

CARBONYL COMPOUNDS EXPOSURES AMONG GASOLINE STATION WORKERS AND THEIR ASSOCIATED HEALTH RISK

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ABSTRACT: The carbonyl compounds exposures were evaluated in twenty gasoline station workers of ten gasoline stations in inner and outer areas of Bangkok, Thailand from February to March 2011. The average worker exposures of formaldehyde, acetaldehyde, propionaldehyde, butyraldehyde, benzaldehyde and valeraldehyde were 14.78, 7.07, 1.36, 1.77, 0.46 and 0.46 µg/m³ respectively, which was not different from their concentrations at roadsides in front of gasoline stations. In addition, they were not significantly different concentrations between exposures in gasoline stations of inner and outer areas. The average lifetime cancer risk of worker exposures for 30 years to formaldehyde and acetaldehyde were determined as the values of 1.89E-05 and 4.32E-06 respectively, in which formaldehyde cancer risk was higher than acceptable criteria defined as 1.00E-06. For non-cancer risk of health effects, the hazard quotients (HQs) of the workers exposures to formaldehyde, acetaldehyde, propionaldehyde, butyraldehyde, benzaldehyde and valeraldehyde were found at the ratio of 0.23, 0.14, 0.03, 0.02, 0.01 and 0.00 respectively. As of these results, the hazard index (HI) was less than 1, indicated that total of carbonyl compounds did not affect to increase risk of other health effects. Most prevalent symptoms were headache, fatigue and throat irritation at 58.9%, 25.0% and 12.5% respectively. All of these symptoms were not associated with carbonyl compounds in this study. In conclusion, mean concentrations of major sources of carbonyl compounds, including formaldehyde and acetaldehyde, found that formaldehyde exposure had cancer risk for gasoline station workers but all carbonyl compounds did not have adverse chronic health effects. The carbonyl compounds exposures were not associated with all symptoms of worker complaints.

Keywords: carbonyl compounds, health risk, exposure, gasoline station

INTRODUCTION

Carbonyl compounds (CCs) contain the carbonyl group, a carbon atom and an oxygen atom linked with a double bond (C=O). The term is commonly used in the restricted sense of aldehydes and ketones, although it actually includes carboxylic acids and derivatives. Primary alcohol is oxidized, distilled out the aldehyde immediately. Volatile organic compounds (VOCs), including carbonyl compounds (CCs), can be emitted from a variety of emission sources which the major sources of directly emitted carbonyls are fuel combustion, mobile sources, and process emissions from oil refineries [1-3]. Some carbonyls are highly reactive and play a critical role in the formation of ozone such as formaldehyde and acetaldehyde [4]. Other carbonyls have adverse chronic and acute health effects. In some cases, carbonyls can be both highly reactive and potentially toxic as carcinogen [5-7]. The sources of CCs in urban areas are expected to be different from those in rural areas due to fugitive emissions of raw materials

and organic solvents [8-12]. Gasoline station is one of importance source of CCs which there is rapidly increasing use of oxygenated gasoline since Thai government introduced using alternative fuels as gasohol [13]. The combustions of oxygenated fuels result in greater aldehyde emissions and contributes to increase ambient concentrations in locales where they are widely used [14, 15]. Interestingly, Environmental Research and Training Centre (ERTC) of Thailand had studied the average general ambient air concentration of formaldehyde and acetaldehyde in 24 hours in Bangkok area during 2007-2008 and found that these substances were increased with the values of 10.0 and 2.9 µg/m³ respectively, which those concentrations at the roadside areas were higher than in residential areas. They had highest level in cold season and lowest in rainy season [16]. Typically, occupational and residential exposures to CCs like formaldehyde and acetaldehyde pose to have cancer risk via inhalation, ingestion and dermal absorption but the most likely route of exposure is by inhalation. Human health effects, carcinogenic and non-carcinogenic, depend upon the chemical hazard, chemical involved, concentration and exposure time [17]. Thus, the

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gasoline worker may be a high risk group to expose to CCs and have adverse chronic health effects. This study aimed to assess average lifetime cancer risk, non-cancer risk and their association between CCs exposure and health effects of symptoms in gasoline station workers.

MATERIALS AND METHODS

Population study

This study was conducted in ten gasoline stations in Bangkok, Thailand from February to March 2011. Six stations were on Sukhumvit road as inner area (a major road in central Bangkok serves as a main commercial street) and four stations were on Bangna road as outer area (a junction road of Bangkok to the Eastern Seaboard Industrial Estates and the eastern part of Thailand to places like Pattaya and Chonburi serve as further growth and development street). All stations were nearby express way and roadside. Fifty-six selected workers were healthy and had worked more than six months. They were provided with the consent form before the study. Permission to conduct cancer risks assessment from human subjects in this study was approved by the Ethical Review Committee for Research Involving Human Research Subjects, Health Science Group, Chulalongkorn University.

Sample and data collections

Ten ambient air samples through the active sampling method were collected by 2,4 DNPH (2,4 dinitrophenylhydrazine) cartridge with flow rate 100 ml/min for 8 hours during work time (6.00 A.M.-2.00 P.M.), in front of gasoline stations nearby roadsides. The cartridges were kept at 4°C during transportation to laboratory and were stored in refrigerator until analysis. Likewise, the personal air exposure to CCs of twenty gasoline workers (two workers for each station) was studied using the same sampling equipment as above mentioned. Fifty-six workers were interviewed their characteristics of age, weight, height, period of working time, working time per week and per day and sign of symptoms during their work shifts.

Air samples analysis

The DNPH cartridge was extracted immediately with acetone free acetonitrile (ACN) after sample collection, and eluted solution were quantified by reverse phase of HPLC with gradient mobile phase 60/40% of ACN/water at flow rate 1.0 ml/min. The detection was ultraviolet (UV) absorption detection at wavelength 360 nm [18]. Triplicate analyses were done for each sample.

Quality control

A solvent blank must be analyzed before any standard and sample was run. The solvent blank must not contain formaldehyde, acetaldehyde. The calibration curve used for quantification consisted of five levels ranging from 0.005 to 0.5 µg/ml. The

standard deviation (SD) of each standard injection was multiply by 3 to obtain IDL (Instrument Detection Limit) and by 10 to obtain LOD (Limit of Detection). The SD of this study range from 0.005 to 0.045 µg/ml. The value of LODs for the quantification of samples was ranged from 0.051 to 0.033 µg/ml with the relative standard deviation (%RSD) varied from 0.077 to 0.279. The slopes and the relative variance (R^2) of each calibration line were

$$\begin{aligned} y &= 1\text{E-}06x - 0.0005, R^2 = 0.9999 \text{ for formaldehyde} \\ y &= 1\text{E-}06x - 0.0005, R^2 = 0.9999 \text{ for acetaldehyde} \\ y &= 2\text{E-}06x - 0.0001, R^2 = 0.9999 \text{ for propionaldehyde} \\ y &= 3\text{E-}06x + 4\text{E-}05, R^2 = 0.9993 \text{ for butyraldehyde} \\ y &= 3\text{E-}06x - 0.0044, R^2 = 0.9963 \text{ for benzaldehyde} \\ y &= 3\text{E-}06x - 0.0024, R^2 = 0.9986 \text{ for valeraldehyde} \end{aligned}$$

Cancer and non-cancer risk calculation

The health risk assessment was focused on chronic exposure to formaldehyde and acetaldehyde in gasoline workers. The main route of exposure was inhalation.

For carcinogenic substances, risk is estimated as the incremental probability of an individual developing cancer over a lifetime as a result of exposure to a carcinogen. Cancer risk can be calculated by Risk Assessment Guidance for Superfund (RAGS) Part A approach as follows;

$$\text{Cancer risk} = \text{CDI} \times \text{CSF}_i$$

$$\text{CDI} = (\text{CA} \times \text{IR} \times \text{ET} \times \text{EF} \times \text{ED}) \times (\text{BW} \times \text{AT})$$

$$\text{EC} = \text{CA} \times \text{ET} \times \text{EF} \times \text{ED} / \text{AT}$$

where; CDI (mg/kg/d) = Chronic Daily Intake

EC (µg/m³) = Exposure Concentration

CA (mg/m³) = Contaminant concentration in Air

IR (m³/h) = Inhalation Rate

BW (kg) = Body Weight

ET (h/d) = Exposure Time

EF (d/y) = Exposure Frequency

ED (y) = Exposure Duration

AT (d) = Averaging Time

Cancer risk > 1.00E-6 means Carcinogenic effects of concern

Cancer risk ≤ 1.00E-6 means Acceptable level

For Non-cancer risk, the risk level is generally expressed as term of the hazard quotient (HQ), which is defined as average daily dose (ADD) multiply with the reference dose (RfD). According to the limitation of RfD values for formaldehyde and acetaldehyde, the ratio of exposure concentration (EC) was divided by RfC, could be applied as mentioned in Risk Assessment Guidance for Superfund (RAGS) Part F approach. The calculation of HQ can be expressed as follows:

$$\text{Hazard Quotient (HQ)} = \text{EC} / \text{RfC}$$

$$\text{EC} = (\text{CA} \times \text{ET} \times \text{EF} \times \text{ED}) / \text{AT}$$

where; HQ > 1 means Adverse non-carcinogenic effects of concern

HQ ≤ 1 means Acceptable level

Table 1 The characteristics of gasoline station workers

Parameter	Men (N=41) Mean $\pm$ S.E. or percentage	Women (N=15) Mean $\pm$ S.E. or percentage	Average Mean $\pm$ S.E. or percentage	P-value*
Age (year)	26.8 $\pm$ 1.3	28.6 $\pm$ 2.3	27.1 $\pm$ 1.1	NS
Weight (kg)	59.7 $\pm$ 1.3	56.8 $\pm$ 3.3	58.9 $\pm$ 1.3	NS
BMI (Kg/m ²)	27.5 $\pm$ 0.6	28.2 $\pm$ 1.5	27.7 $\pm$ 0.6	NS
Period of working time (years)	2.8 $\pm$ 0.4	3.7 $\pm$ 0.4	3.0 $\pm$ 0.4	NS
Working time per week (days)	6.2 $\pm$ 0.1	6.4 $\pm$ 0.1	6.3 $\pm$ 0.1	NS
Working time per day (hours)	10.3 $\pm$ 0.3	9.1 $\pm$ 0.5	10.0 $\pm$ 0.3	<0.05

*Significant difference between men and women

Table 2 Summary CCs levels in the investigated locations

Parameter	Inner Area ($\mu\text{g}/\text{m}^3$) Mean $\pm$ S.E		Outer Area ($\mu\text{g}/\text{m}^3$) Mean $\pm$ S.E		Average level ($\mu\text{g}/\text{m}^3$) Mean $\pm$ S.E		P-value*
Compounds	Gasoline workers	Roadside	Gasoline workers	Roadside	Gasoline workers	Roadside	
Formaldehyde	15.96 $\pm$ 2.70	15.37 $\pm$ 1.75	13.02 $\pm$ 1.88	15.38 $\pm$ 1.39	14.78 $\pm$ 1.77	15.37 $\pm$ 1.13	NS
Acetaldehyde	6.86 $\pm$ 1.34	5.78 $\pm$ 1.16	7.40 $\pm$ 2.04	6.18 $\pm$ 0.24	7.07 $\pm$ 1.07	5.94 $\pm$ 0.68	NS
Propionaldehyde	1.28 $\pm$ 0.14	1.18 $\pm$ 0.78	1.48 $\pm$ 0.48	1.07 $\pm$ 0.14	1.36 $\pm$ 0.19	1.14 $\pm$ 0.12	NS
Butyraldehyde	1.74 $\pm$ 1.10	1.90 $\pm$ 1.21	1.81 $\pm$ 1.06	1.40 $\pm$ 0.81	1.77 $\pm$ 0.74	1.70 $\pm$ 0.76	NS
Benzaldehyde	0.39 $\pm$ 0.24	0.39 $\pm$ 0.24	0.58 $\pm$ 0.33	0.94 $\pm$ 0.62	0.46 $\pm$ 0.19	0.61 $\pm$ 0.28	NS
Valeraldehyde	0.38 $\pm$ 0.25	0.55 $\pm$ 0.38	0.44 $\pm$ 0.30	1.32 $\pm$ 0.76	0.41 $\pm$ 0.18	0.86 $\pm$ 0.38	NS

*Significant difference between inner and outer area

Statistical calculations

All analyses were carried out with SPSS 17.0 statistical software package. The characteristics were described by descriptive statistics. The percentage, mean and standard error values were computed. The comparisons of characteristics and sign of symptoms between genders and the CCs exposure concentrations between investigated locations were evaluated by Independent t-test. Normal distribution was assessed by Kolmogorov-Smirnov test. The correlation between CCs concentrations were computed by Spearman's rho correlation, and association between CCs exposure as independent and symptoms dependent variables were evaluated by logistic regression model with 95% confidence interval (CI). A statistically significant difference was accepted at a *p*-value of <0.05.

RESULTS

Fifty-six gasoline station workers were 41 men and 15 women whose average ages were 27.1 years, weight 58.9 kg, body mass index (BMI) 27.7 kg/m², working for 3 years, working 6.2 days a week and working 10 hours a day. Most of the characteristics were not different between genders except hours of working time which was longer in men than women (Independent t-test, *p*<0.05) (Table 1).

The average concentrations of CCs worker exposures defined as formaldehyde, acetaldehyde, propionaldehyde, butyraldehyde, benzaldehyde and valeraldehyde were 14.78, 7.07, 1.36, 1.77, 0.46 and 0.46 respectively which were not significantly different between inner and outer areas (Table 2). While the average concentrations of formaldehyde,

acetaldehyde, propionaldehyde, butyraldehyde, benzaldehyde and valeraldehyde concentrations at roadside were 15.37, 5.94, 1.14, 1.70, 0.61 and 0.86 $\mu\text{g}/\text{m}^3$ respectively. They were not significant difference between worker exposure and roadside concentrations as well as they were not significant difference between inner and outer areas. Formaldehyde and acetaldehyde ratio of worker exposure and roadside were 2.1 and 2.6.

A good Spearman's rho correlation (*r*) among average concentrations of CCs in worker exposures were found in this study; 0.720 between formaldehyde and acetaldehyde (*p*<0.001), 0.711 between formaldehyde and propionaldehyde (*p*<0.001), 0.759 between acetaldehyde and propionaldehyde (*p*<0.001). In addition, there was a good correlation at roadside 0.715 between acetaldehyde and propionaldehyde (*p*<0.001).

The risk assessment factors are shown in Table 3. The average lifetime cancer risk of formaldehyde and acetaldehyde in gasoline station worker exposures for 30 years were 1.89E-05 and 4.32E-06 while average lifetime cancer risk of formaldehyde and acetaldehyde exposures at roadside were 1.97E-05 and 3.63E-06 which cancer risk of formaldehyde of both sites was higher than acceptable value of 1.00E-06 (Table 4). The cancer risk at roadside was slightly higher than gasoline station. For Non-cancer risk, the hazard quotients (HQs) of formaldehyde, acetaldehyde propionaldehyde, butyraldehyde, benzaldehyde and valeraldehyde in worker exposures were 0.23, 0.14, 0.03, 0.02, 0.01 and 0.00, respectively which the hazard index (HI), summation of HQs, was 0.43. While the HQs of formaldehyde,

Table 3 The risk assessment factors

Parameters	Value	Source of information
Average weight (BW, kg)	58.9	Questionnaires
Exposure time (ET, h/day)	10.0	Questionnaires
Exposure duration time (ED, years)	30	Assume for workers
Average life time (AT, years)	70	EPA ^a
CSF _i of formaldehyde (mg/kg-day) ⁻¹	0.021	OEHHA ^c
CSF _i of acetaldehyde (mg/kg-day) ⁻¹	0.01	OEHHA ^c
RfC of formaldehyde (mg/m ³)	9.83E-03	IRIS ^b
RfC of acetaldehyde (mg/m ³)	9.00E-03	IRIS ^b
RfC of propionaldehyde (mg/m ³)	8.00E-03	EPA ^a
RfC of butyraldehyde (mg/m ³)	15.00E-03	EPA ^a
RfC of benzaldehyde (mg/m ³)	9.00E-03	EPA ^a
RfC of valeraldehyde (mg/m ³)	420.00E-03	EPA ^a

^a Environmental Protection Agency (EPA)^b Integrated Risk Information System (IRIS)^c Office of Environmental Health Hazard Assessment (OEHHA)**Table 4** Average CCs exposures and health risks among gasoline station workers

Parameter	CCs	CDI	EC	Non-cancer	Cancer risk
	compounds	(mg/kg/day)	(mg/m ³)	HQ	
Gasoline station workers	Formaldehyde	9.02E-04	2.35	0.24	1.89E-05
	Acetaldehyde	4.32E-04	1.22	0.14	4.32E-06
	Propionaldehyde		0.23	0.03	
	Butyraldehyde		0.31	0.02	
	Benzaldehyde		0.08	0.01	
	Valeraldehyde		0.07	0.00	
Total Risk				0.44	2.32E-05
Roadside	Formaldehyde	9.38E-04	2.65	0.27	1.97E-05
	Acetaldehyde	3.63E-04	1.03	0.11	3.63E-06
	Propionaldehyde		0.20	0.02	
	Butyraldehyde		0.29	0.02	
	Benzaldehyde		0.11	0.01	
	Valeraldehyde		0.15	0.00	
Total Risk				0.43	2.76E-05

Table 5 Signs of symptoms among gasoline station workers

Symptom	Prevalence n (%)		Total Prevalence (%)	P-value*
	Men (N=41)	Women (N=15)		
Headache	24 (58.5)	9 (60.0)	33 (58.9)	NS
Fatigue	13 (31.2)	1 (6.7)	14 (25.0)	NS
Throat irritation	3 (7.3)	4 (26.7)	7 (12.5)	NS
Nose irritation	2 (4.9)	2 (13.3)	4 (7.1)	NS
Dizziness	0 (0.0)	3 (20.0)	3 (5.4)	NS
Nausea	0 (0.0)	2 (13.3)	2 (3.6)	NS
Depression	0 (0.0)	1 (6.7)	1 (1.8)	NS
Stomachache	0 (0.0)	1 (6.7)	1 (1.8)	NS
Dyspnea	0 (0.0)	1 (6.7)	1 (1.8)	NS

*Significant difference between men and women

acetaldehyde propionaldehyde, butyraldehyde, benzaldehyde and valeraldehyde exposures at roadside were 0.27, 0.11, 0.02, 0.02, 0.01 and 0.00, respectively which the hazard index (HI) was 0.43. HI of worker exposures was closed to exposures at roadsides.

Data collection from 56 gasoline station workers was found that the most prevalence of symptoms were 58.9% headache, 25.0% fatigue and 12.5% throat irritation respectively (Table 5). All symptoms did not have differences between genders. The major sources of CCs including formaldehyde and

acetaldehyde were not associated to all of symptoms.

DISCUSSION

The major sources of CCs exposure of gasoline workers and roadsides were formaldehyde and acetaldehyde as previous studies [16, 19, 20]. The formaldehyde and acetaldehyde ratio of roadside in front of gasoline station in this study was higher levels than general roadside in Bangkok of Moknoy study [16]. It may have pooled CCs sources of volatile gasoline and exhaust vehicles [21-23].

However, Mexico City in Mexico, Cairo in Egypt and Rio de Janeiro in Brazil had much higher concentration levels of formaldehyde and acetaldehyde than in Bangkok roadside environment due to the incomplete combustion of different fuel compositions [24-26]. But their concentrations in Bangkok were higher than Hong Kong, Rome in Italy, Grenoble in France and Denver in USA [19, 27, 28]. There was positive and strong association between formaldehyde and acetaldehyde of worker exposure as well as formaldehyde and propionaldehyde which supported previous studies but only found strong association between acetaldehyde and propionaldehyde at roadside [29]. In addition, the formaldehyde and acetaldehyde ratio of worker exposure and roadside concentration were slightly higher than studies in other countries [30-32] which these ratio changes depending upon the solar flux, vehicular emissions and temperature. Aldehyde concentrations also affect the rate of ozone formation and OH radical consumption [21, 24]. The fuel changes are likely to affect the nature and emission rates of carbonyls in vehicle exhaust and the ratio of formaldehyde and acetaldehyde (F/A) in urban area which may be less than 1 in cases where oxygenated fuels are blended with gasoline [24].

The average lifetime cancer risk of formaldehyde exposure was higher than acceptable value of 1.00×10^{-6} . If they work for longer than 1.7 years, they will have cancer risk of formaldehyde exposure but cancer risk of acetaldehyde was in acceptable limited. It should be concerned of rapidly increasing of gasohol consume in the near future of absolute ethanol like Brazil. For non-cancer risks, HQs of formaldehyde, acetaldehyde, propionaldehyde, butyraldehyde, benzaldehyde and valeraldehyde were less than 1. It means that each of CCs have not adverse effects to the workers. The symptom prevalence of chronic effects were headache, fatigue and throat irritation at 58.9%, 25.0% and 12.5% respectively which had higher prevalence than previous study [33] but they were not associated with CCs exposures. In fact, health risk assessment of workers in this study was underestimated because there were various hazardous air pollutants from petroleum exposures [34]. It should be considered of personal protective equipment (PPE) promotion such as mask for protection or reduction the worker's exposure of pollutants.

In conclusion, the gasoline station workers had cancer risk from CCs of formaldehyde exposure during their work which had good correlation with acetaldehyde and propionaldehyde. But CC exposures had no adverse effects to workers and they were not associated to complain symptoms of the workers.

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