HEALTH RISK ASSESSMENT OF EXPOSURE TO CARBONYL COMPOUNDS IN GASOLINE WORKERS IN BANGKOK, THAILAND

Sucot Nopparatbundit[1], Tanasorn Tunsaringkarn[1], Daisy Morknoy[2], Wattasit Siriwong[1,*]

[1]College of Public Health Sciences, Chulalongkorn University, Bangkok 10330, Thailand
[2]Environmental Research and Training Center, Ministry of Natural Resources and Environment, Pathumthani 12120, Thailand

ABSTRACT: Human risk assessment of volatile organic compounds (carbonyl group) via inhalation exposure route in gasoline station workers was studied during February 2011 in Bangkok, Thailand. The objectives were to measure carbonyl concentrations in gasoline stations and to assess health risk of workers in 4 gasoline stations located in urban and suburban areas of Bangkok. From each gasoline station, 2 workers were randomly recruited. Participants' ages ranged from 18-36 years. The average weight (mean ± SD) was 56.7 ± 8.1 kg. The average working time in urban and suburban areas was 9.3 and 10.0 h/day, respectively. Exposure assessment of gasoline workers was calculated using reasonable maximum exposure (RME) at the 95th percentile. The inhalation intake of carcinogenic carbonyl i.e. formaldehyde and acetaldehyde in workers was in the range of 1.90×10^{-5} to 4.11×10^{-4} mg/kg/day. Risk characterization of cancer was in the range of 2 workers in 10 million to 2 workers in one hundred thousand. For non-carcinogenic carbonyl (formaldehyde, acetaldehyde, benzaldehyde, valeradehyde, propionaldehyde, and butyraldehyde), the inhalation intake of non-carcinogenic carbonyl in workers was in the range of 4.88×10^{-3} to 1.16 µg/m^3. To assess non-carcinogenic health effects, the Hazard Index (HI) was used. The results showed that gasoline workers may not be at risk regarding inhalation exposure of non-carcinogenic health effects because the HI was not greater than the acceptable level (HI < 1).

Keywords: Carbonyl group, Inhalation exposure, Gasoline worker, RME

INTRODUCTION

Carbonyl compounds (CCs), common constituents of the atmosphere, are generally known as toxic for human health. Vehicle emission is an important source of CCs. Atmospheric photochemical reaction is another important source [1]. By product in fuel ethanol, it releases formaldehyde, acetaldehyde [2] among others. Formaldehyde and acetaldehyde are abundant in urban air [3]. Formaldehyde and acetaldehyde are suspected carcinogens [4]. Recently, a working group, convened by the IARC Monographs Programme concluded that formaldehyde is carcinogenic to humans and acetaldehyde is classified by IARC as group 2B, a possible human carcinogen based on sufficient evidence in animals and inadequate evidence in humans [5]. CCs are commonly found in rural and urban atmosphere and are of particular interest due to their potential impact on health [6]. In a large urban area, CCs can be emitted from a variety of emission sources such as motor vehicles and gasoline stations [7]. This study aimed to measure carbonyl concentrations and to assess health risk of gasoline workers in 4 gasoline stations in an urban area (2 stations) and a suburban area (2 stations) of Bangkok. The researcher focused on both carcinogenic and non-carcinogenic CCs such as formaldehyde, acetaldehyde, benzaldehyde, valeradehyde, propionaldehyde, and butyraldehyde.

MATHERIALS AND METHODS

Sampling Methodology: This study was conducted in 4 gasoline stations; two gasoline stations (P1 and P2) located in urban area, Sukhumvit road and nearby, the other two gasoline stations (P3 and P4) located in suburb area, Bangna-Trad road and nearby. This study used a triplicate sampling technique. From each station, two workers were included and an ambient air monitoring station was also operated in front of gasoline station nearby the main road. A total of 36 samples both of workers and roadside places were collected in the early, middle, and end of dry season in February 2011. Each worker was instructed for sample set consisting of one personal air sample within breathing zone. In addition, an interviewer-administered questionnaire was used for collecting socio-demographic data and exposure factors e.g. gender, age, body weight, working hours, and period of working among gasoline workers.

Sampling Analysis: Air sampling were collected by drawing air using a mini pump (Sibata Σ30, japan) through the active2,4-Dinitrophenylhydrazine (DNPH) cartridge (Wako, Japan). Samples were collected for 8 h in all sampling locations at a flow

*Correspondence to: Wattasit Siriwong
E-mail: wattasit.s@chula.ac.th

rate of 100 ml/min. After sampling, the samples were fitted by their cap, and then taken in sample pack to immediately transfer samples to laboratory within 1 hour for elution with 5 ml acetonitrile (HPLC-grade, J/T. Baker, UK). The extract was collected in a 5 ml volumetric flask (grade A SCHOTT DURAN®, 5ml ±0.025 ml) and the final volume adjusted to 5 ml by acetonitrile (HPLC-grade, J/T. Baker, UK). After extraction, the samples were refrigerated (-80 degree Celsius) until the analysis phase. High Performance Liquid Chromatography (HPLC) (model Shimaszu LC-20A) was used to analyze carbonyl samples which were eluted from cartridges. A 20µl aliquot was injected through an autosampler. The HPLC column was Discovery RP Amide C16 reverse column 25cm x 4.6mm, 5 µm. Detector UV-VIS (model SPD-20A) was used at wavelength 360 nm. The gradient mobile phase 55/45% ACN/water and the flow rate was 1 mL/min.

Quality Control (QC)*:* Each sample was triplicate injected by auto sampler into HPLC. The calibration curve used for quantification consisted of five levels ranging from 0.005 to 0.5 µg/ml and $R^2 \geq 0.999$ for all the CCs in this study. Instrument detection limit (IDL) and instrument quantification (IQL) were determined by injection ten replicates of 0.01 µg/ml mix carbonyl standards solution. The SD was calculated and 3SD was set as IDL and 10SD was set as IQL. The IDL in this study was in the range from 0.001 to 0.012 µg/ml; the IQL was in the range from 0.001 to 0.027 µg/ml. The value of IQL of each carbonyl compound was used as the detection limit for the quantification of samples [8]. Mini pumps were calibrated to 0.100 l/min (Dry-cal calibration pump) before every sampling period. QC procedures followed the AOAC international recommendation [9].

Heath risk calculation: The exposure factors from the interview were used to calculate both cancer and non-cancer risk. The major exposure route for CCs in this study was inhalation. The average and 95th percentile daily inhalation intake (Reasonable Maximum Exposure; RME) [10] were calculated for exposure assessment of carcinogenic CCs (equation 1) and non-carcinogenic CCs (equation 2). To characterize risk of both carcinogenic and non-carcinogenic effects, the dose-response or toxicity of each CC was obtained as in Table 1.

Table 1 Lists of RfCs and cancer slope factors (SF)

Compounds	RfC (µg/m3)	Cancer Slope factor (mg/kg/day)$^{-1}$
Formaldehyde	9.8 [11]	0.0455 [12]
Acetaldehyde	9 [13]	0.0077 [12]
Benzaldehyde	9 [11]	-
Valeraldehyde	420 [11]	-
Propionaldehyde	8 [11]	-
Butyraldehyde	15 [11]	-

$$I_{carcinogen} = \frac{CA \times IR \times ET \times EF \times ED}{BW \times AT} \text{ .... (1) [14]}$$

Where I is the inhalation intake (mg/kg/day), CA is the concentration of CCs in the personal air (mg/m^3), IR is the inhalation intake (m^3/h), ET is the exposure time (h/day), EF is the exposure frequency (days/year), ED is the exposure duration (years), BW is the body weight (kg), and AT is the averaging lifetime time (days).

$$I_{non\text{-}carcinogen} = \frac{CA \times ET \times EF \times ED}{AT} \text{ .... (2) [15]}$$

Where I is the inhalation intake (µg/m^3), CA is the concentration of CCs in the personal air (µg/m^3), ET is the exposure time (h/day), EF is the exposure frequency (days/year), ED is the exposure duration (years), and AT is the averaging lifetime time (h).

Table 2 summarizes exposure factors expressed as arithmetic mean and 95th percentile (RME) in the gasoline workers. Each equation, i.e. equation 1 and equation 2, were separately calculated for each area (urban and suburb). Carcinogenic risk characterization was estimated using equation 3 and non-carcinogenic risk characterization is estimated using equations 4 and 5.

Carcinogenic Risk = CSF x I .... (3) [14]

Where I is the inhalation intake (mg/kg/day), and CFS is the cancer slope factor for inhalation (mg/kg/day)$^{-1}$.

Hazard Quotient (HQ) = I/RfC .... (4) [14]

Where I is the Inhalation intake (µg/m^3), and RfC is the reference concentration of non-carcinogen (µg/m^3).

Hazard Index (HI) = ΣHQi .... (5) [16]

Where HI is the sum of hazard quotients, HQi is the summation of HQ of non-carcinogens in each site.

RESULTS

Socio-demographic characteristics: The analysis focused on gender, age,and body weight. There were 21 questionnaire respondents who were the gasoline workers and routine working in outdoor area gasoline station. There were 13 males and 8 females. Worker age was in the range of 18 to 36 years old (mean 24.4 ± 6.4 years old). The body weight (kg) was in the range of 38 to 70 kg (mean 56.7 ± 8.1 kg). Unfortunately, there was nobody using personal protective equipment such as mask and glove.

Table 2 Exposure factors related to carbonyl exposure of gasoline workers

Exposure Factors	Mean	RME	Source
CCs Concentration	Table 3	Table 3	Laboratory analysis
Exposure Time (Urban)	9.33 h/day	12 h/day	Questionnaires
Exposure Time (Suburb)	10 h/day	12 h/day	Questionnaires
Exposure Frequency (All areas)	300 days/year	300 days/years	Gasoline station manager interviewed
Exposure Duration (Urban)	2 years	10 years	Questionnaires
Exposure Duration (Suburb)	1.92 years	5 years	Questionnaires
Averaging Time (All areas)	70 years	70 years	United State Environmental Protection Agency [16]
Body weight (Urban)	56.31 kg	56.31 kg	Questionnaires
Body weight (Suburb)	57.50 kg	57.50 kg	Questionnaires
Inhalation rate	0.83 m^3/h	0.83 m^3/h	United State Environmental Protection Agency [14]

Table 3 Concentration of carbonyl compounds (arithmetic mean ±SD) (μg/m^3) collected from gasoline workers and roadside station (P1, P2, P3, and P4)

Chemical's name	*Urban (P1)*		*Urban (P2)*	
	Gasoline Workers	Roadside	Gasoline Workers	Roadside
Formaldehyde	14.23 ± 1.82	19.55 ±10.57	14.98 ± 4.63	13.96 ± 5.26
Acetaldehyde	5.88 ± 1.94	7.63 ± 6.38	10.13 ± 1.20	8.92 ± 1.11
Acetone	20.76 ± 5.60	11.83 ± 5.37	15.31 ± 4.42	15.22 ± 1.96
Propionaldehyde	1.32 ± 0.44	1.54 ± 0.80	1.90 ± 0.37	1.64 ± 0.14
Crotonaldehyde	0.67 ± 0.30	1.15 ± 0.61	0.66 ± 0.13	0.82 ± 0.26
Butyraldehyde	5.50 ± 2.62	6.14 ± 2.84	4.93 ± 2.51	5.26 ± 0.70
Benzaldehyde	<1.16	<1.16	1.24 ± 0.09	<1.16
Isovaleraldehyde	1.00 ± 0.11	1.36 ± 0.42	1.00 ± 0.10	1.36 ± 0.21
Valeraldehyde	1.40 ± 0.54	1.02 ± 0.53	0.88 ± 0.64	2.28 ± 1.92
o-Tolualdehyde	<2.64	<2.64	<2.64	<2.64
Hexanaldehyde	2.68 ± 1.36	2.28 ± 0.82	2.10 ± 0.72	2.30 ± 0.82
2,5-Dimethylbenzaldehyde	<2.84	<2.84	<2.84	<2.84
	Suburb (P3)		*Suburb (P4)*	
	Gasoline Workers	Roadside	Gasoline Workers	Roadside
Formaldehyde	17.68 ± 9.14	14.70 ± 8.0	13.80 ± 0.95	14.70 ± 2.32
Acetaldehyde	9.17 ± 2.86	6.88 ± 4.42	12.20 ± 0.47	5.82 ± 4.10
Acetone	25.45 ± 17.87	14.78 ± 7.64	12.54 ± 1.66	10.23 ± 3.14
Propionaldehyde	2.88 ± 2.84	1.38 ± 0.72	1.31 ± 0.09	1.22 ± 0.34
Crotonaldehyde	0.71 ± 0.26	0.90 ± 0.62	<0.52	0.80 ± 0.14
Butyraldehyde	4.07 ± 2.96	3.00 ± 1.32	3.16 ± 1.16	2.60 ± 0.27
Benzaldehyde	<1.16	<1.16	<1.16	<1.16
Isovaleraldehyde	1.01 ± 0.15	<0.94	<0.94	<0.94
Valeraldehyde	1.25 ± 1.15	2.50 ± 1.88	<0.52	2.78 ± 0.80
o-Tolualdehyde	<2.64	<2.64	<2.64	<2.64
Hexanaldehyde	3.08 ± 1.62	1.98 ± 0.87	2.36 ± 0.34	1.84 ± 0.36
2,5-Dimethylbenzaldehyde	<2.84	<2.84	<2.84	<2.84

Concentration of CCs in each gasoline station according to gasoline workers and roadside: The concentration of CCs collected from gasoline workers and roadside station (P1, P2, P3, and P4) are shown in Table 3. All stations experienced contamination by formaldehyde, acetaldehyde and acetone. Butyraldehyde was found in 4 gasoline stations.

Risk characterization: Risk characterization of carcinogenic CCs i.e. formaldehyde and acetaldehyde was in the range of 2 workers in 10 million to 2 workers in one hundred thousand (Table 4). Carcinogenic risk was calculated at arithmetic mean and RME levels. The carcinogenic risk in urban area found approximately in range of 2 workers in 10 million to 2 workers in one hundred thousand while suburb area were estimated in range of 3 workers in ten million to 2 workers in one hundred thousand. The carcinogenic risks among gasoline workers were thus slightly different. However, main point is that gasoline workers may be at increased cancer risk in all gasoline stations of this study.

For non-carcinogenic carbonyl (formaldehyde, acetaldehyde, benzaldehyde, valeradehyde, propionaldehyde, and butyraldehyde), the Hazard Index (HI) was used; the results showed that gasoline workers may not be at risk regarding inhalation exposure of non-carcinogenic health because the HI was not greater than the acceptable level (HI < 1) (Table 5).

DISCUSSION

Study findings raise concern about potentially increased cancer risk in workers who are working

Table 4 Results of cancer risks characterization for the gasoline workers in urban station (P1 and P2) and suburb station (P3 and P4)

Chemical's name	Arithmetic mean (μg/m3)	Intake (mg/kg/day)	Cancer Risk	RME (μg/m3)	RME Intake (mg/kg/day)	Cancer Risk
Urban (P1)						
Formaldehyde	14.23	4.60×10^{-5}	2.09×10^{-6}	16.22	3.37×10^{-4}	1.53×10^{-5}
Acetaldehyde	5.88	1.90×10^{-5}	1.46×10^{-7}	6.61	1.37×10^{-4}	1.06×10^{-6}
Urban (P2)						
Formaldehyde	14.98	4.84×10^{-5}	2.20×10^{-6}	19.8	4.11×10^{-4}	1.87×10^{-5}
Acetaldehyde	10.13	3.27×10^{-5}	2.52×10^{-7}	12.28	2.55×10^{-4}	1.96×10^{-6}
Suburb (P3)						
Formaldehyde	17.68	5.75×10^{-5}	2.62×10^{-6}	35.78	3.64×10^{-4}	1.66×10^{-5}
Acetaldehyde	9.17	2.98×10^{-5}	2.30×10^{-7}	13.58	1.38×10^{-4}	1.06×10^{-6}
Suburb (P4)						
Formaldehyde	13.8	4.49×10^{-5}	2.04×10^{-6}	15.3	1.56×10^{-4}	7.08×10^{-6}
Acetaldehyde	12.2	3.97×10^{-5}	3.06×10^{-7}	12.88	1.31×10^{-4}	1.01×10^{-6}

Table 5 Results of non-carcinogenic risk concentration for urban area (P1 and P2) and suburb area (P3 and P4)

Chemical's name	Arithmetic mean (μg/m^3)	Intake (μg/m^3)	HQ	RME (μg/m^3)	RME Intake (μg/m^3)	HQ
Urban (P1)						
Formaldehyde	14.23	1.30×10^{-1}	1.33×10^{-2}	16.22	9.52×10^{-1}	9.72×10^{-2}
Acetaldehyde	5.88	5.37×10^{-2}	5.96×10^{-3}	6.16	3.62×10^{-1}	4.02×10^{-2}
Propionaldehyde	1.32	1.21×10^{-2}	1.51×10^{-3}	2.08	1.22×10^{-1}	1.53×10^{-2}
Butyraldehyde	5.50	5.02×10^{-2}	3.35×10^{-3}	7.88	4.63×10^{-1}	3.08×10^{-2}
Benzaldehyde	1.16	1.06×10^{-2}	1.18×10^{-3}	1.16	6.81×10^{-2}	7.57×10^{-3}
Valeraldehyde	1.40	1.28×10^{-2}	3.04×10^{-5}	2.22	1.30×10^{-1}	3.10×10^{-4}
Hazard Index (HI)			2.53×10^{-2}			1.91×10^{-1}
Urban (P2)						
Formaldehyde	14.98	1.37×10^{-1}	1.40×10^{-2}	19.80	1.16	1.19×10^{-1}
Acetaldehyde	10.13	9.25×10^{-2}	1.03×10^{-2}	12.28	7.21×10^{-1}	8.01×10^{-2}
Propionaldehyde	1.90	1.73×10^{-2}	2.17×10^{-3}	2.42	1.42×10^{-1}	1.78×10^{-2}
Butyraldehyde	4.93	4.50×10^{-2}	3.00×10^{-3}	7.82	4.59×10^{-1}	3.06×10^{-2}
Benzaldehyde	1.24	1.13×10^{-2}	1.26×10^{-3}	1.37	8.04×10^{-2}	8.94×10^{-3}
Valeraldehyde	0.88	8.03×10^{-3}	1.91×10^{-5}	2.18	1.28×10^{-1}	3.05×10^{-4}
Hazard Index (HI)			3.07×10^{-2}			2.56×10^{-1}
Suburb (P3)						
Formaldehyde	17.68	1.66×10^{-1}	1.69×10^{-2}	35.78	1.05	1.07×10^{-1}
Acetaldehyde	9.17	8.61×10^{-2}	9.57×10^{-3}	13.58	3.99×10^{-1}	4.43×10^{-2}
Propionaldehyde	2.88	2.71×10^{-2}	3.38×10^{-3}	8.51	2.50×10^{-1}	3.12×10^{-2}
Butyraldehyde	4.07	3.82×10^{-2}	2.55×10^{-3}	8.72	2.56×10^{-1}	1.71×10^{-2}
Benzaldehyde	1.16	1.09×10^{-2}	1.21×10^{-3}	1.16	3.41×10^{-2}	3.78×10^{-3}
Valeraldehyde	1.25	1.17×10^{-2}	2.80×10^{-5}	3.52	1.03×10^{-1}	2.46×10^{-4}
Hazard Index (HI)			3.37×10^{-2}			2.04×10^{-1}
Suburb (P4)						
Formaldehyde	13.80	1.30×10^{-1}	1.32×10^{-2}	15.30	4.49×10^{-1}	4.58×10^{-2}
Acetaldehyde	12.20	1.15×10^{-1}	1.27×10^{-2}	12.88	3.78×10^{-1}	4.20×10^{-2}
Propionaldehyde	1.31	1.23×10^{-2}	1.54×10^{-3}	1.48	4.34×10^{-2}	5.42×10^{-3}
Butyraldehyde	3.16	2.97×10^{-2}	1.98×10^{-3}	4.32	1.27×10^{-1}	8.45×10^{-3}
Benzaldehyde	1.16	1.09×10^{-2}	1.21×10^{-3}	1.16	3.41×10^{-2}	3.78×10^{-3}
Valeraldehyde	0.52	4.88×10^{-3}	1.16×10^{-5}	0.52	1.53×10^{-2}	3.63×10^{-5}
Hazard Index (HI)			3.07×10^{-2}			1.06×10^{-1}

and continually exposed for a long time in gasoline stations (exposure duration was approximately 5 to 10 years at RME). Calculated risks were somewhat higher in the suburban area than the urban area. Further research is needed to explained this finding. This finding assumed that the risks depended on exposure factors in each gasoline station, such as amount of fuel sold, number of customers in gasoline station, and environmental factors.

Other studies have observed health risks of formaldehyde and acetaldehyde in public placesr such as shopping centers, supermarkets, railway stations, bus stations, furniture stores, ballrooms and offices. Specific risks depended on exposure factors, which differed from place to place [17].

Possible source of CCs to gasoline workers exposure: For any source of CCs, it may come from vehicular emission; most of formaldehyde was released in the ambient background [12]. Gasoline stations are also a source of VOCs, for example formaldehyde and acetaldehyde, that vaporize from fuels containing methanol or ethanol [8]. At the roadside areas, formaldehyde and acetaldehyde in Bangkok have high concentrations as high traffic volumes [8]. In metropolitan areas, formaldehyde is the predominant aldehyde emitted by automobiles [18].

CONCLUSION

This study measured 12 CCs concentration in 4 gasoline stations. Each station was measured by roadside stationary monitoring and monitoring of workers. Questionnaires were used to collect the socio-demographic data and exposure factors. 6 CCs were assessed for potential cancer and non-cancer risks. Study results showed that all studied workers may be at increased risk of cancer, because of the high levels of formaldehyde and acetaldehyde. For non-cancer risk, the results showed that gasoline workers may not at risk regarding inhalation exposure because the HI was not greater than the acceptable level (HI < 1).

Recommendation guideline: In general, it is known that Risk = Hazard × Exposure [10]. Although, we may not reduce the hazard of CCs, but we may reduce exposure by encourage gasoline workers in order to concern about their health. Use of personal protective equipment could conceivably reduce risk, although it was not possible to address this hypothesis in the present study.

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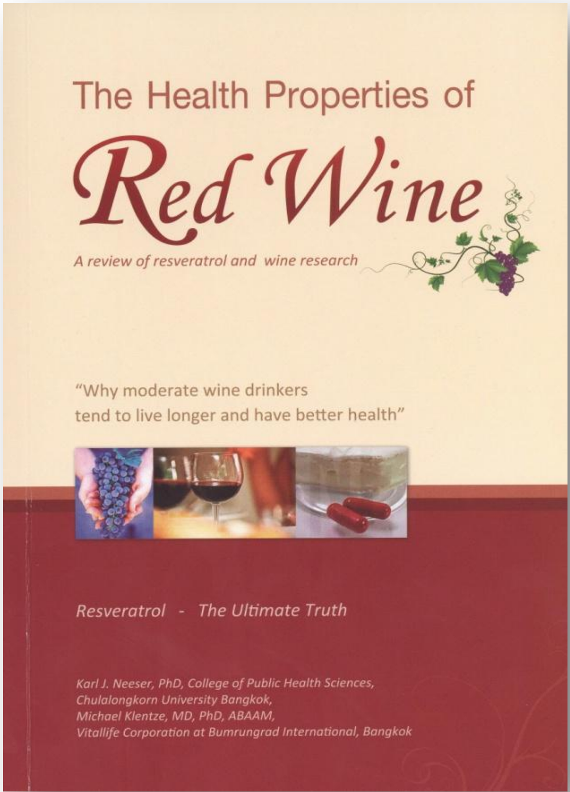

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