Automobile Proximity and Indoor Residential Concentrations of BTEX and MTBE

Diana E. Hun[1,*], Richard L. Corsi[2], Maria T. Morandi[3], and Jeffrey A. Siegel[2]

[1]Oak Ridge National Laboratory, Oak Ridge, TN
[2] University of Texas at Austin, Austin, TX
[3]University of Texas Health Science Center, School of Public Health, Houston, TX

[*]*Corresponding email: hunde@ornl.gov*

SUMMARY
Attached garages have been identified as important sources of indoor residential air pollution. However, the literature lacks information on how the proximity of cars to the living area affects indoor concentrations of gasoline-related compounds, and the origin of these pollutants. We analyzed data from the Relationships of Indoor, Outdoor, and Personal Air (RIOPA) study and evaluated 114 residences with cars in an attached garage, detached garage or carport, or without cars. Results indicate that homes with cars in attached garages were affected the most. Concentrations in homes with cars in detached garages and residences without cars were similar. The contribution from gasoline-related sources to indoor benzene and MTBE concentrations appeared to be dominated by car exhaust, or a combination of tailpipe and gasoline vapor emissions. Residing in a home with an attached garage could lead to benzene exposures ten times higher than exposures from commuting in heavy traffic.

IMPLICATIONS
Residing in a home with an attached garage could lead benzene exposures that have been associated with 17 excess cancers in a population of a million. Strategies to lower exposure to gasoline-related contaminants in homes include improving construction practices to prevent the infiltration of pollutants or incorporating detached garages.

KEYWORDS
Attached garages, benzene, exposure, gasoline, RIOPA.

INTRODUCTION
Various gasoline-related volatile organic compounds (VOCs) have been identified by the U.S. Environmental Protection Agency (EPA) as hazardous air pollutants. Benzene, toluene, ethylbenzene and xylenes (BTEX) vaporize from liquid gasoline, and are emitted in car exhaust and by some consumer products. Benzene has been classified by the U.S. EPA as a known human carcinogen (Group A), and assessments among nonsmoking populations have repeatedly identified benzene as an important contributor to cumulative environmental cancer risk (Hun et al. 2009; Sax et al. 2006). Adverse health effects associated with elevated concentrations of other BTEX components and MTBE have been reported (CalEPA 2009; U.S. EPA 2005). Up until 2000, MTBE was almost always present in gasoline, making it an ideal tracer for gasoline-related exposures.

Even though exposure to BTEX commonly occurs in many microenvironments, personal concentrations for these compounds have been primarily associated with attached garages (Sexton et al. 2007) because of sources within garages and because Americans spend on average nearly 70% of their time in their homes (Klepeis et al. 2001). Sources of BTEX and

MTBE include stored gasoline, gasoline-powered devices, and occasionally consumer products such as paints, adhesives, and paint thinners (DRI 2006; Sack et al 1992). These sources can lead to BTEX levels in garages that are five to 18 times higher than in the adjacent living area of single-family homes (Batterman et al. 2007; Thomas et al. 1993). These contaminants can migrate from attached garages into the occupied space partly because the shared wall between these two areas tends to be among the leakiest components of the house envelope, and because of the presence of heating, ventilation and air conditioning (HVAC) components in some attached garages. Thus, the contribution of sources within garages to indoor concentrations (C_{in}) of BTEX and MTBE has been determined to range from 9 to 85% (Dodson et al. 2008; Graham et al. 2004). In the case of benzene, such contribution can be similar or higher than that of tobacco smoke (Thomas et al. 1993). Consequently, homes with attached garages have been reported to have statistically higher C_{in} for these contaminants than residences that lack this source of pollution (Dodson et al. 2008).

Although the aforementioned work provides compelling evidence that pollutants in attached garages can influence indoor residential environments, data from these studies were based on small samples. Furthermore, attached garages are one of several locations where vehicles are parked at or near homes, and the literature lacks information on how other parking locations affect indoor air quality. To our knowledge, previous researchers have not used MTBE as a tracer to determine if BTEX indoors was related to gasoline sources. In this study we examine how the proximity of parked vehicles next to the living quarters influences indoor concentrations of BTEX and MTBE. To this end, we analyze data from nonsmoking homes that participated in the Relationships of Indoor, Outdoor, and Personal Air (RIOPA) study; and evaluate C_{in} and C_{out} for six cases with different source proximity. We use MTBE measurements to confirm that indoor concentrations for BTEX were influenced by gasoline-related sources, and to determine if pollutants originate from vapor or exhaust emissions. Furthermore, we estimate weekly cumulative exposure to benzene in homes due to vehicles in attached garages and in cars during heavy traffic, and their respective cancer risks.

METHODS

This research is based on an analysis of data from a sample of homes without resident smokers that participated in the RIOPA study. Data were made available by the Health Effects Institute (HEI 2008). Approximately 100 residences volunteered in each of Los Angeles County, California, Elizabeth, New Jersey, and Houston, Texas. Participants in Houston and Elizabeth constitute a convenience sample, while the participants from Los Angeles were a subset from a randomly selected sample of individuals from a previous study.

Weisel et al. (2005) provide a detailed description of the RIOPA field and measurement protocols. From 1999 to 2001, homes were monitored during two 48-hour periods that were about three months apart. MTBE was still in use as a gasoline additive in the three studied cities. Air samples were collected concurrently inside and outside of each home. BTEX and MTBE were monitored using Organic Vapor Monitors (OVM 3500, 3M Company, St. Paul, MN, USA). Concentrations at or below the method detection limit (MDL) were censored by replacement with half the MDL concentrations. In addition to monitoring the air, building characteristics and daily household activity patterns were collected during each of the sampling sessions by means of questionnaires and walkthrough surveys. Air exchange rates (AER) were simultaneously measured using a perfluorocarbon tracer (PFT) method.

Categorical data from the first home visit were usually selected when information from the first and second sampling sessions differed. Averages were calculated when C_{in}, C_{out} and

AER were available for both sessions, because these are dependent variables. Constraints reduced the overall sample size of the RIOPA database from 311 to 114.

Nonparametric analyses were utilized because the data were generally positively skewed. Associations between variables were evaluated with Spearman rank-correlation coefficients (r_s); coefficients were considered statistically significant at $p \leq 0.05$. The Wilcoxon sign-rank test was used to assess differences between paired samples, such as when C_{in} and C_{out} were concurrently measured. The Wilcoxon rank-sum test was utilized to evaluate differences between two independent samples, such as C_{in} from homes with vehicles parked in an attached garage and homes with vehicles in an adjacent carport. Similarly, the Kruskal-Wallis test was used with three or more levels. Differences were considered statistically significant at $p \leq 0.05$. SPSS (version 15.0, SPSS Inc.) was employed for these analyses.

RESULTS

The majority of the residences included in this analysis were located in Houston (HO; n = 55), followed by Los Angeles (LA; n = 38) and Elizabeth (EL; n = 21). These houses were either single-family detached (SFD) structures (n = 99) or manufactured homes (n = 15). Sixty-one of these homes were monitored twice. C_{in} and C_{out} for BTEX and MTBE in these three cities, and their respective MDLs, are summarized in Table 1. In each of the studied cities, correlations between indoor MTBE and indoor BTEX concentrations ($0.45 \leq r_s \leq 0.65$) were statistically significant, with the exception of MTBE and toluene in LA (p = 0.07). These correlations indicate that C_{in} for BTEX partly originated from gasoline-related sources because MTBE is a tracer for this fuel. Table 1 also shows that in general C_{in} was statistically higher C_{out} in Houston and Elizabeth, which indicates that sources were within or close to the living area. In Los Angeles, C_{in} and C_{out} were statistically similar for all compounds but MTBE ($C_{out} > C_{in}$, $p \leq 0.05$) and toluene ($C_{in} > C_{out}$, $p \leq 0.05$). It is not understood why the relationship of C_{in} and C_{out} for MTBE was different than that shown by benzene, ethylbenzene and the xylenes. A possible explanation is house proximity to MTBE production facilities.

Variations in ventilation rates also likely contributed to differences in indoor BTEX and MTBE concentrations among cities. AERs were much lower in Houston (median = 0.48 h^{-1}) than in Los Angeles and Elizabeth (median = 1.1 h^{-1} for both cities). AER appeared to be affected by how households maintained acceptable indoor temperatures; that is, by the amount of time participants used mechanical systems or had the windows open.

The effect of source proximity was investigated by examining the six cases illustrated in Figure 1: single-family detached (SFD) homes with cars in the attached garage (Case 1; n = 14), detached garage (Case 2; n = 7), or adjacent carport (Case 3; n = 34); manufactured homes with cars in adjacent carports (Case 4; n = 15); SFD homes with attached garages but no cars (Case 5; n = 8); and SFD homes without both attached garages and cars (Case 6; n = 36). In these cases, data from LA, EL and HO were combined. Residences in cases 3 and 4 were not combined because their indoor concentrations were statistically different.

Increase in indoor concentrations ($\Delta C = C_{in} - C_{out}$) for all compounds and studied cases are shown in Figure 1. SFD homes with vehicles in attached garages had the highest median ΔC for benzene (1.2 $\mu g/m^3$), toluene (6.4 $\mu g/m^3$), m&p-xylene (2.6 $\mu g/m^3$) and MTBE (2.7 $\mu g/m^3$), and large values for ethylbenzene (0.69 $\mu g/m^3$) and o-xylene (0.91 $\mu g/m^3$). These homes also had the highest median indoor to outdoor concentration ratios (C_{in}/C_{out}): benzene = 2.0, toluene = 2.7, ethylbenzene = 2.1, m&p-xylene = 2.1, o-xylene = 3.3, MTBE = 1.4. The SFD residences with automobiles in carports tended to have the second highest median

Table 1. Indoor and outdoor concentrations (μg/m^3) by city.

Compounds	N	Indoor Mean	Indoor SD	Indoor Med	Indoor %>MDL	Outdoor Mean	Outdoor SD	Outdoor Med	Outdoor %>MDL	Indoor vs. Outdoor[a]
Los Angeles, CA										
Benzene	38	2.14	1.12	2.20	93	2.25	1.39	1.98	83	
Toluene	38	11.2	7.04	9.48	61	8.94	6.31	6.76	39	I*
Ethylbenzene	38	1.92	2.18	1.21	88	1.39	0.75	1.39	85	
m&p-Xylene	38	5.67	7.26	3.94	95	4.08	2.69	3.87	98	
o-Xylene	38	1.97	2.12	1.58	93	1.57	0.92	1.48	90	
MTBE	38	7.81	4.97	6.52	97	9.19	5.71	7.12	98	O*
Elizabeth, NJ										
Benzene	22	1.54	1.13	1.34	88	1.19	0.66	1.15	49	I*
Toluene	22	11.3	9.64	7.46	69	6.65	5.32	3.02	33	I**
Ethylbenzene	22	2.03	3.49	1.07	62	1.10	0.81	0.97	46	
m&p-Xylene	22	5.69	10.8	3.74	92	2.32	1.34	2.28	100	I**
o-Xylene	22	1.77	2.52	1.13	85	0.86	0.43	0.96	82	I**
MTBE	22	4.82	4.13	3.74	79	4.52	4.37	3.92	90	
Houston, TX										
Benzene	58	5.29	4.71	3.60	100	2.80	2.34	2.18	100	I**
Toluene	58	17.4	22.3	10.8	70	5.43	3.45	4.51	41	I**
Ethylbenzene	58	2.67	3.42	1.85	100	1.01	0.73	0.90	95	I**
m&p-Xylene	58	7.69	12.1	5.06	100	2.80	1.96	2.38	99	I**
o-Xylene	58	2.64	4.03	1.85	98	1.03	0.68	0.96	93	I**
MTBE	58	15.3	23.3	6.88	100	10.2	16.8	5.10	95	I*

Abbreviations: MDL, method detection limit; Med, median; MTBE, methyl *tert*-butyl ether.
[a]I: C_{in} were statistically higher than C_{out}; O: C_{out} were statistically higher than C_{in}.
*$0.01 < p \leq 0.05$, **$p \leq 0.01$

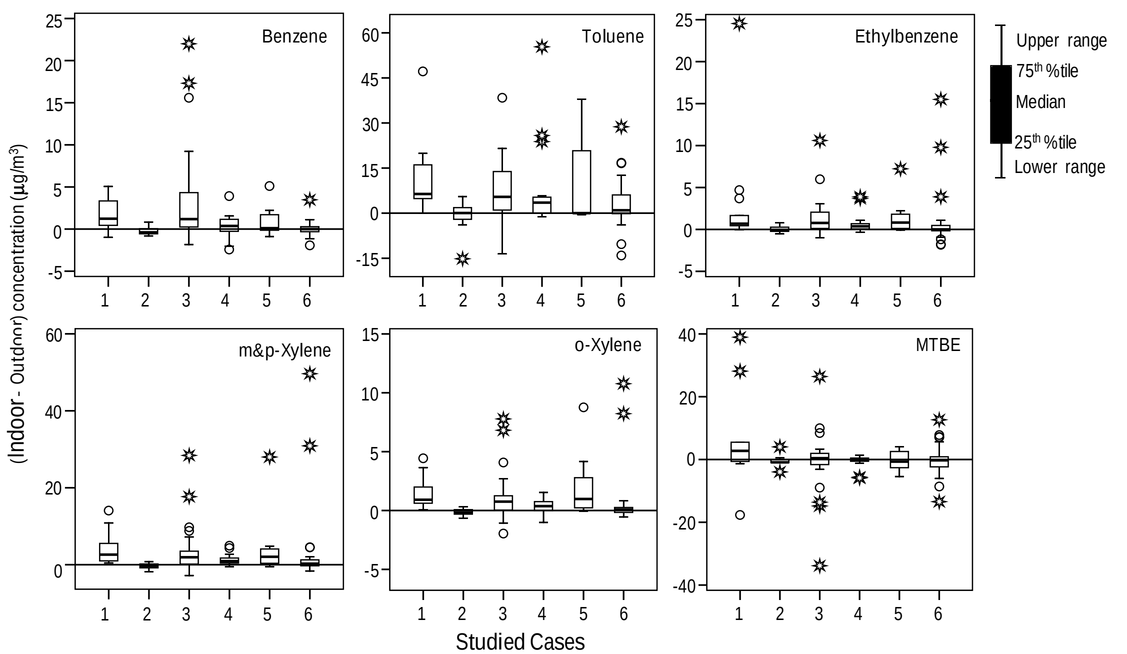

Figure 1. Difference between C_{in} and C_{out} (μg/m^3) for the six cases described in the text. Case 1, n =14 (MTBE = 109 μg/m^3 not shown); Case 2, n = 7; Case 3, n = 34 (MTBE = 103 μg/m^3 not shown); Case 4, n = 15; Case 5, n = 8; Case 6, n = 36. ‘o’ and ‘*’ indicate values between 1.5 and 3, and > 3 times the interquartile range, respectively, from the 25th or 75th percentiles.

ΔC for all VOCs. For the remaining cases, excluding homes with detached garages, median ΔC values were greater than zero for BTEX but not MTBE. Residences with detached garages had median ΔC values that were less than zero ($C_{in} < C_{out}$) for all compounds but toluene. Furthermore, these houses had the lowest median C_{in}/C_{out} ratios for BTEX (0.89 to 1.0), and the same median ratios as homes without cars for MTBE (0.92).

DISCUSSION

Results from the RIOPA investigation and those of others (Dodson et al. 2008; Sax et al. 2006) indicate that BTEX is nearly ubiquitous indoors. In the RIOPA homes, these pollutants partly originated from gasoline-related sources given that MTBE, a VOC emitted almost exclusively by gasoline, was concurrently detected indoors.

We used the ratio of MTBE to benzene indoor concentrations to examine if these compounds originated from gasoline vapors or car exhaust. Low ratios (< 5) indicate that tailpipe emissions are dominant because during combustion the amount of MTBE decreases while benzene is enriched due to toluene and xylene dealkylation (DRI 2006). Alternatively, high ratios (> 10) suggest a significant contribution from evaporative emissions from hot soak, fuel tank "breathing" due to diurnal temperature and barometric changes, and/or fuel system leakage. Our estimates indicate that vehicle exhaust appeared to drive C_{in} in about half of the homes given that median ratios for the six studied cases ranged from 1.5 to 4.2. For most of the homes with ratios above the median, a mixture of tailpipe and gasoline vapor emissions seemed to have influenced indoor concentrations of gasoline-related VOCs because the six studied cases had 80th percentile ratios that did not exceed 7.

To place some of our results into context, we used the RIOPA data to estimate weekly cumulative exposure to benzene in two microenvironments in Houston: homes with vehicles in attached garages and cars driven in a freeway with heavy traffic during commute to and from work. We calculated exposure by multiplying concentration by the exposure time. For homes, we used the mean ΔC for benzene (2.3 $\mu g/m^3$) to better evaluate the effect of nearby sources and assumed that individuals spend 70% of the week in their house (Klepeis et al. 1993). For cars, we assumed a mean in-cabin concentration of 6.1 $\mu g/m^3$ (DRI 2006), an average commute time to work of 26 minutes (U.S. Census 2009), a mean travel time from work equal to the commute time to work, and a five-day work week. Weekly exposure to benzene was 270 $\mu g/m^3 \times$ hour in homes with cars in attached garages, and 26 $\mu g/m^3 \times$ hour in cars during commute to and from work. These increases in benzene concentration due to sources near the living area, particularly vehicles in garages, could contribute to 17 excess cancers per million population in Houston. We calculated cancer risk by multiplying ΔC by the inhalation unit risk factor for benzene (7.8×10^{-6} $m^3/\mu g$) (U.S. EPA 2005).

Methods to reduce indoor residential concentrations of VOCs emitted by parked vehicles next to the living quarters include (1) sealing vertical and horizontal surfaces shared by these two spaces; (2) avoiding placement of HVAC components in the garage; (3) limiting the total air leakage of HVAC components when located in the garage; and (4) maintaining the living area at a higher pressure than that of the garage (ASHRAE 2007). Additionally, the design of new residences could be improved by incorporating detached garages.

CONCLUSIONS

Our evaluation of the RIOPA database supports prior work on the detrimental effects of attached garages on indoor air quality in residences, and provides insight on how variations in the proximity of parked vehicles to the living area affect indoor concentrations of BTEX and

MTBE. Results from our assessment indicate that homes with attached garages were affected the most by cars; these houses generally had the highest median increases in indoor concentrations (relative to outdoor concentrations) for BTEX compounds ($0.69 \le \Delta C \le 6.4$ $\mu g/m^3$). While the ΔC values may appear inconsequential, increases in indoor benzene concentrations can lead to weekly cumulative exposures that are ten times higher than those experienced while commuting in a car in heavy traffic, and to mean excess cancers of 17 per million population. Strategies to reduce exposure to gasoline-related VOCs in homes include sealing surfaces shared by the living quarters and the garage, and improving the design of homes by incorporating detached garages.

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