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International Journal of Environmental Analytical Chemistry

Publication details, including instructions for authors and subscription information: <http://www.tandfonline.com/loi/geac20>

Naphthalene and benzene levels in microenvironments associated with potential exposure: new and old apartments with moth repellents, and cabins of passenger cars

Wan K. Jo^a & Jong H. Lee ^b

^a Department of Environmental Engineering, Kyungpook National University, Daegu 702–701, Korea

b Department of Environmental Management, Kumho PetroChemical Co., Yeosoo, Chun-Nam 555-280, South Korea

Available online: 18 Aug 2011

To cite this article: Wan K. Jo & Jong H. Lee (2011): Naphthalene and benzene levels in microenvironments associated with potential exposure: new and old apartments with moth repellents, and cabins of passenger cars, International Journal of Environmental Analytical Chemistry, 91:15, 1412-1424

To link to this article: <http://dx.doi.org/10.1080/03067310903359468>

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Naphthalene and benzene levels in microenvironments associated with potential exposure: new and old apartments with moth repellents, and cabins of passenger cars

Wan K. Jo^{a*} and Jong H. Lee^b

^aDepartment of Environmental Engineering, Kyungpook National University, n
Daegu 702–701, Korea; ^bDepartment of Environmental Management, Kumho PetroChemical Co., Yeosoo, Chun-Nam 555-280, South Korea

(Received 14 March 2009; final version received 26 August 2009)

A lesser degree of information is available with respect to microenvironments associated with potential exposure to naphthalene, in comparison with other volatile organic compounds. The current study investigated the levels of benzene as well as naphthalene, both in the indoor and outdoor air of apartments and in the cabins of passenger cars. Two groups of 20 apartment buildings (20 new and 20 old) were chosen on the basis of the selection criteria (apartment location and size). In addition, 10 actual commuters were recruited for this study. The equal number of drivers was recruited for the study for comparison of two types of fuels for vehicles (five drivers of gasoline-fuelled and five drivers of diesel-fuelled passenger cars). Indoor naphthalene concentrations were similar between old and new apartments, while the benzene concentrations in new apartments were significantly higher than those of old apartments. The naphthalene concentrations in bedrooms, where wardrobes with moth repellent (MRs) were placed, were significantly higher than those for living rooms where no MRs were present. In turn, these indoor concentrations were significantly higher than outdoor levels. It is noteworthy that the mean and median values of naphthalene measured in the bedrooms exceeded the USEPA RfC (inhalation reference concentration) of $3 \mu g m^{-3}$, and the living room values were close to the RfC, while the residential benzene levels exceeded the European benzene limit of $5 \mu g m^{-3}$. In contrast, the maximum outdoor levels were well below that of the RfC. The use of passenger cars appeared to be a significant daily activity for both naphthalene and benzene exposure. The naphthalene-to-benzene ratios varied with the type of microenvironments. Both the indoor naphthalene and benzene concentrations in the present study were much higher than those of other studies.

Keywords: age of apartment building; in-vehicle concentration; naphthaleneto-benzene ratio; diesel; gasoline

1. Introduction

Individual exposures to naphthalene and benzene have received a great amount of attention due to the adverse health effects of these compounds and their prevalence in various microenvironments. Naphthalene has been classified as a possible carcinogenic substance to humans [1,2], and benzene is a known carcinogen, inducing diseases such

^{*}Corresponding author. Email: wkjo@knu.ac.kr

as leukemia [1]. Homes are an important indoor microenvironment for potential exposure to these hazardous/toxic compounds. Naphthalene is the most abundant polycyclic aromatic hydrocarbon (PAH) found in typical urban air [3]. Benzene is one of several monocyclic aromatic hydrocarbons which may be detected at high concentration levels in both indoor and outdoor environments [4,5]. Several researchers have also reported that naphthalene and benzene levels in urban air were higher than those of rural or suburban air [3,4, 6–9]. These elevated urban air levels have been directly linked to various urban sources such as industrial discharges and the burning of fossil fuels [3,4,10,11]. Atmospheric naphthalene can penetrate indoors, thereby elevating the indoor concentration levels [12]. In addition, indoor naphthalene and benzene levels have been proven to be further elevated by various indoor sources, such as consumer products, cigarette smoke and/or building materials [3,5,6,13].

The use of passenger cars is an important daily activity associated with potential exposure to benzene and naphthalene. Several studies [14–17] have identified the fact that individuals inside vehicles are exposed to elevated levels of benzene and other volatile organic compounds (VOCs) in comparison to the corresponding ambient air levels at nearby monitoring sites. Unlike benzene and other traffic-related pollutants [14,15,18,19], limited information is available regarding in-vehicle exposure to naphthalene. Since naphthalene is a component of both gasoline and diesel fuels [20] and is contained in motor vehicle exhaust emissions [21], in-vehicle naphthalene levels can be elevated by the penetration of evaporative and tailpipe emissions of the motor vehicle or surrounding motor vehicles.

The present study investigated naphthalene and benzene levels in the indoor and outdoor air of apartments with moth repellents (MRs) and the cabins of passenger cars, in order to supplement a lack of research in this area. Since the VOC emission rates from building materials depend on the apartment age [22,23], the surveyed apartments were categorised into new and old dwellings. In addition, a major consumer use of naphthalene in homes is in MRs [10]. A few studies reported that the use of MRs potentially elevated the indoor levels of naphthalene [6,24,25]. As such, this study only surveyed apartments which utilised MRs in bedrooms. Since the chemical emission strength of old MRs may differ from that of recent ones, new information regarding exposure levels to MR emissions may be more appropriate for current risk assessments. This study focused on naphthalene and benzene, because, as compared to other VOCs humans are exposed to, naphthalene has been reported to a lesser degree and benzene has been known to have a higher toxicity compared to other VOCs associated with environmental exposures.

2. Experimental

2.1 Study protocol

The current study measured the concentrations of naphthalene and benzene, both in the indoor and outdoor air of apartment spaces and in the cabins of passenger cars in the city of Daegu, between 6 November 2006 and 13 April 2007. Two groups of 20 apartment buildings (20 new and 20 old) were chosen. The age of the apartments was determined based on the completion date of apartment construction: New apartments are less than six months old after completion of construction (ages, 2 and 5.5 months) and old apartments are between 12 and 18 months old. Since VOC emissions from new residential buildings usually reach peak values within six months, this time period is a reasonable

criterion for the categorisation of new and old apartments associated with VOC levels in residential buildings [26,27]. The criteria regarding apartment selection were as follows: The apartment buildings were located at least 100 metres away from a major roadway so as to minimise the impact of motor vehicle emissions; the apartments should have three or four bedrooms; and MRs should be used in at least one bedroom. All apartments used liquid petroleum gas (LPG) as their primary heating system. LPG was also used for cooking purposes. For each apartment, one 3-hour air sample (between 6:00 pm and 10:30 pm) was collected at breathing height in the middle of a bedroom where MRs were placed (in a wardrobe), and another sample was concurrently collected in the living room where no MRs were present. Simultaneously, one 3-hour air sample was also collected from the outdoor balcony of each apartment. The apartments are constructed with steel reinforced concrete and have 3 to 6 windows. The apartment residents were asked to refrain from smoking and to minimise the opening of doors or windows of the surveyed rooms during the sampling times. In fact, no smoking activities were observed during the sampling procedures. All the smokers among the participating residents confirmed that they usually smoke outside the apartment or on the house balcony during the survey in order to reduce their families' exposure to passive smoking. Although ventilation in the residences was not completely controlled, in most cases (37 of 40 sampling days), the doors and windows were closed during the sampling times. This process was monitored by a trained technician. As such, airflow levels were assumed to be uniform in all residences during the sampling procedure. Since the surveyed apartments had built-in wardrobes, their types and dimensions were similar to each other. In most cases (38 of 40 apartments), there was one built-in wardrobe in the bedroom. Neither the type nor the number of MRs were controlled for this study. Two types of MRs were used by the apartment inhabitants; ball-type (33 apartments) and paper-type products (MRs painted on paper) (7 apartments). The number of MRs employed for a surveyed bedroom were between 2 and 11. The ages of MRs utilised in the surveyed apartments were similar (less than 1 month after purchasing).

Ten commuters were recruited for a pilot-scale study. An equal number of drivers were recruited for the study for comparison of two types of fuels for vehicles (five drivers of gasoline-fuelled and five drivers of diesel-fuelled passenger cars). No factors, such as the car size, type or model year, were controlled for the selection of the test cars. Information regarding the test cars is shown in Table 1. The heights of the gasoline-fuelled cars were also similar, but lower than those of diesel-fuelled cars (sports utility vehicles), whose heights were similar. All the passenger cars were equipped with electronic fuel-injected engines, and used either unleaded gasoline or diesel fuel. Although the test vehicles had not been subject to any precise diagnosis by professional technicians, vehicle owners drove their vehicles without any functioning problems during the entire experimental period. All cars were sampled twice (morning and evening) in a single day over two sampling days. The sampling was conducted during morning and evening commuting hours on weekdays (Monday through Friday). One criterion for the commuter selection was the travel time, between 30 and 60 min for a one-way commute, which was determined during the participants interviews. The samples were collected by a trained technician who occupied the passenger seat. The sampling day was selected on the basis of the technician's personal preference. The commute routes were real rather than hypothetical. The windows and vents of the passenger cars were kept closed for the sampling time, with the temperature levels and blower speeds set to the personal comfort level of the occupants. In order to prevent any interference from tobacco smoke, the participants

Fuel	Identification	Manufacturing	Engine	Model	Odometer
	number (ID)	company	size (cc)	year	reading (km)
Gasoline	2 3 4 5	A B B B B	1800 1800 1600 2000 3500	1998 1996 2006 1998 2003	165,352 181,105 9.371 198,766 75,335
Diesel	6	C	2500	2005	43,163
	7	D	2500	1999	173,310
	8	C	2000	2005	38,778
	9	B	2000	2005	47,325
	10	B	2000	2003	97,443

Table 1. Information on gasoline-fuelled and diesel-fuelled passenger cars surveyed in the current study.

of this experiment were asked not to smoke during the sampling procedures. In addition, there were no previous smoking activities in the survey vehicles. The morning and evening commutes consisted of the first and the second vehicle use of the sampling days, respectively, for all drivers.

2.2 Sampling and analytical methods

Tenax adsorbents (Tenax TA 60/80, Supelco Co.) were utilised for naphthalene and benzene samples [11]. A constant-flow sampling pump (Aircheck Sampler Model 224-PCXR8, SKC Inc.) was connected to a $1/4''$ stainless steel trap containing 0.2 g of a Tenax TA 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 then used as the sample flow rate for all volume calculations. None of the samples deviated by more than 10% from the initial flow rate. A flow rate range of between 150 and 200 mL min^{-1} and 40 and 60 mL min^{-1} was set for the car and residential samples, respectively, based on the relative expected concentrations for each experimental condition.

The target compounds collected in the Tenax Traps were analysed by coupling a thermal desorption system (Perkin Elmer ATD 400) to a gas chromatograph (HP 5890II) and a mass spectrometer (HP MSD5973) (GC/MS) system, within 3 days of collection. Sample traps were stored in shipping containers on the bottom refrigerator in the laboratory. A 30-m-long fused silica capillary column (an internal diameter of 0.32 mm; a film thickness of $1 \mu m$) (Agilent Technologies, HP-5) was used in order to separate the target analytes. Compounds were identified by using both retention times and a Wiley mass spectral library. A spectral search quality of 70% was utilised for the compound selection criteria.

2.3 Quality control

The quality controls (QCs) included laboratory and field blank traps as well as spiked samples. At the beginning of the day, an external standard was analysed daily to check the quantitative response. Both the laboratory blank traps, obtained from each analytical batch, and the field blank traps were analysed to check for any trap contamination; however, no trap contamination was identified. The field blanks were taken to the sampling places, briefly opened, capped and stored with other field samples. When the quantitative response differed more than $\pm 25\%$ from that predicted by the specified calibration equation, a new calibration equation was determined. Seven sampling traps spiked with low concentration standards were used to determine the method detection limits (MDL) of the system. For the lowest sampling volume (4.6 L), the MDLs of the target compounds were 0.01 and 0.13 μ g m⁻³ for naphthalene and benzene, respectively. However, the lack of duplicate field measurements is a limitation of this QC plan.

2.4 Statistical analyses

Statistical analyses were performed by using the SAS Version 9.1 (SAS Institute Inc., Cary, NC, USA). On the basis of log-transformed data, the paired sample means of bedrooms and living rooms (or outdoor air) were analysed by using a paired t-test. The concentration differences between old and new apartments, and between gasoline- and diesel-fuelled cars were analysed using a non-parametric test (Wilcoxon Rank-Sum Test). When the Shapiro-Wilk statistical test indicated that the data were log-normally distributed, median values were also presented. The criterion for significance of the procedures was $p < 0.05$. Spearman correlation coefficients were calculated in order to examine the relationship between the naphthalene and benzene concentrations.

3. Results and discussion

3.1 Indoor and outdoor apartment concentration levels

The concentrations of naphthalene measured in bedrooms, living rooms and the outdoor air of apartments are summarised in Table 2, according to the age of the apartments. No samples were determined to be below MDLs. The naphthalene concentrations in the new apartments were not significantly different from those of the old apartments, confirming that building materials were not a significant indoor source of naphthalene. The naphthalene concentrations in the bedrooms were significantly higher than the living room levels, which in turn were significantly higher than the outdoor levels, for both new and old apartments (criterion for significance, $p < 0.05$). Van Winkle and Scheff [6] reported that several indoor activities are associated with the emission of naphthalene: namely, indoor MR storage, washer/driers in utility rooms, periodic dry cleaning and electric heat. According to their emission factors, MRs represented the second highest source of naphthalene, following the use of electric heat. However, all apartments tested in the present study used liquid petroleum gas (LPG) as their primary heating system as well as for cooking purposes. The use of either washers/driers or dry cleaning activity was not observed during the sampling process. Other studies [6,28] also reported that tobacco smoking is another potential source for naphthalene in homes. However, residents refrained from smoking in the apartments during sampling hours. As such, the storage of MRs in bedrooms is likely to be a potentially significant source regarding the indoor naphthalene levels.

The higher bedroom concentrations, as compared to the living room concentrations, are likely due to the close proximity of MRs. The source proximity effect is supported by Downloaded by [East Carolina University] at 00:29 20 February 2012 Downloaded by [East Carolina University] at 00:29 20 February 2012

a Medians of bedroom- or living room-to-outdoor concentration ratios were calculated using the distribution of individual samples. aMedians of bedroom- or living room-to-outdoor concentration ratios were calculated using the distribution of individual samples.

Mcbride *et al.* [29], who reported that while a source is emitting, the concentrations of SF_6 and CO were influenced by its proximity to the indoor source. MRs in non-monitored bedrooms were not controlled in the present study. However, since the living rooms were located between the bedrooms in all surveyed residences, MRs in non-monitored bedrooms would have a greater influence on the naphthalene levels in the living room than those in the surveyed bedroom. Thus, it is suggested that, even if MRs in non-monitored bedrooms were not controlled in the present study, their effects on the naphthalene levels in the surveyed bedrooms would be insignificant, due to the distance between the bedrooms. Moreover, it is noteworthy that the bedroom naphthalene mean and median values exceeded the USEPA RfC (inhalation reference concentration) guideline of $3 \mu g m^{-3}$, and the living room values were close to the RfC. In contrast, the maximum outdoor levels were well below that of the RfC. Consequently, it is suggested that, in conjunction with average timelines regarding resident activities, residential indoor environments employing recent types of MRs (naphthalene plus unknown herb-scent constituents) as well as old types (primarily naphthalene) are significant for individual exposure to naphthalene, thereby suggesting the need for a control strategy in order to minimise naphthalene exposure in residences.

It is noteworthy that residences which utilised paper-type MRs exhibited lower-range bedroom naphthalene concentrations $(3.48-3.97 \mu g m^{-3})$ as compared to ball-type MRs $(>3.97 \,\mu g \,\text{m}^{-3})$, and that the residence which utilised the highest number of ball-type MR (11) exhibited the maximum bedroom naphthalene concentration level (6.11 μ g m⁻³). A possible explanation for this result is that the naphthalene emission strength from ball-type MRs was greater than that of paper-type MRs, and that more MRs resulted in higher naphthalene emissions.

In contrast to naphthalene, benzene concentrations in the new apartments were significantly higher than those of the old apartments (Table 2), resulting in different ratios of naphthalene-to-benzene indoor concentrations (Table 3). In addition, the correlation of indoor naphthalene and benzene concentrations was not statistically significant in either the new or old apartments (Table 3). The differing ratios point to different benzene emission sources and sinks between the two apartment categories. High benzene emissions or concentrations have often been found in newly-built or renovated buildings [22,23],

	Ratio ^a	Spearman correlation		
Microenvironment		R	p -value	
Apartment, new				
Bedroom	0.27	0.10	0.77	
Living room	0.15	0.28	0.43	
Outdoor	0.10	0.65	0.04	
Apartment, old				
Bedroom	0.55	0.18	0.62	
Living room	0.29	0.42	0.22	
Outdoor	0.10	0.71	0.02	

Table 3. Median ratios of naphthalene to benzene concentrations and their correlations that were determined for apartments.

Note: ^aMedian ratio of naphthalene to benzene concentrations.

since several types of building materials and consumer products, such as textured carpet, carpet glue, liquid detergent, steel wool soap pads and furniture wax, are major indoor sources for benzene levels [30–2]. The benzene concentrations in the bedrooms were similar to those of the living rooms for both the new and old apartments. In agreement with several other studies [5,33], the indoor benzene levels were significantly higher in comparison to the outdoor levels. It is noteworthy that the indoor benzene levels, even in old apartments, were higher than the European limit of $5 \mu g m^{-3}$ (which comes into effect 2010), although they were lower than the RfC guidelines of $30 \,\text{\mu g}\,\text{m}^{-3}$.

The correlations and ratios of the naphthalene and benzene outdoor concentrations are also shown in Table 3. The median ratios of the two compounds were equal between new and old apartments, suggesting similar sources and sinks for outdoor air in the vicinity of the two types of apartments. This suggestion is supported by the finding that the correlation of these two compounds was statistically significant for both the new and the old apartments. The naphthalene-to-benzene outdoor concentration ratios shown in Table 3 (0.10 for both old and new apartments) are higher than those reported for samples taken in Pasadena California by Lu et al. [28]. They reported that the ratios for the ambient samples taken over two days in November 2003 were 0.049 and 0.055, respectively. In addition, the average ratio of motor vehicle emissions as determined in tunnels was 0.033, which was identical to the ratio corresponding to the average gasoline vehicle emissions based on the California Air Resources Board emission inventory. Tunnel studies have determined that the average emission ratio of large numbers of vehicles reflect vehicle emissions under actual traffic conditions [34,35]. As such, the ratio difference between this and Lu et al.'s studies are mainly due to the averaged gasoline vehicle emission ratios.

3.2 In-vehicle levels

The concentrations of both naphthalene and benzene, as measured inside gasoline-fuelled cars, were significantly higher than those of diesel-fuelled cars (Table 4). This pattern of higher levels in gasoline-fuelled cars is likely to be due to the difference in motor vehicle emissions and the height of the vehicles. Since those compounds were more predominant in gasoline fuel than in diesel fuel [20,21,36], their penetration with other unburned fuels into the cabins of vehicles would be greater in gasoline-fuelled vehicles, resulting in elevated in-vehicle levels. Similarly, gasoline engine exhaust emissions exhibited a higher

Notes: Number of samples: $N = 20$ for gasoline-fuelled vehicles; $N = 20$ for diesel-fuelled vehicles. Gasoline/Diesel represents the median concentration ratio of gasoline-fuelled to diesel-fuelled vehicles.

level of naphthalene [37], further elevating naphthalene levels inside gasoline-fuelled cars by means of exhaust entering the vehicle cabin. Two previous studies [15,38] have indicated that the lower the vehicle height, the higher the compound concentrations that were encountered on roads resulting from the vertical gradient of the compound concentrations. This also results in higher levels inside gasoline-fuelled cars, since the heights of gasoline-fuelled cars were lower than those of the diesel-fuelled cars. It should be noted that, although two of the 10 drivers (one diesel-fuelled car and one gasoline-fuelled car drivers) were smokers, the in-vehicle air sampling was conducted at least 30 min after the last recorded smoking by the drivers. As such, the exhaled breath levels of smoking drivers, if detectable at all, would not significantly contribute to the in-vehicle concentration difference between the two types of vehicles.

The mean in-vehicle naphthalene levels in the present study $(3.97 \text{ and } 2.53 \mu\text{g m}^{-3})$ for gasoline-fuelled and diesel-fuelled cars, respectively) were much higher than those in a previous study [39], which reported that mean naphthalene concentrations in bus cabins in Detroit, Michigan were 0.6 and $1.3 \,\mathrm{\upmu}\mathrm{g}\,\mathrm{m}^{-3}$ for morning and afternoon samples, respectively. The in-vehicle naphthalene levels were substantially higher than the mean outdoor air concentrations measured in the present study $(0.41-0.47 \,\mu g \,\text{m}^{-3})$. Similarly, the mean in-vehicle benzene levels (10.6–17.1 μ g m⁻³) were more than two times that of the mean outdoor air concentrations as measured in the present study $(4.5-4.6 \,\mu g \,\text{m}^{-3})$. Consequently, the use of passenger cars appears to be a major daily contributor to both naphthalene and benzene exposure. A broad concentration range, illustrated in Table 4, reflects the combined effects of various driving parameters, such as vehicle type, driving route, driving period, vehicle ventilation, driving speed and fuel composition, as well as local meteorological parameters such as the temperature and dispersion or turbulence variability. In addition, a gasoline-fuelled vehicle of the model year 2006 and with odometer reading of 9,371 km (the newest, and by far the least used vehicle in this study) presented the highest in-vehicle benzene level, but not naphthalene. New vehicles emit more VOCs from the interior materials compared to old vehicles [40]; however, the old vehicles may have larger penetration fractions and evaporative portions. As such, a possible explanation for the high in-vehicle benzene level measured in the newest car is that the interior emissions effect of the new car on in-vehicle benzene levels would outweigh the exterior-to-interior penetration effect of the old cars.

3.3 Comparison of naphthalene and benzene concentrations of apartments with those of other studies

The indoor and outdoor air naphthalene concentrations measured in the apartments and in earlier studies are listed in Table 5. The indoor naphthalene concentrations in the present study were much higher than those in the homes of North Carolina [24] and Chicago [3,6], USA, Birmingham, UK [25], as well as Hangzou, China [13]. Previous studies measured naphthalene levels without considering the presence of MRs, whereas the present study measured them in bedrooms with MRs. As such, the difference between this and previous studies would be due to the emission strength of MRs and/or other potential indoor sources for naphthalene. Other parameters, such as ventilation and other house characteristics, might also have influenced the difference.

As found with the indoor naphthalene concentrations, the indoor benzene concentrations in the present study were much higher than the indoor or personal exposure levels

Table 5. Comparison of naphthalene mean indoor and outdoor concentrations (µg m⁻³) measured by the present and other studies. 3) measured by the present and other studies. Table 5. Comparison of naphthalene mean indoor and outdoor concentrations (μ g m $^-$

Notes: ^aRepresents the concentrations from the present study: indoor and outdoor concentrations were calculated by averaging the matched mean Ű þ ì, concentrations of the new and old apartments.

bd, below analytical detection limit.

^hna, not applicable.
 Median values were presented. concentrations of the new and old apartments.

^bnd, below analytical detection limit.

 c_{na} , not applicable.

dMedian values were presented.

in previous studies [5,41,42]. For example, the mean indoor concentrations were 17.1 and $10.6 \,\mathrm{\upmu}\mathrm{g}\,\mathrm{m}^{-3}$ in the bedrooms of new and old apartments, respectively (Table 2), while they were 2.84 and $6.5 \mu g m^{-3}$ in Michigan homes [5] and European buildings [41], respectively. In addition, D'Souza et al. [42] reported a mean personal benzene concentration of $2.8 \mu g m^{-3}$ in the U.S. National Health and Nutrition Examination Survey.

The outdoor naphthalene levels found in the present study were similar to those of a recent study conducted in Los Angeles and Riverside [11], whereas they were much lower than the outdoor levels of old American studies [43,44]. This difference is likely to be due to reducing naphthalene emissions, such as amounts of industrial discharge, the burning of fossil fuels and naphthalene compositions in fuels [3,4,10,11]. For example, Lu et al. [28] reported that the difference in outdoor levels between older and newer American studies was likely to be due to the introduction of California Phase II-reformulated gasoline (RFG) in the first half of 1996, which caused reductions of 70 to 75% in naphthalene and benzene weight fractions in specific collected gasoline samples [45].

4. Conclusion

The present study investigated naphthalene levels, in conjunction with benzene levels, in two major microenvironments (indoor and outdoor air of apartments with MRs, and cabins of passenger cars) potentially associated with elevated exposure levels. Although the bedroom naphthalene concentrations observed from homes that utilised paper-type MRs were lower than those from homes that utilised ball-type MRs, the concentrations were higher than RfC (3μ g m⁻³). Therefore, recognising the proportion of time people typically spend in residential indoor environments, the use of MRs is a significant source of personal exposure to naphthalene regardless of the types (especially exposure during the time of sleeping in bedroom). Our results also suggest that commuting inside passenger cars is a daily activity significantly contributing to both naphthalene and benzene exposure levels. Moreover, it is noteworthy that the residential benzene levels, even in old apartments, exceeded the European limit of $5 \mu g m^{-3}$.

Acknowledgements

This study could not have been accomplished without the cooperation of 10 car drivers and the residents of the 40 surveyed apartments. The authors would like to thank three graduate students (Mr. C.H. Yang, Mr. J.T. Kim and Miss. M.H. Shin) in the Department of Environmental Engineering, Kyungpook National University, for their sample collecting and/or analyses. We wish to thank the reviewers for their thoughtful corrections and valuable suggestions regarding our paper. This work was supported by the Korea Science and Engineering Foundation (KOSEF) grant funded by the Korea government (MOST) (No. R01-2006-000-10851-0).

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