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Ozone Formation Potential of Oxygenated Hydrocarbons: Phasing-in of Gasohol in Bangkok, Thailand

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Abstract: *The levels of ozone formation potentialfor four oxygenated hydrocarbons, namely formaldehyde, acetaldehyde, acrolein and propionaldehyde, were estimated using photochemical ozone creation potential indices. During the transformation period for the enforcement of gasohol cleaner fuels in Bangkok from 2008 to 2013, the average OFPs of formaldehyde were the foremost levels (480.54/509.16; pre-/post-enforcement; µg.m^{-3}), followed by acetaldehyde OFPs (302.82/289.03), propionaldehyde OFPs (48.86/43.53) and acrolein OFPs (30.38/33.20) subsequently. The Mann-Whitney U-test was conducted to determine the significance of gasohol on the quantities of the four carbonyl OFPs and atmospheric ozone. The results suggested that the maximum ozone concentrations (135.53/160.42;µg.m^{-3}) for Bangkok and its vicinity areas were significantly increased after the compliance of gasohol fuels, whereas the OFP levels of four carbonyl compounds were statistically insignificant (p< 0.05, 2-tailed).*

Keywords: *Photochemical ozone creation potential, ozone formation potentialoxygenated hydrocarbons, carbonyl VOCs, gasohol fuels*

I. Introduction

Urban ozone pollution is one of the most important environmental problems in Bangkok [1] affected by rapid urbanization, economic development and increases in number of transport vehicles [2-3]. Oxygenated volatile organic compounds (VOCs), as hazardous air pollutants (HAPs) [4] and ozone precursors [5-6], are emitted into the atmosphere extensively. Oxygenated VOCs and ozone are not only the major species of photochemical smog but also the important human health hazards, exclusively to the respiratory system [7]. On-road vehicles and industrial sources are typically the major anthropogenic sources of global environmental concerns [8-9]. Additionally, Thailand VOCs emission was high, about 14% of all countries in South East Asia (SEA), added by road transport, whereas 96.84% of greenhouse gases containing non-methane volatile organic compounds (NMVOCs) released by Thailand's transport sector was from road mode [10].

In Bangkok and its adjacent areas, Thailand, the maximumlevels for 1-hour average of ground level ozone have been exceeded the standard (100 ppb) [11] from 1996 to 2013 [1, 12-13]. In addition, the national promotion programs for phasing out of methyl tertiary butyl ether (MTBE) in gasoline, which has been contaminated in in surface and groundwaters, and phasing in of ethanol-gasoline blended in 2004 [14] and gasohol cleaner-fuel under the enforcementfor Euro IV emission standards in 2012 [15], have been applied as the environmental control strategies. These may have possibly been the additional emission sources for urban ozone precursors because the levels of oxygenated hydrocarbons emitted from gasohol are higher than gasoline fuels [14, 16-17].

The objective of this research is to evaluate the levels for regional ozone formation potentials (OFPs) of oxygenated VOCs ozone precursorsand atmospheric ozone in Bangkok and its vicinity, Thailand during the transformation period of gasoline-to-gasohol usage. This was achieved using the framework of Mann-Whitney U-test to collate the levels of surface ozone and OFPs of target aldehyde compounds from 2008 to 2013.

II. MaterialsandMethods

2.1 Study area

The study sites for evaluating the potential of surface ozone formation were consisted of three inner districts and one outer district of Bangkok comparable to its suburban area in PathumThani province, located at the north-east of Bangkok, showed in Table 1 and Fig. 1. The study stations, with a distance of 50 kilometers (approximately 31 miles), were selected based on a year-round wind direction at upwind and downwind, which mainly impacted by SW and NE monsoon of the center of Bangkok.

Table 1. OFPsand ozone monitoring sites

Monitoring Station*		Geographic Location	Province
(A)	Environmental Research &Training Center, ERTC	14°03' N 100°43' E	Residential/General PathumThani
(B)	Chokchai Police-Station, CC4	13°48' N 100°36' E	Urban Bangkok
(C)	DinDaeng National Housing Authority, DD	13°46' N 100°33' E	Urban Bangkok
(D)	King Chulalongkorn Memorial Hospital, CUH	13°44' N 100°32' E	Urban Bangkok
(E)	Bansomdej Chaopraya Rajabhat University, BSD	13°44' N 100°29' E	Residential/General Bangkok

*The (F) & (G) sites in Bangkok University at Rangsit (BU) and Thonburi Power Sub-Station (ThBri) were employed as the alternative stations of (A) and (D) stations for air quality data other than oxygenated VOCs samples data respectively.

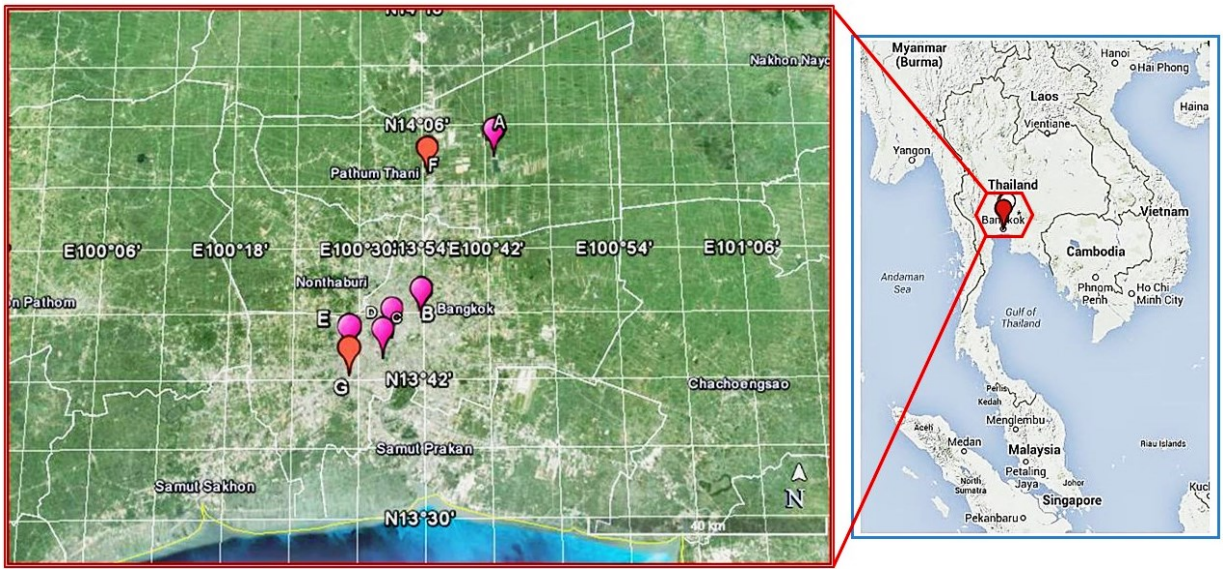

Fig.1. Map for OFPs and ozone monitoring sites: (A) ERTC, (B) CC4, (C) DD, (D) CUH, (E) BSD, (F) BU and (G) ThBri

2.2 Data acquisition

All monitoring data, including four carbonyl compounds, specifically formaldehyde, acetaldehyde, acrolein and propionaldehyde, and ozone were acquired from Pollution Control Department (PCD), Thailand. The four compounds of oxygenated VOCs were actively sampled onadsorbentcartridge coated with 2,4-dinitrophenylhydrazine (2,4-DNPH) and quantified the hydrazone derivatives by high performance liquid chromatography (HPLC) equipped with ultraviolet (UV) detection [18]. In addition, ozonewas determined by continuous chemiluminescence detection, was based on the U.S EPA ambient standard method [19]. The data of daily maximum 1-hour average of ozone samples wasextracted correspondingly to monthly oxygenated VOCs samples data.

2.3 Ozone formation potentials

The OFPs of four oxygenated VOCs were estimated from the photochemical ozone creation potential (POCP) method, which was developed by Derwent et al. [20] and the POCP indices were quantified from the formation of ozone relatively to the capacity of ethylene using photochemical trajectory model allowing for long range transport, as expressed in equation (1-2) and listed in Table 2 [21-23].

$$POCP_i = \frac{O_{3i} - O_{3basecase}}{O_{3ethylene} - O_{3basecase}} \times 100 \quad (1)$$

$$OFP_i\ [\mu g \cdot m^{-3}] = C_{VOC_i}\left(\mu g \cdot m^{-3}\right) \times POCP_i \quad (2)$$

Where $O_{3basecase}$ refers to the ozone mixing ratio along the trajectory in the base case, O_{3i} with an additional of the i^{th} VOC species, $O_{3ethylene}$ refers to that with the same mass of ethylene, OFP_i, C_{VOCi} and $POCP_i$ are the ozone formation potential, the concentration and the photochemical ozone creation potential coefficient of i^{th} VOC respectively.

Table 2. POCP indices for target oxygenated VOCs [22]

VOCs	POCP
Formaldehyde	46
Acetaldehyde	55
Acrolein	90.4
Propionaldehyde	72

2.4 Statistical analysis

The Mann-Whitney U-test was used to compare the significant differences ($p < 0.05$) in the intervals before (Jan 2008 to Jun 2012) and after (Jul 2012 to Dec 2013) the compliance of gasohol as cleaner-fuels [24-26]. All data was examinedusing IBM SPSS Statistics 22.

III. ResultsandDiscussion

The monthly OFPs of aldehyde compounds and maximum ozone concentrations obtained in this study are shown in Fig. 2-3 and Table 3-4. In orderto compare the differences of sample means for the quantities of monthly maximum ozoneand oxygenated VOCs' OFPs in the two periods of time, before and after the enforcement of gasohol application as cleaner fuels for Euro IV emission standards. Ethanol blended fuels have been legally enforced to be the cleaner gases in Bangkok since July 2015 [15], whereas gasohol fuels have been used as an alternatively additive for MTBE in Thailand since 2004 [14].

3.1 OFP levels of oxygenated hydrocarbons

Table 3 presented the OFP levels of oxygenated VOCs (formaldehyde, acetaldehyde, acrolein and propionaldehyde) were varied across sites. However, the OFP levels of carbonyl compounds in core areas of Bangkok trended to be greater than the OFP levels in suburb areas, according to high density of vehicular emission sources in the central districts of Bangkok [27-28]. In addition, there were upward tendencies for the OFP levels of formaldehyde over almost all of areas around Bangkok after the enforcement of gasohol fuels, while other species were diverse across the sites. The formaldehyde OFPs were the highest levels, followed by the acetaldehyde OFPs and appeared to have similar trends to ground level ozone concentrations(in Fig. 2), due to they were the major carbonyl species released from biomass and biofuels oxidation which created from exhaust gases [29-30]. Moreover, the emission of carbonyls VOCs (e.g. formaldehyde, acetaldehyde) from ethanol blended fuels were higher than from gasoline fuels [14, 16-17, 31].

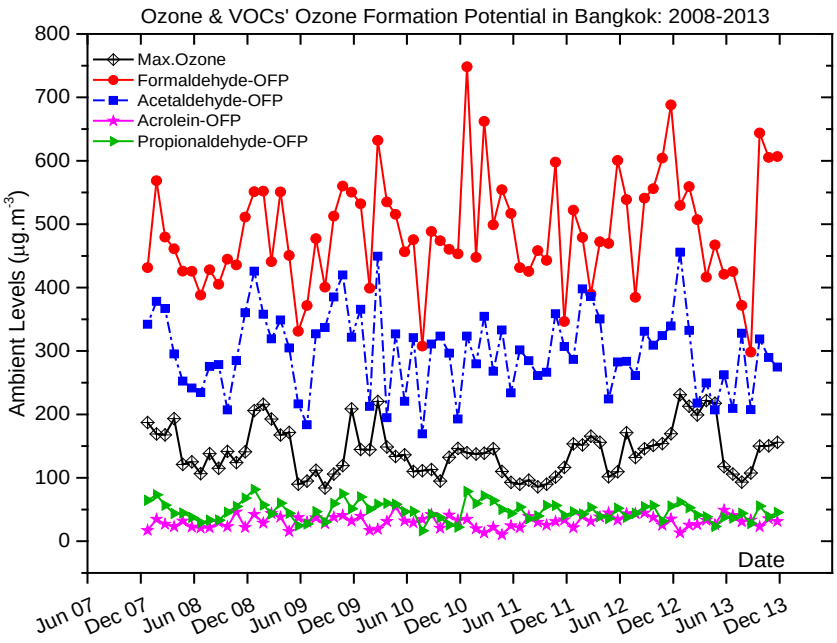

Fig.2. Levels of ozone and the ozone formation potentials of target carbonyls compounds in Bangkok (2008-2013)

Table3. The average monthly levels of ambient ozone and VOCs' OFPs during 2008 to 2013

Site	Variable (μg.m^{-3})	Pre-enforcement Mean ±SD*, n = 54	Post-enforcement Mean ±SD, n = 18
ERTC	Max. ozone	149.59 ± 53.30	184.22 ± 77.84
	Formaldehyde-OFP	397.60 ± 120.24	351.21 ± 166.09
	Acetaldehyde-OFP	254.04 ± 93.27	221.34 ± 94.16
	Acrolein-OFP	29.71 ± 18.31	28.40 ± 16.17
	Propionaldehyde-OFP	40.37 ± 23.15	21.92 ± 8.15
CC4	Max. ozone	122.96 ± 43.40	139.56 ± 43.92
	Formaldehyde-OFP	510.28 ± 130.52	618.84 ± 206.20
	Acetaldehyde-OFP	366.33 ± 133.09	340.35 ± 110.30
	Acrolein-OFP	19.07 ± 19.07	16.79 ± 20.85
	Propionaldehyde-OFP	53.19± 22.97	47.58 ± 29.84
DD	Max. ozone	108.34 ± 45.53	126.11 ± 39.55
	Formaldehyde-OFP	580.78 ± 179.27	594.81 ± 197.57
	Acetaldehyde-OFP	326.68 ± 89.63	324.13 ± 103.57
	Acrolein-OFP	37.69 ± 21.36	44.02 ± 21.42
	Propionaldehyde-OFP	55.00 ± 19.66	37.56 ± 17.69
CUH	Max. ozone	148.37 ± 51.58	178.89 ± 56.13
	Formaldehyde-OFP	549.72 ± 104.50	533.82 ± 137.52
	Acetaldehyde-OFP	315.89 ± 95.72	306.17 ± 125.82
	Acrolein-OFP	33.49 ± 19.38	44.45 ± 24.84
	Propionaldehyde-OFP	55.52 ± 17.64	47.91 ± 29.12
BSD	Max. ozone	148.39 ± 51.16	173.33 ± 56.00
	Formaldehyde-OFP	364.32 ± 97.93	447.09 ± 154.62
	Acetaldehyde-OFP	251.16 ± 91.59	253.17 ± 164.70
	Acrolein-OFP	31.93 ± 15.25	32.34 ± 11.02
	Propionaldehyde-OFP	40.24 ± 26.24	62.62 ± 27.31
BKK**	Max. ozone	135.53 ± 35.72	160.42 ± 42.00
	Formaldehyde-OFP	480.54 ± 81.59	509.16 ± 105.41
	Acetaldehyde-OFP	302.82 ± 65.41	289.03 ± 62.36
	Acrolein-OFP	30.38 ± 9.38	33.20 ± 9.13
	Propionaldehyde-OFP	48.86 ± 14.86	43.52 ± 10.71

*SD= standard deviation, Grubbs' test statistic, G (0.05, 72) < 3.1

**BKK = Bangkok (average from 5 sites), n = 72 for each site (72 months), 5 sites,

The Mann-Whitney U-test, abridged in Table 4, was employed to evaluate the hypothesis that the levels of monthly maximum ozone and the aldehyde-VOC OFPs (formaldehyde, acetaldehyde, acrolein and propionaldehyde) after the compliance of gasohol cleaner fuels would higher, on the average, than their levels before the compliance of gasohol fuels. It was observed that the monthly maximum ozone concentrations for the BSD site and overall Bangkok area were significantly increased after the compliance of gasohol cleaner fuels. Whereas, the OFP levels of four carbonyl compounds were varied at different sites: (1) Formaldehyde OFPs were statistically significant in the core district sites of CC4 and BSD; (2) Acetaldehyde OFPs were insignificantly changed for all sites; (3) Acrolein OFPs were substantially raised up in the site of CUH; (4) Propionaldehyde OFPs were noticeably enlarged in the stations of ERTC, DD and BSD ($p < 0.05$, 2-tailed). By the findings of this study, the enforcement of gasohol fuels was probablyone of the factors caused to rise the levels of aldehyde OFPs and maximum ozone. According to emission products of ethanol gasoline blended fuels, for example, formaldehyde is reactively to form ozone [32-33], acrolein is one the most efficient ozone producers [34] and carbonyls compounds (formaldehyde, acetaldehyde, propionaldehyde) are observed to form ozone [35-36].

Table 4. Result for test of significant differences using Mann-Whitney U-test

Site	Variable (μg.m^{-3})	Pre-enforcement	Post-enforcement	Mann-Whitney, $n_1 = 54, n_2 = 18$, $p < 0.05$ (2-tailed)*	
		Mean rank	Mean rank	U	Z
ERTC	Max. ozone	34.41	42.78	373.00	-1.47
	Formaldehyde-OFP	38.36	30.92	385.50	-1.31
	Acetaldehyde-OFP	38.35	30.94	386.00	-1.30
	Acrolein-OFP	36.77	35.69	471.50	-1.90
	Propionaldehyde-OFP	40.82	23.53	252.50	-3.04*
CC4	Max. ozone	34.51	42.47	378.50	-1.40
	Formaldehyde-OFP	33.35	45.94	316.00	-2.21*
	Acetaldehyde-OFP	38.20	31.39	394.00	-1.20
	Acrolein-OFP	37.90	32.31	410.50	-1.00
	Propionaldehyde-OFP	38.46	30.61	380.00	-1.38

Table 4. Result for test of significant differences using Mann-Whitney U-test (Cont'd)

Site	Variable (µg.m^{-3})	Pre-enforcement	Post-enforcement	Mann-Whitney, $n_1 = 54, n_2 = 18$, $p < 0.05$ (2-tailed)*	
		Mean rank	Mean rank	U	Z
DD	Max. ozone	34.16	43.53	359.50	-1.65
	Formaldehyde-OFP	36.31	37.06	475.50	-0.14
	Acetaldehyde-OFP	37.19	34.44	449.00	-0.48
	Acrolein-OFP	35.02	40.94	406.00	-1.04
	Propionaldehyde-OFP	40.91	23.28	248.00	-3.10*
CUH	Max. ozone	33.74	44.78	337.00	-1.94
	Formaldehyde-OFP	37.69	32.94	422.00	-0.83
	Acetaldehyde-OFP	38.06	31.83	402.00	-1.09
	Acrolein-OFP	34.05	43.86	353.50	-1.72*
	Propionaldehyde-OFP	38.39	30.83	384.00	-1.33
BSD	Max. ozone	33.80	44.61	340.00	-1.90*
	Formaldehyde-OFP	32.94	47.17	294.00	-2.50*
	Acetaldehyde-OFP	36.81	35.56	469.00	-0.22
	Acrolein-OFP	36.31	37.06	476.00	-0.13
	Propionaldehyde-OFP	32.63	48.11	277.00	-2.72*
BKK	Max. ozone	33.14	46.58	304.50	-2.36*
	Formaldehyde-OFP	34.91	41.28	400.00	-1.12
	Acetaldehyde-OFP	37.87	32.39	412.00	-0.96
	Acrolein-OFP	34.78	41.67	393.00	-1.21
	Propionaldehyde-OFP	38.56	30.33	375.00	-1.44

3.2 Levels of maximum ozone

The levels of monthly maximum ozone over five sites and overall areas of Bangkok were from 112.78 to 158.25 µg.m^{-3}, displayed in Fig.3. The lowest peak ozone concentration was found at DD site, the inner district of Bangkok and the highest level by BU site, the alternative site of ERTC, in PathumThani Province (downwind site during November and April) located at the NE suburb of Bangkok. These suggested that the mechanisms of photochemical reactions required time to cook and yield ozone with long range transport at downwind suburb area. These were similarly to the previous reports, revealing that the area of Bangkok and the adjacent provinces were impacted by SW and NE monsoon [30, 37]. However, the ozone levels of the other suburban sites, namely BSD and the alternative site of CUH (ThBri) were also higher than the ozone concentrations of inner districts of Bangkok. These advised that the two SW suburban sites of Bangkok, where were closely to the gulf of Thailand, were associated with vehicular and industrial emission sources in SamutPrakan and SamutSakhon provinces coupled with additional effects from sea breeze. Mixing of chlorine (Cl) in sea salt and urban NO_x could availably create decisive conditions to produce nitryl chloride ($ClNO_2$), which cooperated in photodecomposition during the daytime yielded chlorine radicals, regenerated nitrogen dioxide (NO_2) and ozone production might raise from their reactions [38-39]. The maximum ozone concentrations in the post-enforcement period of gasohol fuels were found to be greater than the pre-enforcement period for all sites, summarized in Table 3.

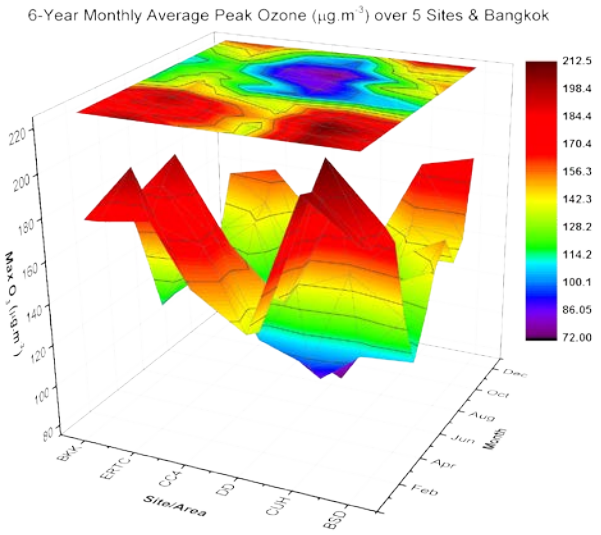

Fig.3. 3Dplot of 6-year monthly averages of maximum ozone concentrations over 5 sites and overall areas for Bangkok and its vicinity.

IV. Conclusion

Photochemical ozone creation potential indices was used to estimate the levels of ozone formation potential of four carbonyl VOCs: formaldehyde, acetaldehyde, acrolein and propionaldehyde in order to investigate the effect of the enforcement of gasohol cleaner fuels in Bangkok from 2008 to 2013 on the ground level ozone. The average OFPs of formaldehyde and acetaldehyde were the first two main species come after by propionaldehyde and acrolein respectively. The effect of gasohol enforcement on the levels of the four carbonyl OFPs and ambient ozone was examined using the Mann-Whitney U-test. From the results of this study, the compliance of gasohol might be one of significant factors affected on the maximum ozone concentrations in Bangkok and the adjacent area. However, the OFP levels were not significantly observed to be increased by the gasohol enforcement.

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