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Automobile proximity and indoor residential concentrations of BTEX and MTBE

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ABSTRACT

Attached garages have been identified as important sources of indoor residential air pollution. However, the literature lacks information on (1) how the proximity of cars to the living area affects indoor concentrations of gasoline-related compounds, such as benzene; and (2) the origin of these pollutants, i.e., vapor or tailpipe emissions. We analyzed data from the Relationships of Indoor, Outdoor, and Personal Air (RIOPA) study to evaluate indoor (C_{in}) and outdoor (C_{out}) concentrations for 114 residences with cars either in an attached garage, a detached garage or carport, or without cars. Results indicate that single-family detached homes with cars in attached garages were affected the most by parked vehicles, followed by homes with vehicles in carports. Concentrations in homes with cars in detached garages were similar to those in residences without cars. Low ventilation rates exacerbated C_{in} in homes with attached garages. In general, the contribution from gasoline-related sources to indoor benzene and MTBE concentrations appeared to have been dominated by car exhaust, or by a combination of tailpipe and gasoline vapor emissions. Residing in a home with an attached garage could lead to benzene exposures that are an order of magnitude higher than exposures from commuting in a car in heavy traffic, with a risk of 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 into the living quarters or incorporating detached garages.

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1. 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 risk assessments among nonsmoking populations have repeatedly identified benzene as an important contributor to cumulative environmental cancer risk [1–4]. Adverse health effects associated with elevated concentrations of other BTEX components and methyl tert-butyl ether (MTBE) have been reported [5,6]. Up until 2000, MTBE was emitted almost entirely by 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 [7,8] because of sources within garages and because Americans spend on average nearly 70% of their time in their homes [9]. Sources of BTEX and MTBE include stored gasoline, gasoline-powered devices (e.g., automobiles and lawn mowers), and occasionally consumer products such as paints, cleaners, detergents, adhesives, paint thinners and oils/lubricants [10–12]. 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 [13,14].

Air 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 [15], and because of the presence of heating, ventilation and air conditioning (HVAC) components in some attached garages [16]. Batterman et al. [13] estimated that about 6.5% of the whole-house air exchange rate can originate from the attached garage. Thus, the contribution of sources within garages to indoor concentrations of BTEX and MTBE has been determined to range from 9 to 85% [13,17,18]. In the case of benzene, such contributions can be similar or higher than that of tobacco smoke. Thomas et al. [14] concluded that mass transfer rates of benzene from the garage

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to the living area ranged from 24 to 26,000 $\mu\text{g}/\text{h}$ in three homes, whereas a home with three smokers had indoor source strengths for benzene that varied from 150 to 13,000 $\mu\text{g}/\text{h}$. Given that BTEX and MTBE can migrate into the living quarters, homes with attached garages have been reported to have statistically higher indoor concentrations for these contaminants than residences that lack this source of pollution [17].

Although the aforementioned work provides compelling evidence that pollutants in attached garages can infiltrate and 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. The literature lacks information on how other parking locations that vary the proximity of cars to the living area affect indoor concentrations of BTEX and MTBE. To our knowledge, previous researchers have not used MTBE as a tracer to determine if indoor concentrations of BTEX were related to gasoline sources. In this study we examine how the proximity of parked vehicles next to the living quarters affects 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. In particular, we evaluate indoor and outdoor concentrations for six cases: single-family detached (SFD) homes with cars in the (1) attached garage, (2) detached garage, or (3) adjacent carport; (4) manufactured homes with cars in adjacent carports; (5) SFD homes with attached garages but no cars; and (6) SFD homes without both attached garages and cars. 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 compare the RIOPA concentrations with those from a fixed monitoring site. Last, 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.

2. Methodology

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 [19]. 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. [20] provide a detailed description of the RIOPA field and measurement protocols. From 1999 to 2001, homes were monitored during two 48-h periods that were approximately three months apart. MTBE was still in use as a gasoline additive in the three studied cities during the RIOPA study. 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) for a compound were censored by replacement with half the MDL concentrations. Sampling quality control measures involved the evaluation of analytical and measurement precision. Analytical precision was utilized as a measure of instrumental reproducibility and was estimated using the pooled coefficient of variation (CV) of replicate sample analyses. Measurement precision was an indicator of method reproducibility and was estimated using the pooled CV of collocated sample concentrations. For MTBE, the compound used as a tracer for gasoline, the CV for analytical precision was 8.6% ($n = 44$) and the CV for measurement precision was 22% ($n = 151$) [20]. 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 that employed perfluorinated methylcyclohexane as the tracer gas. Air exchange rates were reported as time-averaged values for the sampling period.

In the analysis of the RIOPA dataset, categorical data from the first home visit were usually selected when information from the first and second sampling sessions differed. Averages were calculated when indoor (C_{in}) and outdoor (C_{out}) concentrations, and AER were available for the two monitoring sessions, because these are dependent variables that describe a single household.

Constraints reduced the overall sample size of the RIOPA database from 311 to 114. Apartments ($n = 108$) and single-family attached homes ($n = 11$) were not included in the evaluation because pollutants from adjacent dwellings can infiltrate through shared walls and affect the measured concentrations. Households where someone smoked during a sampling period ($n = 1$) or that had gasoline-powered devices other than vehicles inside the house ($n = 16$) were excluded to limit the assessment on the effects from parked vehicles. Ventilation rates greater than 5 h^{-1} ($n = 2$) were also excluded because the PFT method is unreliable at these values. Residences where volumes were recorded to be less than 80 m^3 ($n = 3$) were not included because it is highly probable that these values were not correct. Houses where information on the location of the parked car was missing or where vehicles were parked in different locations during each sampling period ($n = 36$) were excluded from the analysis. Only homes with measurements for C_{in} , C_{out} and AER were evaluated. Missing information and other constraints further reduced the dataset size by 20 homes.

Nonparametric statistical 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 indoor and outdoor concentrations that were concurrently measured. The Wilcoxon rank-sum test was utilized to evaluate differences between two independent samples, such as indoor concentrations 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.

Indoor concentrations were used to evaluate associations between BTEX and MTBE. The net increase in indoor concentration ($\Delta C = C_{in} - C_{out}$) was utilized to isolate the effect from indoor or nearby sources (e.g., cars in attached garages or carports) on C_{in} , and to examine how the proximity of these sources affected indoor concentrations. The percent contribution of sources within or close to the living area to indoor concentrations (I_{cont}) was calculated by dividing ΔC by C_{in} . This contribution was assumed to be zero in homes where ΔC was negative (i.e., $C_{out} > C_{in}$). Source strength was not estimated because vehicles, the main source of benzene and MTBE in the studied homes, were not in the occupied space. Furthermore, the garage volume was not reported in the RIOPA study.

3. 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. Indoor

Table 1
Indoor and outdoor BTEX and MTBE concentrations ($\mu\text{g}/\text{m}^3$) by city.

Compound	MDL		Indoor				Outdoor				Indoor vs. Outdoor ^a	
	n	Mean	n	Mean	SD	Median	% > MDL	Mean	SD	Median		% > MDL
Los Angeles, CA	118		38									
Benzene		0.73		2.14	1.12	2.20	93	2.25	1.39	1.98	83	
Toluene		7.30		11.2	7.04	9.48	61	8.94	6.31	6.76	39	I*
Ethylbenzene		0.50		1.92	2.18	1.21	88	1.39	0.75	1.39	85	
m&p-Xylene		0.75		5.67	7.26	3.94	95	4.08	2.69	3.87	98	
o-Xylene		0.36		1.97	2.12	1.58	93	1.57	0.92	1.48	90	
MTBE		0.59		7.81	4.97	6.52	97	9.19	5.71	7.12	98	O*
Elizabeth, NJ	76		21									
Benzene		0.97		1.59	1.13	1.40	88	1.23	0.65	1.21	50	I*
Toluene		6.04		11.7	9.69	7.55	71	6.83	5.39	3.02	34	I**
Ethylbenzene		0.72		2.11	3.55	1.07	63	1.13	0.82	1.02	47	
m&p-Xylene		0.50		5.93	11.0	3.99	92	2.39	1.33	2.30	100	I**
o-Xylene		0.33		1.85	2.56	1.14	87	0.88	0.42	0.98	82	I**
MTBE		0.87		4.84	4.23	3.50	79	4.49	4.47	3.91	89	
Houston, TX	211		55									
Benzene		0.54		5.45	4.77	3.64	100	2.86	2.39	2.29	100	I**
Toluene		6.35		17.4	22.7	10.4	72	5.51	3.51	4.51	43	I**
Ethylbenzene		0.18		2.72	3.50	1.82	100	1.01	0.75	0.90	94	I**
m&p-Xylene		0.54		7.87	12.3	5.10	100	2.84	1.98	2.41	99	I**
o-Xylene		0.25		2.70	4.12	1.86	98	1.05	0.68	0.96	93	I**
MTBE		0.39		15.5	23.7	6.88	100	10.4	17.1	5.28	96	I*

Abbreviations: MDL, method detection limit; MTBE, methyl *tert*-butyl ether.

* $0.01 < p \leq 0.05$, ** $p \leq 0.01$.

^a I: indoor concentrations were statistically higher than outdoor concentrations; O: outdoor concentrations were statistically higher than indoor concentrations.

and outdoor concentrations for BTEX and MTBE in these three cities, and their respective MDLs, are summarized in Table 1. Toluene had the lowest percentage of indoor (61%) and outdoor (34%) concentrations greater than the MDL, likely because of high toluene background levels in the charcoal pads of the OVMS [21]. At least 79% of the indoor concentrations and 47% of the outdoor concentrations for the remaining compounds were above their respective MDLs, with the exception of indoor ethylbenzene concentrations in Elizabeth (63%).

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.

Statistical comparisons between C_{in} and C_{out} were used to examine source location. Table 1 indicates that in the Houston homes C_{in} was statistically higher than C_{out} for all VOCs. This suggests that sources were within or close to the living area, which was the case for 93% of the households that reported having a parked vehicle nearby during the study. Residences in Elizabeth had indoor and outdoor MTBE concentrations that were not statistically different; only 5% of these homes had cars. However, C_{in} for benzene, toluene and the xylenes were statistically higher than C_{out} , which implies that indoor sources for these VOCs were more important than ambient mobile sources. 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 indoor and outdoor concentrations for MTBE ($C_{out} > C_{in}$, $p \leq 0.05$) was different than that shown by benzene, ethylbenzene and the xylenes ($C_{out} \approx C_{in}$, $p \leq 0.05$). Possible explanations include house proximity to MTBE production facilities, interactions with liquid water on cooling coils or dehumidifiers (MTBE is far more water soluble than any of the BTEX compounds), or uncertainties in measurements. However, these results indicate that outdoor gasoline-related sources were driving indoor concentrations in LA homes for every contaminant but toluene,

even though 47% of the residences had a vehicle next to the occupied space. Discrepancies among cities in terms of the percentage of homes that had parked cars near the living area during the sampling period may explain why Houston generally had the highest median C_{in} values for all VOCs, whereas Elizabeth usually had the lowest concentrations.

Variations in ventilation rates (Table 2) 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). Low AERs limit the dilution of contaminants generated in attached garages or within the occupied space with fresh air. No associations were observed between the age of the house and ventilation rates; however, AER appeared to be affected by how households maintained acceptable indoor temperatures. The mean (median) percent of the sampling period in which households reported to have conditioned the air (P_{cond}) was 50% (50%) in Houston homes, while much lower values of 4.4% (0%) and 7.6% (0%) were observed in LA and EL, respectively. Conversely, the percent of monitoring time in which windows were reported to have been open (P_{window}) was much lower in Houston (mean = 11%, median = 0%) than

Table 2
Air exchange rates (h^{-1}) by city, and by building type and location of parked car.

Case	n	AER (h^{-1})		
		Mean	SD	Median
City				
Los Angeles, CA	38	1.45	1.11	1.06
Elizabeth, NJ	21	1.49	1.21	1.06
Houston, TX	55	0.68	0.60	0.48
Studied cases				
1: SFD home, car in attached garage	14	0.48	0.25	0.50
2: SFD home, car in detached garage	7	0.81	0.29	0.65
3: SFD home, car in carport	34	0.82	0.65	0.58
4: Manufactured home, car in carport	15	1.14	0.91	0.77
5: SFD home, no car in attached garage	8	1.30	1.36	0.71
6: SFD home, no car and no attached garage	36	1.56	1.27	1.03

Abbreviations: AER, air exchange rate; SFD, single-family detached.

in Elizabeth (mean = 18%, median = 0%) and Los Angeles (mean = 39%, median = 26%). Statistically significant correlations were observed between AER and P_{cond} in Houston ($r_s = -0.27$), and between AER and P_{window} in LA ($r_s = 0.62$) and HO ($r_s = 0.39$). Outdoor conditions likely influenced the behavior of the participants, and consequently, P_{cond} , P_{window} and AER in these cities. The median outdoor temperature for the homes that were visited once was the highest in Houston (26 °C, $n = 23$) followed by Los Angeles (19 °C, $n = 20$) and Elizabeth (11 °C, $n = 10$); about an equal number of residences participated in each season.

The influence of parked cars and ventilation rates on indoor concentrations of BTEX and MTBE was further evaluated. Because of the small sample sizes, data from LA, EL and HO were combined. Residences with vehicles next to the living area during the sampling period ($n = 70$) had indoor concentrations that were statistically higher than in homes without such sources ($n = 44$) for all VOCs but toluene and m&p-xylene. The ratio of median C_{in}

values in homes with and without cars ranged from 1.1 (m&p-xylene) to 2.0 (benzene).

The increase in indoor concentrations due to sources near the living quarters was estimated by subtracting C_{out} from C_{in} (ΔC). This increase was higher in homes with cars than in residences without automobiles for all pollutants; however, these results were not statistically significant for MTBE ($p = 0.12$). Variations in the proximity of parked vehicles may have influenced the MTBE results. Median ΔC values ranged from $-0.01 \mu\text{g}/\text{m}^3$ (MTBE) to $4.7 \mu\text{g}/\text{m}^3$ (toluene) when cars were present, and from $-0.37 \mu\text{g}/\text{m}^3$ (MTBE) to $0.71 \mu\text{g}/\text{m}^3$ (toluene) in homes without automobiles. Ventilation rates in homes with vehicles (median = 0.59 h^{-1}) were statistically lower than in residences without vehicles (median = 1.0 h^{-1}), which exacerbated the effect from the presence of nearby sources. Seventy-three percent of the homes with cars were in Houston, which explains their low AERs.

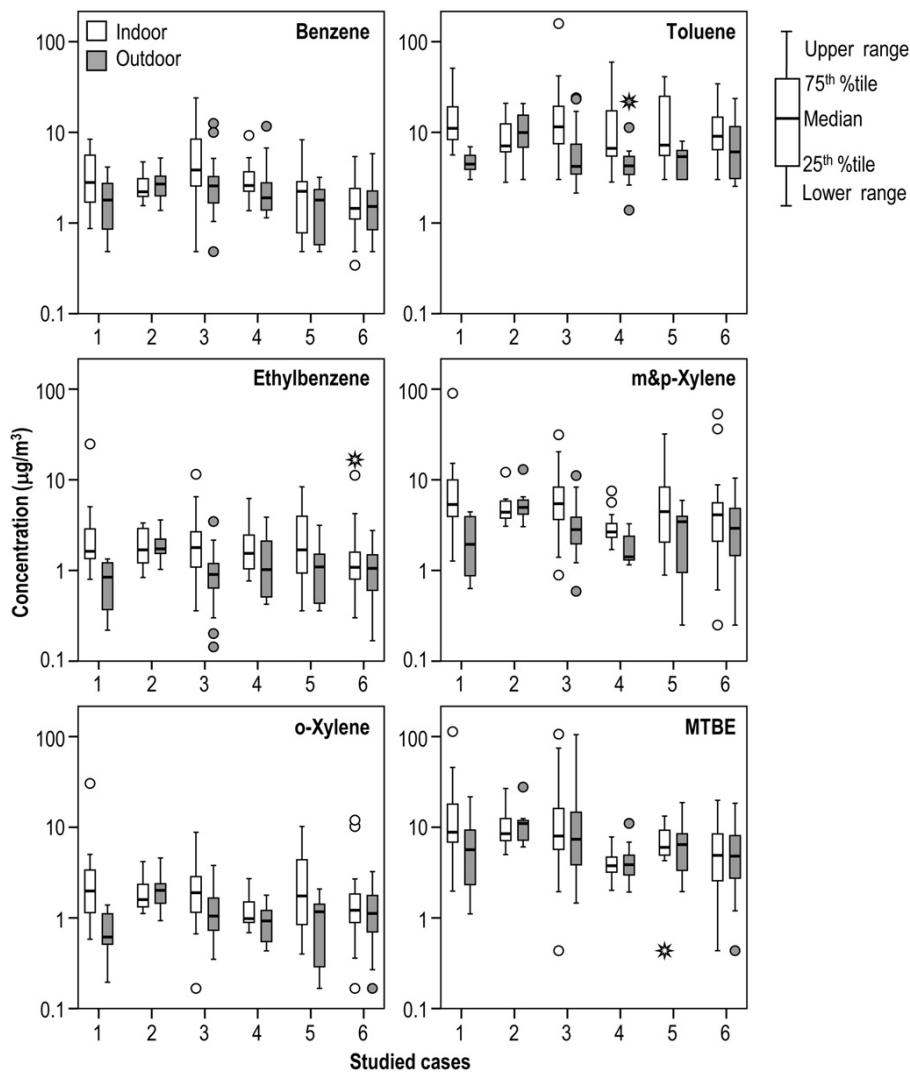


Fig. 1. Indoor and outdoor concentrations ($\mu\text{g}/\text{m}^3$) for six cases: single-family detached (SFD) homes with cars in the attached garage (Case 1; $n = 14$), detached garage (Case 2; $n = 7$), or adjacent carport (Case 3; $n = 34$); manufactured homes with cars in adjacent carports (Case 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; $n = 36$). 'o' and '*' indicate values between 1.5 and 3, and >3 box lengths, respectively, from the 25th or 75th percentiles.

The effect of source proximity was investigated by examining the six cases illustrated in Fig. 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; $n = 36$). Residences in cases 3 and 4 were not combined because their indoor concentrations were statistically different. In general, C_{in} for BTEX compounds and MTBE were statistically significantly correlated ($0.34 \leq r_s \leq 0.86$) in all of the studied cases, but in cases 4 and 5 where indoor BTEX concentrations appear to have been dominated by non-gasoline-related sources. Single-family detached homes with cars in attached garages were the only case where C_{in} was statistically higher than C_{out} for all pollutants. The SFD homes with cars in attached garages or carports had the highest median

C_{in} for all compounds. In contrast, households without both attached garages and vehicles had the lowest or the second lowest median C_{in} values for all VOCs but toluene. Indoor concentrations in homes with cars in attached garages were likely negatively affected by low house ventilation rates (median = 0.5 h^{-1}) as indicated in Table 2. Their low AERs were due to the fact that 10 out of these 14 homes were located in Houston. Residences with vehicles in detached garages were the only case where C_{in} and C_{out} were statistically similar for all contaminants, although the small sample size ($n = 7$) may have influenced the inability to detect a statistical difference.

Increase in indoor concentrations (relative to outdoors) for all compounds and studied cases are shown in Fig. 2. Single-family detached homes with vehicles in attached garages had the highest median ΔC for benzene ($1.2 \mu\text{g}/\text{m}^3$), toluene ($6.4 \mu\text{g}/\text{m}^3$), m&p-xylene ($2.6 \mu\text{g}/\text{m}^3$) and MTBE ($2.7 \mu\text{g}/\text{m}^3$), and relatively

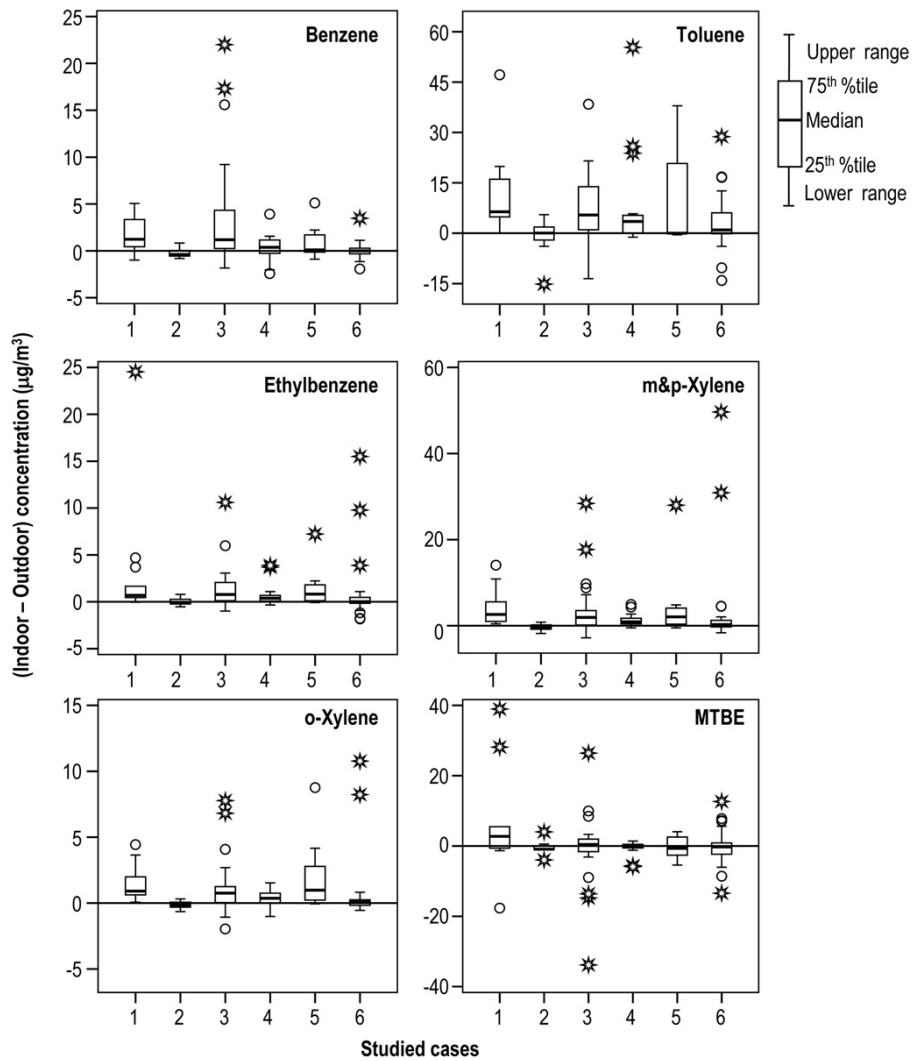


Fig. 2. Difference between indoor and outdoor concentrations ($\mu\text{g}/\text{m}^3$) for six cases: single-family detached (SFD) homes with cars in the attached garage (Case 1; $n = 14$; m&p-xylene = $89 \text{ mg}/\text{m}^3$, o-xylene = $30 \text{ mg}/\text{m}^3$ and MTBE = $109 \text{ mg}/\text{m}^3$ were not included for clarity), detached garage (Case 2; $n = 7$), or adjacent carport (Case 3; $n = 34$; toluene = $155 \text{ mg}/\text{m}^3$ and MTBE = $103 \text{ mg}/\text{m}^3$ were not included for clarity); 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; $n = 36$). 'o' and 'x' indicate values between 1.5 and 3, and >3 times the interquartile range, respectively, from the 25th or 75th percentiles.

large values for ethylbenzene ($0.69 \mu\text{g}/\text{m}^3$) and o-xylene ($0.91 \mu\text{g}/\text{m}^3$). These homes also had the highest median indoor to outdoor concentration ratios ($C_{\text{in}}/C_{\text{out}}$): 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 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 ($C_{\text{in}} < C_{\text{out}}$) for all compounds but toluene. Furthermore, these houses had the lowest median $C_{\text{in}}/C_{\text{out}}$ ratios for BTEX (0.89–1.0), and the same median ratios as homes without cars for MTBE (0.92).

Fig. 3 shows the cumulative distribution curve for the percent contribution of sources within or close to the occupied space to indoor concentrations (I_{cont}) for four of the six studied groups; cases 2 and 5 were omitted because of their small sample size. In general, I_{cont} for most compounds was the highest in residences with automobiles in attached garages; median values ranged from 30% (MTBE) to 58% (o-xylene). The SFD homes and manufactured homes with cars in carports had the next highest I_{cont} . Their median values were relatively similar, varying from 4% (MTBE) to 49% (ethylbenzene), and from 0% (MTBE) to 37% (both xylenes), respectively. Homes without both attached garages and cars typically had the lowest indoor contributions; the median ranged from 0% (MTBE) to 34% (toluene).

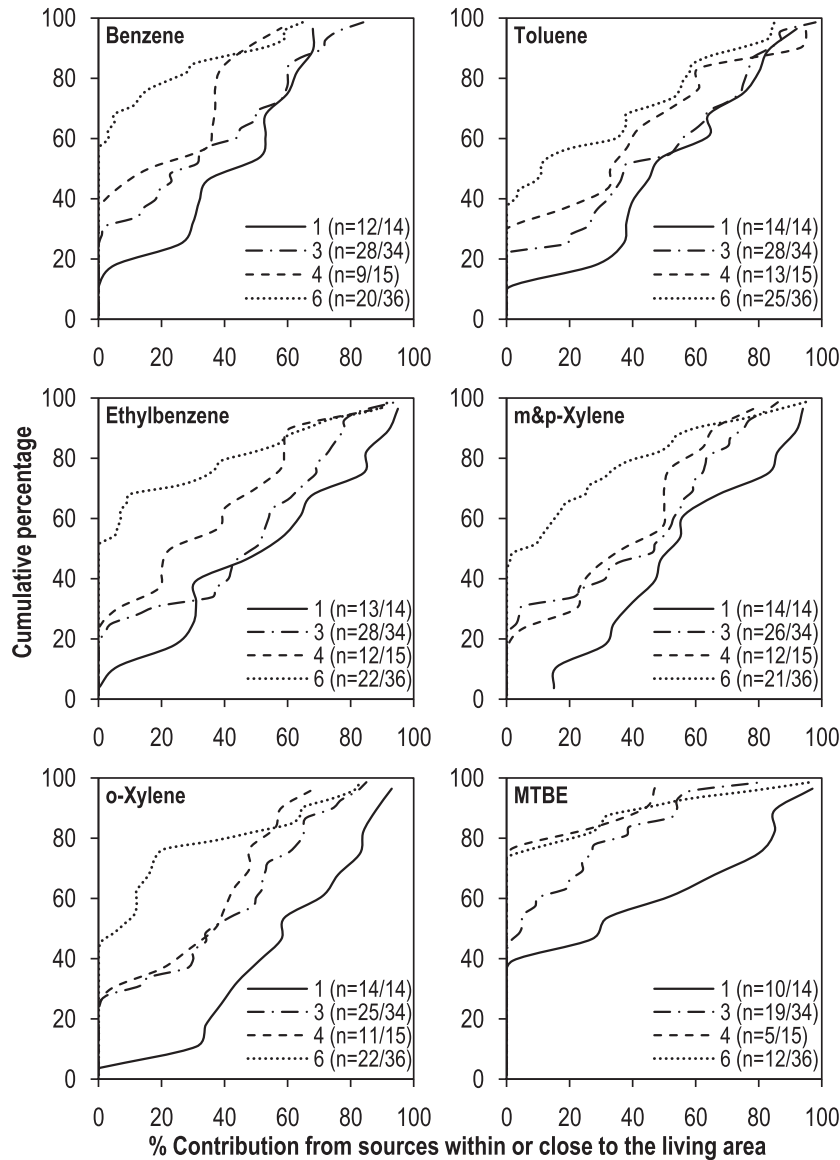


Fig. 3. Cumulative distribution functions for the percent contribution from indoor sources to indoor concentrations for four cases (n = number of homes where indoor concentrations were higher than outdoor concentrations/total number of homes.): single-family detached (SFD) homes with cars in the attached garage (Case 1), or adjacent carport (Case 3); manufactured homes with cars in adjacent carports (Case 4); and SFD homes without both attached garages and cars (Case 6). Cases 2 and 5 were excluded because of small sample size.

4. Discussion

Results from the RIOPA investigation and those of others [17,22] 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. Outdoor sources in urban areas contribute considerably to background concentrations of BTEX and MTBE; however, parked vehicles adjacent to residential living areas can exacerbate C_{in} . Batterman et al. [13], Dodson et al. [17], and Thomas et al. [14] monitored single-family homes and determined that BTEX and MTBE concentrations in attached garages can exceed C_{in} by an order of magnitude. Batterman et al. [13] estimated the median contribution from sources in the garage to indoor BTEX concentrations to range from 47% (toluene) to 65% (benzene) using field measurements and multi-zonal mass-balance models to approximate the airflow between these two areas. Dodson et al. [17] reported comparable median garage contributions (i.e., 30% for toluene to 44% for m&p-xylene) after following a similar procedure. Our results were also comparable, with I_{cont} ranging from 43% (benzene) to 58% (o-xylene) in SFD homes with vehicles in attached garages; however, our estimates do not distinguish between emissions within the living area or the garage. With regard to MTBE, our estimates for I_{cont} (median = 30%) were similar to those from Dodson et al. [17] (median = 32%). Variations that affected these results include the amount of air that infiltrated from the garage into the living space, house air exchange rates, and source strengths within the garage, living area and outdoors.

In addition to being emitted by mobile sources, ethylbenzene and xylene isomers tend to be concurrently emitted by paint-related products [11]. This may explain the high statistically significant correlations among indoor concentrations for these VOCs that we noted in homes without vehicles ($r_s > 0.79$), and that Jia et al. [23] observed using personal concentrations for the population at large ($r_s > 0.92$). Since toluene is found in a wider variety of consumer products, such as cleaners, paints, polishes and adhesives [11,12], homes without cars had indoor concentrations for toluene that had lower correlation coefficients with the other BTEX compounds ($r_s \cong 0.45$). However, these results may have also been affected by the low percentage of toluene measurements above the MDL.

We used the ratio of MTBE to benzene indoor concentrations to examine if these compounds originated from gasoline vapors or car exhaust. Low MTBE/benzene ratios indicate that tailpipe emissions are dominant because during combustion the amount of MTBE decreases while benzene is enriched due to toluene and xylene dealkylation [10]. Alternatively, high MTBE/benzene ratios 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. Investigators at the Desert Research Institute (DRI) [10] reported ratios for various microenvironments in Houston (freeway = 1.7–2.9, in-cabin underground garage = 2.4–3.2, in-cabin refueling = 25–42, and outdoor refueling = 29–56), car exhaust (Houston = 0.44–1.4, Los Angeles = 0.43–1.1), and liquid gasoline (Houston = 13–18, Los Angeles = 12–20).

Our estimates for MTBE/benzene ratios are shown in Fig. 4; we excluded nine houses that had indoor benzene measurements that were both below the MDL and less than $1 \mu\text{g}/\text{m}^3$. 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. Evaporative emissions were substantial in four households where MTBE/benzene values

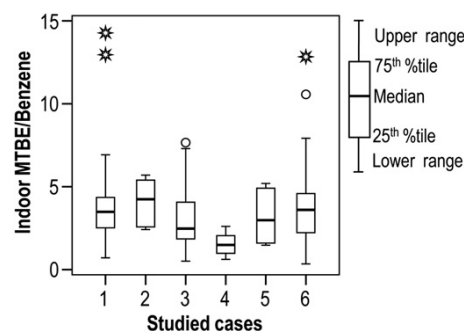


Fig. 4. Ratio of MTBE to benzene indoor concentrations for six cases: single-family detached (SFD) homes with cars in the attached garage (Case 1; $n = 14$), detached garage (Case 2; $n = 7$), or adjacent carport (Case 3; $n = 33$); manufactured homes with cars in carports (Case 4; $n = 15$); SFD homes with attached garages but no cars (Case 5; $n = 6$); and SFD homes without both attached garages and cars (Case 6; $n = 30$). Nine homes were excluded where indoor benzene concentrations were both lower than the MDL and less than $1 \mu\text{g}/\text{m}^3$. ‘o’ and ‘+’ indicate values between 1.5 and 3, and > 3 times the interquartile range, respectively, from the 25th or 75th percentiles.

were greater than 11. Two of these were SFD homes with cars in the attached garage. The other two residences did not have cars next to the living area; we speculate that these participants failed to report the presence of indoor gasoline sources given that indoor MTBE concentrations were greater than C_{out} by at least $7.8 \mu\text{g}/\text{m}^3$.

Since air pollutants emitted by mobile sources are commonly assessed by governmental agencies using data from fixed stations, we examined how measurements from RIOPA-TX compared to those from such stations. We obtained ambient concentrations for these compounds from the Texas Commission on Environmental Quality (TCEQ), which operated one monitoring station near the sampled homes using canisters and reported time-averaged 24-h concentrations that were collected every six days [24]. We paired data from RIOPA with the TCEQ concentration measured the week before the home was sampled. Outdoor concentrations for ethylbenzene, toluene and m&p-xylene from RIOPA were statistically higher than those reported by TCEQ, probably because of sources near the residences that were not detected by the fixed monitoring site or because of differences in meteorology between the sample sites [25]. Only outdoor o-xylene concentrations from TCEQ were statistically higher than those from RIOPA. Indoor BTEX and MTBE concentrations from the RIOPA homes were statistically higher than ambient levels from TCEQ. For benzene and MTBE, residences with cars in attached garages ($n = 10$) had median indoor concentrations (benzene = 3.8 , MTBE = $11 \mu\text{g}/\text{m}^3$) that showed the highest discrepancy from their respective median TCEQ values (benzene = 2.4 , MTBE = $6.3 \mu\text{g}/\text{m}^3$).

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 parked in attached garages and cars driven on a freeway with heavy traffic during commute to and from work. We selected Houston because 71% of the RIOPA homes that correspond to the first microenvironment were located in this city. We calculated exposure by multiplying concentration by the exposure time. For homes, we used the mean ΔC for benzene ($2.3 \mu\text{g}/\text{m}^3$) to better evaluate the effect of nearby sources and assumed that individuals spend 70% of the week in their house [9]. These sources were likely dominated by parked vehicles because there were not tobacco smokers and other gasoline-powered devices in the homes. For cars, we assumed a mean in-cabin concentration of $6.1 \mu\text{g}/\text{m}^3$ [10], an average commute time to work of 26 min [26], a mean travel time from work equal to the commute time to work, and a five-day work

week. Weekly exposure to benzene was $270 \mu\text{g}/\text{m}^3 \times \text{h}$ in homes with cars in attached garages, and $26 \mu\text{g}/\text{m}^3 \times \text{h}$ in cars during commute to and from work. These results indicate that even though increases in indoor concentrations due to vehicles in attached garages are relatively small, the fact that we spend a large amount of time in our homes can lead to exposures to benzene that are ten times higher than what we experience in more severe microenvironments that we typically frequent such as heavily congested highways. Additionally, 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 ($7.8 \times 10^{-6} \text{ m}^3/\mu\text{g}$) [6]. The estimated risk would be much greater if we had calculated the cumulative effect of cancer-related air contaminants that are found in vehicle emissions (e.g., 1,3-butadiene, formaldehyde and acetaldehyde). The EPA benchmark for exposure to potential carcinogens is 1 per million.

Methods to reduce indoor residential concentrations of VOCs emitted by parked vehicles next to the living quarters need special attention because 55% of single-family homes and manufactured homes in the U.S. have an attached garage or carport [27]. ASHRAE Standard 62.2 [28] describes measures to prevent the migration of pollutants from attached garages into the occupied area in new housing, although these are also applicable to existing residences. These recommendations 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, especially when located in the garage; and (4) maintaining the living area at a higher pressure than that of the garage. It is not evident that carports are a good alternative to attached garages; SFD homes with cars in carports had relatively high median ΔC values (e.g., benzene = $1.2 \mu\text{g}/\text{m}^3$, MTBE = $0.42 \mu\text{g}/\text{m}^3$). Conversely, manufactured homes with vehicles in carports had much lower median ΔC values (e.g., benzene = $0.38 \mu\text{g}/\text{m}^3$, MTBE = $-0.10 \mu\text{g}/\text{m}^3$). Various factors could have affected these results such as the location of windows and doors with respect to the carport, the number of parked cars, and meteorological conditions. Infiltration of pollutants into the living quarters can be limited by tightening the house envelope, specifically close to the carport. In addition to the measures just described, the design of new residences could be improved by incorporating detached garages. Our results indicate that homes with detached garages had minimal increases in indoor concentrations of BTEX and MTBE, which suggests that in addition to cars, non-gasoline-related sources for BTEX may have been stored in the detached garage where they were not as likely to affect the occupied space.

Our assessment provides evidence that gasoline-related VOCs from parked cars next to residential living areas, particularly in attached garages, can deteriorate indoor air quality. However, further research is needed where there is more control over the houses that are examined. Our evaluation involved an uneven number of residences for the six cases we examined because we were restricted by the information reported in the RIOPA dataset. Furthermore, this research should be accompanied by estimations of airflow rates from attached garages into the occupied space to quantify the contribution of parked vehicles to indoor concentrations of contaminants.

5. 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 of six parking cases indicate that homes with attached garages were affected the most by cars. The effect from the presence of nearby sources was exacerbated by the fact that most of the homes with cars in attached garages were located in Houston; these residences tended to have low ventilation rates because of regional climatic conditions. The median percent contribution of sources within or close to the living area to C_{in} in these residences ranged from 30 to 58%. Moreover, houses with attached garages generally had the highest median increases in indoor concentrations (relative to outdoor concentrations) for BTEX compounds ($0.69 \leq \Delta C \leq 6.4 \mu\text{g}/\text{m}^3$). 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 cancer estimates that are 17 times higher than the EPA benchmark of one per million. Strategies to reduce exposure to gasoline-related VOCs in homes include sealing surfaces shared by the living quarters and the garage, and not placing components of the air conditioning system in the garage. Furthermore, our results suggest that improving the design of homes by incorporating detached garages could be an alternative solution.

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References

- [1] Hun DE, Siegel JA, Morandi MT, Stock TH, Corsi RL. Cancer risk disparities between Hispanic and non-Hispanic white populations: the role of exposure to indoor air pollution. *Environ Health Perspect* 2009;117(12):1925–31.
- [2] Loh MM, Levy JJ, Spengler JD, Houseman EA, Bennett DH. Ranking cancer risks of organic hazardous air pollutants in the United States. *Environ Health Perspect* 2007;115(8):1160–8.
- [3] Payne-Sturges DC, Burke TA, Breyse P, Diener-West M, Buckley TJ. Personal exposure meets risk assessment: a comparison of measured and modeled exposures and risks in an urban community. *Environ Health Perspect* 2004;112(5):589–98.
- [4] Sax SN, Bennett DH, Chillrud SN, Ross J, Kinney PL, Spengler JD. A cancer risk assessment of inner-city teenagers living in New York City and Los Angeles. *Environ Health Perspect* 2006;114(10):1558–66.
- [5] CalEPA. Air toxics hot spots program risk assessment guidelines. Part II: technical support document for the describing available cancer potency factors. Berkeley, CA: California Environmental Protection Agency (CalEPA), Office of Environmental Health Hazard Assessment (OEHHA), Air Toxicology and Epidemiology Section, http://oehha.ca.gov/air/hot_spots/2009/TSDCancerPotency.pdf; 2009 [accessed 09.11.09].
- [6] U.S. EPA. Integrated Risk Information System (IRIS). U.S. Environmental Protection Agency (U.S. EPA), <http://cfpub.epa.gov/ncea/iris/index.cfm>; 2005 [accessed 31.12.08].
- [7] Sexton K, Mongin SJ, Adgate JL, Pratt GC, Ramachandran G, Stock TH, et al. Estimating volatile organic compound concentrations in selected microenvironments using time-activity and personal exposure data. *J Toxicol Environ Health* 2007;70:465–76.
- [8] Symanski E, Stock TH, Tee PG, Chan W. Demographic, residential, and behavioral determinants of elevated exposures to benzene, toluene, ethylbenzene, and xylenes among the U.S. population: results from 1999–2000 NHANES. *J Toxicol Environ Health* 2009;72:903–12.
- [9] Klepeis NE, Nelson WC, Ott WR, Robinson JP, Tsang AM, Switzer P, et al. The National Human Activity Pattern Survey (NHAPS): a resource for assessing exposure to environmental pollutants. *J Expo Anal Environ Epidemiol* 2001;11:231–52.
- [10] DRI. Section 211(B) Tier 2 high end exposure study of conventional and oxygenated gasoline. Reno, NV: Desert Research Institute (DRI); 2006.
- [11] Sack TM, Steele DH, Hammerstrom K, Remmers J. A survey of household products for volatile organic compounds. *Atmos Environ* 1992;26A(6):1063–70.
- [12] Nazaroff WW, Weschler CJ. Cleaning products and air fresheners: exposure to primary and secondary air pollutants. *Atmos Environ* 2004;38:2841–65.
- [13] Batterman S, Jia C, Hatzivasilis G. Migration of volatile organic compounds from attached garages to residences: a major exposure source. *Environ Res* 2007;104(2):224–40.

- [14] Thomas KW, Pellizzari ED, Clayton CA, Perritt RL, Dietz RN, Goodrich RW, et al. Temporal variability of benzene exposures for residents in several New Jersey homes with attached garages or tobacco smoke. *J Expo Anal Environ Epidemiol* 1993;3(1):49–73.
- [15] Emmerich SJ, Gorfain JE, Howard-Reed C. Air and pollutant transport from attached garages to residential living spaces – literature review and field tests. *Int J Ventil* 2003;2(3):265–76.
- [16] Gammage RB, White DA, Gupta KC. Residential measurements of high volatility organics and their sources. In: Proceedings from the 3rd international conference on indoor air quality and climate – indoor air 1984. Stockholm, Sweden; 1984. p. 157–162.
- [17] Dodson RE, Levy JI, Spengler JD, Shine JP, Bennett DH. Influence of basements, garages, and common hallways on indoor residential volatile organic compound concentrations. *Atmos Environ* 2008;42:1569–81.
- [18] Graham LA, Noseworthy L. Contribution of vehicle emissions from an attached garage to residential indoor air pollution levels. *J Air Waste Manage Assoc* 2004;54:563–84.
- [19] HEI. Relationships of Indoor, Outdoor, and Personal Air (RIOPA) database. Boston, MA: Health Effects Institute (HEI), <http://riopa.aer.com>; 2008 [accessed 15.11.08].
- [20] Weisel CP, Zhang J, Turpin BJ, Morandi MT, Colome S, Stock TH, et al. Relationships of Indoor, Outdoor, and Personal Air (RIOPA): part I. Collection methods and descriptive analyses. HEI Report No. 130 (Pt. 1). Boston, MA: Health Effects Institute; 2005. NUATRC Report No. 7, Houston, TX: National Urban Air Toxics Research Center.
- [21] Chung CW, Morandi MT, Stock TH, Afshar M. Evaluation of a passive sampler for volatile organic compounds at ppb concentrations, varying temperatures, and humidities with 24-hr exposures: 2. Sampler performance. *Environ Sci Technol* 1999;33:3666–71.
- [22] Sax SN, Bennett DH, Chillrud SN, Kinney PL, Spengler JD. Differences in source emission rates of volatile organic compounds in inner-city residences of New York City and Los Angeles. *J Expo Anal Environ Epidemiol* 2004;14:S95–109.
- [23] Jia C, D'Souza J, Batterman S. Distributions of personal VOC exposures: a population-based analysis. *Environ Int* 2009;34:922–31.
- [24] TCEQ. Monitoring operations division, monitoring data management & analysis section. Austin, TX: Texas Commission on Environmental Quality (TCEQ); 2003.
- [25] Smith LA, Stock TH, Chung KC, Mukerjee S, Liao XL, Stallings C, et al. Spatial analysis of volatile organic compounds from a community-based air toxics monitoring network in Deer Park, Texas. USA. *Environ Monit Assess* 2007;128:369–79.
- [26] U.S. Census Bureau, http://www.census.gov/Press-Release/www/releases/archives/american_community_survey_acs/004489.html; 2009 [accessed 01.12.09].
- [27] U.S. EIA. Residential energy consumption survey, 2005 housing characteristics tables. U.S. Energy Information Administration (U.S. EIA), http://www.eia.doe.gov/emeu/recs/recs2005/hc2005_tables/hc2owneroccupied/pdf/tablehc2.2.pdf; 2005 [accessed 11.01.10].
- [28] ASHRAE. Standard 62.2-2007. Ventilation and acceptable indoor air quality in low-rise residential buildings. Atlanta, GA: American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE); 2007.