Risk assessment of exposure to volatile organic compounds in the holy city of Makkah

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Abstract

Millions of pilgrims arrive in Makkah every year to perform Hajj and/or Umrah. The increase of pilgrim numbers is accompanied by the increase of their daily activities as well as the increase in the demands of transportation means. Consequently, considerable quantities of either gaseous or solid pollutants are emitted to the atmosphere. The present study focuses on volatile organic compounds (VOCs) where there is a lack of studying VOCs as air toxins and it is considered as the first of its kind in Saudi Arabia. The goals of the current work aimed to: I) develop adequate air monitoring technology (methodology) for VOCs; II) assess the life time of cancer and non-cancer risk; and III) enhance research capacity on VOCs air pollution and health risk analysis.

The ambient VOCs samples (during Hajj season 1431H) were collected from different sites (Al-Shbaikah, Al-Aziziah and Mina). Fourteen (14) VOCs compounds (benzene, toluene, ethylbenzene, p-xylene, m-xylene, o-xylene, 1,2,4-trimethylbenzene, vinyl chloride, styrene, chlorotolune, 1,2-dichlorobenzene, 1,3-dichlorobenzene, 1,2,4-trichlorobenzene and 1,2,3-trichlorobenzene) were targeted in this study. The mean concentrations of most abundant group of VOCs; Benzene, Toluene, Ethylbenzene, and Σ Xylene in El Shebaka were 8.11; 15.85; 9.20; 8.45 $\mu g/m^3$, respectively during day time and 31.44; 37.80; 42.03 and 38.71 $\mu g/m^3$ during night time, respectively. In Al-Aziziah, the values measured were 11.75; 9.42; 48.8; and 33.49 $\mu g/m^3$ during day time, meanwhile, during night time were 4.78; 6.67; 8.45; and 18.56 $\mu g/m^3$, respectively. The concentrations of benzene, toluene, ethylbenzene, xylenes in the air were significantly higher than other species.

The lifetime cancer and noncancer risks for the population groups exposed to VOCs were also assessed in detail. Noncancer risk was estimated in relation to

WIT Transactions on Ecology and The Environment, Vol 162, © 2012 WIT Press
www.witpress.com, ISSN 1743-3541 (on-line)
doi:10.2495/EID120541

the reference hazard level of 1.0. It was found that benzene may be the most important cause of both cancer and noncancer risk.
Keywords: ambient volatile organic compounds (VOCs), Makkah, Hajj, air quality.

1 Introduction

Urban air pollution is rapidly becoming an environmental problem of public concern during the last few decades [1]. Volatile organic compounds (VOCs) are organic chemicals and a major group of pollutants, and receive increasing concern worldwide [2]. However, all organic compounds contain carbon, and organic chemicals are the basic chemicals found in all living things and in all products derived from living things. VOCs include gasoline, industrial chemicals such as benzene, solvents such as toluene and xylene, and perchloroethylene (principal dry cleaning solvent) [2]. Sources of VOCs emission are both anthropogenic and natural. The major anthropogenic sources in the urban environment are vehicle exhaust, gasoline evaporation, emissions from the use of solvents, and gas leakage from natural gas and liquefied petroleum gas (LPG) [3].

VOCs are hazardous air pollutants, which cause various acute health problem as well as carcinogenic risk. Under this situation, the current study considers VOCs as an important issue in ambient environmental matters. National Environmental authorities have instructed plans to implement countermeasure for VOCs [3–9]. In addition, the presence of VOCs in the atmosphere plays an important role in the formation of ground level ozone, photochemical oxidants and smog episodes [10]. Exposure to the ambient air pollution poses a high risk to urban population. The sensitive groups (children and elderly) are certainly more vulnerable from the resulting health effects [8, 11, 12]. Besides, many street vendors who are working on the streets exposes them to continuous air pollution from traffic including toxic pollutants such as PAHs, VOCs and fine particulate matters ($PM_{2.5}$) [12]. These people thus have much higher exposure to ambient air pollution both in terms of concentration and time period [1].

Many studies, [1, 7, 8, 13–24], were conducted and examined the occupational exposure to VOCs from gasoline vapor emissions. Accordingly, it is common that the workers are exposed to highly elevated VOCs levels compared with ambient levels by inhalation, ingestion, and dermal contact. Most of the toxicants assessed are VOCs that remain as gases when emitted into the air. These compounds are not subject to appreciable deposition to soil, surface waters, or plants. Therefore, human exposure does not occur to any appreciable extent via ingestion or dermal exposure.

The current study is conducted in three locations in the city of Makkah (Al-Shbaikah, Al-Aziziah, and Mina) during Hajj season 1431H. This study is considered the first of its kind to be performed in Saudi Arabia to evaluate concentration and risk assessment. Herein only four VOC compounds (Benzene, Toluene, Ethylbenzene and Xylene (BTEX)) are being considered.

2 Materials and methods

2.1 Site description

The Holy City of Makkah is located at 80 km from the Red Sea (Latitude 19° 25' 21'' North Meridian 46° 49' 39"); the average elevation is approximately 277 m above sea level. The city is situated between mountain ranges, which control the contemporary expansion of the urban areas; however and due to its importance the city has dense population and nearly accommodates 1,700,000 [25]. The city centre is around the Holy Mosque (Haram), which altitude is lower than most of the city. Three sampling locations were selected in the present study as shown in Fig. 1. The ambient air samples were collected from the selected sites (Al-Shbaikah near to Haram; Al-Aziziah; and Mina) to represent different activities during Hajj season of 1431H.

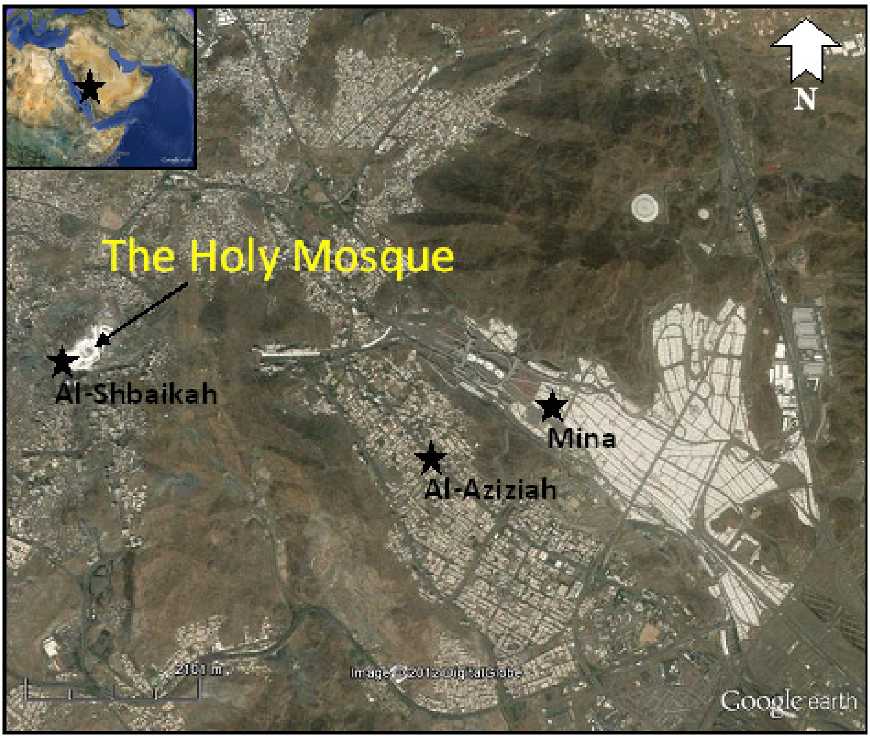

Figure 1: Map of Makkah showing the sampling locations.

2.2 Sampling and analysis

Ambient VOCs samples were collected according to a standard method developed by using activated charcoal tubes (ORBOTM-32 activated coconut charcoal (20/40)) for the collection [26, 27]. The collection tubes (Supelco, Inc., Bellefonte, PA) contained 150 mg of coconut charcoal were sub-divided into two portions of 100 and 50 mg: the front portion of 100 mg was used to collect the VOCs, while the 50 mg backup section was intended to determine if solvent breakthrough occurred from the front section. Air was drawn through the charcoal tubes, using a pump calibrated to draw 0.20 L/min. After sampling, the tubes were closed and wrapped in foils and identified by labels. Maximum duration time in refrigerator was 5 days, separated from other samples. During the sampling procedure, one charcoal tube was opened at the sample site and then the ends capped, which served as a blank. The samples and blanks tubes were put into special plastic bags that were tightly closed and kept at -10 °C in a freezer until analysis, not more than 15 days. BTEX compounds were extracted from activated charcoal using 1 ml carbon disulphide (CS_2). Operational parameters were optimized and quantitative recovery was obtained using CS_2 as the extraction solvent [26, 27]. Determination of BTEX compounds was carried out by gas chromatography using a flame ionization detector (GC/FID).

2.3 Health risk assessment

The health risk assessment focused on chronic exposure to compounds that may cause cancer or other toxic effects, rather than on acute toxicity. The main exposure route of interest was inhalation [6]. The inhalation intake was calculated by averaging daily intake over the exposure period. The carcinogenic and non-carcinogenic intakes of VOCs for gas service station workers were calculated as shown in eqn. (1):

$$I = (C \times ET \times EF \times ED) / AT \qquad (1)$$

where I is the inhalation intake (μg/m^3), C is the concentration of the compound in the personal air sample (μg/m^3), ET is the exposure time (hr/day), EF is the exposure frequency (days/year), ED is the exposure duration (years), and AT is an average lifetime (years). Inhalation exposure is always related to exposure frequency, duration, and quantity (dose) and activity pattern. The inhaled compounds were assumed to be totally absorbed for risk calculations in some studies [6, 28]. In this assessment, risk estimates for VOCs with a cancer endpoint were expressed in terms of the probability of developing cancer from a lifetime of continuous exposure to VOCs. The lifetime cancer risk was estimated using eqn. (2):

$$\text{Cancer risk} = I\ (\mu g/m^3) \times \text{cancer unit risk factors}\ (\mu g/m^3)^{-1} \qquad (2)$$

The non-cancer risk is expressed in terms of the hazard quotient (HQ) as shown in eqn. (3), which is the estimated ground level concentration divided by the reference exposure level (REL) for a single substance and a particular endpoint. The REL is an exposure level at, or below which, no non-cancer adverse health effect is anticipated to occur in a human population exposed for a specific duration [13]. The non-cancer health impacts were expressed as the hazard index (HI) as shown in eqn. (4), which is determined by calculating the HQ for a compounds and summing all of the HQ at investigated location [29].

$$HQ = I\ (\mu g/m^3)\ /\ RELs\ (\mu g/m^3) \quad (3)$$

$$HI = HQ_1 + HQ_2 + HQ_3 + \ldots\ HQ_n \quad (4)$$

Table 1 summarizes the exposure and risk assessment factors Risk characterization required combining the estimated exposure concentrations with toxicity data and toxicity values for BTEX compounds to provide a quantitative estimate of the potential health impacts. The potential for adverse effects was increased as exposures further exceed the reference dose.

Table 1: The exposure and risk assessment factors.

Exposure settings		Value	Source
Exposure time (ET)		24 h /day	Surveys
Exposure frequency (EF)		15 day /year	
Exposure duration : carcinogenic (ED)		2 year	
Exposure duration : non-carcinogenic (ED)		2 year	
Average life time : carcinogenic (AT)		70 year	[13]
Average life time : non-carcinogenic (AT)		70 year	
Cancer Unit Risk Factors for BTEX		2.9E-05 ($\mu g/m^3$) -1	
Chronic Inhalation Reference Expose Levels (RELs)	benzene	6.0E+01 $\mu g/m^3$	
	toluene	3.0E+02 $\mu g/m^3$	
	ethyl benzene	2.0E+03 $\mu g/m^3$	
	xylene	7.0E+02 $\mu g/m^3$	

Inhalation rates were taken into account when studying dose-response relationships and in developing the screening values. Exposure doses from inhalation of BTEX in air were calculated as shown in eqn. (5):

$$D = (C\ X\ IR\ X\ EF)\ /\ BW \quad (5)$$

where, D is exposure dose (mg/kg/day), C is contaminant concentration (mg/m^3), IR is intake rate (m^3/day), EF is exposure factor (unit less) and BW is body weight (kg). Default air intake rates were: 4.5 m^3/day for infant (less than 1 year), 10 m^3/day for child (6-8 years), 12 m^3/day for girl (12-14years), 15 m^3/day for boy (12-14 years), 11.3 m^3/day for female (19-65+ years) and 15.2 m^3/day for male (19-65+ years) [30].

3 Results and discussion

There was a lack on the information and data in Saudi Arabia regarding to VOCs concentration and level in ambient air, therefore, the present study was designed to fill the gap and suggested further research studies for such new hazardous air pollutants in the holy city. Consequently, the current study was surveyed some published data and literature to draw a picture about the levels of VOCs in ambient air in different parts of the world.

Fourteen (14) VOCs compounds (benzene, toluene, ethylbenzene, p-xylene, m-xylene, o-xylene, 1,2,4-trimethylbenzene, vinyl chloride, styrene, chlorotolune, 1,2-dichlorobenzene, 1,3-dichlorobenzene, 1,2,4-trichlorobenzene and 1,2,3-trichlorobenzene) were targeted in this study. But risk assessment calculation was concerned on only BTEX compounds. Statistical analysis of the most abundant group of VOCs (BTEX); Benzene, Toluene, Ethylbenzene, and Xylene in the three studied locations are shown in the table 2.

Table 2: The mean VOCs concentrations (μg/m^3) at investigated sites.

site		Al-Shbaikah	Al-Aziziah	Mina	Mean	Min.	Max.
Benzene	am	8.1	11.8	3.2	7.7	3.2	11.8
	pm	31.4	4.8	34	23.4	4.8	34
Toluene	am	15.9	9.4	8.1	11.1	8.1	15.9
	pm	37.8	6.7	52.4	32.3	6.7	52.4
Ethyl benzene	am	9.2	18.8	6.2	11.4	6.2	18.8
	pm	42	8.5	35.5	28.7	8.5	42
Xylene	am	2.8	11.2	1.9	5.3	1.9	11.2
	pm	12.9	6.2	21.5	13.5	6.2	21.5
Total VOCs	am	47.3	91.7	26.9	55.3	26.9	91.7
	pm	197.4	51.6	219.9	156.3	51.6	219.9

Max.; maximum value; Min.: minimum value; am: day-time; pm: night-time

Table 2 shows that BTEX concentrations were higher at Mina than those found in Al-Shbaikah and Al-Aziziah during night-time and lower during day-time. The observed levels of BTEX in Al-Shbaikah was due to the high traffic density, since this site was very close to the holy mosque surrounded by many street intersections and roads leading to Haram in addition to the most commercial and heaviest traffic areas in Makkah. The maximum concentrations of total VOCs were found at Mina site (219.9 μg/m^3) during night-time. The higher levels of VOCs in Mina area was due to the highest activity of Hajj in the valley where pilgrims move to Mina during day-time and reached to the highest density at night-time. Compared to Al-Shbaikah and Al-Aziziah, total VOCs concentrations in Mina during day-time of Hajj days were the lowest at Mina area. In contrast, the concentrations of total VOCs were highest than other two sites during night-time of Hajj days. These concentrations were mostly caused by traffic. Therefore, the increase in traffic density will increase both exhaust and vehicle evaporative emission [14, 31].

In this study, most VOCs Concentrations were higher than other concentration reported in different countries. In China (Beijing city), the mean mass concentration of total VOCs was 132.6 μg/m^3. This value was mostly lower than most of the current results [21]. In general, comparison between the organic compounds obtained in this study was extremely higher than later research.

Table 3 expresses the VOCs risk assessment calculations at investigated sites. In this table, it can be noticed that the mean inhalation intake of VOCs were 159.9, 232.2, 205.1 and 96.7 μg/m^3 for Benzene, Toluene, Ethyl benzene and Xylene, respectively. The inhalation intakes of VOCs varied from place to place. The minimum inhalation intake of VOCs was found at Mina site (19.0 μg/m^3) during day-time. While, the maximum inhalation intake of VOCs was found at Al-Shbaikah site (432.3 μg/m^3) during night-time.

Table 3: The VOCs risk assessment calculations at investigated sites.

	site	Al-Shbaikah		Al-Aziziah		Mina		
parameter	VOCs	am	pm	am	pm	am	pm	Mean
inhalation intake (ug/m^3)	benzene	83.4	323.4	120.9	49.2	32.5	350.0	159.9
	toluene	163.0	388.8	96.9	68.6	83.0	538.9	223.2
	Ethyl benzene	94.6	432.3	193.4	86.9	63.8	365.3	206.1
	Xylene	29.0	132.7	114.8	63.7	19.0	221.1	96.7
cancer risk	benzene	0.002	0.009	0.004	0.001	0.001	0.010	0.005
	toluene	0.005	0.011	0.003	0.002	0.002	0.016	0.006
	Ethyl benzene	0.003	0.013	0.006	0.003	0.002	0.011	0.006
	Xylene	0.001	0.004	0.003	0.002	0.001	0.006	0.003
	Total	0.011	0.037	0.015	0.008	0.006	0.043	0.020
Hazard Quotient	benzene	1.39	5.39	2.01	0.82	0.54	5.83	2.66
	toluene	0.54	1.30	0.32	0.23	0.28	1.80	0.74
	Ethyl benzene	0.05	0.22	0.10	0.04	0.03	0.18	0.10
	Xylene	0.04	0.19	0.16	0.09	0.03	0.32	0.14
	HI	2.02	7.09	2.60	1.18	0.88	8.13	3.65

Also, Table 3 shows that cancer risk of BTEX ranged between 0.001-0.013 (at Al-Shbaikah), 0.001-0.005(at Al-Aziziah) and 0.001-0.016 (at Mina). Hazard Quotient (HQ) for noncancer risk was found to be 2.66 for Benzene, 0.74 for Toluene, 0.10 for Ethyl benzene and 0.14 for Xyelene. The ratio of the potential exposure to the BTEX was anticipated to show that there were no adverse effects. Moreover, if the calculated HQ was equal or less than 1 ($HQ \leq 1$), then no adverse health effects were expected as a result of exposure. However, if the HQ was greater than 1 ($HQ > 1$), then adverse health effects were possible. Therefore, the HQ cannot be translated to a probability that adverse health effects would occur and it was unlikely to be proportional to risk. It was especially important to note that an HQ exceeding 1 does not necessarily mean that adverse effects will occur.

The risk of health effects from inhaling BTEX were classified into: short-term (acute) exposures as hours to days or long-term (chronic) exposures as years to even lifetime. Breathing low levels of VOCs for long periods of time may increase some people's risk of health problems. Several studies, [4, 5, 8, 14, 21, 26, 32–40] suggest that exposure to VOCs may make symptoms worse in people who have asthma or are particularly sensitive to chemicals. These were much different exposures than occupational exposures to VOCs. Common symptoms of exposure to VOCs included: i) Short-Term (Acute) to high levels of VOCs

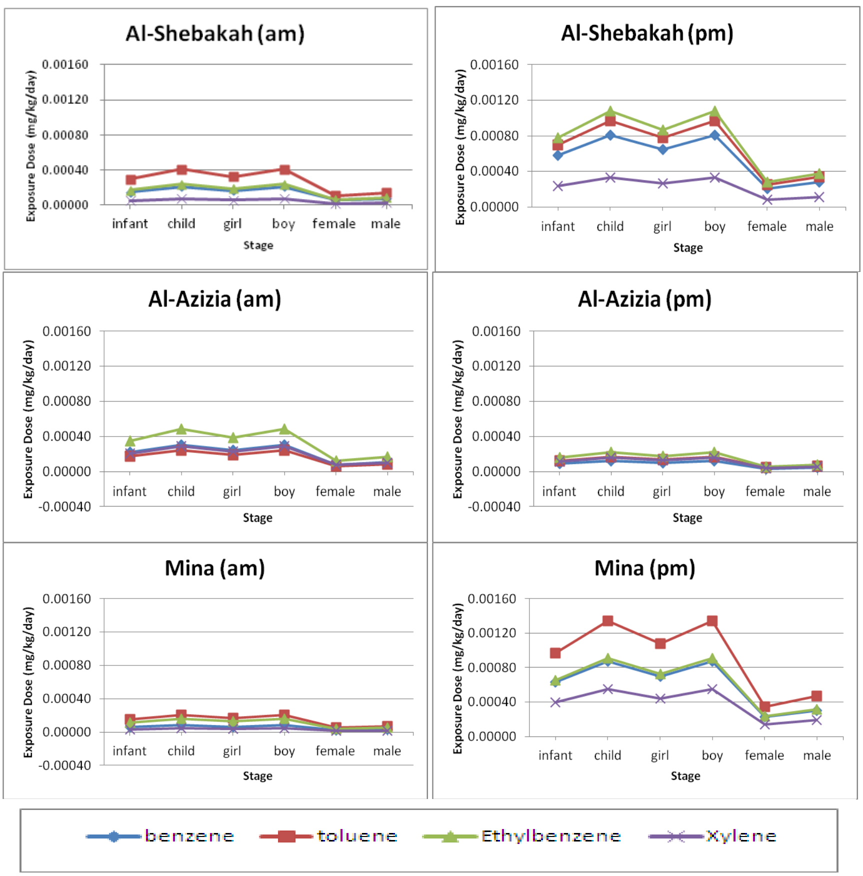

Figure 2: Exposure dose (mg/kg/day) at investigated sites (Al-Shbaikah, Al-Aziziah and Mina) for different stages (infant, child, girl, boy, female and male).

(Eye, nose and throat irritation, Headaches, Nausea/Vomiting, Dizziness, immune effects in infants or children and Worsening of asthma symptoms. ii) Long-Term (Chronic) to high levels of VOCs Increased risk of Cancer, Liver damage, Kidney damage and Central Nervous System damage [8, 11, 12].

Fig. 2 shows that the exposure dose at investigated sites. It was found that exposure dose ranged between 0.00003 to 0.00097 mg/kg/day for infant (less than 1 year), from 0.00005 to 0.00135 mg/kg/day for child (6-8 years), from 0.00004 to 0.00108 mg/kg/day for girl (12-14 years), from 0.00005 to 0.00135 mg/kg/day for boy (12-14 years), from 0.00001 to 0.00035 mg/kg/day for female (19-65+ years) and from 0.00002 to 0.00047 mg/kg/day for male (19-65+ years). Moreover, this figure was highlighted that the maximum exposure dose for female (0.00001 mg/kg/day), was recorded in Mina site during day-time. While, the minimum exposure dose for child (0.00135 mg/kg/day), was recorded in the same site (Mina) during night-time.

4 Conclusions

The current study indicated that exposure to aromatic VOCs is mainly influenced by different activities in the holy city. Moreover, risk assessment and exposure dose for BTEX in the city of Makkah was found to be within guidelines levels. Therefore, it is highly important to study risk assessment (inhalation intake, cancer risk and noncancer risk) for VOCs levels in indoor where people spend most of their time at home, offices, malls and closed buildings. International studies and Environmental Protection Agency (EPA) found concentrations of VOCs in indoor air commonly to be 2 to 5 times greater than an outdoor air and sometimes far greater. Also, it is recommended to study VOCs over one complete year indoor and outdoor to investigate the seasonal variations.

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