

## Assessing the Levels and Health Risk of Atmospheric Formaldehyde in Makkah, Saudi Arabia

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### Abstract

Atmospheric Formaldehyde (HCHO) was monitored in four sites on the Holy Mosque of Makkah, Saudi Arabia during August, 2013. The daily mean concentrations of HCHO were ranged from 1.09-18.92  $\mu\text{g}/\text{m}^3$ . The levels of HCHO were significantly higher than the permissible exposure limit (0.042  $\mu\text{g}/\text{m}^3$ ) of the Occupational Safety and Health Administration (OSHA). However, it were not exceeded the recommended exposure limit of the National Institute for Occupational Safety and Health (20  $\mu\text{g}/\text{m}^3$ ) and Egyptian law 4/1994 (0.37  $\text{mg}/\text{m}^3$ ). Spatial variations of HCHO concentrations most probably due to variations in local meteorology and traffic flow, which was considered the main source of emissions. Exposure doses for various age groups were estimated, which ranged from 0.000004 to 0.000259  $\text{mg}/\text{kg}/\text{day}$ . Maximum exposure dose was recorded for boys (age 12-14 years) and children (6-8 years) and minimum for females (19-65 years).

**Keywords:** formaldehyde; vehicle emissions; risk assessment; The Holy Mosque; Makkah.

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### 1. Introduction

Formaldehyde (HCHO) is the simplest form of the organic compounds known as aldehyde. In urban areas formaldehyde is emitted by combustion sources mainly vehicles emissions; it poses a threat to human health. At concentrations lower than Recommended Exposure Limit (REL) (20  $\mu\text{g}/\text{m}^3$ ) of the National Institute for Occupational Safety and Health (NIOSH), HCHO can cause irritation in the respiratory tract, eyes and skin, whereas it is considered a toxic substance at concentrations higher than REL (OSHA, 1988). It is also reported that long-term exposure to HCHO can lead to cancer (OSHA, 1988). Human exposure to HCHO is listed by the US Environmental Protection Agency (USEPA) as a priority for regular assessment (USEPA, 2006).

HCHO emissions have been investigated by several authors (Proctor and Hughes, 1978; Blackwell *et al.*, 1981; NRC, 1981; Preuss *et al.*, 1985; Harrison and Perry, 1986; WHO, 1989). They concluded that HCHO is a primary product of fuel combustions, such as traffic emissions. In addition, they reported that atmospheric concentrations of HCHO in the close vicinity of industrial outlets or in areas of heavy smog ranged from 0.005 to 0.6 ppm (0.0061-0.74  $\text{mg}/\text{m}^3$ ) (Proctor and Hughes, 1978; Blackwell *et al.*, 1981; NRC, 1981; Preuss *et al.*, 1985; Harrison and Perry, 1986; OSHA, 1988; WHO, 1989). Vaught (1991) reported that a major source of HCHO in urban air was

incomplete combustion of hydrocarbon fuels used in power plants and vehicles. The International Agency for Research on Cancer (IARC, 1995) has reported that HCHO was one of the volatile compounds that formed in the early stages of decomposition of plant residues in the soil, and occurred naturally in fruit and other foods. Seinfeld and Pandis (1998) mentioned that atmospheric HCHO has a lifetime of approximately 4 hours (Seinfeld and Pandis, 1998).

The California Environmental Protection Agency (CEPA) reported that ambient HCHO concentrations were 1.9-4.7  $\mu\text{g}/\text{m}^3$  in Los Angeles (CEPA, 1999). However, Khoder *et al.* (2000) reported much higher concentration of HCHO (40  $\mu\text{g}/\text{m}^3$ ) in Cairo, Egypt (Khoder *et al.*, 2000). In Minnesota, USA Pratt *et al.* (2000) reported that HCHO concentrations ranged from 0.8 to 2.9  $\mu\text{g}/\text{m}^3$  (Pratt *et al.*, 2000). Environment Canada (2001) reported similar levels in remote areas of Canada (EC, 2001). Nguyen *et al.* (2001) and Grosjean *et al.* (2002) reported that HCHO concentrations ranged between 10.7 and 32  $\mu\text{g}/\text{m}^3$  in Rio de Janeiro, and 2.8  $\mu\text{g}/\text{m}^3$  in São Paulo, Brazil (Nguyen *et al.*, 2001; Grosjean *et al.*, 2002). Maître *et al.* (2002) mentioned that the concentration of HCHO in the centre of Grenoble, France was 14 and 21  $\mu\text{g}/\text{m}^3$  in summer and winter, respectively. In Los Angeles, short-term measurements (2 hours) showed that HCHO levels range was 7-20  $\mu\text{g}/\text{m}^3$  in urban and 11-15  $\mu\text{g}/\text{m}^3$  at roadside monitoring sites (EC, 2003). In contrast, short

term measurements in Sacramento showed somewhat lower ambient concentrations in urban areas (2-4  $\mu\text{g}/\text{m}^3$ ) and roadside locations (4-6  $\mu\text{g}/\text{m}^3$ ) (EC, 2003). Furthermore, the Canadian National Air Pollution Surveillance system showed that the HCHO concentration was 2.4  $\mu\text{g}/\text{m}^3$  in urban areas and 1.5  $\mu\text{g}/\text{m}^3$  in rural areas (EC, 2003). Pohanish (2002) and IARC (2004) mentioned that HCHO was used in a number of industrial processes and was released during the curing of certain adhesives, especially those used in laminated wood products and household products. Zielinska *et al.* (2004) reported that HCHO levels in Arizona was 0.8-4.4  $\mu\text{g}/\text{m}^3$  in urban areas and 1.3  $\mu\text{g}/\text{m}^3$  in rural areas. Chiu *et al.* (2005) reported that HCHO levels in Taiwan ranged from 4.8 to 10.9  $\mu\text{g}/\text{m}^3$ . Furthermore, Báez *et al.* (2003) found that HCHO concentrations ranged from 5 to 44  $\mu\text{g}/\text{m}^3$  in urban areas in Mexico City. Also, in the U.S., the National-Scale Air Toxics Assessment (NATA) reported that the annual mean concentrations of atmospheric HCHO ranged from 0 to 49  $\mu\text{g}/\text{m}^3$ , with an overall national mean concentration of (4.3  $\mu\text{g}/\text{m}^3$ ) (USEPA, 2006b).

IARC (2005) showed that short-term exposure of HCHO at levels between 0.5 and 1 ppm (0.62-1.23  $\text{mg}/\text{m}^3$ ) led to irritations of eyes, nose and throat. HCHO is a sensitizer and can cause allergic contact dermatitis and asthma. Exposure to formaldehyde level of up to 3 ppm (3.69  $\text{mg}/\text{m}^3$ ) is unlikely to provoke asthma in an un-sensitized individual. In contrast, exposure to HCHO concentration < 0.02-5 ppm (< 0.03-6.15  $\text{mg}/\text{m}^3$ ) has shown to cause transient, reversible declines in lung function, but there is no evidence that HCHO induces a chronic decrement in lung function (IARC,

2005).

The main aim of the current work was to assess the levels of HCHO, mainly emitted by traffics, and investigate its effects on human health at the Holy Mosque in Makkah.

## 2. Materials and methods

### 2.1. Site description

Samples were collected in the south squares of the Holy Mosque, Makkah, Saudi Arabia (Fig.1). Brief descriptions of the monitoring sites are given in Table 1.

The concentrations of atmospheric HCHO are measured for a short term (2 hours) at four locations in the south squares of the Holy Mosque, Makkah, Saudi Arabia. The results of this study are compared with the results obtained in other countries and with the permissible exposure limit (PEL) for HCHO in urban areas set by WHO (World Health Organization), Environment Canada, National Institute for Occupational Safety and Health (NIOSH) and Occupational Safety and Health Administration (OSAH).

### 2.2. Sampling and analysis of formaldehyde

Four sampling sites were selected in the south squares of the Holy Mosque, Makkah, Saudi Arabia (as shown in Fig. 1). HCHO concentration was measured for a short-term (2 hours) during August, 2013. A calibrated vacuum pump was used to collect air samples at the rate of 2 L/min. The concentration of total



Figure 1. Showing sampling sites in the south squares of the Holy Mosque, Makkah, Saudi Arabia.

Table 1. Sampling sites and their characteristics

Site No.	Brief description
Site 1.	Upper small market tunnel, in center of the south squares of the Holy Mosque. Generally a very busy location in terms of prayers. High density of prayers was present during sampling period.
Site 2.	Upper small market tunnel, downwind from the south squares of the Holy Mosque and situated near bathrooms. Had a high density of prayers during sampling period.
Site 3.	Upper small market tunnel, upwind from the south squares of the Holy Mosque. Had high density of prayers during sampling period.
Site 4.	Lower south squares of the Holy Mosque. Likewise a busy location and had a high traffic density during sampling period.

aldehyde was calculated in terms of HCHO (Perry and Young, 1977). Air was passed (2 L/min) through a glass bubbler sampler containing 50 ml of absorbing solution. The absorbed HCHO was determined instantaneously calorimetrically at 628 nm using UV/VIS 1800 Spectrophotometer, Shimadzu, with a blank reagent as a reference. HCHO in ambient air was collected in a 0.05% aqueous solution of 3-methyl-2-benzothiazolone hydrozone hydrochloride (MBTH). The resulting azine was then oxidised by oxidising reagent (ferric chloride - sulphamic acid solution) to form a blue cationic dye in acid solution which can be measured calorimetrically at 628 nm. The determination of HCHO was carried out by the colorimetric method by adding 2 ml of oxidising reagent to 10 ml of an absorbing sample solution. The blank solution was prepared by the same procedure using unexposed absorbing reagent. The flask was allowed to stand for 12 minutes until complete colour was developed. The absorbance was measured at 628 nm against the blank. The concentration of HCHO expressed as  $\mu\text{g}/\text{m}^3$  was calculated from the standard curve and volume of air.

### 2.3. Health risk assessment

The health risk assessment focused on chronic exposure to HCHO, which are related to long term health impacts, such as cancer or other toxic effects, rather than on acute toxicity. The main exposure route of interest was inhalation (Muller *et al.*, 2003). The inhalation intake was calculated by averaging daily intake over the exposure period. To calculate the inhalation intake this study follows the methodology developed by CEPA (2003) as shown in eqn. (1):

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

Where I is the Inhalation intake ( $\mu\text{g}/\text{m}^3$ ), C is the Concentration ( $\mu\text{g}/\text{m}^3$ ) of the compound (here HCHO), 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 Time (lifetime in 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 (Muller *et al.*, 2003; Hoddinott and Lee, 2000). In this study risk assessment for formaldehyde was expressed in terms of the probability of developing cancer assuming continuous lifetime exposure to HCHO. The lifetime cancer risk was estimated using eqn. (2) (CEPA, 2005):

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

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 (CEPA, 2005). The non-cancer health impacts were expressed as the hazard index (HI) as shown in eqn. (4), which is the sum of HQs at various locations (CEPA, 2003).

$$HQ = I (\mu\text{g}/\text{m}^3) / \text{RELS } (\mu\text{g}/\text{m}^3) \quad (3)$$

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

Generally no health effects are likely at a HQ value less than or equal to 1 ( $HQ \leq 1$ ), however at a HQ value greater than 1 ( $HQ > 1$ ), there is a possibility that adverse health effect will occur. Table 2 summarises various exposure and risk assessment factors, adopted in this study.

Inhalation rates were taken into account when studying dose-response relationships and in developing the exposure doses. Exposure doses from inhalation of HCHO were calculated as shown in eqn. (5) (EPA, 1997):

Table 2. The exposure and risk assessment factors

Exposure settings	Value	Reference
Exposure time (ET)	2 h /day	
Exposure frequency (EF)	8 day /year	Surveys
Exposure duration: carcinogenic (ED)	2 year	
Exposure duration: non-carcinogenic (ED)	2 year	
Average life time: carcinogenic (AT)	70 year	
Average life time: non-carcinogenic (AT)	70 year	(CEPA, 2005)
Cancer Unit Risk Factors for BTEX	0.000006 ( $\mu\text{g}/\text{m}^3$ ) <sup>-1</sup>	
Chronic Inhalation Reference Exposure Levels (RELs)	94 $\mu\text{g}/\text{m}^3$	

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

Where, D is exposure Dose (mg/kg/day), C is HCHO Concentration ( $\text{mg}/\text{m}^3$ ), IR is Intake Rate ( $\text{m}^3/\text{day}$ ), EF is Exposure Factor (unit less) and BW is Body Weight (kg). Air intake rates used in this study were: 4.5  $\text{m}^3/\text{day}$  for infants (less than 1 year), 10  $\text{m}^3/\text{day}$  for children (6-8 years), 12  $\text{m}^3/\text{day}$  for girls (12-14 years), 15  $\text{m}^3/\text{day}$  for boys (12-14 years), 11.3  $\text{m}^3/\text{day}$  for females (19-65+ years) and 15.2  $\text{m}^3/\text{day}$  for males (19-65+ years) (EPA, 1997).

Risk for exposure doses from inhalation of HCHO in air were calculated as shown in eqn. (6) (PCS, 2005):

$$\text{Risk} = D \times \text{CSF} \quad (6)$$

Where, D is exposure Dose (mg/kg/day), CSF is Cancer Slope Factor of formaldehyde ( $2.1 \times 10^{-2}$ ) ( $\text{mg}/\text{kg}/\text{day}$ )<sup>-1</sup> (PCS, 2005).

### 3. Results and Discussion

Results showed that the mean concentrations ( $\mu\text{g}/\text{m}^3$ ) of HCHO recorded during the study period were (2.86-18.92), (1.64-10.42), (1.09-9.75) and (3.4-18.74) at site 1, 2, 3 and 4, respectively (Table 3). Even the minimum concentration (1.09  $\mu\text{g}/\text{m}^3$ ) of HCHO in the current study was higher than the Permissible Exposure Limit (PEL) recommended by Environment Canada (0.042  $\mu\text{g}/\text{m}^3$ ) (EC, 1999a), and WHO (0.05  $\mu\text{g}/\text{m}^3$ ) (WHO, 1977). However, HCHO levels were lower than recommended exposure limit of the National Institute for Occupational Safety and Health (20  $\mu\text{g}/\text{m}^3$  in 8-hour) (OSHA, 1988) and the Egyptian law 4/1994 (0.37  $\text{mg}/\text{m}^3$ ) (EEAA, 1994).

The relatively high concentrations of HCHO measured in the current study could be attributed to the location of sampling sites, where generally high traffic density inside the tunnel and around the Holy Mosque can be observed. The area is also surrounded by several very busy roads, for example Al-Kalil Ibrahim and the

Holy Mosque roads, where significant amount of air pollutants, including HCHO are emitted by the road traffics.

These results are in agreement with Zheng and Fang (2000) and Ravindra *et al.* (2006), who concluded that the lower atmospheric mixing heights, lower temperature, decreased photo-chemical oxidation, and traffic and soil volatilization contribute to the higher concentration of atmospheric pollutants during winter (Zheng and Fang, 2000; Ravindra *et al.*, 2006). Larsen and Larsen (1998), EC (2001) and AENV (2006) concluded that the lower concentrations of HCHO in summer could be explained by its short half-lifetime in hot weather due to reaction with sunlight (photolysis).

In Makkah the major factor responsible for the high emission of HCHO is probably the higher number of visitors to the Holy Mosque in Ramadan (Ummah season) that leads to higher traffic flow inside small market tunnel and on roads around the Holy Mosque. The high rate of emission coupled with low wind speeds and the frequent inversions in the sampling sites can result in high load of local air pollution. The Holy Mosque area has a very poor dispersion rates due to the presence of tall buildings and narrow streets around it, which results in the re-circulation of the pollutants like a typical street canyon.

Table 3 shows the HCHO risk assessment calculations at the 4 sampling sites. In this table, it can be noticed that the inhalation intake (I) of HCHO ranged (1.31-8.65), (0.75-4.76), (0.50-4.47) and (1.55-8.57)  $\mu\text{g}/\text{m}^3$  at site 1, 2, 3 and 4, respectively during the period of study. The inhalation intakes of HCHO varied from site to site, demonstrating significant spatial variations due to micro-climatic factors. The minimum inhalation intake of HCHO was found at site-3 (0.5  $\mu\text{g}/\text{m}^3$ ) on 7<sup>th</sup> August 2013, whereas the maximum inhalation intake of HCHO was recorded at site-1 (8.65  $\mu\text{g}/\text{m}^3$ ) on 14<sup>th</sup> August 2013.

Furthermore, table 3 shows that cancer risk of HCHO ranged from 0.000003 to 0.0000052 at sampling sites during the period of study. Hazard Quotient (HQ)

for non-cancer risk ranged between 0.01 and 0.09 during the period of study. In addition, hazard index (HI) was found to be 0.44, 0.24, 0.21, and 0.42, at site - 1, 2, 3, and 4, respectively. HQ values were less than 1 ( $HQ < 1$ ), indicating that no adverse health effects were likely as a result of HCHO exposure.

Exposure to HCHO may make symptoms worse in people who have asthma or are particularly sensitive to chemicals. Common symptoms of exposure to HCHO include: i) Short-Term (Acute) effects caused by high levels of HCHO include eye, nose and throat irritation, headaches, nausea/vomiting, dizziness, immune effects in infants or children and worsening of asthma symptoms. ii) Long-Term (Chronic) effects due to high levels of HCHO include increased risk of cancer, liver damage, kidney damage and central nervous system (ACGIH, 1991; 1994; 2003; CEP, 1993; Edgerton *et al.*, 1989; NIOSH, 1996; 2003; WHO, 1989; 2002; AQGE, 2000; IGWQ, 2003; Nasralla and Albar, 2005; EHSTSG, 2007; Khoder, 2007; Liu *et al.*, 2008).

Table 3 shows the average exposure dose for different population groups, which ranged (0.000040-0.000154), (0.000021-0.000083), (0.000019-0.000073) and (0.000038-0.000149) mg/kg/day at site- 1, 2, 3, and 4, respectively. Maximum exposure dose was

recorded for boys and children (0.000259 mg/kg/day) at site-1 on 14<sup>th</sup> August 2013, whereas the minimum exposure dose recorded for females was 0.00004 mg/kg/day at site-3 on 7<sup>th</sup> August 2013. The average risks of exposure doses for various age groups are depicted in Fig. 2. This figure shows that the average risk was higher for boys and children (0.000032) and lower for females (0.000008).

Table 4 shows a comparison between mean concentrations of HCHO ( $\mu\text{g}/\text{m}^3$ ) reported in different studies around the world. The levels of HCHO demonstrated significant variations across the World, most probably due to variations in emissions sources, local topography and meteorological conditions. Correlation analysis between HCHO concentrations versus temperature and relative humidity showed negative correlation between temperature and HCHO concentrations and positive correlation between relative humidity and HCHO. The values of correlation coefficients for temperature and relative humidity were 0.52 and 0.58, respectively. High temperature and low relative humidity increase photo-dissociation of HCHO, which most probably decrease its atmospheric concentrations (Larsen and Larsen, 1998; AENV, 2006).

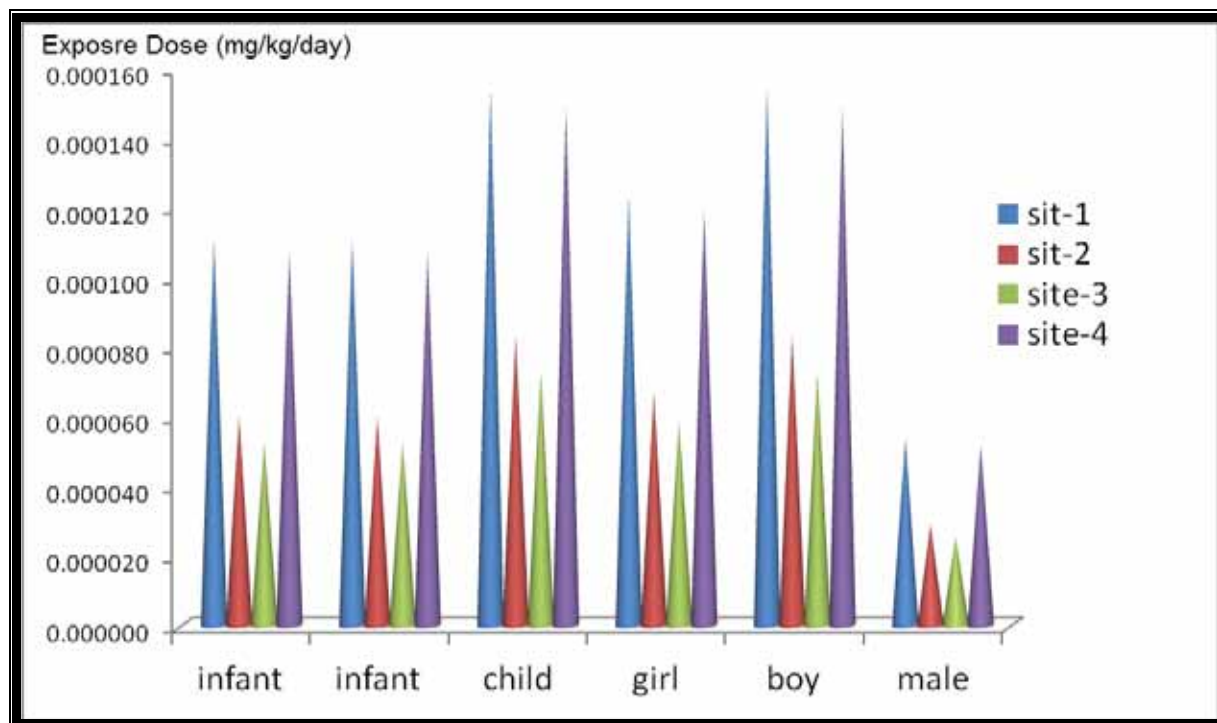


Figure 2. Risk of formaldehyde at the 4 sampling sites for different population groups, where infants (male and female) < 1 year, children (male and female) 6 – 8 years, girls and boys 12 – 14, and male and female 19 – 65 years.

Table 3. HCHO risk assessment calculations in four sampling sites at south squares of the Holy Mosque, Makkah city, Saudi Arabia

Site	Date	Conc. ( $\mu\text{g}/\text{m}^3$ )	I ( $\mu\text{g}/\text{m}^3$ )			Cancer risk	HQ	ED (mg/kg/day)			
			Infant	Child	Girl			Boy	Female	Male	
1	7 Aug. 2013	2.86	1.31	0.00008	0.01	0.00028	0.00039	0.00031	0.00039	0.00010	0.00010
	8 Aug. 2013	6.7	3.06	0.00018	0.03	0.00066	0.00092	0.00073	0.00092	0.00024	0.00030
	9 Aug. 2013	8.81	4.03	0.00024	0.04	0.00087	0.00121	0.00097	0.00121	0.00031	0.00040
	10 Aug. 2013	11	5.03	0.00030	0.05	0.00108	0.00151	0.00121	0.00151	0.00039	0.00050
	11 Aug. 2013	11.27	5.15	0.00031	0.05	0.00111	0.00154	0.00124	0.00154	0.00040	0.00050
	12 Aug. 2013	13.11	5.99	0.00036	0.06	0.00129	0.00180	0.00144	0.00180	0.00046	0.00060
	13 Aug. 2013	17.3	7.91	0.00047	0.08	0.00171	0.00237	0.00190	0.00237	0.00061	0.00080
	14 Aug. 2013	18.92	8.65	0.00052	0.09	0.00187	0.00259	0.00207	0.00259	0.00067	0.00090
	7 Aug. 2013	1.64	0.75	0.00004	0.01	0.00016	0.00022	0.00018	0.00022	0.00006	0.00010
	8 Aug. 2013	4.1	1.87	0.00011	0.02	0.00040	0.00056	0.00045	0.00056	0.00015	0.00020
	9 Aug. 2013	4.32	1.97	0.00012	0.02	0.00043	0.00059	0.00047	0.00059	0.00015	0.00020
	10 Aug. 2013	4.91	2.24	0.00013	0.02	0.00048	0.00067	0.00054	0.00067	0.00017	0.00020
	11 Aug. 2013	5.9	2.70	0.00016	0.03	0.00058	0.00081	0.00065	0.00081	0.00021	0.00030
	12 Aug. 2013	7.77	3.55	0.00021	0.04	0.00077	0.00106	0.00085	0.00106	0.00027	0.00040
13 Aug. 2013	9.47	4.33	0.00026	0.05	0.00093	0.00130	0.00104	0.00130	0.00034	0.00050	
14 Aug. 2013	10.42	4.76	0.00029	0.05	0.00103	0.00143	0.00114	0.00143	0.00037	0.00050	

I: inhalation intake, HQ: Hazard Quotient, ED: Exposure Dose

Table 3. HCHO risk assessment calculations in four sampling sites at south squares of the Holy Mosque, Makkah city, Saudi Arabia (Continue)

Site	Date	I		Cancer risk	HQ	ED (mg/kg/day)					
		Conc. ( $\mu\text{g}/\text{m}^3$ )	( $\mu\text{g}/\text{m}^3$ )			Infant	Child	Girl	Boy	Female	Male
3	7 Aug. 2013	1.09	0.50	0.000003	0.01	0.000011	0.000015	0.000012	0.000015	0.000004	0.000010
	8 Aug. 2013	2.88	1.32	0.000008	0.01	0.000028	0.000039	0.000032	0.000039	0.000010	0.000010
	9 Aug. 2013	3.88	1.77	0.000011	0.02	0.000038	0.000053	0.000043	0.000053	0.000014	0.000020
	10 Aug. 2013	4.37	2.00	0.000012	0.02	0.000043	0.000060	0.000048	0.000060	0.000015	0.000020
	11 Aug. 2013	4.92	2.25	0.000013	0.02	0.000049	0.000067	0.000054	0.000067	0.000017	0.000020
	12 Aug. 2013	7.01	3.20	0.000019	0.03	0.000069	0.000096	0.000077	0.000096	0.000025	0.000030
	13 Aug. 2013	8.56	3.91	0.000023	0.04	0.000084	0.000117	0.000094	0.000117	0.000030	0.000040
	14 Aug. 2013	9.75	4.46	0.000027	0.05	0.000096	0.000134	0.000107	0.000134	0.000034	0.000050
	7 Aug. 2013	3.4	1.55	0.000009	0.02	0.000034	0.000047	0.000037	0.000047	0.000012	0.000020
	8 Aug. 2013	7.1	3.25	0.000019	0.03	0.000070	0.000097	0.000078	0.000097	0.000025	0.000030
	9 Aug. 2013	8.42	3.85	0.000023	0.04	0.000083	0.000115	0.000092	0.000115	0.000030	0.000040
	10 Aug. 2013	10.16	4.64	0.000028	0.05	0.000100	0.000139	0.000111	0.000139	0.000036	0.000050
	11 Aug. 2013	10.46	4.78	0.000029	0.05	0.000103	0.000143	0.000115	0.000143	0.000037	0.000050
	12 Aug. 2013	11.56	5.28	0.000032	0.06	0.000114	0.000158	0.000127	0.000158	0.000041	0.000060
13 Aug. 2013	16.99	7.77	0.000047	0.08	0.000168	0.000233	0.000186	0.000233	0.000060	0.000080	
14 Aug. 2013	18.74	8.57	0.000051	0.09	0.000185	0.000257	0.000205	0.000257	0.000066	0.000090	

I: inhalation intake, HQ: Hazard Quotient, ED: Exposure Dose

Table 4. Mean concentrations ( $\mu\text{g}/\text{m}^3$ ) of HCHO reported in different studies around the world.

Location	Country	City	Sampling time	Conc.	Reference
	Saudi Arabia	Makkah	Aug. 2013	1.09-18.92	The current study
	Egypt	Cairo	Mar.-Aug. 1999	7.1-40	Khoder <i>et al.</i> , 2000; Sin <i>et al.</i> , 2001; Hu <i>et al.</i> , 2008
	Algeria	Algiers	2000	5.2-27.1	Hu <i>et al.</i> , 2008
	Japan	Osaka	2004	2.33	Sin <i>et al.</i> , 2001; Dutta <i>et al.</i> , 2009
	India	Kolkata	2008	19.75	Dutta <i>et al.</i> , 2009
	China	Hong Kong	2002	4.17	Sin <i>et al.</i> , 2001; Ho <i>et al.</i> , 2002; Hu <i>et al.</i> , 2008; Dutta <i>et al.</i> , 2009
	Finland	Hyytiälä	2004	0.48	Sin <i>et al.</i> , 2001; Dutta <i>et al.</i> , 2009
	Greece	Athens	2003	39.02	Dutta <i>et al.</i> , 2009
	Norway	Drammen	2000	8.9	Ofstedal <i>et al.</i> , 2003
	Spain	Madrid	1996	4.7-20	Hu <i>et al.</i> , 2008
	Sweden	Uppsala	1998	1.8	Sakai <i>et al.</i> , 2004
	USA	California	2002	2-4.3	CalEPA, 1997; Sin <i>et al.</i> , 2001; Dutta <i>et al.</i> , 2009
	Brazil	Rio de Janeiro	2 003	66.63	Sin <i>et al.</i> , 2001; Montero <i>et al.</i> , 2001; Corrêa <i>et al.</i> , 2003; Dutta <i>et al.</i> , 2009
	Mexico	Mexico City	Mar.-May 1995	2-63	Sin <i>et al.</i> , 2001; Baez <i>et al.</i> , 2003; Hu <i>et al.</i> , 2008
Roadside sites	China	HKPU, Hong Kong	Jan. to Feb. 2002	4.7	Sin <i>et al.</i> , 2001
	Brazil	Rio de Janeiro	Dec. 1998 to Jan. 2001	18.3	Hu <i>et al.</i> , 2008
	Finland	Eastern	May, Jul., Jan.	13.2	Hu <i>et al.</i> , 2008
Central reservation highway	Finland	Kuopio	2000	38.04	Dutta <i>et al.</i> , 2009
	Germany	Schauinsland	September 1992	0.006-3.22	Slemr <i>et al.</i> , 1996; Sin <i>et al.</i> , 2001
	Denmark	Lille Valby	May-July 1995	1.5	Sin <i>et al.</i> , 2001
Semi-rural and rural sites	Sweden	Birkenes	January 1992-December 1995	1.0	Sin <i>et al.</i> , 2001
	Canada	Central Ontario	July-August 1998	2.0	Sin <i>et al.</i> , 2001



#### 4. Conclusion

Assessing the levels of atmospheric HCHO is vital because of its effect on human health. This paper analyses HCHO concentration and its potential health impacts at four monitoring sites in the south squares of the Holy Mosque, Makkah, Saudi Arabia. Results showed that HCHO concentrations were higher than the exposure limits suggested by the Environment Canada and WHO. Furthermore, the concentrations were higher than the permissible exposure limit of Occupational Safety and Health Administration and lower than recommended exposure limit of the National Institute for Occupational Safety and Health. Considering emissions sources in the surrounding areas, traffic seem to be the main source of HCHO emission, especially the vehicles inside the small market tunnel near the monitoring sites. HQ values were lower than 1 (HQ < 1), suggesting no adverse health effects were likely to occur at the present level of HCHO concentration. Furthermore, the estimated average risk of formaldehyde was higher for children and boys, and lower for females. This investigation is based on limited data for a short period of time; therefore further work is required to provide a greater temporal and special coverage around the city of Makkah. Furthermore, source apportionment of various sources of HCHO and other air pollutant is required to identify the sources of emissions and quantify their contributions, which will be helpful in devising an air quality management plan for Makkah.

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#### References

ACGIH (American Conference of Governmental Industrial Hygienists). Documentation of the threshold limits values and biological exposure indices. 6<sup>th</sup> ed. Cincinnati, OH, 1991.

ACGIH (American Conference of Governmental Industrial Hygienists). Threshold limit values for chemical substances and physical agents and biological exposure indices. Cincinnati, OH, 1994.

ACGIH (American Conference of Governmental Industrial Hygienists). Guide to Occupational Exposure Values. Cincinnati, OH, 2003.

AENV (Alberta Environment). Assessment report on formaldehyde for developing ambient air quality objectives. Toxico-Logic Consulting Inc. February 2006.

AQGE (Air Quality Guidelines for Europe). World Health Organization Regional Office for Europe (WHO), Regional Publications, European Series, No. 91, 2<sup>nd</sup> ed. 2000.

Báez A, Padilla H, García R, Torres M del C, Rosas I, Belmont R. Carbonyl levels in indoor and outdoor air in Mexico City and Xalapa, Mexico. *Science of the Total Environment* 2003; 302(1-3): 211-26.

Blackwell M, Kang H, Thomas A, Infante P. Formaldehyde: evidence of carcinogenicity. *American Industrial Hygiene Association* 1981; 42(7): A34-A38.

CalEPA (California Environmental Protection Agency). Technical support document for the determination of noncancer chronic reference exposure levels. Draft for Public Comment. Office of Environmental Health Hazard Assessment, Berkeley, CA. 1997.

CEP (Canadian Environmental Protection Act). Priority substance list, assessment report, Xylenes. Government of Canada, Environment Canada, Health Canada. Canada Communication Group Publishing, 1993.

CEPA (California Environmental Protection Agency). Determination of acute reference exposure levels for airborne toxicants [monograph on the Internet]. Acute toxicity summary, formaldehyde, 1999 [last Updated, March 1999]. Available from: [http://www.oehha.ca.gov/air/acute\\_rels/pdf/50000A.pdf](http://www.oehha.ca.gov/air/acute_rels/pdf/50000A.pdf).

CEPA (California Environmental Protection Agency). The air toxics hot spots program guidance manual for preparation of health risk assessments. California, USA. 2003.

CEPA (California Environmental Protection Agency). Technical support document for describing available cancer potency factors. California, USA. 2005.

Chiu KH, Sree U, Tseng SH, Wu CH, Lo JG. Differential optical absorption spectrometer measurement of NO<sub>2</sub>, SO<sub>2</sub>, O<sub>3</sub>, HCHO and aromatic volatile organics in ambient air of Kaohsiung Petroleum Refinery in Taiwan. *Atmospheric Environment* 2005; 39(5): 941-55.

Corrêa SM, Martins EM, Arbilla G. Formaldehyde and acetaldehyde in a high traffic street of Rio de Janeiro, Brazil. *Atmospheric Environment* 2003; 37(1): 23-29.

Dutta C, Som D, Chatterjee A, Mukherjee AK, Jana TK, Sen S. Mixing ratios of carbonyls and BTEX in ambient air of Kolkata, India and their associated health risk. *Environmental Monitoring and Assessment* 2009; 148(1-4): 97-107

EC (Environment Canada). Canadian Environmental Protection Act - Priority Substances List - Supporting document for the environmental assessment of formaldehyde. Hull, Quebec, Environment Canada, Commercial Chemicals Evaluation Branch. 1999.

EC (Environment Canada). Canadian Environmental Protection Act, 1999. Priority Substances List Assessment Report: Formaldehyde, Ottawa, Ontario. 2001.

EC (Environment Canada). Annual Data Summary for 2002 [monograph on the Internet]. National Air Pollution Surveillance (NAPS) Network. Environment Canada. Ottawa, Canada. 2003. Available from: [http://www.etc-cte.ec.gc.ca/publications/naps/naps2002annual\\_updated.pdf](http://www.etc-cte.ec.gc.ca/publications/naps/naps2002annual_updated.pdf).

- Edgerton SA, Holdren MW, Smith DL, Shah JJ. Inter-urban comparison of ambient volatile organic compound concentrations in U.S. cities. *Journal of Air Pollution and Control Association* 1989; 39(5): 729-32.
- EEAA (Egyptian Environmental Affairs Agency). Egypt Environmental Protection Law. No. 4, 1994.
- EHSTSG (Environmental, Health, Safety, Transport and Storage Guidelines). Styrene Monomer, Belgium. 2007.
- EPA (Environmental Protection Agency). Exposure factors handbook [monograph on the Internet]. 1997. Available from: <http://www.epa.gov/ncea/pdfs/efh/front.pdf>.
- Grosjean D, Grosjean E, Moreira LF. Speciated ambient carbonyls in Rio de Janeiro, Brazil. *Environmental Science and Technology* 2002; 36(7): 1389-95.
- Harrison RM, Perry R. Handbook of air pollution analysis. 2<sup>nd</sup> ed. Chapman and Hall, London, UK. 1986.
- Ho KF, Lee SC, Louie PKK, Zou SC. Seasonal variation of carbonyl compound concentrations in urban area of Hong Kong. *Atmospheric Environment* 2002; 36(8): 1259-65.
- Hoddinott KB, Lee AP. The use of environmental risk assessment methodologies for an indoor air quality investigation. *Chemosphere* 2000; 41(1-2): 77-84.
- Hu M, Wu Z, Slanina J, Lin P, Liu S, Zeng L. Acidic gases, ammonia and water-soluble ions in PM<sub>2.5</sub> at a coastal site in the Pearl River Delta, China. *Atmospheric Environment* 2008; 42(25): 6310-20.
- IARC (International Agency for Research on Cancer). Monographs on the evaluation of carcinogenic risks to humans. Wood Dust and Formaldehyde, Lyon, 1995; Volume 62: 217-362.
- IARC (International Agency for Research on Cancer). Formaldehyde as carcinogenic to humans. Press Release no. 153 (15-6-2004), Lyon, France. 2004.
- IARC (International Agency for Research on Cancer) [homepage on the Internet]. 2005. Available from: <http://www.cie.iarc.fr/htdocs/announcements/vol88.htm>.
- IGWQ (Interim Guidance on the Water Quality) Regulations 2000 (England) and the Water Supply (Water Quality) Regulations 2001 (Wales). Drinking Water Inspectorate, 2003.
- Khoder MI. Ambient levels of volatile organic compounds in the atmosphere of Greater Cairo. *Atmospheric Environment* 2007; 41(3): 554-66.
- Khoder MI, Shakour AA, Farag SA, Abdel Hameed AA. Indoor and outdoor formaldehyde concentrations in homes in residential areas in Greater Cairo. *Journal of Environmental Monitoring* 2000; 2(2): 123-26.
- Larsen JC, Larsen PB. Chemical carcinogens. *In: Air pollution and health* (Eds: Hester RE, Harrison RM). Issues in Environmental Science and Technology, Royal Society of Chemistry, Cambridge, UK. 1998.
- Liu Y, Shao M, Lu S, Chang CC, Wang JL, Fu L. Source apportionment of ambient volatile organic compounds in the Pearl River Delta, China: Part II. *Atmospheric Environment* 2008; 42(25): 6261-74.
- Maître A, Soulat JM, Masclet P, Stoklov M, Marquès M, de-Gaudemaris R. Exposure to carcinogenic air pollutants among policemen working close to traffic in an urban area. *Scandinavian Journal of Work, Environment and Health* 2002; 28(6): 402-10.
- Montero L, Vasconcellos PC, Souza SR, Pires MA, Sanchez-Ccoyllo OR, Andrade MF, Carvalho LR. Measurements of atmospheric carboxylic acids and carbonyl compounds in São Paulo City, Brazil. *Environmental Science and Technology* 2001; 35(15): 3071-81.
- Muller E, Diab RD, Binedell M, Hounsoume R. Health risk assessment of kerosene usage in an informal settlement in Durban, South Africa. *Atmospheric Environment* 2003; 37(15): 2015-22.
- Nasralla M, Albar O. Particulate in Makkah atmosphere, the custodian of the two Moly Mosques Institute for Hajj Research, Annual Report, Makkah, Saudi Arabia. 2005.
- Nguyen HTH, Takenaka N, Bandow H, Maeda Y, de-Oliva ST, Botelho MMF, Tavares TM. Atmospheric alcohols and aldehydes concentrations measured in Osaka, Japan and in São Paulo, Brazil. *Atmospheric Environment* 2001; 35(18): 3075-83.
- NIOSH (National Institute for Occupational Safety and Health). Method 2549 in the NIOSH Manual of Analytical Methods. 4<sup>th</sup> ed. US Department of Health and Human Services, Center for Disease Control, NIOSH, Cincinnati, OH. 1996.
- NIOSH (National Institute for Occupational Safety and Health) Chemical listing and documentation of revised IDLH values [homepage on the Internet]. 2003. Available from: <http://www.cdc.gov/niosh/intrid14.html>
- NRC (National Research Council). Committee on aldehydes, formaldehyde and other aldehydes. National Academy Press, Washington DC. 1981.
- Oftedal B, Nafstad P, Magnus P, Bjørkly S, Skrandal A. Traffic related air pollution and acute hospital admission for respiratory diseases in Drammen, Norway 1995-2000. *European Journal of Epidemiology* 2003; 18(7): 671-75.
- OSHA (Occupational Safety and Health Guideline). OSHA Guideline for formaldehyde. *In: Occupational safety and health administration guidelines for chemical hazards*. Cincinnati, National Institute for Occupational Safety and Health, 1988.
- PCS (Pacific Cleaners Site). Evaluation of follow-up indoor air sample taken at Randy's nutrition center, adjacent to pacific cleaners site, City of Olympia, Thurston County, Washington, EPA Facility ID: WAD988479838. 2005.
- Perry R, Young RJ. Handbook of air pollution analysis. 1977.
- Pohanish RP. Sittig's handbook of toxic and hazardous chemicals and carcinogens. 4<sup>th</sup> ed. Noyes Publications/William Andrew Publishing, Norwich, NY, 2002; 1179-82.

- Pratt GC, Palmer K, Wu CY, Oliaei F, Hollerbach C, Fenske MJ. An assessment of air toxics in Minnesota. *Environmental Health Perspectives* 2000; 108(9): 815-25.
- Preuss PW, Dailey RL, Lehman ES. Exposure to formaldehyde. *In: Formaldehyde (Ed: Turoski V). Analytical Chemistry and Toxicology (Advances in Chemistry Series, Vol. 210), Washington DC, American Chemical Society, 1985; 247-59.*
- Proctor NH, Hughes, JP. *Chemical hazards of the workplace*, J. B. Lippincott, Philadelphia. 1978.
- Ravindra K, Bencs L, Wauters E, de-Hoog J, Deutsch F, Roekens E, Bleux N, Berghmans P, Grieken RV. Seasonal and site specific variation in vapor and aerosol phase PAHs over Flanders (Belgium) and their relation with anthropogenic activities. *Atmospheric Environment* 2006; 40(4): 771-85.
- Sakai K, Norbäck D, Mi Y, Shibata E, Kamijima M, Yamada T, Takeuchi Y. A comparison of indoor air pollutants in Japan and Sweden: formaldehyde, nitrogen dioxide, and chlorinated volatile organic compounds. *Environmental Research* 2004; 94(1): 75-85.
- Seinfeld JH, Pandis SN. *Atmospheric chemistry and physics: from air pollution to climate change*. John Wiley & Sons Inc, New York, NY. 1998.
- Sin DWM, Wong YC, Louie PKK. Trends of ambient carbonyl compounds in the urban environment of Hong Kong. *Atmospheric Environment* 2001; 35(34): 5961-69.
- Slemr J, Junkermann W, Volz-Thomas A. Temporal variations in formaldehyde, acetaldehyde and acetone and budget of formaldehyde at a rural site in southern Germany. *Atmospheric Environment* 1996; 30(21): 3667-76.
- USEPA (Environmental Protection Agency USA). *Integrated Risk Information System (IRIS): Formaldehyde [homepage on the Internet]*. 2006. Available from: <http://www.epa.gov/iris/subst/0419.htm>.
- USEPA (Environmental Protection Agency USA). *Technology transfer network: 1999 national-scale air toxics assessment [homepage on the Internet]*. 2006b. Available from: <http://www.epa.gov/ttn/atw/nata1999>. Accessed 2006b January 26.
- Vaught C. *Locating and estimating air emissions from sources of formaldehyde (Revised)*. (Report No. EPA-450/4-91-012; US NTIS PB91-181842), Research Triangle Park, NC, Environmental Protection Agency WHO (1989) *Formaldehyde (Environmental Health Criteria 89)*, International Programme on Chemical Safety. Geneva, 1991.
- WHO (World Health Organization). *Environmental health criteria: formaldehyde [homepage on the Internet]*. International Programme on Chemical Safety, Geneva, Switzerland. 1989. Available from: <http://www.inchem.org/documents/ehc/ehc/ehc89.htm>.
- WHO (World Health Organization). *Indoor air quality: organic pollutants. Report on a WHO Meeting, Berlin, EURO Reports and Studies 111, Copenhagen, 1989.*
- WHO (World Health Organization). *Formaldehyde, Concise International Chemical Assessment Document 40*. 2002.
- Zheng M, Fang M. Correlations between organic and inorganic species in atmospheric aerosols. *Environmental Science and Technology* 2000; 34(13): 2721-26.
- Zielinska B, Sagebiel J, Arnott WP, Rogers CF, Kelly KE, Wagner DA, Lighty JS, Sarofim AF, Palmer G. Phase and size distribution of polycyclic aromatic hydrocarbons in diesel and gasoline vehicle emissions. *Environmental Science and Technology* 2004; 38(9): 2557-67.

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