297–314 (1989).
13. Ozkanyah H, Ryan PB, Wallace LA, Nelson WC, Behar JV. Source and emission rates of organic chemical vapors in homes and buildings. In: Indoor Air 87: Proceedings of the Fourth International Conference on Indoor Air Quality and Climate, 17–21 August 1987, West Berlin: Institute for Water, Soil, and Air Hygiene, 1987:17–21.
14. Tancrede M, Wilson R, Zeise L, Crough EAC. The carcinogenic risk of some organic vapors indoors: a theoretical survey. Atmos Environ 21(10):2187–2205 (1987).
15. Fishbein L. An overview of environmental and toxicological aspects of aromatic hydrocarbons. I: Benzene. Sci Total Environ 40:189–218 (1984).
16. WHO. Environmental Health Criteria, No 150. Geneva: World Health Organization, 1993.
17. Bono R, Arossa W, Russo R, Scarsatone E, Castello D, Gilli G. Environmental tobacco smoke and urinary cotinine in a group of adolescents. J Environ Sci Health A29(7):1439–1449 (1994).
18. Bono R, Russo R, Arossa W, Scarsatone E, Gilli G. Involuntary exposure to tobacco smoke in a population of adolescents. Urinary cotinine and environmental factors. Arch Environ Health 51(2):127–131 (1996).
19. Gilli G, Bono R, Scarsatone E. Volatile halogenated hydrocarbons in urban atmosphere and in human blood. Arch Environ Health 45(2):101–106 (1990).
20. Gilli G, Bono R, Scarsatone E, Guerrini E. Geographical and temporal patterns of air-borne and personal 1,1,1-trichloroethene exposure in Piedmont region (Italy). Sci Total Environ 116:261–268 (1992).
21. SAS/STAT User’s Guide. Version 6, 4th ed. Vols 1–2. Cary, NC: SAS Institute, 1992.
22. Gilli G, Scarsatone E, Bono R. Benzene, toluene and xylene in air, geographical distribution in the Piedmont region (Italy) and personal exposure. Sci Total Environ 148(49–56 (1994).
23. Wallace LA, Pellizzi E, Harwell TD, Perritt R, Ziegenfuß R. Exposure to benzene and other volatile compounds from active and passive smoking. Arch Environ Health 42(5):272–279 (1987).
24. Lagoría S, Tagesson C, Forastiere F, Iavarone I, Axelson O, Carere A. Exposure to benzene and urinary concentrations of 8-hydroxydeoxyguanosine, a biological marker of oxidative damage to DNA. Occup Environ Med 51:739–743 (1994).
25. Butcher M, Lehner R, Angerer J, GC/MS determination of N-phenylvaline, a possible marker for benzene exposure in human hemoglobin by the "N-alkyl Edman Method." Arch Occup Environ Health 65(6):411–414 (1994).
26. Harkov R, Kebekus B, Bozzelli W, Lioy PJ, Daisey J. Comparison of selected volatile organic compounds during the summer and winter at urban sites in New Jersey. Sci Total Environ 38:259–274 (1984).
27. Sexton K, Westberg H. Nonmethane hydrocarbon composition of urban and rural atmospheres. Atmos Environ 18(6):1125–1132 (1984).
28. Clark AI, McIntyre AE, Lester JN, Perry R. Ambient air measurements of aromatic and halogenated hydrocarbons at urban, rural, and motorway locations. Sci Total Environ 39:265–279 (1984).
29. Shah J. Inter-urban comparison of ambient volatile organic compound concentrations in U.S. cities. J Am Pollut Contr Assoc 39(5):729–732 (1989).