Formaldehyde and Aromatic Volatile Hydrocarbons in the Indoor Air of Egyptian Office Buildings

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Abstract

Indoor and outdoor measurements of formaldehyde and the aromatic hydrocarbons: benzene, toluene, ethylbenzene, m,p-xylene and o-xylene (BTEX) were conducted at six offices in a suburban area in Giza, Egypt, during the winter 2003–04 season. The mean indoor level of formaldehyde was 59.79 ppb in offices where there was no smoking and 85.01 ppb in offices where people smoked. The highest mean level of formaldehyde in offices without smoking was in a new office, 90 ppb, and the lowest, 36.16 ppb, was in an old office. During the period of study, in 13.33%, 20% and 16.67% of samples from non-smoking, smoking and total offices, respectively, the level of formaldehyde exceeded the ASHRAE standard of 0.1 ppm. The mean indoor concentrations of benzene, toluene, ethylbenzene, m,p-xylene and o-xylene were 4.32, 25.06, 3.60, 9.14 and 4.38 ppb, respectively, in the six offices. The mean indoor levels in non-smoking offices were lower, and in offices with smoking, higher than the overall mean. The highest mean levels of BTEX in non-smoking offices were found in a new office, whereas the lowest concentrations were recorded in an old office. Significant positive correlations were found between the indoor concentrations of BTEX inside the six offices, except benzene in non-smoking offices. Benzene had only weak positive correlations with these compounds.

Introduction

Indoor air quality (IAQ) is an important issue because people spend most of their time in indoor environments where there may be more pollutants and at higher concentrations than outdoors [1–3]. In particular, the levels of volatile organic compounds (VOCs) found indoors are generally reported to be greater than outdoors [4–7]. Consequently IAQ is liable to influence human health more than outdoor air quality. Poor IAQ has been linked to a number of symptoms, which the world health organisation has defined together as Sick Building Syndrome (SBS) [8]. A relation between indoor (VOCs) and SBS has been shown in some offices and schools [8–11]. The common indoor pollutant formaldehyde (HCHO) is

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known to be toxic and irritating to the respiratory tract, eyes and skin [12] and is carcinogenic to humans at high concentrations [13]. Aromatic VOCs in the urban environment mostly arise from the fuels used in motor vehicles [14,15] and these can infiltrate buildings [16]. The major compounds are benzene, toluene, ethylbenzene and the isomeric xylenes (a mixture commonly abbreviated as BTEX) [4,17]. Of these, benzene is a known carcinogen while toluene and xylene are toxic at high concentrations [1,18,19]. WHO [20] has estimated that lifetime exposure of urban populations to benzene concentrations of $1.7 \mu g \cdot m^{-3}$, may result in 10 cases of leukaemia per million inhabitants.

The sources and levels of the VOCs in indoor air vary depending on the type of building. Major sources are construction materials, furnishings, cosmetics and textiles, paints, carpet, architectural finishes, insulation, fabrics and paper, varnishes and solvents, adhesives, cleaning compounds and combustion by-products [21–26]. Combustion, particularly tobacco smoking, but also photocopying or laser printing on paper, strongly influence the indoor concentrations of VOCs [27,28].

HCHO levels were found to range from 5.4 to 25 ppb, in a call centre office in the San Francisco Bay Area, CA [29]. A level of 27 ppb was measured in an office building [30]. Levels of *o*-xylene from 0.30 to 1.40 ppb and of *m,p*-xylene from 0.93 to 4.60 ppb have been measured in 12 office buildings in California [31]. The average concentrations of benzene, toluene, ethylbenzene, *m,p*-xylene and *o*-xylene were 3.94, 21.31, 1.75, 5.36 and 3.33 ppb, respectively, in 12 offices in Korea [32]. In 10 offices in Hong Kong, the average levels of individual BTEX compounds were 2.53 ppb for benzene, 14.08 ppb for toluene, 1.68 ppb for ethylbenzene, 4.34 ppb for *m,p*-xylene and 1.26 ppb for *o*-xylene [33]. In addition, Guo et al. [34] found that the mean concentrations of individual BTEX compounds ranged from 1.38 ppb for benzene to 12.53 ppb for toluene in six offices in Hong Kong.

Greater Cairo (Cairo, Giza and Shoubra El-Khiema) houses over 11.3 million inhabitants and is the source of most of the polluting activities in Egypt. It is one of the most polluted megacities in the world. Previous studies on IAQ in homes and offices in Greater Cairo have focused on inorganic pollutants like sulphur dioxide, nitrogen dioxide, ammonia, ozone, total oxidants and suspended particulate matter and its chemical composition. However, there is a lack of information on the indoor levels of VOCs.

The present study aimed to reduce that lack by evaluating the indoor concentrations of HCHO and BTEX in six offices in a suburban area in Giza, Egypt, and to compare these concentrations with ambient air levels. In addition, the factors that had an effect on the indoor HCHO and BTEX concentrations were measured.

Materials and Methods

Sampling Sites and Period

A narrow strip of Giza Governorate runs along the western side of the Nile, opposite the city of Cairo. Indoor and outdoor measurements of HCHO and BTEX were made at six different offices in a suburban area in Giza (Kafr Tohormos), Egypt. Information concerning the sampling sites is shown in Table 1. The indoor samples were collected at approximately 1.5 m above floor and in the centre of each office. At the same time outdoor samples were also collected nearby. Sampling was conducted during the winter months (December 2003 to February 2004). To facilitate evaluation of the contribution of environmental tobacco smoke (ETS) to BTEX concentrations, three smoking and three non-smoking offices were selected. Smoking offices were defined as those in which at least one smoker worked, and in which smoking occurred during sampling. In an unventilated room with no apparent source of VOCs, the concentrations of BTEX were also measured before and after smoking to evaluate the contribution of ETS to

Table 1. Characterization of the sampling sites in Giza

Site	Age	Floor	Size (m²)	Smoking allowed
Office 1	14 years	2nd	12	No
Office 2	7 years	3rd	16	No
Office 3	6 months	2nd	30	No
Office 4	7 months	2nd	27	Yes
Office 5	6 years	3rd	24	Yes
Office 6	12 years	2nd	21	Yes

All offices have natural ventilation.

BTEX levels. The HCHO samples were taken from 08.30 to 15.30 h local time (7 h), whereas the BTEX samples were taken from 09.00 to 14.00 h local time (5 h).

Sampling and Analytical Methods
Formaldehyde

Aldehydes in the air were collected in a 0.05% aqueous solution of 3-methyl 2-benzothiazolone hydrazone hydrochloride (MBTH) [35]. Air samples were collected in glass bubblers with a coarse fritted inlet containing 35 ml of 0.05% MBTH solution, using a pump calibrated to draw 0.5 L·min⁻¹. The resulting azine was then oxidised by ferric chloride-sulphamic acid solution to a blue product which was quantitated at 628 nm using spectrophotometry. The concentration of total aldehydes was calculated in terms of HCHO. This method is relatively free from interference and its collection efficiency is 84% [35].

Aromatic Volatile Compounds (BTEX)

Air samples were sucked through charcoal where the VOCs were adsorbed, following the NIOSH standard method [36]. The collection tubes contained 150 mg of coconut charcoal sub-divided into two portions of 100 mg and 50 mg. The front portion (100 mg) was used to collect the VOCs, while the 50 mg backup section was used to determine if solvent breakthrough occurred. Air was drawn through the charcoal tubes, using a pump calibrated to draw 0.2 L·min⁻¹. After sampling, the charcoal tube was removed and open ends were tightly closed using special caps to avoid loss of sample. During the sampling procedure, one charcoal tube was opened at the sample site and then the ends capped, which served as a blank. The sample and blank tubes were put into special plastic bags that were tightly closed and kept in a freezer until processed. Before analysis, all samples and blanks were taken from the freezer, contents of both sections of the adsorber tubes were emptied into sample vials and 0.75 ml of carbon disulphide as extraction solvent was added. Vials were shaken mechanically and then left for at least 1 h to obtain the analyte. Aliquot volumes of samples and blanks were then analysed using gas chromatography with flame ionisation detection.

Statistical Analysis

The correlation coefficient (r) and the correlation significant t-test were determined [37]. Student's t-test was used to estimate the significant difference between the mean concentration in different offices and between indoor and outdoor concentrations [37].

Results

The indoor and outdoor concentrations of HCHO at the six offices are shown in Table 2. Indoor HCHO concentrations were all significantly higher than the corresponding levels outdoors with the exception of office 1. The overall mean level indoors was significantly higher than that outdoors ($p < 0.001$) with an I/O ratio for the six offices of 2.73. In non-smoking offices levels were highest in the newest office: office 3, 6 months old and lowest in the oldest: office 1, 14 years old. The difference was statistically significant ($p < 0.001$). A similar situation was found in the smoking offices. Mean indoor levels of HCHO were 59.79 and 85.01 ppb in non-smoking and smoking offices, respectively, and the difference was statistically significant ($p < 0.001$). Moreover, the mean I/O values were 2.10 at non-smoking offices and 3.45 at smoking offices. The frequency percentage distribution of HCHO concentrations inside the six offices is represented graphically in Figure 1. From this figure, it can be seen that 13.33%, 20% and 16.67% of the concentrations in non-smoking, smoking and total offices, respectively, exceeded the maximum HCHO concentration (100 ppb, 0.1 ppm) set by ASHRAE.

Statistics for indoor and outdoor concentrations of BTEX at the six offices are summarised in Table 3. Toluene and m,p-xylene were the most abundant compounds, followed by o-xylene and benzene. The levels of BTEX in 3 smoking offices and 3 non-smoking offices are compared in Table 4. Mean indoor concentrations were higher than those in non-smoking offices. Significant differences ($p < 0.05$) were found between mean concentrations in smoking and non-smoking offices for all BTEX compounds. To examine further the influence of smoking on BTEX levels the I/O concentration ratios of BTEX at smoking and non-smoking offices were compared. Figure 2
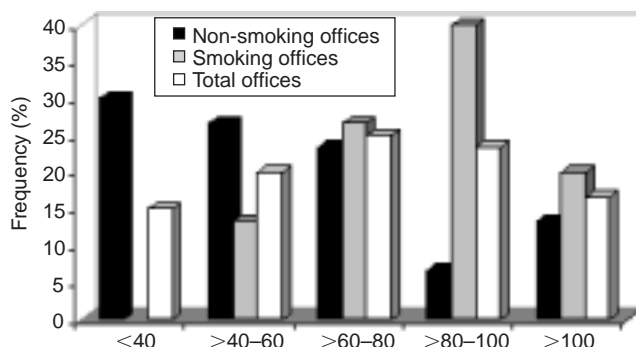

Fig. 1. Frequency distribution percentage of formaldehyde concentrations measured inside the six offices.

Table 2. Summary statistics for indoor and outdoor concentrations of formaldehyde (ppb) at the six different offices

Site	Indoor						Outdoor						I/O ratio
	N	Min.	Max.	Median	Mean	SD	N	Min.	Max.	Median	Mean	SD	
Office 1	10	28.50	47.80	35.58	36.16	6.90	10	26.50	39.50	31.27	32.54	4.91	1.11
Office 2	10	37.56	73.70	54.30	53.20	13.41	10	24.50	37.80	29.19	30.48	5.29	1.74
Office 3	10	66.40	115.50	87.25	90.00	20.05	10	18.64	27.40	21.43	22.26	3.41	4.04
Office 4	10	81.51	135.80	99.18	103.36	20.61	10	16.50	25.40	19.37	20.15	3.43	5.13
Office 5	10	64.40	111.50	80.09	82.18	16.76	10	23.46	35.70	27.20	28.50	4.75	2.88
Office 6	10	52.75	95.80	67.25	69.50	15.79	10	20.50	31.50	24.44	25.28	4.10	2.75

N: number of samples; Min.: minimum; Max.: maximum; SD: standard deviation; I/O: indoor/outdoor.

Table 3. Summary statistics for indoor and outdoor concentrations of BTEX at the six different offices

BTEX (ppb)	Indoors					Outdoors					I/O ratio
	Min.	Max.	Median	Mean	SD	Min.	Max.	Median	Mean	SD	
Benzene	1.08	12.42	3.71	4.32	2.81	2.45	7.22	4.40	4.53	1.32	0.95
Toluene	7.33	46.71	25.22	25.06	10.43	7.73	20.78	12.16	12.95	3.53	1.93
Ethylbenzene	0.97	7.58	3.30	3.60	1.77	1.19	3.45	2.06	2.15	0.56	1.67
m,p-xylene	2.36	18.84	8.73	9.14	3.99	2.82	10.32	5.71	6.03	1.69	1.52
o-xylene	1.29	8.31	4.35	4.38	1.77	1.43	3.56	2.27	2.25	0.57	1.95

Min.: minimum; Max.: maximum; SD: standard deviation; I/O: indoor/outdoor.

Table 4. Comparison of the BTEX concentrations measured inside non-smoking and smoking offices

BTEX (ppb)	Non-smoking offices					Smoking offices					S/N ratio
	Min.	Max.	Median	Mean	SD	Min.	Max.	Median	Mean	SD	
Benzene	1.08	6.04	2.05	2.29	1.22	3.29	12.42	6.00	6.36	2.46	2.78
Toluene	7.33	39.11	19.64	20.31	10.47	18.07	46.71	28.66	29.81	8.11	1.47
Ethylbenzene	0.97	6.41	2.66	2.87	1.68	2.22	7.58	3.98	4.34	1.56	1.51
m,p-xylene	2.36	14.83	6.96	7.48	3.59	4.51	18.84	10.74	10.81	3.73	1.45
o-xylene	1.29	7.13	3.17	3.64	1.71	2.73	8.31	5.06	5.12	1.52	1.41

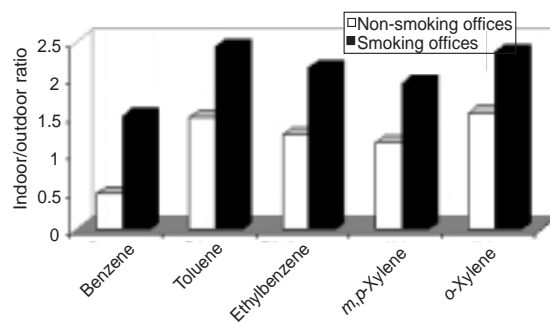Min.: minimum; Max.: maximum; SD: standard deviation; S/N: smoking/non-smoking.

Fig. 2. Comparison of mean indoor/outdoor concentration ratios of BTEX at non-smoking and smoking offices.
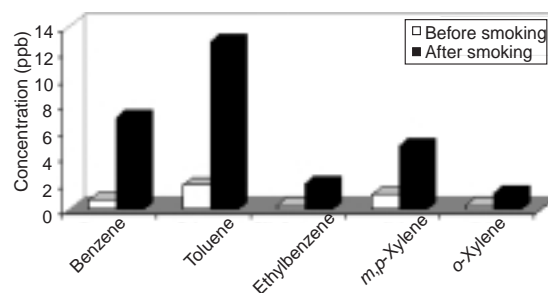

Fig. 3. Concentrations of BTEX before and after smoking inside an unventilated room with no apparent source of VOCs.

shows the comparison of mean I/O concentration ratios of BTEX at offices with and without smokers. The concentrations of BTEX were also measured in an unventilated room with no apparent source of VOCs before and after four cigarettes smoked by a human volunteer (Figure 3). The concentrations of benzene, toluene, ethylbenzene, *m,p*-xylene and *o*-xylene were 7, 12.88, 1.98, 4.86 and 1.22 ppb after smoking, respectively, i.e. 10, 7, 5.66, 4.42 and 3.48 times higher respectively than those recorded before smoking (background level).

The indoor concentrations of BTEX inside new (6 months old) and old (14 years old) non-smoking offices are represented graphically in Figure 4. Significantly higher BTEX levels were found in new offices ($p < 0.05$). The new/old concentration ratios were 1.96, 3.25, 3.46, 2.83 and 2.68 for benzene, toluene, ethylbenzene, *m,p*-xylene and *o*-xylene, respectively. In addition, the I/O values were 0.93, 3.38, 2.59, 2.20 and 2.97 at new offices, whereas they were 0.30, 0.70, 0.58, 0.65 and 0.83 at old offices for benzene, toluene, ethylbenzene, *m,p*-xylene and *o*-xylene, respectively (Figure 5). The effect of using cleaning products in non-smoking offices is shown in Figure 6.

The correlation coefficients between the indoor concentrations of the BTEX compounds in non-smoking and smoking offices are shown in Table 5.
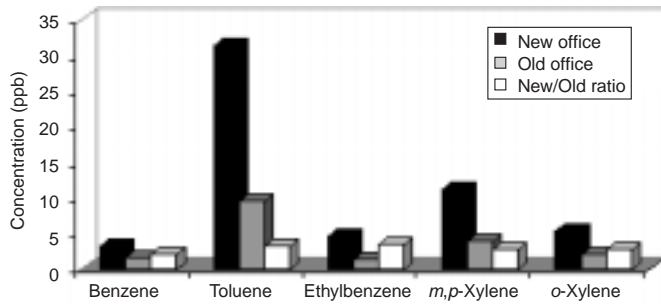
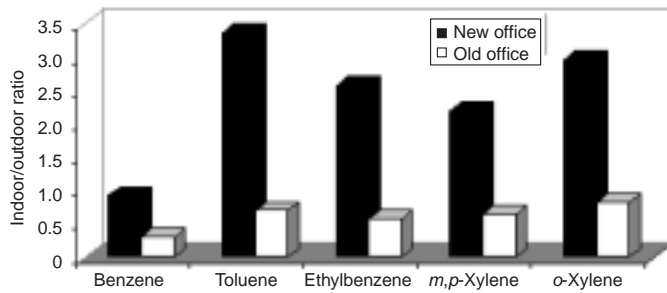**Fig. 4.** Comparison of the indoor concentrations of BTEX inside new and old non-smoking.

Fig. 5. Comparison of mean indoor/outdoor concentration ratios of BTEX at new and old non-smoking offices.
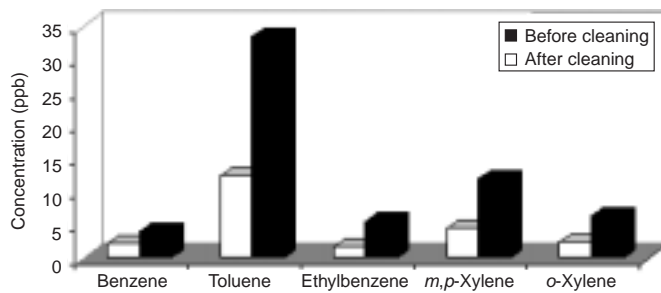

Fig. 6. Concentrations of BTEX found in non-smoking office before and after using cleaning products.

Discussion

The levels of HCHO in office buildings are dependant on many factors such as the surface area of materials (wood products, carpet and insulation), fabrics and paper that can emit HCHO, the age of the building, tobacco smoking, temperature, relative humidity and ventilation rate. The higher HCHO levels in offices 3 and 4 are almost certainly due to the higher emission rate from new building materials which is in agreement with previous studies. These had shown that the highest HCHO

Table 5. Correlation coefficients between different BTEX concentrations inside non-smoking and smoking offices

	Non-smoking offices					Smoking offices				
	Benzene	Toluene	Ethylbenzene	m,p-xylene	o-xylene	Benzene	Toluene	Ethylbenzene	m,p-xylene	o-xylene
Benzene	1					1				
Toluene	0.47	1				0.85[a]	1			
Ethylbenzene	0.42	0.78[a]	1			0.82[a]	0.91[a]	1		
m,p-xylene	0.43	0.81[a]	0.97[a]	1		0.86[a]	0.96[a]	0.90[a]	1	
o-xylene	0.33	0.84[a]	0.94[a]	0.94[a]	1	0.83[a]	0.94[a]	0.93[a]	0.96[a]	1

[a]Significant (p < 0.001).

concentrations were found in newly renovated classrooms [38] and in new buildings [39,40]. The extensive use of new pressed wood furniture and smoking increase the indoor levels of HCHO [41].

Both qualitatively and quantitatively the results reported here are in agreement with other studies including that of Lee et al. [41] who found that the indoor levels of HCHO were higher than outdoor concentrations at ten offices in Hong Kong. Also, the I/O concentration ratios of HCHO ranged from 1.44 to 10.7 in a call centre office in the San Francisco Bay Area, CA [29]. In the present study, the average concentration of HCHO inside the six offices was higher than that reported by Hodgson et al. [29] and Cavalcante et al. [30].

Regardless of age the higher levels of HCHO in smoking offices indicate that tobacco smoking increases the indoor concentrations of this compound. Previous studies had shown that the HCHO levels had increased to more than 82 ppb in a 30 m^3 room after smoking two cigarettes [42]. Also, higher HCHO concentrations were found in smoking homes [43].

The indoor HCHO concentration in non-industrial settings should not exceed 0.1 ppm (100 ppb) (American Society of Heating, Refrigerating, and Air Conditioning Engineers (ASHRAE)) [44]. From Figure 1 it can be seen that, 13.33%, 20% and 16.67% of the concentrations in non-smoking, smoking and total offices, respectively, exceeded this recommended maximum.

The levels of BTEX measured at the six offices were higher than those reported in comparative studies [31–34,45]. Also, the mean indoor concentrations of toluene, ethylbenzene and the isomeric xylenes were higher than outdoor levels, and the differences were statistically significant ($p < 0.001$). The mean I/O values greater than unity clearly indicates that indoor sources of these compounds must be present. The outdoor mean concentration of benzene was slightly higher than the indoor level, and no statistical difference was found between the means, showing no important source inside the offices. This result is in agreement with Guo et al. [34] who found an I/O value for benzene of 0.88 in offices.

Vehicular emission is the major source of outdoor VOCs in urban areas. The relative ratios of different aromatic compounds can be very useful in identifying VOCs emission sources [46]. The ratios of benzene to toluene in various cities around the world has been measured to range from 0.25 to 0.50 for ambient air [46]. Studies on vehicular exhaust generally report a ratio of benzene to toluene of 0.5 [47,48]. In the present study, the mean ratio of benzene to toluene in the ambient air was 0.35, which clearly implies that vehicular emission is the dominant source of outdoor VOCs. The variation in the benzene to toluene ratio in this study and those found in the different countries of the world may be due to the differences in fuel composition and vehicle types, particularly their use of catalytic converters, in various cities around the world. On the other hand, benzene to toluene ratios in indoor environments are more variable than for ambient air due to the wide variety of potential indoor sources and the complexity of the indoor environment [31,32,49]. In the present study, benzene to toluene ratios inside the six offices was 0.17, which suggests additional indoor sources. This result is in agreement with Baek et al. [32] who found that indoor benzene to toluene ratios was 0.16 in offices.

VOC levels vary in office buildings depending upon factors such as: the age of the building; the nature of work carried out; occupant density; type and number of equipment used; smoking; the amount and type of cleaning products used; the building ventilation rate; and outdoor concentrations. A probable effect of smoking is the higher levels of BTEX measured in smoking offices compared to non-smoking offices. The difference in levels was significant ($p < 0.05$) for all BTEX compounds. Further support for the influence of smoking on BTEX levels was made by comparing I/O concentration ratios of BTEX at smoking and non-smoking offices (Figure 2). The I/O ratios were higher at offices where smoking occurred than offices without smokers. The concentrations of BTEX in an unventilated room with no apparent source of VOCs were appreciably increased after four cigarettes were smoked by a human volunteer. The mean level of ethylbenzene in the expired air of smokers has been found to be 2 to 3 times higher than in that of non-smokers [50]. Benzene is also found as a constituent of tobacco smoke and its levels are generally higher in houses where there are smokers [51]. A significant amount of benzene, toluene and C_8 alkylbenzene are found in side-stream cigarette smoke [52]. Other work has shown that tobacco smoking increases the levels of benzene, toluene and *m,p*-xylene [43].

Greater concentrations of BTEX were measured inside the newer buildings. This probably reflects the greater emission of these compounds from new building materials. Others have found that the level of VOCs is particularly high in new buildings [53]. New and renovated buildings contain high levels of VOCs which decay appreciably over about 6 months [40].

Another source of BTEX is the products used for

cleaning which was demonstrated by measurements made before and after using them in a non-smoking office. This result is in agreement with Kim et al. [45] who found that higher concentrations of benzene, toluene, ethylbenzene, *m,p*-xylene and *o*-xylene were recorded after cleaning products were used in homes. The frequency of house cleaning has also been identified as a cause of higher benzene, toluene and ethylbenzene concentrations in homes [34].

When the correlation coefficients between the indoor concentrations of BTEX compounds in non-smoking and smoking offices (Table 5) are examined, significant positive correlations are seen between the concentrations of toluene, ethylbenzene, *m,p*-xylene and *o*-xylene in non-smoking offices. On the other hand, benzene was only weakly positively correlated with toluene, ethylbenzene, *m,p*-xylene and *o*-xylene. This is suggestive of the contribution of smoking to benzene levels. Significant positive correlations were found between the concentrations of individual BTEX in smoking offices. Similarly, others have found significant positive correlations between the concentrations of individual aromatic hydrocarbons in offices [33,45].

Conclusion

The mean indoor HCHO concentration has been found to be higher than the outdoor level at six offices where measurements were made. Of the many measurements made inside the six offices, 13.33%, 20% and 16.67% of the concentrations in the non-smoking, smoking and total offices, respectively, exceeded the maximum concentration of 0.1 ppm formaldehyde recommended by ASHRAE. Similarly, the mean indoor concentrations of toluene, ethylbenzene and the isomeric xylenes in all the offices were higher than outdoor levels, again indicating indoor sources. The exception was benzene whose indoor mean concentration was lower than the outdoor level. Maximum indoor concentrations of HCHO and BTEX were found in new non-smoking offices, while the minimum concentrations were recorded in an old office. Formaldehyde and BTEX levels in smoking offices were higher than those found in non-smoking offices. Higher concentrations of the VOCs were found after using cleaning products. These results indicate that the age of the office building, smoking activities and using of cleaning products are the major factors affecting the indoor levels of HCHO and BTEX.

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