Exposure of the General Population to Gasoline

by Gerald G. Akland

This paper summarizes the currently available information on gasoline exposure to the general population. In general, the largest contribution to the time weighted exposures results from exposures while indoors, which are influenced by the outside air, indoor sources, and attached garages. Personal activities, including refueling and commuting, contribute significantly higher exposures but last for only a small portion of the 24-hr time weighted average. The highest exposed group includes those individuals living near large service stations and those with contaminated water supplies.

Introduction

The many benefits of our modern, developing, industrial society are accompanied by certain hazards. Careful assessment of the relative risk of existing and new manmade environmental hazards is necessary to establish sound regulatory policy. There is an increasing awareness of the need for exposure information as part of this riskbased approach to environmental standard setting. This need is equal for exposure information as it relates to gasoline exposures. Because gasoline contributes to virtually everyone's daily exposure as a result of traffic emissions, there is clearly a need to understand the significance of this exposure as it relates to public health. Accordingly, it is not surprising that the sponsors of the International Symposium on the Health Effects of Gasoline wanted an overview of gasoline exposures to the general population. In particular, the organizers wanted information about what people were being exposed to, when they were exposed, where and for how long, and who made up the "sensitive population subgroups" exposed to all the numerous constituents of gasoline. With this information, the health scientists could debate the toxicological importance of the many constituents of gasoline. This information could then provide a basis for understanding the health risks of gasoline and help prepare us for the inevitable changes in the fuel composition already in progress, including the change to clean fuels. It is therefore the objective of this paper to summarize the available general population exposure information. This task has been made easier as a result of a contract effort by ENVIRON Corporation of Arlington, Virginia, funded by the American Petroleum Institute. The results of this contract as well as other research were presented in a conference about 1 year ago sponsored by the American Petroleum Institute on Exposures to Gasoline (1).

Exposure Concepts

"Exposure" has been used in many different ways when applied to environmental or human health effects studies. In this paper, exposure is defined as the joint occurrence of two events: The pollutant of a measurable concentration is present at a particular location at a particular time, and the person is present at the same time and location. Exposure is characterized by who is exposed, to what pollutant, how the exposure occurred (through air, water, soil, food), where the exposure occurred (route-inhalation, ingestion, dermal contact) and when (time pattern). This definition is consistent with the definition given by the National Academy of Science report (2) and the recently completed EPA Guidelines on Exposure Assessment (3). Total human exposure accounts for all exposures to a specific contaminant regardless of environmental medium or route of entry (4).

Traditionally, air pollutant exposure has meant exposure to ambient air. Environmental scientists have recognized that measurements at outdoor monitoring sites provide a severely limited estimate of personal exposure. Concerns about the adequacy of ambient measurements have been increased by our recent comprehension of the impact of the proportion of time that most individuals spend indoors. Because we spend most of our time indoors (approximately 90%, i.e., 21.5 hr), it follows that the pollutant levels experienced within these microenvironments weigh heavily on one's total exposure for a given pollutant (5). No matter how toxic a chemical might be, if people do not come into contact with the chemical, there is no exposure or public health risk from that chemical.

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Exposure assessment is a critical component of the risk assessment. An exposure assessment includes the identification of the appropriate sources, microenvironments with potentially high concentrations, and factors that when combined with the emissions products result in increased risks to the exposed human populations and ecosystems. The major elements of an exposure assessment are: a) identifying and quantifying source emissions and transformation products (source characterization and fate); b) understanding the movement of pollutants to exposed populations (both humans and ecosystems) and identifying exposure route(s); c) determining the concentration of the substance in various media and microenvironments; d) assessing exposure from all sources; e) determining the magnitude, duration, frequency, and probability of exposure and the percentage of a population exposed above specified levels of health or ecosystem concern; and f) determining the amount of a pollutant to which the subject is exposed actually enters the subject.

In the absence of exposure information, the typical exposure assessment has relied on standard assumptions, such as a 70-kg man who spends 24 hr each day of his life on his porch being exposed to the ambient air (commonly referred to as the "porch potato"). Air exposure is estimated by "exposure models," which are generally air pollution dispersion models designed to provide a reasonable estimate of a pollutant concentration at a fixed point in space. In general, this is the state of exposure science today for most pollutants under regulatory consideration. Within this list of pollutants is the complex, constantly changing mixture we call gasoline. The available information on gasoline exposures is summarized in the following sections.

Gasoline Exposures

As highlighted by the Workshop on Gasoline Exposures, little is known about personal exposures for even the inhalation pathway. What we have learned is primarily the result of TEAM studies (6) using benzene as the surrogate chemical for automotive emissions. Unfortunately, TEAM studies do not provide much information needed to understand exposures to gasoline. For example, Table 1 lists nine microenvironments or activities identified at the workshop as needing additional exposure data. TEAM studies provide us with some information about exposures at home, at work (nonoccupational), and outside ambient exposures. Other microenvironments (worker exposures, underground leaks, fenceline community exposures, and parking

Table 1. Ranking of exposure microenvironments/activities based on data need and potential for maximum exposure.

Occupational exposures
Homes with contaminated water source
Fenceline community exposures
Home exposures
General population at work
In-transit exposure
Parking garages
Urban air (ambient exposures)

garages) are not specifically addressed in the TEAM study design, or the data are severely limited by time integration of the sampling methodology (e.g., 12-hr sample integration [exposures at service stations and exposures in transit]).

In contrast, we know a lot about the chemistry and automotive emissions of gasoline. For example, liquid gasoline is a complex mixture that consists of many constituents that vary from company to company, region to region, season to season, and year to year. The use of aromatic compounds in gasoline has increased over the past decade to boost automobile performance. In 1980, U.S. unleaded gasoline contained approximately 22% aromatic hydrocarbons. Typical blends of unleaded gasoline today contain about 33% (7). One aromatic compound in gasoline, benzene, is generally present in small amounts (<5% by volume, typically about 1.5%).

Gasoline vapor has significantly lower amounts of aromatic compounds than the liquid phase of gasoline. For example, at 25°C under equilibrium conditions, gasoline vapor contains approximately 2% aromatic compounds (8). Benzene typically averages less than 1% (0.7%). Benzene will be the primary compound characterized in this paper because it is the compound that has the most information by which to judge human exposures to gasoline-related emissions.

Motor vehicles emit organic compounds from a variety of sources, generally categorized according to tailpipe, parking (diurnal and hot soak), and running loss (evaporative and refueling emissions). The relative contribution of each depends on fuel characteristics and vehicle operating conditions (e.g., speed and temperature). Light-duty gasoline vehicles are responsible for more than 90% of the motor vehicle nonmethane hydrocarbon (NMHC) emissions. Approximately 50% of the emissions occur from the tailpipe during routine operations, 45% is due to evaporative losses, and 5% while refueling. Between 1975 and 1990, fleet average NMHC emissions decreased more than 70%, of which 4% or less is benzene.

Mobile sources account for 85% of the total benzene emissions in the United States. Of this amount, 70% comes from exhaust, 14% from evaporative emissions, and 1% from motor vehicle refueling (9). Opportunities for human exposure to gasoline and gasoline combustion byproducts are abundant. Whereas relatively few people are occupationally engaged in the production, distribution, and storage of gasoline where there may be greater opportunities for high exposure (both in time and concentration levels), virtually the entire population is exposed nonoccupationally through exposures to gasoline engine emission products found in the ambient air or while driving their vehicles. In addition, a large portion of the U.S. population (about 110 million) engages in refueling their vehicles, others live downwind of refineries and major storage and transfer facilities, have homes with attached garages where evaporation of gasoline vapors may seep into the homes, and may find their source of drinking water has been contaminated from leakage of gasoline into the water supply. These nonoccupational exposures to gasoline generally occur through inhalation (air), with the exception of exposures caused by underground leaks, which may lead to multimedia exposures. The exposures measured in each of the above-mentioned microenvironments (ambient air, refueling, fenceline, parking garages, etc.) will be described based on the available information.

Inhalation Exposures

Ambient (Outdoor) Concentrations

Ambient levels become the reference to which additional concentration levels are added. For example, concentration levels within the indoor microenvironment (described below) are some function of the outdoor levels plus the contribution from the indoor sources and activities. Accordingly, the ambient concentration levels are the first microenvironment of concern in this paper.

Several investigators have measured benzene over the past 20 years. Shah (10) summarized a database consisting of over 5400 measurements he had compiled wherein the average value was 3.3 ppb, which is 1.3 ppb larger than the average reported by Wallace (6). Wallace (11) offers an explanation for these differences as a result of differences in sampling locations (TEAM outdoor measurements were primarily in residential areas instead of the commercial center city data reported by Shah). A more recent EPA investigation of 6 A.M.-9 A.M. average concentrations in U.S. cities showed the median of the median values of the 39 cities to be 2 ppb in 1984–1986, with median values by city ranging from less than 1 ppb to 5.7 ppb (12). Sexton and Westberg (13) reported 6 A.M.-9 A.M. geometric mean values ranging in seven cities from 6 ppb in Houston, Texas, to 2.7 ppb in Washington, DC. In contrast, rural areas have geometric values of less than 0.5 ppb. These results clearly establish the importance of the automobile as a source of benzene values. Furthermore, a factor of three about the average benzene concentration might be expected from urban area to urban area depending on the sample location and time of sample collection and/or the availability of additional stationary source emissions (e.g., concentrated oil refinery and petrochemical operations as in Houston, Texas).

Concentration Levels Near Service Stations, Bulk Terminals, and Refineries

Benzene exposures to people living in the vicinity of service stations, bulk plants, bulk terminals, and refineries may be increased above average ambient levels when the wind direction distributes emissions that may occur from these facilities to the nearby residences. However, in the TEAM studies conducted in Bayonnne and Elizabeth, New Jersey, people living within 1 km of chemical and petroleum refineries did not have elevated exposures as compared to those people living more than 1 km from these sources (14). In contrast, a number of microenvironmental studies have been conducted that characterize gasoline vapor exposures at service stations. In particular, the Petroleum Association for Conservation of the Canadian Environment (15,16) collected and analyzed samples at the

boundaries of service stations. They reported mean concentration levels of 146 ppb in the summer and 461 ppb in the winter, with corresponding maxima of 2277 and 5417 ppb, respectively. These results should be considered as of very short-term duration and are likely to be higher than those observed at residences located adjacent to the service station properties.

To obtain a more realistic estimate of actual concentrations at the residence, the data were adjusted based on findings by Bond et al. (17) that concentrations away from the pump were typically 2% of the concentrations at the pump, with a maximum of about 10%. This would result in indoor levels of about 6 ppb and outdoor levels of 540 ppb for people living right at the boundary of the service station property. However, these values are probably extremely high when viewed by data published by the Northeast States for Coordinated Use Management (NESCAUM) (18), which estimated annual benzene levels to average 0.04 ppb with an upper limit of 0.16 ppb for residents in neighborhoods near (within 30-200 m downwind) service stations. Because these values are below typical outdoor values in the United States, it is clear that we have great uncertainty in our exposure estimates.

There is little information available in the literature about gasoline vapors in the vicinity of bulk plants (there are some 15,000 in the United States). Exxon (19) published fenceline measurements of benzene vapor concentrations at four refinery plant complexes located in the United States, showing that the concentrations were 1 to 4 ppb. Westberg and Lamb (8) reported 24-hr ambient benzene concentrations at distances greater than 1 km from the refinery perimeter to range from 1 to 4.3 ppb, the typical urban monitoring average concentration. This confirms Wallace's conclusion that refineries and chemical manufacturing plants do not contribute to one's total exposure in any discernible way (14).

Indoor Residential Concentrations

Several indoor air studies of volatile organic compounds, including benzene, have been carried out. In the United States, Wallace and Pellizzari (20) reported indoor air concentrations in homes without smokers averaged 3.1 and 1.5 ppb during the fall and winter and spring and summer, respectively. In contrast, homes with smokers averaged 5.3 and 1.6 ppb, over the same time periods, respectively, with an average increase of approximately 1.2 ppb benzene in homes with smokers. These findings are similar to those obtained in homes in Holland (21) and West Germany (22). The few measurements of benzene in office buildings generally have shown no increase in indoor air levels above those found in the outdoor air.

There are a number of consumer products, surface coatings, and materials found indoors that emit benzene vapors, such as latex paints, adhesives, marking pens, and rubber products. Even though each of these materials and products emit small amounts, when combined they account for about 20% of the total population exposure to benzene (10) because of the amount of time spent near these sources.

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Residential Garages

There is little information in the literature that documents the levels of benzene concentrations inside garages. One study (23) indicated that garage levels were 16 times higher in the summer than outdoor levels and three times higher in the winter. The concentrations reported were 24 ppb in the summer and 8 ppb in the winter. The authors conclude that evaporative emissions from automobiles and the storage of fuels and solvents in the garage were the likely sources for the higher levels (23). There also may be evaporative contributions from other mechanical devices often found in the garages, such as power lawn mowers and chain saws. The extent to which these may add to the total concentrations measured was not quantified.

Other Routes of Exposure

Drinking Water

Surveys of the U.S. drinking water supplies have rarely found benzene levels above the detection limit. Letkiewicz et al. (24) estimated that 99.8% of all the groundwater systems contain either no benzene or levels below 0.5 µg/L; an additional 118 systems were estimated to have levels between 0.5 and 5 µg/L, and no systems were expected to exceed 5 µg/L. Of the 11,202 surface water systems, all were expected to have benzene levels below 0.5 μg/L. However, discharges into the soil or groundwater have become an important concern for many state and local health officials. Because there are approximately 1.4 million underground storage tanks, with some 20 to 35% leaking (25), the potential for contamination of the water supply can be a significant concern for exposures. The affected water can enter a house through the plumbing (faucets, commodes, showers/baths, washing machines, and dishwashers), which can lead to exposures through ingestion of water or food, inhalation of vapors released from sources of water in the house, and dermal contact via bathing/showering or in other uses of water such as washing dishes. Benzene levels in homes where there has been a contaminated water source have been measured, but the levels are generally less than 1 ppb (18). However, severely contaminated homes may have levels in excess of 20 ppb, with peak concentrations in the bathroom while showering in excess of 160 ppb. Peak concentrations while showering represent a 6-fold increase over the drinking water concentration (18).

Auto-related Activities

Exposure to automobile exhaust while in an automobile and pumping gas results in increased personal exposure to benzene. (In addition, benzene vapors are emitted from parked cars after being driven, which has an impact on concentrations in garages, as noted above.) Available information about each of these microenvironments/activities is described below.

Self-serve Automobile Refueling. Exposures to gasoline and its vapors at self-serve stations result from: *a*) vapors displaced from the filler tubes and gas tanks

during refueling, b) fuel spills during refueling, c) loss of vapors from vented underground storage tanks, and d) evaporative and tailpipe emissions from other automobiles present. Exposures can occur during refueling and during nonfueling time spent at the service station. Based on studies summarized by NESCAUM (18), the range of average benzene levels was 164 to 1100 ppb, with upper limits of 4200 ppb. The Petroleum Association for Conservation of the Canadian Environment (15,16) conducted a study in five Canadian cities (Halifax, Montreal, Toronto, Calgary, and Vancouver) in the summer of 1985 and the winter of 1986. Benzene concentrations ranged up to 7070 ppb with an average of 1130 ppb for the summer, and in excess of 10,000 ppb with an average of 1250 ppb for the winter.

The U.S. EPA (26) estimated in 1984 that approximately 70% of the gasoline was dispensed by self-service pumps (a percentage that is likely low compared to today). With at least 158 million U.S. automobile drivers, this translates into more than 110 million people being exposed while engaged in self-serve automobile refueling, which usually lasts 2 min or less. NESCAUM (18) has estimated that the annual number of station visits is 70, with the average time at a station lasting 5 min. This results in an average yearly time of 350 min (about 6 hr) at a service station. NESCAUM provides an extreme case of 262 min per year (18), which is likely to be low for multicar families with one individual performing the fueling activity.

Auto Travel. Benzene exposures increase in proportion to the time spent in the car. Wallace (11) calculated that while in the car, exposures increased by a factor of 3-4 over normal exposures (15-20 ppb). The average commute time one way is about 40 min, which results in about 30 ppb during each commute. In-vehicle concentrations of benzene were also recorded in a study conducted in the South Coast Air Basin of California (27). During the trips that averaged 33 min in duration (typical of local commuters), in-vehicle levels averaged 13 ppb, 2-4 times the ambient levels measured at fixed sites. These levels were higher in the winter (16 ppb) than in summer (10 ppb), with maximum levels recorded over 80 ppb during the study. It was noted that when average speeds were below 25 mph, the concentrations were significantly higher (by 4 ppb). In another study of in-transit exposures, Chan et al. (28) observed lower benzene levels in their simulated study of travel around the EPA facility in Research Triangle Park, North Carolina (3-4 ppb average, with maximum levels of 15 ppb), which were about six times the value measured at fixed sites throughout the area.

Other Activities. Exposure to gasoline and gasoline vapors can occur through other activities, although actual concentrations are generally not known. For example, it is known that people receive relatively higher exposures to carbon monoxide while using chain saws and other gasoline-powered appliances in and around the home. Therefore, it is reasonable to assume that exposures to the other gasoline emissions, including benzene, are likely. The actual exposure levels are not known; however, a doubling of the ambient levels would be a first approximation to the exposure levels experienced while operating the equipment.

Timeweighted Attached Refueling, garage, In transit, exposure, Indoor, 1320 min Outdoor 1440 min 5 min 80 min 5 min 16.0 13 630 6.7 Urban environments Average 2780 1000 33.0 6 Maximum 48 6 16.0 300 13 630 14.7 Near gas station (fenceline) Average 540 588 5400 80 1000 617 Maximum Showering^a Home Contaminated groundwater 1500 16.0 3 13 630 42.0 20 Average Maximum 200 6000 48 6 80 1000 272

Table 2. Estimated daily benzene exposures based on microenvironmental concentrations (ppb).

Synthesizing Exposure Information

Driven by the fact that health officials must use data whether it is good or bad, I will calculate what Wallace (11) describes as an "exposure budget" for the average non-occupationally exposed individual and the maximally exposed individual. This budget will be estimated based on the data available for each of the previously discussed microenvironments and activities that are influenced by gasoline sources. This calculation does not include the influence of other sources of benzene, most notably the effect of smoking. For the individuals who smoke or who spend time in microenvironments that are occupied or have been occupied in the recent past by active smokers, there would be an increased total benzene exposure. Other activities such as painting, working with solvents, etc., would likewise increase these exposure levels.

Table 2 summarizes the results of this analysis, which is the calculation of concentrations in the microenvironments listed across the top of the table multiplied by the estimate of time people spend in these microenvironments. It can be seen that the average person is likely to have an average exposure of no more than 6.7 ppb, but if the residence has a contaminated water supply from a leaking gasoline source, then the average exposure could be about six times greater, to 42 ppb. In the extreme, the highest exposure might exceed 270 ppb if there were the underground water source, or 600 ppb for an individual who might live within the immediate property of a very large gasoline station.

Estimates such as these are imprecise and especially vulnerable to errors in the measurements (or mathematical estimates) that were used to calculate the exposures. However, within the constraints and assumptions given, it provides a ballpark upper exposure limit, and it also serves to emphasize just how important some microenvironments potentially can be for the total contribution to one's exposure. In the benzene example, if only the outdoor concentrations were increased by a factor of 10, the resulting impact on total exposure would be less than 10%. However, changes in the indoor concentrations would result in a more comparable change in total exposure because the general population spends over 90% of its time indoors.

Participants at the Workshop on Gasoline Exposures recognized these difficulties and came up with a list of recommendations for future research to address many of the noted deficiencies. In particular, they recommended

Table 3. Recommendations for future research.

Develop a critically reviewed database

Refine exposure pathway analyses to include total exposure measurements and source contributions

Improve the understanding of sources contributing to the shortand long-term variation in ambient levels

Validate and define the limitations of models for both short- and longterm exposures

Standardize methods for sampling and analysis

Enhance our information about how people spend their time in microenvironments that would receive higher exposures

Improve the scientific understanding of the exposure impacts of additives and reformulations

Develop the scientific understanding that would relate exposure to biological indicators

Refine exposure measures used in epidemiological studies and health assessments

Evaluate the effectiveness of public policies to reduce exposures

that a critically reviewed database and a compendium of methods for sampling and laboratory analysis for measuring gasoline exposures should be developed. The complete list of recommendations is provided in Table 3.

Summary and Conclusions

There is a direct relationship between the fuel consumed for transportation and the resulting emissions. Because gasoline is the major fuel used currently and in the foreseeable future, it is necessary to understand the potential exposures to the general population from the use of gasoline. This is especially important to us as we consider the potential impact on exposures that could develop from the United States switching from this fuel to alternative forms of energy, such as methanol- or ethanol-based fuels. In addition, this understanding is important on a global basis, because the United States alone consumes about 35% of the world's transport energy (29).

There is a nearly complete database on the chemical composition of gasoline and the resulting automotive emissions. Likewise, there is a database that contains some information about the magnitude and extent of occupational exposures for those engaged in the refining and distribution of gasoline. Unfortunately, the information about general population exposures is incomplete, nonexistent, and often what is available is inconsistent with other data. However, the limited information on benzene, which

[&]quot;While showering (5 min).

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is one of the constituents of gasoline, is presented as an example by which to judge the relative importance of several of the identified major microenvironments and activities that increase exposures to gasoline emissions. The analysis showed, for example, that average benzene exposures should be about 7 ppbv. If the individuals happen to have a contaminated groundwater supply, their exposures could average over 40 ppb. The extreme portion of the distribution (the most uncertain part of the distribution) indicates that individuals who live near a gas station under the worst measured concentrations could experience maximum exposures over 600 ppb, which is about a factor of 100 greater than the average benzene exposures.

The uncertainty of these estimates cannot be overemphasized. The concluding message is to appreciate the need for complete, accurate exposure information in the process of determining risks related to any pollution source. It would be extremely unfortunate and costly to the consumer if we should develop a national policy for mitigating gasoline exposures based entirely on our understanding of outdoor ambient exposures. Or, similarly, it would be most unfortunate if we attempt to regulate based on our exposures while at a service station. Clearly, the activities, locations, the amount of time spent while engaging in these activities, and the concentrations within these microenvironments are needed before a scientifically defensible exposure assessment for gasoline can be undertaken. Fortunately, this workshop can help spread this message to all the scientists engaged in risk assessment, so that we can encourage the gathering of appropriate exposure data in the future.

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