

Characterization and Health Risk Assessment of Volatile Organic Compounds in Gas Service Station Workers

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Abstract

Gas service station workers who work near volatile organic compounds (VOCs) sources, such as gasoline vapor emissions, and motor vehicle exhausts, may be exposed to highly elevated VOCs levels. This study investigates air samples from gas service stations in Thailand to evaluate the health risks following inhalation exposure. Personal air samplings were obtained at nine gas service stations in Chonburi, Thailand from October to December 2007. The concentrations of benzene, toluene, ethylbenzene, xylenes, and hexane in the air from the workplaces were significantly higher than in a control group of office workers ($p < 0.05$). However, all VOCs in these air samples were lower than TWA limit of Thailand and the OSHA standard. Samples of urine, collected after 8-h work periods which were analyzed for VOCs metabolites, including t,t muconic acid, hippuric acid, mandelic acid and m-hippuric acid, demonstrate that the average levels of metabolites in gas service station workers and in controls were close, except for t,t muconic acid of gas service station workers which displayed higher levels than the in the controls. The lifetime cancer and noncancer risks for the workers exposed to VOCs were also assessed. Results show that all nine gas service stations in this study had a elevated lifetime cancer risk ranging from 53 to 630 per million, thus exceeding the "normal" risk of 1 per million. For noncancer risks, the levels in all gas stations ranged between 0.03 and 0.4, which is well below the reference hazard level of 1.0. Benzene may the most important cause of both cancer and noncancer risk followed by 1,3 butadiene.

Keywords: volatile organic compounds; VOCs; gas service station workers; health risk assessment

1. Introduction

Volatile organic compounds (VOCs), associated with gasoline vapor emissions and motor vehicle exhaust, are pollutants of concern because of their toxicity (IARC, 1987; USEPA, 1990a). Many studies have been conducted on occupational exposure to VOCs from gasoline vapor emissions (Jo and Song, 2001; Hartle and Young, 1977; Kearney and Dunham, 1986).

These studies reported that the workers 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. Significant exposure to these volatile organic toxicants emitted into the air only occurs through the inhalation pathway (Cal/EPA, 2003), and has therefore gained the attention of researchers. Currently, benzene is classified as a human carcinogen,

and 1-3 butadiene, chloroform, trichloroethylene and 1-4 dioxane are classified as possible human carcinogens (USEPA, 1998).

2. Materials and Methods

2.1. Sampling and Analysis

This study was conducted in 9 gas stations located in Chonburi Province, Thailand from October to December 2007. The workers included 15 males and 12 females, all of whom had volunteered to take part in the study. They were 15-34 years old, and they spent about 8 hours per day at gas service stations. Urine and personal air samples were collected at nine gas service stations, and in each gas station three workers were included. A total of 27 samples were thus collected. Three office workers volunteered as controls for this study; these persons were not in direct contact with VOCs. Each worker was instructed to collect a sample set consisting of one personal air sample within breathing zone and one spot urine sample after the work shift. In addition, each of them were asked to fill

in a questionnaire containing information, such as age, sex, work-shift, and work duration.

2.1.1. Analysis of Air Samples

Air samples were collected by diffusion into a tube type diffusive sampler, made of stainless steel (Markes, Markes International Ltd., United Kingdom) packed with carbo pack B 60/80 mesh. The tube was attached to the clothes within 30 cm from the nose (“breathing zone”). The analyses were carried out essentially according to the Instruction Manual TO-17 (USEPA, 1999). Figure 1 shows the chromatogram of an air sample containing VOCs. The quantitative analysis of the VOCs was performed using the calibration curves at six concentrations. The quality assurance/quality control (QA/QC) program included laboratory and field blank samples. An external standard was analyzed daily.

2.1.2. Analysis of Urine Samples

Urine samples (10 ml each) were collected in a plastic container, sealed and stored at -20 °C until analysis. Under these conditions, samples can be stored for 2 months. The urine samples were analyzed by HPLC–UV (model HP1100, Hewlett Packard, Germany) in order to determine concentrations of t,t

muconic acid (at 254 nm), hippuric acid, mandelic acid and m-hippuric acid (at 210 nm). For this purpose we followed the methods by Boogard *et al.* (1996) and NIOSH (2003). All measured values were divided by the concentration of urinary creatinine, as analyzed by an analyzer for clinical chemistry, model Stardust MC15 (Diasys diagnostic system, Germany).

2.2. Health Risk Calculation

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. The inhalation intake was calculated by averaging daily intake over the exposure period. The carcinogenic and noncarcinogenic intakes of VOCs for gas service station workers were calculated as

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

where I is the inhalation intake ($\mu\text{g}/\text{m}^3$), C is the concentration of the compound in the personal air sample ($\mu\text{g}/\text{m}^3$), 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

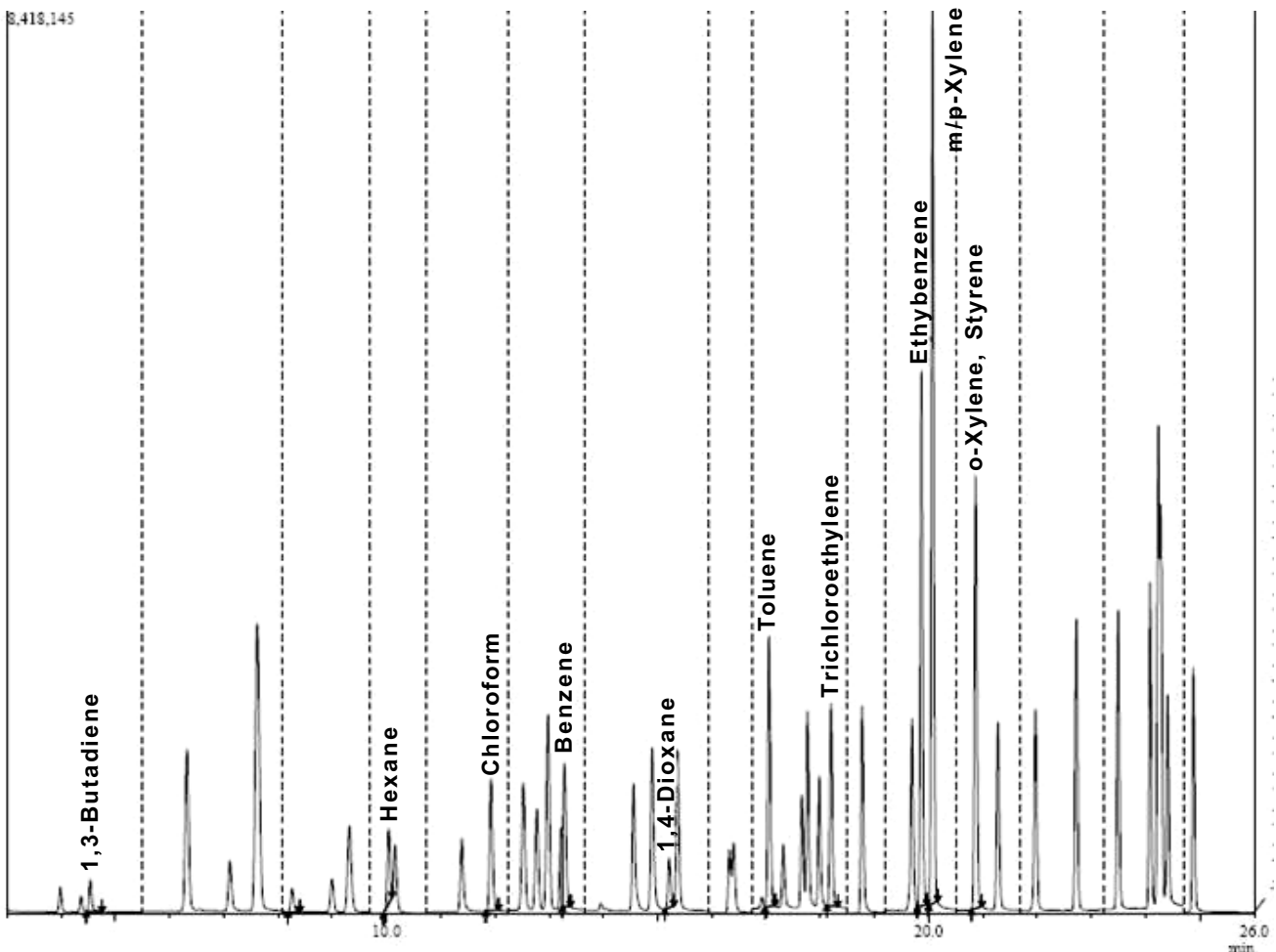


Figure 1. The chromatograms of the VOCs from GC-MS.

Table 1. The exposure and risk assessment factors

Exposure settings	Value	Source of exposure settings
Exposure time	8 h /day	Questionnaires
Exposure frequency	300 day /year	Questionnaires
Exposure duration :carcinogenic	2 year	Questionnaires
Exposure duration : noncarcinogenic	2 year	Questionnaires
Average life time : carcinogenic	70 year	Cal/EPA, 2005
Average life time : noncarcinogenic	70 year	Cal/EPA, 2005

lifetime (years). Inhalation exposure is always related to exposure frequency, duration, and quantity (dose) and activity pattern. To simplify the exposure and risk assessment, several assumptions regarding individual exposures were made based on the questionnaire data and professional judgment. Exposure to VOCs was based on the average 8 hrs (full shift) time weighted average (TWA) concentration, the summation throughout the workday of the product of the concentrations, and the time periods for the concentration encountered in each time interval and were averaged over an 8 h standard workday. The inhaled compounds were assumed to be totally absorbed for risk calculations in some studies (Hoddinott and Lee, 2000; Muller *et al.*, 2003). Table 1 summarizes the exposure and risk assessment factors

Risk characterization requires combining the estimated exposure concentrations with toxicity data to provide a quantitative estimate of the potential health impacts. 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 the equation

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

The noncancer risk is expressed in terms of the hazard quotient (HQ), 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 noncancer adverse health effect is anticipated to occur in a human population exposed for a specific duration (Cal/EPA, 2005). The noncancer health impacts were expressed as the hazard index (HI), which is determined by calculating the HQ for a compounds and summing all of the HQ at a specific location.

$$\begin{aligned} \text{HQ} &= I (\mu\text{g}/\text{m}^3) / \text{RELs } (\mu\text{g}/\text{m}^3) \\ \text{HI} &= \text{HQ}_1 + \text{HQ}_2 + \text{HQ}_3 + \dots + \text{HQ}_n \end{aligned}$$

For a given airborne toxic compound, exposures below the reference level ($\text{HI} < 1$) are unlikely to be associated with adverse health effects (Cal/EPA, 2003). The potential for adverse effects increases as exposures further exceed the reference dose. Table 2 summarizes the toxicity values for compounds of concern.

Table 2. The toxicity values for compounds of concern

Compound	RELs ($\mu\text{g}/\text{m}^3$)	Cancer Unit Risk Factors ($\mu\text{g}/\text{m}^3$) ⁻¹	USEPA Class
benzene	6.0E+01	2.9E-05	A
toluene	3.0E+02		D
ethylbenzene	2.0E+03		D
xylene	7.0E+02		D
styrene	9.0E+02		-
1,3-butadiene	2.0E+01	1.7E-04	B2
carbendisulfide	8.0E+02		D
hexane	7.0E+03		D
chloroform	3.0E+02	5.3E-06	B2
trichloroethylene	6.0E+02	2.0E-06	B2
1,4-dioxane	3.0E+03	7.7E-06	B2

RELs = Chronic Inhalation Reference Exposure Levels

Ref: Cal/EPA, 2005

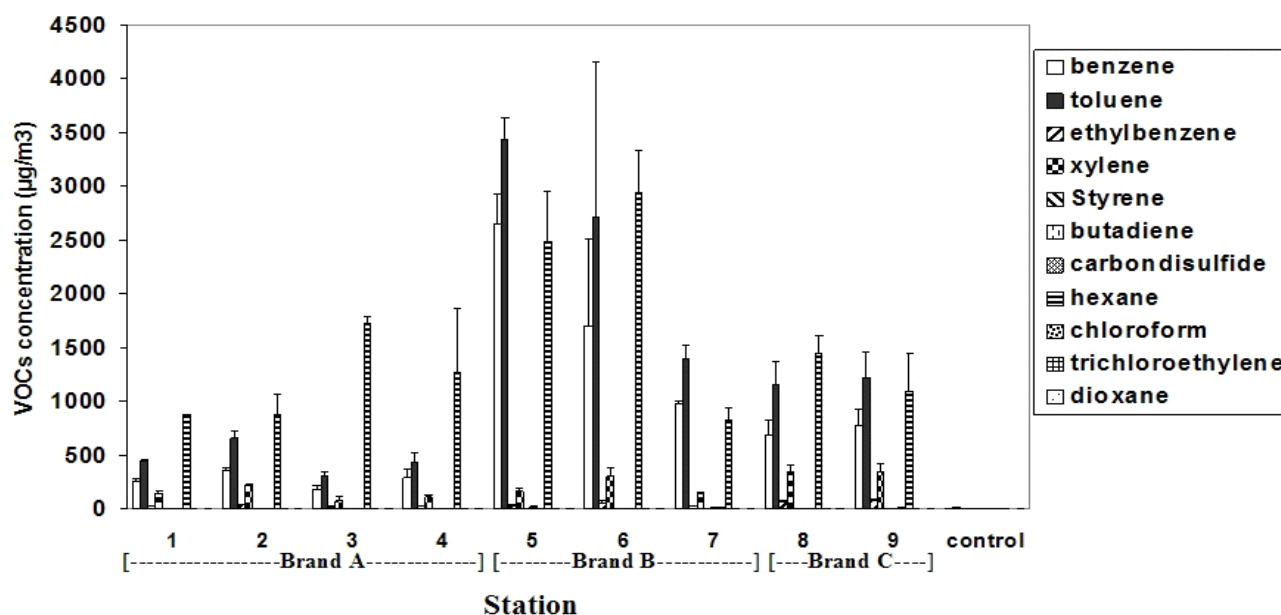


Figure 2. The volatile organic compound (VOCs) concentrations ($\mu\text{g}/\text{m}^3$) in the personal air samples from gas service station workers, and from controls (office workers) in 8 h. worktime

3. Results

3.1. Characterization and Exposure Assessment

The measurement of volatile organic compounds (VOCs) in the personal air sampling of gas service station workers, and in the controls (office workers) are summarized in Table 3 and Figure 2. The average concentrations of VOCs found were in decreasing order: hexane, toluene, benzene, xylene, ethylbenzene, 1,3-butadiene, carbon disulfide, chloroform, styrene, 1,4-dioxane and trichloroethylene. At all service stations, the workers' air samples displayed significantly (<0.05) higher concentrations of benzene, toluene, xylene, ethylbenzene and hexane than in the air samples from the controls. Personal air samples from three brands of gasoline in this study (brands A, B and C) were compared using one way ANOVA. The results show that the concentrations of benzene, toluene and ethylbenzene were significantly different between each brand ($p<0.05$), and were highest in brand B and lowest in brand A, as shown in Table 4.

3.2. Health Risk Assessment

Health risk assessment for workers in the nine gas service stations was assessed using personal air sampling data. In this study, benzene, 1,3-butadiene, chloroform, trichloroethylene and 1,4-dioxane were assessed as human carcinogens (USEPA, 1998). The results for the average lifetime cancer risk was 210 per million as shown in Table 5. Risks less than 1 in a million are typically considered to be well below a level of concern, and risks above 100 per million are

typically considered sufficient for action or intervention to take place. In the US, EPA final national contingency plan for Superfund site remediation, the EPA codified a range of acceptable risks (*i.e.*, 1×10^{-4} - 1×10^{-6}) as a basis for the cleanup and protection of human health at Superfund sites (US EPA, 1990b).

The cancer risks were highest at the gas service station 5, 630 per million, and lowest at the gas service station 3, 53 per million. The cancer risk was highest from benzene, 200 per million and the second was 1, 3 butadiene, 13 per million. The cancer risk was highest from brand B, the second was brand C and the third was brand A (421, 181 and 71 per million, respectively), as shown in Table 6.

The average lifetime noncancer risk, the HI, for all gas service stations was 0.156, below the reference levels. In this study, exposure to compounds with a $\text{HI} \leq 1.0$ is considered not likely to result in adverse noncancer health effects over a lifetime of exposure. If a $\text{HI} > 1.0$, then some possibilities exist that noncancer effects may occur, although an $\text{HI} > 1.0$ does not indicate a definite effect. The noncancer risks was highest at the gas service station 5, $\text{HI} = 0.447$, and lowest at the gas service station 3, $\text{HI} = 0.038$. The noncancer risk was highest from benzene, $\text{HQ} = 0.114$ and the second was 1, 3 butadiene, $\text{HQ} = 0.034$, as shown in Table 7.

3.3. Biological Exposure Indices: BEI

This study assessed the gas service station workers' exposure to benzene, toluene, ethylbenzene and xylene, by determining the biological exposure indices (BEI) for t,t muconic acid, hippuric acid, mandelic acid and m-hippuric acid in urine. The results

Table 3. The volatile organic compounds (VOCs) concentrations ($\mu\text{g}/\text{m}^3$) in the personal air samples from gas service station workers (n = 3/station), and from the controls in 8 hrs. worktime

station	benzene		toluene		ethylbenzene		xylene		styrene		1,3-butadiene	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
1	265.2*	17.7	446.8*	6.4	29.4*	4.9	146.5*	31.4	4.0	4.0	6.3	2.6
2	357.7*	29.6	655.2*	75.5	42.2*	4.1	216.7*	23.2	8.4*	0.8	3.8	0.2
3	181.8*	41.1	305.7*	37.0	18.8*	6.0	82.7*	33.9	0.6	0.6	8.6*	0.3
4	286.2*	79.4	437.6*	77.2	23.5*	3.1	113.8*	14.1	0.0	0.0	9.0*	0.4
5	2649.5*	280.2	3424.1*	207.4	38.0*	4.2	168.7*	21.5	0.0	0.0	20.4*	2.0
6	1702.6*	805.7	2710.3*	1449.1	64.9*	16.6	309.7*	76.7	0.5	0.5	8.9*	0.0
7	971.3*	28.7	1394.5*	124.5	31.7*	3.6	139.7*	19.8	1.4	1.5	11.1*	2.5
8	693.8*	128.0	1155.2*	215.6	71.2*	11.1	353.4*	51.8	2.0	0.8	9.2*	1.1
9	779.9*	140.1	1222.7*	232.1	76.4*	16.9	346.4*	76.8	5.1*	1.5	11.3*	0.5
Mean 9 station	876.4	822.8	1305.8	1122.9	44.0	22.1	208.6	106.0	2.4	3.0	9.8	4.6
Min	140.7		268.7		12.8		48.8		0.00		3.7	
Max	2929.7		4142.1		91.5		415.3		9.2		22.4	
control	2.1*	3.6	9.1*	6.4	0.0*	0.00	0.00*	0.00	0.0*	0.0	4.0	0.0
Min(control)	0.0		2.1		0.0		0.00		0.0		3.7	
Max(control)	6.3		14.6		0.0		0.00		0.0		4.3	

* significantly different (p<0.05)

Table 3. (continued) The volatile organic compounds (VOCs) concentrations ($\mu\text{g}/\text{m}^3$) and SD in the personal air sampling of gas service station workers (n = 3/station), and control (office workers) in 8 hrs. worktime

station	carbendisulfide		hexane		chloroform		Trichloroethylene		1,4-dioxane	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
1	4.2	0.2	875.1*	6.1	4.3*	1.5	0.5	0.5	0.0	0.0
2	2.7	2.3	879.1*	187.7	5.5*	0.3	0.8	0.7	0.0	0.0
3	5.7	0.5	1731.8*	61.2	5.9*	0.2	0.0	0.0	0.0	0.0
4	4.6	0.3	1266.7*	595.7	3.9*	2.0	0.0	0.0	0.0	0.0
5	4.6	0.3	2475.3*	474.3	3.7*	0.9	0.0	0.0	11.0*	1.5
6	4.8	0.5	2940.3*	388.9	6.9*	0.2	0.0	0.0	0.0	0.0
7	11.8	6.9	829.2*	107.1	1.5	1.5	0.0	0.0	0.0	0.0
8	4.6	0.0	1450.3*	160.1	4.9*	0.3	0.0	0.0	3.9	6.7
9	14.9	2.6	1100.5*	344.2	0.0	0.0	0.0	0.0	0.0	0.0
Mean 9 station	6.4	4.5	1505.4	770.0	4.1	2.3	0.1	0.4	1.7	4.1
Min	0.0		676.2		0.0		0.0		0.0	
Max	18.7		3328.9		7.1		1.2		12.6	
control	3.6	1.2	0.4*	0.5	0.0	0.00	0.0	0.0	0.0	0.0
Min(control)	2.1		0.0		0.0		0.0		0.0	
Max(control)	4.3		0.9		0.0		0.0		0.0	

* significantly different (p<0.05)

Table 4. The volatile organic compound (VOC) concentrations ($\mu\text{g}/\text{m}^3$) in the personal air samples of gas service station workers. Comparisons of three difference brands

Compound	Brand A	Brand B	Brand C
	Station 1-4	Station 5-7	Station 8-9
benzene	272.7*	1774.5*	736.8*
toluene	461.3*	2509.7*	1188.9*
ethylbenzene	28.5*	44.8*	73.8*
xylene	139.9	206.0	349.9
styrene	3.2	0.7	3.6
1,3-butadiene	6.9	13.5	10.3
carbendisulfide	4.3	7.1	9.8
hexane	1188.2	2081.6	1275.4
chloroform	4.9	4.1	2.5
trichloroethylene	0.3	0.0	0.0
1,4-dioxane	0.3	0.0	0.0

* significantly different ($p < 0.05$)

are shown in Table 8. Biological monitoring data demonstrate that the average level of metabolites in gas service station workers and control were close, except for t,t muconic acid of gas service station workers which was higher than in the controls.

4. Discussion

This study demonstrated that gas service station workers were exposed to higher VOCs levels than workers who were not direct contact with VOCs. This result is in agreement with a Korean study, which found that the concentrations of benzene, toluene, ethylbenzene and xylene (BTEX) in personal air samples were significantly correlated to the concentrations in breath samples. The breath concentrations were significantly higher in gas service station workers than in the controls. (Jo and Song, 2001). Furthermore, the breath benzene of the gas service station workers ($41 \mu\text{g}/\text{m}^3$) was 3.8 times higher than that of the housewives ($10.9 \mu\text{g}/\text{m}^3$) (Jo and Moon, 1999), 21.5 times higher than that of the residents of Los Angeles ($1.9 \mu\text{g}/\text{m}^3$) (Wallace *et al.*, 1991), and 3.4 times higher than that of residents of New Jersey ($12.0 \mu\text{g}/\text{m}^3$) (Wallace *et al.*, 1987).

Table 6. The average lifetime cancer risk assessment for gas service station workers of three difference brand (per million)

Compound	Brand A	Brand B	Brand C
benzene	61.9	402.8	167.3
1,3-butadiene	9.2	17.9	13.6
chloroform	0.2	0.2	0.1
trichloroethylene	0.0	0.0	0.0
1,4-dioxane	0.0	0.2	0.1
total	71.3	421.1	181.1

The concentrations of BTEX in the present study (876.4 , 1305.8 , 44.0 , and $208.6 \mu\text{g}/\text{m}^3$, respectively) were higher than in the study from Taegu, Korea (72.1 , 126.0 , 12.1 , and $50.7 \mu\text{g}/\text{m}^3$, respectively) (Jo and Song, 2001), the Bangkok study (174 and $296 \mu\text{g}/\text{m}^3$, respectively) (Lekcharernkul, 2006) and in a medium-sized city in northern Italy ($44.0 \mu\text{g}/\text{m}^3$) (Carrieria *et al.*, 2005). The concentrations of TEX in our study were lower than the study in southern Finland (2200 , 260 , and $1100 \mu\text{g}/\text{m}^3$, respectively) (Vainiotalo *et al.*, 2006). However all VOCs in personal air samples were lower than TWA limit of the Thailand and the OSHA standard (OSHA, 1997). The urinary levels of t,t muconic acid in this study ($215 \mu\text{g}/\text{g}$ creatinine) were close to that of the Italian study ($171 \mu\text{g}/\text{g}$ creatinine) (Carrieria *et al.*, 2005).

The gas service station 5 had the highest level of both the lifetime cancer risk and non cancer risk, while the gas service station 3 had the lowest risks. It is interesting to note that gas station 5 sold brand B, whereas station 3 sold brand A. It may be speculated that the differences are due to differences in the proportions of benzene and toluene in the different brands of gasoline as discussed in a study from Bangkok (Lekcharernkul, 2006). Obviously, sales volumes should also have an impact.

Epidemiologic studies and case studies provide clear evidence of a causal association between

Table 5. The results for the average lifetime cancer risk assessment of gas service station workers (per million)

	Stations									Mean	control	
	1	2	3	4	5	6	7	8	9			
Cancer Risk												
Benzene	60.2	81.2	41.3	65.0	601.5	386.5	220.5	157.5	177.0	199.6	0.5	
1,3-butadiene	8.3	5.0	11.4	12.0	27.1	11.9	14.7	12.3	15.0	13.0	5.3	
chloroform	0.2	0.2	0.2	0.2	0.2	0.3	0.1	0.2	0.0	0.2	0.0	
trichloroethylene	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
1,4-dioxane	0.0	0.0	0.0	0.0	0.7	0.0	0.0	0.2	0.0	0.1	0.0	
total	68.7	86.5	53.0	77.1	629.4	398.7	235.3	170.2	192.1	212.9	5.8	

Table 7. The results for the average lifetime non cancer risk assessment of gas service station workers

	Stations									Mean	control
	1	2	3	4	5	6	7	8	9		
Non Cancer Risk (HQ)											
benzene	0.035	0.047	0.024	0.037	0.346	0.222	0.127	0.091	0.102	0.114	0.000
toluene	0.012	0.017	0.008	0.011	0.089	0.071	0.036	0.030	0.032	0.034	0.000
ethylbenzene	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
xylene	0.002	0.002	0.001	0.001	0.002	0.003	0.002	0.004	0.004	0.002	0.000
styrene	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
1,3-butadiene	0.002	0.001	0.003	0.004	0.008	0.003	0.004	0.004	0.004	0.004	0.002
carbonylsulfide	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
hexane	0.001	0.001	0.002	0.001	0.003	0.003	0.001	0.002	0.001	0.002	0.000
chloroform	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
trichloroethylene	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
1,4-dioxane	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
HI	0.052	0.069	0.038	0.055	0.448	0.304	0.170	0.130	0.144	0.157	0.002

HQ = Hazard quotient is defined as the ratio of the estimated exposure of an individual to the reference dose.

HI = Hazard index is determined by calculating HQ for a compound and summing all of the HQ at a specific location.

Table 8. The average level of metabolites in urine samples from gas service station workers and controls

Compound (metabolite)	gas service station workers (N = 27)	control (N = 3)
benzene (t,t muconic acid) ($\mu\text{g/g}$ creatinine)	215.15	73.22
toluene (hippuric acid) (g/g creatinine)	0.28	0.23
ethylbenzene (mandalic acid) (g/g creatinine)	0.02	0.01
xylene (m-hippuric acid) (g/g creatinine)	0.06	0.02

exposure to benzene and acute nonlymphocytic leukemia (ANLL) and also suggest evidence for chronic nonlymphocytic leukemia (CNLL) and chronic lymphocytic leukemia (CLL). Other neoplastic conditions that are associated with an increased risk in humans are blood disorders such as preleukemia and aplastic anemia, Hodgkin's lymphoma, and myelodysplastic syndrome (MDS). These human data are supported by animal studies. Benzene is a known human carcinogen based upon evidence presented in numerous occupational epidemiological studies. Significantly increased risks health risk should be considered. The gas service stations should reconsider the use of vapor recovery systems, which are mandatory in many other countries, and strict measures to control the gasoline emission during the gasoline transmission to storage and transport trucks.

Moreover, there should be research and development leading to a decrease in the amount of benzene and other hazardous substances. Studies about other dimensions of health risk from VOCs should be conducted. These studies should include the effects on hematopoiesis, chromosomal aberrations and abnormal fetal development.

5. Conclusion

Gas service station workers who work near VOCs sources, such as gasoline vapor emissions and motor vehicle exhausts, exposed to higher VOCs levels than control persons working in offices who did not directly contact with VOCs. However, all VOCs in personal air samples were lower than the TWA limit of the Thailand and the OSHA standard. The urinary metabolites after 8 hours work period, including t,t muconic acid, hippuric acid, mandalic acid and m-hippuric acid in gas service station workers and in controls were close, except for t,t muconic acid in gas service station workers which was higher than in the controls. The results of the health risk assessment show that all nine gas service stations in this study had a lifetime cancer risk exceeding 1 per million because of the high levels of benzene and 1,3 butadiene. For noncancer risk, levels of all VOCs were below the reference levels in all gas service stations. Benzene may be the most important cause of both cancer and noncancer risk, followed by 1,3 butadiene.

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