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Ambient air's volatile organic compounds and potential ozone formation in the urban area, Bangkok, Thailand

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ABSTRACT

Aims: The study were evaluated for the presence of the ambient air volatile organic compounds (VOCs) (benzene, toluene, ethylbenzene and xylene [BTEX] and carbonyl compounds [CCs]) at the university, roadside and residential areas in order to assess the applicability for prediction of ozone formation in ambient air of these in urban area, Bangkok, Thailand. **Materials and Methods:** The ambient air VOCs levels were evaluated by the gas chromatography/flame ionization detector for BTEX analysis and the high-performance liquid chromatography/UV-visible detector for CCs analysis. **Results:** The comparison of VOCs levels in those areas showed that the roadside area had the highest VOCs levels (of toluene and benzene levels), while the residential area had the lowest of benzene level. However, the benzene levels of all studied-areas were higher than ambient air quality standard. The contributions of hydrocarbons to local ozone formation potential were evaluated in terms of maximum incremental reactivity. Our results showed that the largest contributors to ozone production in Bangkok were toluene and formaldehyde (possibly from the emission of biofuel motor vehicle exhaustion). The roadside area also had the highest ozone formation potential. The aromatic hydrocarbon was the major contribution to anthropogenic emissions of VOCs. However, anthropogenic emissions and photochemistry are mainly transported VOCs to ozone formation. **Conclusion:** There was the highest VOCs level at roadside area and also was the largest amount of ozone level from chemical reactions. Therefore, it is a need for more research and the environmental protection policy because it may have serious health risk from these pollutants in these areas.

KEY WORDS: Ambient air, ozone formation, urban areas, volatile organic compounds

INTRODUCTION

Ozone, one of the oxidized products, was produced by a complex chemical reaction between volatile organic compounds (VOCs) and nitrogen oxides (NOx) in the presence of sunlight. It is a major environmental concern because of its adverse impacts on human health [1] and on crops and forest ecosystems [2,3]. Most of average ozone level reaches its maximum in winter and its minimum during the rainy season. Human expose to ozone by ways of inhalation and contact between a person and from the pollutant in the microenvironments where they spend their time. Exposure to ozone has been linked to a number of respiratory health effects, including significant decreases in lung function, inflammation of airways, and caused several symptoms such as cough and pain when breathing deeply [4-7]. In Asia, ground-level ozone concentrations are alarmingly high in some large metropolitan areas, as demonstrated in many countries such as China, Japan, Korea, Taiwan and Thailand [8]. Many researchers have reported the rising ground-level ozone in the range of 0.5-2.0% per year over the mid-latitudes of the Northern Hemisphere and in some areas exceeding the standard level (of 100 part per billions [ppb]) [9]. The ground-level ozone was not associated with death, cardiovascular and respiratory diseases (Relative risk [RR] = 1.009, 95% confidence interval [CI] = 0.911, 1.117; RR = 1.239, 95% CI = 0.901, 1.705; RR = 1.157, 95% CI = 0.791, 1.692) [10]. All of the relative risks were for 10 ppb increase in ground-level ozone concentration.

However, in Bangkok, Ruangdej and Chaosuansreecharoen [10] found that ground-level ozone was marginally associated with death from unclassified causes (RR = 1.234, 95% CI = 0.998, 1.525).

Air quality in urban areas is affected mainly by photochemical oxidants [11] and ozone concentrations showed a relatively correlation with traffic densities [12]. Motor vehicle exhaustion, industrial emissions, and chemical solvents are the major anthropogenic sources of these chemicals [13,14]. Vehicle emissions constitute the major source of ozone precursors in Bangkok [15]. For the meteorological parameters in metropolitan areas such as in Bangkok, the monsoonal rain and wind play significant effects on the characteristics of ground-level ozone [16]. The VOCs in the atmosphere are regularly oxidizing to ozone by reaction with hydroxyl radicles in the presence of NOx in present of sunlight [5,17]. Maximum incremental reactivity (MIR) was the most commonly used to developed the express the ozone formation potential from VOCs [18].

Thus, this study evaluated ambient air VOCs of benzene, toluene, ethylbenzene and xylene (BTEX) and carbonyl compounds (CCs) in the university, roadside and residential areas and assessed the applicability for prediction of ozone formation in ambient air of these in an urban area, Bangkok, Thailand.

MATERIALS AND METHODS

Study Sites

Pathumwan district of central Bangkok was chosen as the study areas: Roadside, university and residential areas. The roadside areas were on the Rama I, Phaya Thai and Henry Dunant roads, which with high traffic congestion during the day. The Chulalongkorn University was selected as the university area. It located on those roadside areas and surrounded by several department stores (Ma Boon Klong Center, Siam Paragon and Siam Discovery Stores, and Siam Square shopping center). The residential areas were the five low-income communities (of Wat Pathumwanaram, Bonkaipattana, Soi Prajane, Salakhin, and Chaochoocheep) which located around the roadside and university areas [Figure 1].

Ambient Air Sample Collections and Analyses

A total of 31 ambient air samples were collected (6 from the university; 10 from roadside and 15 from residential areas) from 16 different sites in Pathumwan district, from February to June 2013. The active sampling method was chosen to collect the ambient air sample by using of a 2,4-dinitrophenylhydrazine cartridge (for CCs) and activated charcoal tube (for BTEX) connected to a low flow rate air pump (1.0 mL/min). The device was set up for 8 h (8.00 am-16.00 pm). Both cartridge and charcoal tube were kept at 4°C and stored in the refrigerator. The BTEX analysis was done by the gas chromatography/flame ionization detector procedure as described by Tunsaringkarn *et al.* [19]. The CCs analysis was done by the high-performance liquid chromatography/UV-visible detector according to the Method TO-11A [20]. The methodology analyses were validated through determination of the limits of detection, linearity, precision and accuracy. The calibrations were conducted

Figure 1: Study Sites Map O = University areas: 1 = Satit Chulalongkorn Demonstration school, 2 = Entrance-exit Gate 1, 3 = Entrance-exit Gate 2, 4 = Entrance-exit Gate 3, 5 = Entrance-exit Gate 4, 6 = Checkpoint- Faculty of Science, • = Roadside areas: 1 = Community Pharmacy Clerkship, 2 = Bangkok Bank ■ Public Company Limited, 3 = Krung Thai Bank Public Company Limited, 4 = Faculty of Science, 5 = Siam Commercial Bank Public Company Limited, = Residential areas: 1 = Wat Pathumwanaram community, 2 = Bonkaipattana community, 3 = Soi Prajane community, 4 = Salakhin community, 5 = Chaochoocheep community

using standard solutions of five concentrations with the coefficients of determination (R^2) above 0.999. The percentages of relative standard deviations (% RSD) were < 0.178. Each sample analysis was done by triplicate.

The permission to conduct this study was approved by the Ethical Review Committee for Research Involving Human Research Subjects, Health Science Group, Chulalongkorn University with COA No. 089/2012.

Statistical Analysis

All analytical measurements were performed in duplication to give value with a standard error. All analyses were carried out with Statistics Package for the Social Sciences Version 17 (Chulalongkorn University License [IBM Microsoft, New York, USA]). Descriptive statistical analysis was evaluated on concentrations of ambient air concentrations. The mean difference levels between areas were evaluated by one-way ANOVA (lithium dodecyl sulfate -*post-hoc* multiple comparisons). A statistically significant difference was accepted at $P < 0.05$ as other medical studies. The ozone formation potentials were calculated by the following equation [18,21]:

$$\text{Ozone } (O_3) = \text{VOCs } (\mu g/m^3) \times \text{MIR } (gO_3/gVOCs)$$

RESULTS

Occurrence and Distribution of Ambient Air VOCs

Our results of the ambient VOCs concentrations of university, roadside and residential areas in central Bangkok area were

89.07, 200.16 and 84.67 $\mu g/m^3$ respectively [Table 1], with the average of 124.25 $\mu g/m^3$. The highest of the average total VOCs level (200.16 $\mu g/m^3$) was recorded at the roadside areas. However, they were not significant differences, when compared to the other areas. Most average ambient VOCs concentrations were benzene, toluene, ethyl benzene, m,p-xylene, o-xylene, formaldehyde and acetaldehyde at 18.59, 77.41, 4.33, 4.13, 3.16, 8.57 and 4.27 $\mu g/m^3$ respectively.

VOCs (toluene:benzene [T/B]; xylene:benzene [X/B]; xylene:ethyl benzene [X/EB]; formaldehyde acetaldehyde [F/A]; acetaldehyde:propionaldehyde [A/P]) ratios

Of the total 5 VOCs ratios (T/B, X/B, X/EB, F/A and A/P) measured in this study, the residential areas possessed 3 highest ratios of the T/B (18.11), X/B (2.91)and X/EB (2.88), while the university areas had one of F/A (2.43)ratio and the roadside had one of A/P (6.86) ratio [Table 2]. The average T/B ratios of university, roadside and residential areas were 1.29, 5.35, and 18.11 respectively with the average ratio of 4.19. The residential areas were also found to have the highest X/B (2.91) and X/EB (2.88) ratios among the 3-studied areas, while the average ratios of both ratios were 0.39 and 1.68. The F/A ratios of university, roadside and residential areas were 2.43, 1.92, and 1.59 respectively with the average ratio of 2.00. While the A/P ratios were 4.46, 6.86, and 5.50 respectively with the average ratio of 5.77.

Ozone Formation Potential

The total ozone formation potentials in the university, roadside and residential areas were 365.06, 932.70, and 436.36 $\mu g/m^3$, respectively [Table 3]. Most of ozone formation of the university, roadside and residential areas came from toluene, formaldehyde, acetaldehyde, m,p-xylene, o-xylene, benzene and ethyl benzene at 371.57, 81.04, 27.90, 32.24, 24.12, 13.39, and 13.16 $\mu g/m^3$ respectively.

DISCUSSION

Motor vehicles emit millions of pounds of hazardous pollutants into the air each year in the United State, including VOCs [22]. The petroleum constituents of primary interest to human health have been the aromatic hydrocarbons (i.e. benzene, toluene, ethyl benzene, and xylenes-BTEX) poly-nuclear aromatic hydrocarbons and gasoline additives. Fuel combustion also emits carbon monoxide, benzene, acetaldehyde, formaldehyde, and diesel particulates. Benzene, formaldehyde, and diesel particulates are the major components of vehicle exhaust that drive the estimated inhalation risks in urban areas [23]. The most abundance of VOCs in ambient air were toluene and benzene, which were the same results as reported by Barletta *et al.* [24] and Tiwari *et al.* [25]. Among the BTEX, the toluene was the most abundance levels of all areas, however, the highest toluene levels were recorded from the roadside areas. The toluene level at residential area was lower than the roadside area because it is restricted the area for all motor vehicles. Therefore, the toluene emission from motorcycles was less than those of the other areas [26,27]. Ho and Lee [28] and Kourtidis *et al.* [29] reported that the amounts of benzene, toluene, ethylbenzene, emitted from motor vehicles, were high in traffic areas. While, Bravo *et al.* [30] studied in Mexico City and found that benzene and m,p-xylene at the roadside and university areas were significant higher than the residential areas. Most of the residential areas were nearly main roads of Rama I, Rama IV, Phyathai, Charu Muang and expressway of Sirat (Toll road) and Chaloem Maha Nakhon with a high number of motor vehicles a day [31]. However, Han and Naeher [32] explained this situation that the traffic VOCs decreases drastically as the distance from the main traffic roads. However, ambient air benzene levels in all areas were higher than ambient air quality standards (1.17 $\mu g/m^3$) [33] and higher than the previous study at the roadside of Bangkok in 2007 by Laowagul *et al.* [34]. For the examination of the CCs, the ambient air formaldehyde and acetaldehyde levels were the most abundance and with the highest levels recorded in the roadside

Table 1: Ambient VOCs concentration ($\mu g/m^3$) at 21 locations in Pathumwan area

VOCs	Mean±SE			
	University area (n=6)	Roadside area (n=10)	Residential area (n=15)	Average
Benzene	26.90±3.51**	25.50±3.58$^{\#\#}$	3.37±3.23**,$^{\#\#}$	18.59±2.79
Toluene	34.74±13.33	136.45±65.56	61.04±12.21	77.41±32.15
Ethylbenzene	5.47±3.13	4.11±0.87	3.41±1.00	4.33±0.97
M, p-Xylene	2.75±1.39**	2.98±0.76$^{\#\#\#}$	6.67±2.96**,$^{\#\#\#}$	4.13±1.18
O-Xylene	2.53±1.13	3.80±0.80	3.14±1.25	3.16±0.60
Formaldehyde	8.45±1.53	14.11±3.25$^{\#}$	3.14±2.19$^{\#}$	8.57±1.77
Acetaldehyde	3.48±0.51	7.34±1.79	1.98±1.65	4.27±0.95
Propionaldehyde	0.78±0.22	1.07±0.25	0.36±0.30	0.74±0.14
Crotonaldehyde	0.29±0.22*	0.00±0.00*,$^{\#\#}$	0.48±0.50$^{\#\#}$	0.26±0.11
Butyraldehyde	0.34±0.18	0.42±0.40	0.31±0.48	0.36±0.20
Benzaldehyde	0.12±0.11	0.93±0.43	0.31±0.08	0.45±0.22
Isovaleraldehyde	0.00±0.00	0.01±0.00	0.06±0.22	0.02±0.02
Valeraldehyde	0.84±0.38	0.75±0.36	0.09±0.12	0.56±0.21
o-Tolualdehyde	0.12±0.12	0.45±0.38	0.06±0.08	0.21±0.18
m, p-Tolualdehyde	0.08±0.08	0.93±0.63	0.09±0.29	0.37±0.31
Hexaldehyde	1.16±0.56	0.61±0.33	0.64±0.39	0.80±0.23
2,5Dimethyl benzaldehyde	1.02±0.17	1.31±1.31	6.56±0.38	0.83±0.62
Total	89.07±9.98	200.16±32.78	84.67±14.56	124.25±18.64

*,$^{\#}$Significant different between different areas at $P<0.05$, **,$^{\#\#}$Significant different between different areas at $P<0.01$, $^{\#\#\#}$Significant different between different areas at $P<0.001$, VOCs: Volatile organic compounds, SE: Standard error

areas. These pollutants were mainly from combustion emission from biofuel which was rapidly increasing in Bangkok [35-37].

The T/B ratio of the residential area was much higher than the average of 4.19. The T/B ratio is a tool for characterizing the distance from vehicle emission sources [38]. The residential area recorded the lowest benzene level and about averaged-toluene level. The X/B ratio is used to indicate the possibility of air mass transported, while the X/EB ratio is used to identify the degree of evolution of photo-oxidation reaction. The X/B and X/EB ratios were high in the residential area indicated that there were low amount of air mass from the motor vehicles (transportation) and photo-oxidation from the nearby main roads and highways [39]. While both ratios were lower in the university and roadside areas, these indicated that there was higher air mass transportation with higher photochemical activity to ozone formation at roadside area. The small X/B and X/EB ratio revealed that the photo-chemical reaction were active [25].

Formaldehyde and acetaldehyde are emitted by vehicles that use the oxygenated fuels such as methanol and ethanol [40]. The high F/A ratio in the university area referred to low-used of ethanol as automotive fuel which supported data from Brazil where high levels of acetaldehyde in urban air reflect the nationwide use of ethanol fuel [41]. The residential areas were normally polluted from the vehicle's emission from the main roads and express highways. The average F/A ratio appeared in this study was in ranged of the average F/A reported in other studies [42-44]. The A/P ratio is generally used to indicate the anthropogenic origin of ambient carbonyls, since the propionaldehyde is believed to be associated only with anthropogenic emissions [45]. This ratio is typically found to be high in a rural area, but low in the urban area [46]. The A/P ratios in our study were high in the roadside areas which demonstrated that the lifetime of acetaldehyde exceeds that of propionaldehyde with respect to photolysis reactions [17]. The large photochemical production of acetaldehyde at high temperatures and strong solar radiation may be counterbalanced rapid loss by photolysis [47,48]. Our result was similar to the study in Rome by Possanzini *et al.* [43] but a decreasing trend in VOC levels was observed in Rome urban air during 2011 suggesting the effectiveness of European directives on air quality [49].

Table 2: Comparison of VOCs ratios in different areas

Area	VOCs ratio				
	T/B	X/B	X/EB	F/A	A/P
University area	1.29	0.20	0.96	2.43	4.46
Roadside area	5.35	0.27	1.65	1.92	6.86
Residential area	18.11	2.91	2.88	1.59	5.50
Average	4.19	0.39	1.68	2.00	5.77

VOCs: Volatile organic compounds, T/B: Toluene: benzene, X/B: Xylene: benzene, X/EB: Xylene: ethyl benzene, F/A: Formaldehyde: acetaldehyde, A/P: Acetaldehyde: propionaldehyde

The ozone formation potential is calculated by the multiplication of the VOCs concentrations by the MIR coefficient [18,21]. The total concentrations of VOCs in the university, roadside and residential areas were 88.05, 199.46, and 85.15 $\mu g/m^3$, respectively. While, the total ozone formation potentials in the university, roadside and residential areas were 365.06, 932.70 and 436.36 $\mu g/m^3$, respectively. Our results indicated that the ozone levels of all 3 studied-areas were higher than air quality standard limited (100 $\mu g/m^3$) [50]. Among the 3 studied-areas, the roadside areas exhibited the highest ozone concentration (199.46 $\mu g/m^3$) and the ozone formation potential (932.70 $\mu g/m^3$) which the most chemical reactions transformed toluene and formaldehyde. The presence of toluene and formaldehyde are normally the main pollutants to ozone formation. Results of the measurements of toluene and formaldehyde levels in the roadside area in this study were the highest (136.45 and 14.11 $\mu g/m^3$) among the 3 other studied-areas. Thus, the toluene and formaldehyde may contribute to the overall risk associated with population exposure to toxic

Table 3: Ozone formation potential with respect to MIR coefficient in 3 studied areas

VOCs	MIR coefficient[a]	Concentration ($\mu g/m^3$)			O_3 formation potential[b] ($\mu g/m^3$)		
		University area	Roadside area	Resident area	University area	Roadside area	Resident area
Benzene	0.72	26.9	25.5	3.37	19.37	18.36	2.43
Toluene	4.80	34.74	136.45	61.04	166.75	654.96	292.99
Ethylbenzene	3.04	5.47	4.11	3.41	16.63	12.49	10.37
M, p-Xylene	7.80	2.75	2.98	6.67	21.45	23.24	52.03
O-Xylene	7.64	2.53	3.8	3.14	19.33	29.03	23.99
Formaldehyde	9.46	8.45	14.11	3.14	79.93	133.48	29.70
Acetaldehyde	6.54	3.48	7.34	1.98	22.76	48.00	12.95
Propionaldehyde	7.08	0.78	1.07	0.36	5.52	7.58	2.55
Crotonaldehyde	9.39	0.29	0	0.48	2.72	0.00	4.51
Butyraldehyde	5.97	0.34	0.42	0.31	2.03	2.51	1.85
Benzaldehyde	−0.67	0.12	0.93	0.31	−0.08	−0.62	−0.21
Isovaleraldehyde	4.97	0.00	0.01	0.06	0.00	0.05	0.30
Valeraldehyde	5.08	0.84	0.75	0.09	4.26	3.81	0.46
o-Tolualdehyde	−5.09	0.12	0.45	0.06	−0.61	−2.29	−0.31
m, pTolualdehyde	−0.59	0.08	0.93	0.09	−0.05	−0.55	−0.05
Hexaldehyde	4.35	1.16	0.61	0.64	5.05	2.65	2.78
Total formation	-	88.05±10.25	199.46±33.74	85.15±14.98	365.06±10.25	932.70±162.62	436.34±72.31

[a]Fanizza *et al.*[21] and Carter [18], [b]Ozone (O3)=VOCs ($\mu g/m^3$)×MIR ($gO_3/gVOCs$). VOCs: Volatile organic compounds, MIR: Maximum incremental reactivity

air pollutants. Therefore, it applied to the conditions in which outdoor airs in the roadside areas have large amounts of exhaust vehicle emission and photo-oxidation in atmospheric especially in daytime [51,52]. However, there are many other factors influenced the amounts of pollutants from the tailpipe emission as well as made the spatial variation even greater, such as fuel chemistry, traffic density, driving conditions, meteorological conditions (temperature, wind, etc.) and city buildings [53-56].

Ambient ozone trends are influenced by year-to-year changes in meteorological conditions, population growth, VOCs ratio, the ratio of the concentration of nitric oxide (NO) and nitrogen dioxide (NO_2)-NOx ratio, and by changes in emissions from ongoing control measures [57]. Emissions of the NOx and VOCs may increase by 95% and 65%, respectively, mainly driven by the expected increasing in road traffic volume. On the other hand, ammonia, mainly emitted from agriculture, is projected to double by 2030 in East Asia [58]. From our results, all the ratio ozone formation potentials of 3 studied-areas (university:roadside:resident = 1:2.6:1.2 [365.06:932.70:436.34]) had potential to produce ozone over the air-standard limitation. These lead to the high potential in which people have to expose to the high amount of ozone. The increasing volume of ozone might be directly affected the health conditions. Since, ozone can induce respiratory symptoms (chest tightness, wheezing, or shortness of breath) including coughing, throat irritation, pain, burning, or discomfort in the chest when taking a deep breath, lung function decrements and airways inflammation [59]. Higher daily ozone concentrations are associated with increased asthma attacks, increased hospital admissions, increased daily mortality, and other markers of morbidity. The consistency and coherence of the evidence for effects upon asthmatics suggests that the ozone can make asthma symptoms worse and can increase sensitivity to asthma triggers.

CONCLUSIONS

In the most dense-population areas in Bangkok, like our 3 studied-areas, people who work or live very close to the main roadsides and highways must have risk of serious health effects from air pollutants, especially from the carcinogenic pollutant liked benzene. This study demonstrated the usefulness of ambient air VOCs levels screening and ozone formation potential in difference urban studied-areas. We need to do more extensive researches and begin to exercise the environmental protection policy.

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