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Methyl-Tertiary Butyl Ether(MTBE) and BTEX Inside and Outside Apartments with Different Construction Age

Wan–Kuen Jo and Jong–Hyo Lee*

Department of Environmental Engineering, Kyungpook National University, Daegu 702-701, Korea
**Division of Environmental Management, Kumho Petroleum Co., Yeosoo, Korea*

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Abstract

Only limited information is available on the measured exposure levels of residents according to the construction age of apartments. As such, present study was conducted to measure and to compare the bedroom, living-room, and outdoor air levels of MTBE and benzene, toluene, ethyl benzene and m,p-xylene(BTEX) in both newer and older apartments. For both newer and older apartments, all the compounds except for MTBE showed significantly higher levels in bedrooms or living-rooms as compared to the outdoor concentrations. The ratio of bedroom or living-room median concentration to outdoor concentration was close to 1 for MTBE, whereas it was larger than 1 for other target compounds. It was also found that the bedroom and living-room appeared to have similar indoor sources and sinks for BTEX, but not for MTBE. The median concentration ratios of the newer apartments to the older apartments ranged from 1.63 to 1.81, depending upon the compounds. In contrast, the MTBE concentrations did not differ significantly between the newer and older apartments, thereby suggesting that although newer buildings could emit more VOCs, this is not applicable to all VOCs. Conclusively, the findings of present study should be considered, when designing exposure studies associated with VOC emissions in buildings and/or managing indoor air quality according to construction age of buildings.

Key Words : Newer, Older, Bedroom, Living-room, Outdoor

1. Introduction

Exposure to volatile organic compounds(VOCs) has been a subject of concern in urban areas because of the prevalence of these compounds in both indoor and outdoor environments and because of their adverse health effects. In particular, aromatic VOCs such as benzene, toluene, ethyl benzene and xylene(BTEX) have been detected at high concentration levels in urban atmosphere[1~3]. The atmospheric BTEX can penetrate indoors, thereby influencing the indoor concentration levels[4]. In addition, there are various indoor sources of BTEX and other VOCs such as building materials and consumer products[5~7]. BTEX are toxic(e.g. benzene) or potentially toxic to humans(e.g. toluene, ethyl benzene, and xylene)[8,9].

Meanwhile, MTBE is added to gasoline to reduce the motor vehicle exhaust emission of carbon monoxide(CO) and increase octane ratings[10~13]. According to the Korean Petroleum Corporation, the gasoline manufactured by all five Korean petroleum companies contains 6 to 8% MTBE. However, the addition of MTBE can elevate ambient and indoor MTBE levels, thereby increasing personal exposure to this compound[14~16]. Exposure to this compound is of particular concern because of its toxicity and is potentially linked to ailments such as headaches, dizziness, irritated eyes and nausea[17~19]. Moreover, MTBE may also be acutely

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Corresponding Author : Wan-Kuen Jo, Department of Environmental Engineering, Kyungpook National University, Daegu 702-701, Korea
Phone: +82-53-950-6584
E-mail: wkjo@knu.ac.kr

toxic[19~21], chronic noncancer toxic[17,22], and carcinogenic[18]. However, the U.S. Environmental Protection Agency(EPA) continues to maintain that MTBE is a safe and effective oxygenate addition to fuels, based on previous test results of toxicological and epidemiological studies on the health effects of MTBE. This controversy surrounding the issue thus implies the need for further studies on MTBE exposure and its related health effects. These characteristics of MTBE and BTEX warrant the measurement of these pollutants to supplement exposure data that can be linked to health risk.

In Korea, many city residents live in apartment buildings. According to the Korea National Statistical Office, about 22 million people live in the eight largest cities and about half of them live in apartments. Several VOCs have been suspected to cause "sick-building syndrome" such as headache, eye irritation, unusual fatigue, nausea, and asthmatic symptoms[23~25]. In particular, it was well known that newer building materials emitted more VOCs as compared to older ones[26,27]. As such, it can be expected that residents living in newer apartments be exposed to more VOCs those living in older apartments. Nevertheless, limited information only is available to confirm this hypothesis. Even the hypothesis was confirmed, information on the measured exposure levels, not on the estimated levels, for both apartment residents is still required to better link exposure levels to health effects. Consequently, the current study was conducted to measure and to compare the indoor and outdoor air levels of MTBE and BTEX in both newer and older apartments.

2. Materials and Methods

2.1. Experiment protocol

This study measured the concentrations of MTBE and BTEX in the outdoor and indoor air of apartments according to construction age. Fifty-six homes in 15 apartment buildings that satisfied the experimental criteria were approached for permission to measure the air levels inside their apartments. The 56 homes consisted of 31 newer apartments(constructed within 6 months) and 25 older apartments(constructed between 12 and 18 months before). The criteria were as follows: the apartment buildings should be high-rise apartment buildings with 10 or more stories; the buildings should be located at least 100 meters away from any gas stations so as to minimize the impact of service stations on the apartment ambient VOC levels; and the exteriors of apartments should not have been painted within at least 1 year before the study. Only residents of 12 homes(5 newer apartments and 7 older apartments) in 5 apartment buildings granted permission to measure the air levels. Finally, five-newer and five-older apartments were surveyed in the present study. None of the residents smoked during the sampling periods. The apartments were constructed with concrete and iron frames. All the apartments used liquid petroleum gas(LPG) as their primary heating system. LPG was also used for cooking.

One newer apartment and one older apartment were concurrently surveyed. One 8-hour outdoor air samples were collected outside the apartment porches(between 8 a.m. and 8 p.m.). Concurrently, one indoor air samples were collected in both a living-room and a bedroom. A constant-flow sampling pump was connected to a 1/4" stainless steel trap containing 0.3 g of Tenax and 0.4 g of Carboxen 569 adsorbent. The sampling pump was calibrated by a mass flow meter prior to and following the collection of each sample. The average of these two rates was used as the sample flow rate in all the volume calculations. No samples departed by more than 10% from the initial flow rate in the current study. A flow rate of about 20 mL/min was set for the current study. The sample flow rate was determined based on the relative expected concentrations for each experimental condition.

MTBE and BTEX trapped on the Tenax TA/Carboxen 569 trap were analyzed by coupling a thermal desorption system(TDS, Tekmar Model Aerotrap 6000) to a gas chromatography(GC, Varian 3400CX) with a flame ionization detector(FID) using a

0.53-mm-ID by 60-m-length and 5.0 μm phase film thickness SPB-5 capillary column(Supelco Co.). The adsorbent trap was thermally desorbed at 250°C for 10 minutes, and the target compounds cryo-focussed at -120°C on a cryo trap. The cold trap was rapidly heated to 250°C and flushed to the Cryo-focusing Module(CM) of the Aerotrap system cooled down to -120°C to refocus the target compounds. Then the CM was heated to 225°C and flushed to transfer the target compounds to GC. The initial oven temperature of the GC was set to 35°C for 5 minutes and ramped at 4°C/min to 200°C for 5 minutes. A preliminary study confirmed that there were not co-eluting substances for the target compound analysis. The quantitative analysis of the target compounds was performed by using the calibration curves of a minimum of five concentrations. The concentrations were prepared by injecting 11.1 to 1110 ng of MTBE standard prepared in water and 10.5 to 1050 ng of each of BTEX prepared in methanol into a flash evaporation system(FES) to transfer the MTBE and BTEX to a trap. The injected amounts were within the operational range for analysis.

2.2. Quality assurance/quality control

The quality control/quality assurance for MTBE and BTEX measurements included laboratory and field blank traps, spiked samples, and duplicate measurements of integrated samples. At the beginning of the day, a laboratory blank trap and a field blank trap were analyzed to check trap contamination. No trap contamination was identified in any traps. A known amount of an MTBE standard prepared in water and of a BTEX standard prepared in methanol were directly injected into a trap to transfer the target compounds to the GC through the TDS in order to check the quantitative response. When the quantitative response differed more than ± 25% from that predicted by a specified calibration equation, a new calibration equation was determined. Seven sampling traps spiked with 11.1 ng of MTBE and 10.5 ng of BTEX standards using a FES were used to determine the method detection limits(MDLs) of the system. The MDLs determined were 0.8 to 2.1 μg/m^3 for the target VOCs.

3. Results and Discussion

3.1. Indoor and outdoor concentrations

The concentrations of six target compounds measured in bedroom, living-room, and outdoor air of newer and older apartments are summarized in Tables 1 and 2, respectively. Since the distributions of all the results for the indoor and outdoor air concentrations were right-skewed with higher arithmetic means than median values, it would seem that the data were log-normally distributed. However, a statistical test of normality(Shapiro-Wilk statistics) did not indicate whether the data were normally or log-normally distributed. Toluene was the most abundant VOC in both indoor and outdoor air. The indoor air levels of the target compounds were compared to the matched concurrent out-

Table 1. Summary of concentrations (μg/m^3) of MTBE and BTEX in bedrooms, living-rooms, and outdoor of newer apartments

Compound	Bedroom			Living-room			Outdoor		
	Median	Mean	Range	Median	Mean	Range	Median	Mean	Range
MTBE	4.3	4.6	0.8-7.7	4.5	4.7	0.5-8.1	4.1	4.4	0.7-8.0
Benzene	10.3	13.3	5.7-28.3	8.7	9.9	1.1-23.6	2.5	2.9	0.6-4.3
Toluene	56.8	63.5	18.5-132	44.9	50.8	15.4-113	19.2	23.7	2.9-37.8
Ethyl benzene	6.3	6.8	1.3-9.4	3.7	3.8	0.6-6.1	1.7	1.8	0.7-3.1
m,p-Xylene	9.2	9.7	1.7-21.7	7.8	8.1	1.2-17.3	3.3	3.5	0.8-5.4

Note. Number of samples: N = 30 for each environment.

door air levels using the nonparametric Wilcoxon test. For both newer and older apartments, all the compounds except for MTBE showed significantly higher levels in bedrooms or living-rooms as compared to the outdoor concentrations. For instances, the median values for the bedroom samples, which were collected in newer apartments, ranged from 6.3 to 56.8 $\mu g/m^3$, and those for living-room samples ranged from 3.7 to 44.9 $\mu g/m^3$, while those for the outdoor samples ranged from 1.7 to 19.2 $\mu g/m^3$, depending upon compounds. The higher indoor levels compared to the outdoor levels indicate that the sources of the target compounds were present inside the homes. This result indicates that the strength of potential indoor sources outweighed that of outdoor source for the indoor levels of target compounds. Major sources of the target compounds inside the home include cigarette smoke, paint, solvents, paint thinners, and combustion sources$^{1,5)}$. For this study, however, combustion processes are not significant sources of target compounds since any cigarette smoking was not observed during the experimental periods. In addition, the fuels used in kerosene, liquid propane gas(LPG), butane gas, and/or electric heaters and stoves, according to the Korean Petroleum Association, did not contain any of

Table 2. Summary of concentrations ($\mu g/m^3$) of MTBE and BTEX in bedrooms, living-rooms, and outdoor of older apartments

Compound	Bedroom			Living-room			Outdoor		
	Median	Mean	Range	Median	Mean	Range	Median	Mean	Range
MTBE	4.5	4.7	0.6-8.3	4.1	4.6	0.7-7.5	4.2	4.5	0.6-8.3
Benzene	5.8	6.0	1.5-8.7	5.1	5.3	1.3-8.2	2.8	3.1	0.7-4.7
Toluene	33.1	34.0	9.7-89	25.4	26.8	8.7-69	21.7	25.4	3.8-41.2
Ethyl benzene	3.5	3.9	0.7-5.6	2.7	2.8	0.6-5.1	1.5	1.6	0.6-2.7
m,p-Xylene	5.4	5.6	0.9-9.3	4.1	4.5	0.7-8.6	2.9	3.3	0.7-4.6

Note. Number of samples: N = 30 for each environment.

Table 3. Correlation of concentrations of target compounds between bedroom, living-room and outdoor air of newer apartments

Compound	Bed-Living		Bed-Outdoor		Living-Outdoor	
	R^2	*P*-value	R^2	*P*-value	R^2	P-value
MTBE	0.78	<0.0001	0.83	<0.0001	0.85	<0.0001
Benzene	0.55	<0.005	0.03	<0.63	0.05	<0.53
Toluene	0.68	<0.0001	0.15	<0.31	0.12	<0.26
Ethyl benzene	0.47	<0.01	0.02	<0.71	0.11	<0.22
m,p-Xylene	0.44	<0.05	0.07	<0.42	0.05	<0.49

Table 4. Correlation of concentrations of target compounds between bedroom, living-room and outdoor air of older apartments

Compound	Bed-Living		Bed-Outdoor		Living-Outdoor	
	R^2	*P*-value	R^2	*P*-value	R^2	P-value
MTBE	0.71	<0.0001	0.68	<0.0001	0.74	<0.0001
Benzene	0.48	<0.01	0.13	<0.29	0.25	<0.11
Toluene	0.63	<0.0001	0.11	<0.23	0.08	<0.56
Ethyl benzene	0.37	<0.05	0.04	<0.65	0.14	<0.15
m,p-Xylene	0.41	<0.05	0.09	<0.51	0.07	<0.53

Note. R^2 adjusted R-square coefficient *P*-value, statistical significance.

the target compounds. The other known major sources of target compounds were not observed in any of the homes. The presence of indoor sources for the target compounds are further supported by the statistically significant relationship between bedroom and living-room concentrations. The adjusted R^2 of BTEX ranged from 0.44 to 0.55 for newer apartment(Table 3), and it ranged from 0.37 to 0.63 for older apartments(Table 4). This relationship indicates that the bedroom and living-room appeared to have similar indoor sources and sinks for BTEX[28)]. In contrast, the adjusted R^2 of BTEX ranged from 0.02 to 0.15 and 0.05 to 0.12 for the relationships between bedroom and outdoor concentrations and between living-room and outdoor concentrations of newer apartments, respectively, and it ranged from 0.04 to 0.13 and 0.07 to 0.25 for those of older apartments. These relationships were not statistically significant, suggesting that the sources and sink mechanisms for BTEX are different between bedroom or living-room and outdoor air[28)].

It is noteworthy that the bedroom or living-room concentrations of MTBE were similar to the outdoor concentrations. The ratio of bedroom or living-room median concentration to outdoor concentration was close to 1 for MTBE, whereas it was larger than 1 for other target compounds(Table 5). This finding suggests that there would not be any potential indoor source for MTBE. Moreover, this suggestion is supported by previous studies[13~15)], which reported that the major source for MTBE is motor vehicle emissions- major outdoor source- in most urban areas. Meanwhile, for MTBE, both the relationship between bedroom and living-room concentrations and between bedroom or living-room and outdoor air concentrations were statistically significant, regardless of the construction age of apartments, with the adjusted R^2 of between 0.78 and 0.85 for newer apartment and between 0.68 and 0.74 for older apartments. This high relationship indicates that there would be a same source and sink mechanism for MTBE in bedroom, living-room, and outdoor air. The most potential source for MTBE in urban areas is known as motor vehicle emissions[14~16)].

Table 5 also allows the comparison of bedroom and living-room concentrations for the target compounds. Regardless of the construction age of apartments, for BTEX the ratios of bedroom concentrations to liv-

Table 5. Ratio of bedroom to living-room median concentrations, and ratio of bedroom or living-room median concentrations to outdoor median concentrations in newer and older apartments

Compound	Newer apartment			Older apartment		
	Bed/Living	Bed/Out	Living/Out	Bed/Living	Bed/Out	Living/Out
MTBE	0.96	1.05	1.09	1.09	1.07	0.98
Benzene	1.18	4.12	3.48	1.14	2.07	1.82
Toluene	1.27	2.96	2.33	1.30	1.53	1.17
Ethyl benzene	1.70	3.70	2.18	1.25	2.33	1.80
m,p-Xylene	1.14	2.79	2.36	1.20	1.86	1.41

Table 6. Median concentrations ($\mu g/m^3$) of MTBE and BTEX inside apartments according to construction age

Compound	Newer	Older	Newer/Older	*P*-value
MTBE	4.5	4.3	1.05	<0.95
Benzene	9.8	5.4	1.81	<0.001
Toluene	52.3	29.5	1.77	<0.001
Ethyl benzene	4.9	3.0	1.63	<0.01
m,p-Xylene	8.3	4.6	1.80	<0.001

Note. Number of samples: N = 60 for each of the two types of apartments; *P*-value, statistical significance.

Table 7. Correlation matrix of target compounds in newer and older apartments

Apartment	Compound	MTBE	Benzene	Toluene	Ethyl benzene	m,p-Xylene
Newer	MTBE	1.00				
	Benzene	0.08				
	Toluene	0.12	0.69**			
	Ethyl benzene	0.18	0.47*	0.70**		
	m,p-Xylene	0.10	0.53*	0.47*	0.42*	1.00
Older	MTBE	1.00				
	Benzene	0.05				
	Toluene	0.09	0.25			
	Ethyl benzene	0.14	0.22	0.17		
	m,p-Xylene	0.17	0.11	0.09	0.21	1.00

Note. Statistical significance: *, 0.05, **, 0.005.

ing-room concentrations were larger than 1, whereas the ratio of MTBE was close to 1. The higher BTEX concentrations in bedrooms are likely due to the relatively less space volume of bedrooms. Plus, the use of apartment doors which are more closely located to living-rooms might dilute living-room air. In contrast to BTEX, since the major source for MTBE is motor vehicle emissions and there are no potential indoor sources[1,14,28)], the penetration of ambient MTBE into the interiors of apartments would not cause any significant difference between bedroom and living-room concentrations.

3.2. Newer apartments vs. older apartments

Table 6 presents the median concentrations of MTBE and BTEX inside apartments according to construction age. The median values were obtained by combining the bedroom and living-room concentrations for both the newer and older apartments. As expected, the BTEX concentrations measured in newer apartments were significantly higher than that of older apartments. The median concentration ratios of the newer apartments to the older apartments ranged from 1.63 to 1.81, depending upon the compounds. In contrast, the MTBE concentrations did not differ significantly between the newer and older apartments. The BTEX concentration differences between the newer and lower apartments would mainly be attributed to the emission rate of indoor sources such as bulding finishing materials and furniture[1,6,7,26,27)]. The present finding is consistent with the result of Järnström et al.'s study[26)], which have reported that the VOC concentrations inside new finished buildings decreased as time passed. For MTBE, since the bedroom and living-room levels are not mainly influenced by indoor sources, but outdoor motor vehicle emission source[14~16)], there would be no significant concentration difference between newer and older apartments.

Meanwhile, more strong correlation between target compounds were observed for certain target compounds in newer apartments as compared older apartments (Table 7). MTBE did not have any significant correlation with BTEX, regardless of the construction age of apartments. However, BTEX had significant correlation among themselves in newer apartments. Although BTEX showed some correlation among themselves in older apartments, the correlations were not statistically significant. As such, stronger concentration correlations between BTEX appear to occur under the conditions of higher emission rates in newer apartments. However, future studies are recommended to confirm this assertion, since we could not get all information necessary for it.

4. Conclusions

The current study measured and compared the indoor and outdoor air levels of MTBE and BTEX in both newer and older apartments. For both newer and older apartments, all the compounds except for MTBE showed significantly higher levels in bedrooms or living-rooms as compared to the outdoor concentrations. This result indicates that the strength of potential indoor sources outweighed that of outdoor source for the indoor levels of target compounds. It was found that the bedroom and living-room appeared to have similar indoor sources and sinks for BTEX, but not for MTBE. While the BTEX concentrations measured in newer apartments were significantly higher than that of older apartments, the MTBE concentrations were not significantly different. Accordingly, it is suggested that although newer buildings can emit more VOCs, this is not applicable to all VOCs. The present findings can be employed as basic data in designing exposure studies associated with VOC emissions in buildings and/or managing indoor air quality according to construction age of buildings.

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