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Potential Risk Comparison of Formaldehyde and Acetaldehyde Exposures in Office and Gasoline Station Workers

Tanasorn Tunsaringkarn*, Wattasit Siriwong *, Tassanee Prueksasit**
Saowanee Sematong *, Kalaya Zapuang*, Anusorn Rungsiyothin*

*College of Public Health Sciences, Chulalongkorn University, Bangkok 10330, Thailand.
** Department of General Science, Faculty of Science, Chulalongkorn University, Bangkok, 10330 Thailand

Abstract- Assessment of exposures of formaldehyde and acetaldehyde that related to urinary biomarkers among office and gasoline station workers was conducted. Our results revealed that the evaluation of ambient air formaldehyde and acetaldehyde in official were significantly higher than in gasoline stations; 28.72 to 11.31 µg m^{-3} and 7.85 to 3.34 µg m^{-3} respectively (*independence t-test*, $p<0.01$). The sum average lifetime cancer risk of official and gasoline stations workers exposure to formaldehyde and acetaldehyde were determined as the values of 2.51E-05 and 1.30E-05, which were higher than an acceptable criteria defined as 1.00×10^{-6}. For non-cancer health effects, sum of the hazard quotients (hazard index, HI) of exposures to these two substances in office and gasoline station workers were found at the ratio of 0.40 and 0.21. Both HIs were lower than minimal level which indicated that there is no risk of other health effects. Ambient air formaldehyde was strongly and significantly correlated to ambient air acetaldehyde (r=0.907, $p<0.01$) as well as urinary formaldehyde was significantly correlated to urinary acetaldehyde (r=0.498, $p<0.01$). Urinary formaldehyde did not show correlation to ambient air formaldehyde but urinary acetaldehyde was significantly correlated to ambient air acetaldehyde (r=0.429, $p<0.01$). In addition, urinary formic acid was not related to any exposures**.** Results of this study supported that urinary acetaldehyde may be used as biomarkers for bio-monitoring in human exposures.

Index Terms- Exposure, Formaldehyde, Acetaldehyde, Urinary biomarker

I. Introduction

Formaldehyde and acetaldehyde are member of aldehyde group which considered as human carcinogens- and are classified as B1 and B2 [1-2]. Chronic inhalation of these chemicals can lead to eye irritation, respiratory complaints and nervous disorders [3]. The combustion of oxygenated fuels resulted in greater aldehyde emissions and contributed to the increasing ambient concentrations [4-5] especially in the gasoline station areas [6]. The most likely route of aldehyde's exposure is by inhalation, but other routes, for examples ingestion and dermal absorption, are possible. Exposures to these types of aldehydes could lead to a chance of cancer risk. Possibility of inhalation formaldehyde and acetaldehyde may exert toxics at the sites which transport via blood circulation system [7]. Human health effects against any carcinogenic and non-carcinogenic depend upon the chemical hazard, chemical involved, concentration and exposure time [8]. However, several studies reported that formaldehyde and acetaldehyde concentration levels in the indoor were higher than outdoor [9-11]. Higher doses may overwhelm the metabolic capacities and unmetabolites may remain in urine. The study on formaldehyde and acetaldehyde exposure assessments of official and gasoline workers in Thailand has been limited. Therefore, it was our main objective to assess whether or not there is any risk in these two chemicals by monitoring the urinary metabolites as the biomarkers for formaldehyde and acetaldehyde exposures.

II. Materials and Methods

A. Population Study

The survey population consisted of 74 workers: 33 official workers of Chulalongkorn University and 41 gasoline workers from 6 gasoline stations in Pathumwan district, Bangkok, Thailand. The study was conducted from May to July 2010. Permission to conduct study in human subjects was approved by the Ethical Review Committee for Research Involving Human Research Subjects, Health Science Group, Chulalongkorn University.

B. Sample Collections

Exposure to ambient air formaldehyde and acetaldehyde were monitored during work shift of 8 hours (6.00 A.M.-2.00 P.M.) by active sampling with 2, 4 dinitrophenylhydrazine (2, 4-DNPH) cartridges at flow rate 100 ml min^{-1}. All air samples were kept at 4^{o}C during transportation to laboratory. The DNPH cartridges were extracted immediately after sample collection and analyzed by high performance liquid chromatography/ultraviolet-visible detector (HPLC/UV-VIS).

For urine samples, seventy-four (74) urine samples were collected in glass bottles and kept at 4^{o}C during transportation to laboratory, stored at -20^{o}C and later analyzed by gas chromatography/flame ionize detector (GC/FID).

C. Ambient Air Analyses

Fourteen ambient air formaldehyde and acetaldehyde (12 from gasoline stations and 2 from offices) were collected by DNPH cartridges, extracted with acetronitrile and analyzed by HPLC with UV-VIS detector at 360 nm [12]. The mobile phase included 45% water and 55% acetonitrile with flow rate 1 ml min^{-1} and incubated at 40^{o}C. Analysis of the ambient air samples was performed at Environmental Research and Training Centre (ERTC) of Thailand.

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D. Urinary Analyses

The urinary analyses of formaldehyde, acetaldehyde and formic acid were analyzed by GC/FID [13]. The determinations of urinary creatinine (Cr) were performed by spectrophotometry method [14]. All urinary analyses performed at a standard laboratory (Special Laboratory, Bangkok, Thailand). All measured values were corrected by urinary Cr concentrations for clinical chemistry analysis.

E. Cancer and Non-cancer Risk Calculation

The cancer risk was calculated by multiplying chronic Daily Intake (CDI) by inhalation cancer slope factor (CSF_i) and Hazard quotient (HQ) for non-cancer calculated by dividing exposure concentration (EC) by inhalation reference concentration (RfC) as followed equations:

Cancer risk = $CDIxCSF_i$;

Where; CDI = (CAxIRxETxEFxED)/(BWxAT)

CDI (mg kg^{-1} day^{-1}) = Chronic Daily Intake; CSF_i (mg kg^{-1} $day^{-1})^{-1}$ = Inhalation cancer slope factor; CA (mg m^{-3}) = Contaminant concentration in air; IR (m^3 hr^{-1}) = Inhalation rate (0.875 m^3 hr^{-1} assumed for adult); BW (kg) = Body weight (60.0 kg, average body weight of workers); ET (hrs day^{-1}) = Exposure time (8 hr day^{-1} for workers); EF (days $year^{-1}$) = Exposure frequency (350 d yr^{-1} for gasoline workers and 269 d yr^{-1} for official workers); ED (years) = Exposure duration (30 years for workers); AT (days) = Averaging time (70 yrs x 365 days)

Cancer risk > $1.00x10^{-6}$ means Carcinogenic effects of concern; Cancer risk ≤ $1.00x10^{-6}$ means Acceptable level

For non-cancer risk assessment

HQ = EC/RfCx1000; where; EC = (CAxETxEFxED)/AT

EC (µg m^{-3}) = Exposure concentration ; RfC (mgm^3) = Inhalation reference concentration

HQ > 1 means Adverse non-carcinogenic effects of concern; HQ ≤ 1 means Acceptable level

F. Statistical analyses

All statistical analyses were carried out with SPSS 17.0 statistical software package. Normal distribution was assessed by Kolmogorov-Smirnov test. The mean values of comparative variables were computed by descriptive statistics. The correlations between ambient air and urinary biomarkers were computed by Spearman's rho correlation. A statistically significant difference was accepted at a *p*-value of <0.05.

III. RESULTS

The characteristic and biological measurements of formaldehyde and acetaldehyde in ambient air and urine of office and gasoline station workers were shown in Table 1. Mean age and number of male and female workers in both groups were not difference. Both of the ambient air formaldehyde and acetaldehyde concentrations in offices were significantly higher than those in gasoline stations (*independence t-test*, $p<0.001$). The average levels of urinary acetaldehyde analyses of gasoline station workers were significantly lower than in the office workers (*independence t-test*, $p<0.01$). While, the urinary formic acid analyses in office and gasoline station workers were higher in female than male workers with a significantly higher in female than in the male gasoline station workers (*independence t-test*, $p<0.05$). Whereas, the World Health Organization (WHO) acceptable limits on urinary Cr concentrations is between 0.3 and 3.0 g L^{-1} [15].

Results of averages lifetime cancer risk of office and gasoline station workers whom exposed to formaldehyde and acetaldehyde were as in Table 2. The averages lifetime cancer risk of formaldehyde in both groups were higher than acceptable criteria of 1.00 x 10^{-6}. Thus, cancer risk summations of official and gasoline station workers were at 2.51x 10^{-5} and 1.30 x 10^{-5}. The non-cancer risk, the hazard quotient (HQ) of formaldehyde and acetaldehyde exposure in office workers were found at the ratio of 0.31 and 0.09 while in gasoline station workers were 0.16 and 0.05 respectively. Both HQs and hazard index (HI) in office and gasoline station workers were 0.40 and 0.21 which were lower than one (HI<1).

Analyses of ambient air formaldehyde were strongly correlated to ambient air acetaldehyde and urinary acetaldehyde while urinary formaldehyde was correlated to urinary acetaldehyde as shown in Table 3. Moreover, ambient air acetaldehyde was correlated to urinary acetaldehyde. However, both ambient air formaldehyde and acetaldehyde were not correlated to urinary formic acid.

IV. DISCUSSION

Our results revealed that both office and gasoline station workers had potential cancer risk of exposure to formaldehyde. For those of gasoline station workers whom worked outdoor, they were directly exposed to the oxygenated gasoline and automobile [16]. The previous study found that the ambient concentrations of formaldehyde at five roadsides and residential areas in Bangkok were 11.53 and 9.65 µg m^{-3} while ambient concentrations of acetaldehyde at roadsides and residential areas were 3.51 and 3.11 µg m^{-3} respectively [17]. Both ambient air formaldehyde and acetaldehyde concentrations at roadside areas were closed to their concentrations at gasoline station areas in this study. While, the office workers who normally worked inside, they were directly exposed to indoor and building materials including particleboard and plywood, wood finishes, furniture, textiles, consumer products [18]. Our results showed lower indoor formaldehyde and acetaldehyde levels than the results obtained by Ongwandee et al [19] study in office building in Bangkok as well as other results in other countries such as China [20], Egypt [21], Mexico [22]. Both ambient air formaldehyde and acetaldehyde indoor concentrations of offices were significantly higher than outdoor concentrations of gasoline stations ($p<0.001$). In addition formaldehyde/acetaldehyde ratio of offices was 3.7 while formaldehyde/acetaldehyde ratio of gasoline station was 3.4 which supported indoor carbonyl compounds were higher than that of outdoor [23]. However, indoor concentration of formaldehyde and acetaldehyde are lower than permissible exposure limit (PEL) of 0.75 ppm (0.92 mg m^{-3}) for formaldehyde and 200 ppm (360.33 mg m^{-3}) for acetaldehyde average over 8-hr work shift [24].

The indoor–outdoor (I/O) relationships showed that the I/O ratios were 2.5 (28.72/11.31) for formaldehyde and 2.4 (7.85/3.34) for acetaldehyde in ambient air of indoor office and outdoor gasoline stations. Our results supported some previous studies [25-26] especially of Ongwandee et al. [19] found that I/O relationships of office buildings in Bangkok were 3.5 (35.5/10.1) for formaldehyde and 5.7 (17.1/7.9) for acetaldehyde.

They also suggested that indoor sources are more important contributors to the indoor levels than outdoor sources such as infiltration of oxygenated gasoline and vehicle exhaust. Moreover, formaldehyde levels can be higher in indoor air than in outdoor air. Important determinants of indoor air levels include the sources of the formaldehyde, the age of the source materials, temperature, humidity, and ventilation rates [27].

The averages lifetime cancer risk of office workers exposed to indoor air formaldehyde and acetaldehyde could be simply translated as 2.5 cancerous cases per 100,000 and 1.3 cancerous cases per 100,000 for the gasoline station workers over lifetime 70-year [28]. It was possible that other environmental exposures during the remaining part of the day may aggravate both the actual cancer and non-cancer risk. The risk estimates assumed that the workers spend their life time (70 years) exposed to various hazardous air pollutants in their working areas [29]. The risk values estimated only inhalation dose with absolute absorption into the body, which means that the cumulative risk estimates for cancer and non cancer health effects may be under estimated [30].

However, the HI for non-cancer risk, of formaldehyde and acetaldehyde were bellowed the reference levels (HI < 1). Thus, all workers did not have adverse health effects from formaldehyde and acetaldehyde exposures.

Averages urinary formaldehyde and acetaldehyde concentrations of office workers were higher than those found in the gasoline workers but the urinary acetaldehyde showed significant higher in office workers. In addition, urinary formaldehyde was not correlated to ambient air formaldehyde but urinary acetaldehyde was significantly correlated to ambient air acetaldehyde while urinary acetaldehyde was significantly correlated to ambient air formaldehyde and urinary formaldehyde. However, formaldehyde can undergo rapid chemical changes immediately after absorption and is much more rapidly dialyzed by the kidneys and excreted through the urinary bladder than by respiration [31] but it reacts rapidly with serum protein and few protein in urine [32]. There were significantly and strongly correlated between ambient air formaldehyde and acetaldehyde as well as urinary formaldehyde was correlated to urinary acetaldehyde. In addition, urinary acetaldehyde was correlated to urinary formaldehyde. Thus, urinary formaldehyde may be correlated to ambient air formaldehyde in higher exposure concentration. Similar finding was done by Taranenko and Efimova studied [33] urinary formaldehyde from the pediatric population in the town of Shelekhov, of the Irkutsk Region and found significant correlation to air concentration of living rooms. Mautempo et al. [34] investigated the potential of using the formic acid for the biological monitoring of aldehyde exposure and report that the formaldehyde is metabolized in the body to formic acid and may be excreted in the urine which formic acid, which have a potential use as a biomarker of formaldehyde exposure in work environments. However, the amount of formaldehyde exposure had to be considered since if the formaldehyde exposure was less than 500 μg m^{-3} it did not significantly affect upon formic acid shift [35]. Our data exhibited the same results that ambient air formaldehyde and acetaldehyde levels in both groups investigated were not correlated to urinary formic acid level. However, the urinary formic acid in female gasoline workers was significantly higher than male. These high formic excretion and acidosis might be signaled the potential of inducing nephrotoxicity [36-37] so, female gasoline worker may had higher potential of this effect.

V. CONCLUSION

We concluded that the office workers had higher risk of formaldehyde and acetaldehyde exposures than gasoline workers. The urinary aldehyde may be used as non-invasive biomarkers for human exposures by inhalation.

Table 1: Characteristic and biological measurements in formaldehyde and acetaldehyde exposed workers

Parameter	Official Workers (N=33) Mean±SE	Gasoline Workers (N=41) Mean±SE	*p-value**
Age (year)	35.8±1.5	32.3±1.8	*NS*
Male N(%)	16(48.5)	23(56.1)	-
Female N(%)	17(51.5)	18(43.9)	-
Ambient air formaldehyde (μg m^{-3})	28.72±1.15	11.31±0.20	*<0.001*
Male	11.30±0.22	26.44±2.12	
Female	31.14±0.00	11.33±0.35	
Ambient air acetaldehyde (μg m^{-3})	7.85±0.07	3.34±0.09	*<0.001*
Male	7.72±0.12	3.25±0.11	
Female	7.99±0.00	3.47±0.35	
Urinary formaldehyde (mg gCr^{-1})	50.13±13.41	28.79±3.01	*NS*
Male	60.47±25.15	26.64±3.79	
Female	39.14±7.64	31.82±4.93	
Urinary acetaldehyde (mg gCr^{-1})	90.92±17.71	40.93±3.93	*<0.01*
Male	65.83 ±8.04	37.19±4.66	
Female	117.58±34.85	45.99±6.74	
Urinary formic acid (mg gCr^{-1})	20.91±4.04	32.00±5.87	*NS*
Male	16.66 ± 6.12	20.93 ± 4.08[a]	
Female	25.44 ± 5.17	49.71± 12.76[a]	

*significant difference between gasoline and office workers
[a]significant difference between sex of workers at *p<0.05*

Table 2: Average life time cancer risk and hazard quotients (HQ) assessments for official and gasoline workers

Parameter	CDI (mg kg^{-1} day^{-1})	Average Life Time Cancer Risk	EC (μg m^{-3})	HQ
Office Official Workers				
Formaldehyde	1.06E-03	2.22E-05	3.02	0.31
Acetaldehyde	2.89E-04	2.89E-06	0.83	0.09

Total Risk		2.51E-05	-	0.40
Gasoline station Workers				
Formaldehyde	5.42E-04	1.14E-05	1.55	0.16
Acetaldehyde	1.60E-04	1.60E-06	0.46	0.05
Total Risk		1.30E-05	-	0.21

Table 3: Spearman's rho correlations between parameters

Para-meter	Am. F	Am. A	U. F	U. A	U. FA
Am.F	1	0.907 *(p<0.01)*	0.097 *(p=0.423)*	0.424 *(p<0.01)*	-0.139 *(p=0.255)*
Am.A		1	0.094 *(p=0.438)*	0.429 *(p<0.01)*	-0.026 *(p=0.829)*
U.F			1	0.498 *(p<0.01)*	-0.003 *(p=0.981)*
U.A				1	-0.006 *(p=0.962)*
U.FA					1

Note: Am. = Ambient air, F = formaldehyde,
A = acetaldehyde, FA = formic acid, U. = urinary

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AUTHORS

First Author – Tanasorn Tunsaringkarn (M.Sc, Biochemistry): Corresponding author, she worked as researcher, assistant dean and an executive board member at College of Public Health Sciences (CPHS), Chulalongkorn University. She is a member of the Asia Pacific Academic Consortium of Public Health (APACPH). Her research interests include environmental health and toxicology. Her research interests include environmental health and toxicology. She has more than 50 papers in scientific journals as well as presentations in seminar and conference proceedings. College of Public Health Sciences, Chulalongkorn University, Institute Building 2-3, Soi Chulalongkorn 62 Phyathai Rd., Bangkok 10330, Thailand.
E-mail: tkalayan@chula.ac.th

Second Author – Wattasit Siriwong (Ph.D., Environmental Management): He is a faculty member, Assistant Dean, and an executive board member of the College of Public Health Sciences (CPHS), Chulalongkorn University. He is an active researcher in the field of environmental health and also a member of the editorial board of International Journal of Occupational and Environmental Health and Journal of Health Research. College of Public Health Sciences, Chulalongkorn University, Institute Building 2-3, Soi Chulalongkorn 62 Phyathai Rd., Bangkok 10330, Thailand.
E-mail: Wattasit.S@chula.ac.th

Third Author - Tassanee Prueksasit (Ph.D., Urban Engineering; Environmental Engineering): She is a lecturer and her research interests are the air pollution of some organic compounds including VOCs (BTEX, carbonyl compounds) and polycyclic aromatic hydrocarbons (PAHs), and of particulate matters, and health risk assessment of the exposure to such air pollutants. Department of General Science, Faculty of Science, Chulalongkorn University, Phyathai Rd., Bangkok 10330, Thailand
E-mail: Tassanee.C@chula.ac.th

Fourth Author - Saowanee Sematong (M.Sc., Ecology): She worked as researcher for College of Public Health Science, Chulalongkorn University. Her research interests include breastfeeding, environmental impact assessment and toxicology. College of Public Health Sciences, Chulalongkorn University Institute Building 2-3, Soi Chulalongkorn 62 Phyathai Rd., Bangkok 10330, Thailand
E-mail: Saowanee.Se@chula.ac.th

Fifth Author - Kalaya Zapuang (B.Ed)**:** She is a research-staff at College of Public Health Sciences (CPHS), Chulalongkorn University. College of Public Health Sciences, Chulalongkorn University Institute Building 2-3, Soi Chulalongkorn 62 Phyathai Rd., Bangkok 10330 Thailand
E-mail: Zkalaya@chula.ac.th

Sixth Author - Anusorn Rungsiyothin (B.Ed): He is a research-staff in laboratory of College of Public Health Sciences (CPHS), Chulalongkorn University. College of Public Health Sciences, Chulalongkorn University Institute Building2-3, Soi Chulalon gkorn 62 Phyathai Rd., Bangkok 10330 Thailand
E-mail:ranusorn@chula.ac.th