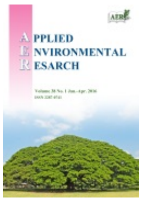
App. Envi. Res. 43(4) (2021): 14-28

Applied Environmental Research

Journal homepage : http://www.tci-thaijo.org/index.php/aer



Ozone Formation Potential of Ambient Volatile Organic Compounds at Roadside in Bangkok, Thailand

Narita Fakkaew[1,2], Surat Bualert[1,2,*], Thunyapat Thongyen[1,3], Thitima Rungratanaubon[1,2]

[1] The Monitoring of Microclimate and Air Pollution in Thailand Project, Kasetsart University, Bangkok 10900, Thailand
[2] Department of Environmental Science, Faculty of Environment, Kasetsart University, Bangkok 10900, Thailand
[3] Department of Environmental Technology and Management, Faculty of Environment, Kasetsart University, Bangkok 10900, Thailand
[*] Corresponding author: surat.b@ku.ac.th

Article History
Submitted: 3 February 2021/ Revision received: 1 May 2021/ Accepted: 5 May 2021/ Published online: 14 September 2021

Abstract

Volatile organic compounds (VOCs) play an important role in atmospheric chemistry due to their high reactivity—reacting photochemically with oxides of nitrogen (NO_x) in the presence of solar radiation forming tropospheric ozone (O_3). Each VOC species have different effects on ozone formation according to the rates and pathways of their reactions. The objective of this study aims to examine ozone formation from the estimation of ozone formation potential (OFP). The observation of 29 VOCs species was carried out in the urban area near the roads of Bangkok, Thailand. Measurements were carried out during the dry season, from 16th February to 15th March, 2018. The air samples were analyzed using gas chromatography flame ionization detector (GC-FID). The results showed that toluene had the highest VOCs concentration followed by propane, and carbon tetrachloride (CCl_4). The average ratio of benzene to toluene (B/T) and toluene to benzene (T/B) indicate that both toluene and benzene emitted from industrial area and vehicular emission. Ratio of m/p-xylene to benzene (m/p-X/B) indicate that BTEX emitted far from the source. The ozone formation potential indicated that toluene was the main VOC contributing to the total ozone formation. High VOCs concentration in monitoring site was influenced by vehicular sources and the sea breeze brought the pollutants back to the land.

Keywords: Ozone formation potential; Urban area; Bangkok; Volatile organic compounds; Vehicle exhausts

Introduction

Volatile organic compounds (VOCs) are organic compounds found in the atmosphere. Sources of VOCs is emitted from anthropogenic (e.g., vehicular exhaust, solvent usage, common cooking gas mixtures, industry) and biogenic

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https://doi.org/10.35762/AER.2021.43.4.2

sources (e.g., vegetation, soil and earth activity) [1] depend on the element and activity in specific areas. In urban areas some of the major sources of VOCs is from vehicular exhaust emissions on roadsides include toluene, propane, and *i*-pentane [2]. VOCs and their oxidation products are harmful to human health, as VOC species have both acute and chronic effects on human health [3–4]. VOCs also plays an important role as a precursor of tropospheric ozone (O_3) which is a secondary pollutant generated by photochemical reaction of VOCs and NOx in the presence of sunlight [5–6]. O_3 is a greenhouse gas and is associated with cardiovascular mortality in the summer and winter seasons [7]. In Bangkok, Chon Buri, and Rayong Province, Thailand, the hourly O_3 concentration exceeded the Thailand National Ambient Air Quality standard (NAAQs) during the dry season (summer and winter) [8–9]. At Kasetsart University, located in Bangkok, Thailand, the KU-Tower is 117-m used for measurement of meteorology and air pollution. Attached to the tower are tubes for trace gas sampling that was installed on the tower at 30, 75 and 110 m above the ground. The O_3 monitoring data is taken at 30 m during 2016 to 2018. The results found that all the hourly maximum O_3 concentration exceeded the 1-h O_3 standard (100 ppb) during the dry season with the frequently during the summer being 96% and winter 4% as the frequently of high O_3 concentration about 0.8 % per year. Dry season is high O_3 concentration due to the strong solar intensity [10], lower relative humidity, lower wind speed, less cloud and the chemical regime of O_3 formation was VOC-sensitive showed that the VOCs is a precursor controls O_3 production by the reaction of VOC and hydroxyl radical (OH) [11]. However, during the wet season the O_3 concentration was low because of the higher relative humidity, lower temperature, weaker solar intensity and cloudier sky [12]. Zhang et al. studied on O_3 and O_3 precursors in Thailand founded that the O_3 concentration at stations are located far from city center is higher than city center because of O_3 dispersion as this is based on its precursors and the longer time in the NO_x cycle to react with the OH radical which often takes about several hours [13].

Ozone production is produced by specific hydrocarbon and is dependent on its particular oxidation mechanism. It can be influenced by concentrations of VOCs, and different species have different contributions to the ozone photochemical formation [14]. Each VOC species have different OH loss rates therefore different effects on ozone formation [15]. Wang et al. studied the ozone formation potential (OFP) in Zhoushan, finding that type of VOCs species was high emitted (e.g., *n*-butane, *m/p*-xylene, *i*-pentane) from source differs from high OFP (e.g., *m/p*-xylene, *o*-xylene, ethylene) [16]. Aromatics contributed the most to ozone formation in Zhoushan, but alkanes contributed the most to ozone formation in Wuhan [17]. Nevertheless, the chemical reaction activity was lower than other VOC species. Toluene was high in abundance and reaction with the highest OFP in the suburban (industry and resident) in the North of Bangkok site. This site is located about 25 km from the monitoring site in this study (Kasetsart University) [18]. Ozone formation is strongly related to meteorological parameters such as temperature, solar radiation, and wind speed [13]. The OFP is used as an index to quantify the potential of VOC compound in ozone production and used to assess the roles of species in the process of ozone formation.

This study focuses on determining the concentrations of roadside VOCs and the source profiles of VOCs based on BTEX species; benzene, toluene, ethylbenzene, and *m/p*-xylenes ratios and the chemical reactivity of each VOCs species to the formation of ozone by ozone formation potential.

Materials and methods

1) Monitoring site and measuring plan

Measurements were conducted at the Faculty of Environment (13.55°N and 100.57°E), Kasetsart University located in the north of Bangkok, the capital city of Thailand. The city is an urban site with a high air pollution level that is mainly generated from vehicle traffic which is mainly emitted from motor vehicles include gasoline engines accounted for 56%, followed by diesel engines at 28%, and compressed natural gas (CNG) at 16% with other sources such as residential area, power plants, industries, incinerators, and biomass/residue burning. [15, 17–19]. This monitoring site is near two roads, with toll way above an 8-lane main road located about 420 m on the west side of the monitoring site, and a 2-lane about 28 m on the north side of the monitoring site (Figure 1).

Measurements were carried out in the dry season, during the 16th February to 15th March 2018 as the local summer starts from 16th February to 15th May and local winter starts from 16th October to 15th February in Thailand [19–20]. Air sampling and real-time monitoring of VOCs, NO_x, and O_3 was conducted 30 m from ground level on the rooftop of the Faculty of Environment, Kasetsart University. This observation detects 29 VOCs species, according to previous studies [13, 21].

2) Instrumentation

2.1) Measurement of VOCs

VOC species were continuously analyzed by online VOCs monitoring system, which involves a detection technology is thermoelectric cooling system to control the temperature of the sample trap through to gas chromatograph with flame ionization detector (GC-FID) system (Chromato-Sud airmoVOC C2-C6 and airmoVOC C6-C12, France). For the measurement of C2-C6, the sample passes through three Peltier-cooled traps (-8 ºC). Then, the tube is heated, and thermo-desorption is fixed at 300 ºC, followed by separation on an Al_2O_3/Na_2SO_4 column (id = 0.53 mm, length = 25 m). A flame ionization detector (FID) is used to quantify the species present. For the measurement of heavier hydro-carbons (C6-C12), the sample is passed through a single trap, then thermos-desorption is fixed at 380 ºC, followed by separation in a capillary column (MXT 30 CE) (id = 0.28 mm, length = 30 m). The detection is then done with the FID. This instrument includes one auto-calibration unit, which uses three internal permeation tubes with standard compounds, for auto-calibration at 2.00 pm. every day. The FID was the standard method thus data of VOCs concentration at 2.00 pm. were excluded. These instruments can measure 86 VOCs, with a time resolution of 30 min and a minimum detection limit of the ppt level. The concentration of every half hour interval was calculated to obtain hourly mean values.

2.2) Measurement of ozone (O_3) and oxide of nitrogen (NO_x)

Ambient O_3 was measured by Thermo Scientific^{TM} Model 49*i* and NO_x was measured by Thermo Scientific^{TM} Model 42*i* as designated by the United States Environmental Protection Agency (US EPA). The minimum detection limits were 1 ppb for O_3 and 0.4 ppb for NO_x. The measurement method operates on the principle that O_3 molecules absorb UV light at a wavelength of 254 nm and NO_x was measured by chemiluminescence detector. The O_3 and NO_x analyzer calibration by auto-calibration unit as operated for 23 hours per day and reported the results every hour.

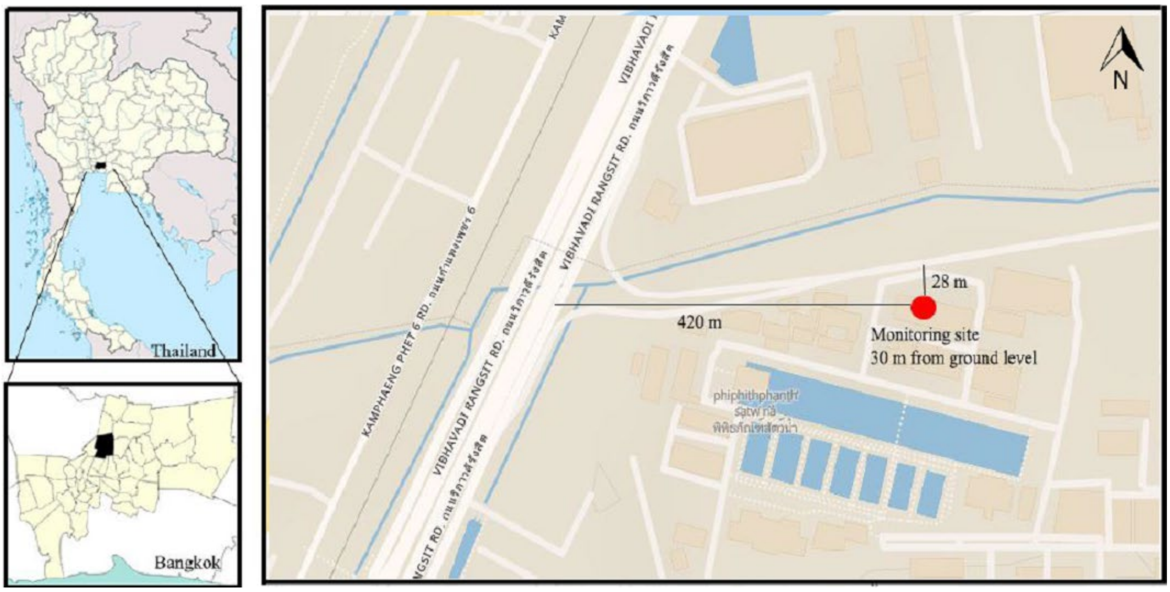


Figure 1 Map of the monitoring site (red circle) in Bangkok, Thailand.

Results and discussion

1) Environmental conditions and gas concentrations

The averaged diurnal variations of meteorological (temperature and relative humidity), total VOCs (TVOCs), NO$_x$, and O$_3$ concentration were separated into weekday and weekend categories in the summer from 16th February to 15th March 2018. The temperatures observed in weekday and weekend were hardly different during the sampling period. The minimum and maximum temperatures were in weekday (weekend) of 23 (23) ºC and 33 (33) ºC, respectively. The relative humidity values were in the range of 43–89% in weekday and 39–90% in weekend. The winds were not calmer. The maximum wind speed was 3.97 m s^{-1} and 3.08 m s^{-1} in weekday and weekend, respectively. The wind direct blows from the southwest (Figure 2) which is the behavior of wind direction inside the boundary layer [20].

The TVOCs, NO$_x$, and O$_3$ concentration in the weekdays and weekends were between 0.03–1.69 and 0.07–1.11 µg m^{-3}, 4.99–95.88 and 6.11–75.77 ppb, and 3.28–102.78 and 7.03–71.11 ppb, respectively. The patterns of TVOCs, NO$_x$ and O$_3$ concentration on weekdays and weekend were quite similar. The concentrations of VOCs and NO$_x$ were high in the morning (6:00–9:00 a.m.) and evening (4:00–9:00 p.m.) which are related to the rush hour with almost all pollution emitted from traffic sources [22–23], inversely varying from the O$_3$ concentration as O$_3$ is generated from the photo radical reaction of VOCs and NO$_x$ [13] as shown in Figure 3. The high O$_3$ concentration in the afternoon due to its precursors (VOCs and NO$_x$) that are related to the stronger UV radiation and higher photochemical reaction under the boundary layer [10]. The correlation coefficient between TVOCs and NO$_x$ in weekdays (r=87) was higher than the weekend (r=72) as this indicates that the TVOCs and NO$_x$ are emitted from same source (with both concentrations of TVOCs and NO$_x$ in weekday is higher than weekend).

This result is similar to that reported previously in Bangkok [24], Kuwait [25], China [26], and Hanoi [23]. The case of Kuwait, it was found that high BTEX concentrations in the morning (07.00–8.00 am.) and during rush (7.00–9.00 pm.) in the evening.

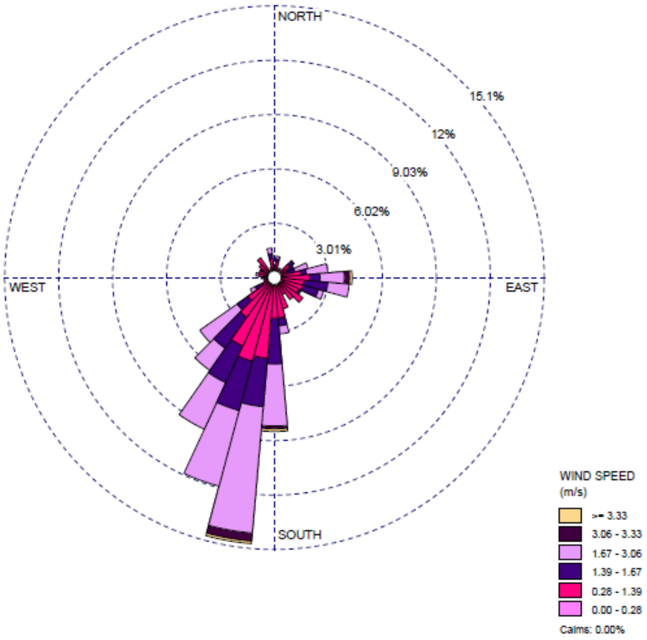
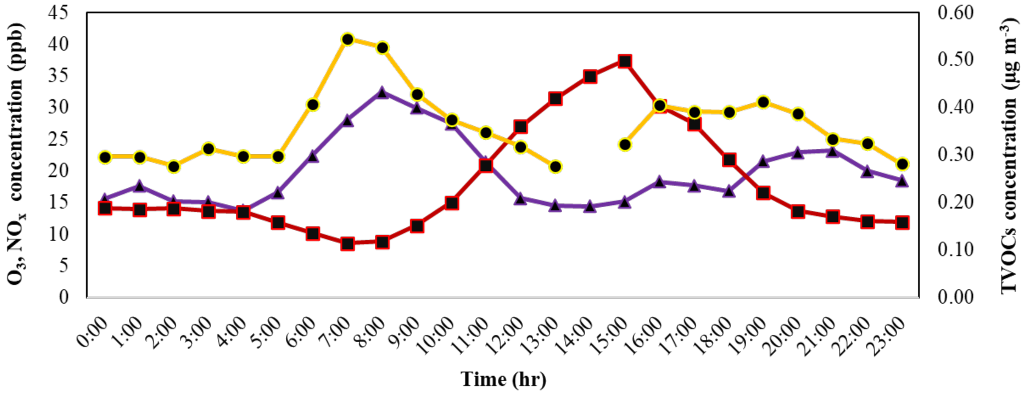


Figure 2 Wind rose diagram at monitoring site.

(a) weekday



(b) weekend

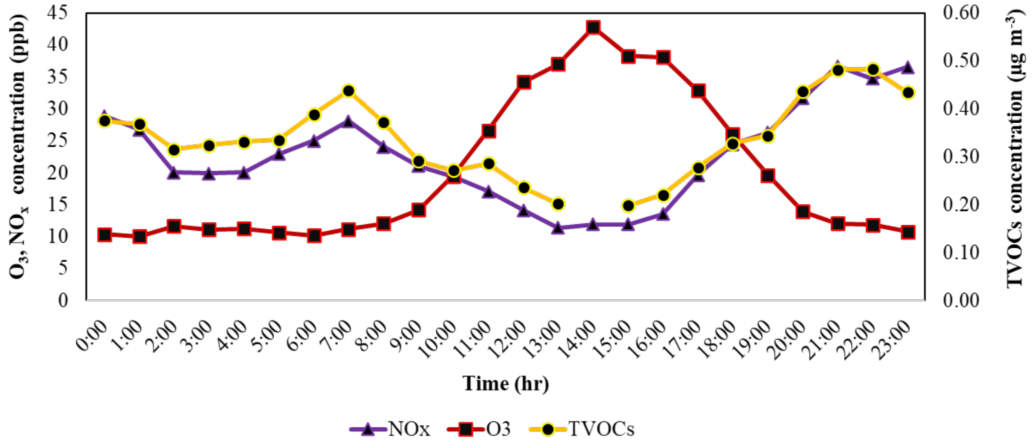


Figure 3 Diurnal variations of TVOCs, NO_x, and O_3 concentrations in a) weekday and b) weekend during 16th February to 15th March 2018 in Bangkok, Thailand.

2) Volatile organic compounds (VOCs)

Twenty-nine VOCs species were measured 23 hours per day (Table 1). In this study, we found that the concentrations of VOCs in weekday and weekend are hardly different during the sampling period. The average concentrations in weekday and weekend were 0.36± 0.25 μg m^{-3} and 0.34±0.21 μg m^{-3}. The most abundant species in weekday (weekend) was found to be toluene 3.32±2.29 (3.11±2.93) μg m^{-3}, followed by propane 2.70±2.11 (2.88±2.05) μg m^{-3}, and carbon tetrachloride (CCl_4) 2.16±2.75 (1.43± 1.71) μg m^{-3}. Standard deviation for each VOCs is wide because of the monitoring site is far from the source so the concentration of VOCs lower than detection limit causing the SD range to be wide. Toluene emitted from vehicular combustion, fossil fuel combustion, gasoline evaporation [27], and industrial activities [28]. While Propane is a typical tracer for liquefied petroleum gas (LPG), fuel evaporation, common cooking gas mixtures [4]. CCl_4 is relatively stable in the environment and sources of CCl_4 from residential areas or industry. It is used mainly as an intermediate in the synthesis of chlorinated solvents [29], chlorinated tap water, and precursor to refrigerants. Saeaw et al. studied source apportionment of VOCs in roadside, Dindaeng District in Bangkok found that the CCl_4 is chemical found in background in ambient air [30]. The case of Spain found that CCl_4 in the urban area influenced by the surrounding industry and indoor sources, chlorine-bleach products used as cleaning agents [31].

The top three VOCs with high concentration in the monitoring site indicate that VOCs emitted from anthropogenic sources including toluene and propane emitted from vehicular sources [32] and CCl_4 emitted from industrial area.

3) BTEX species

BTEX species; Benzene, toluene, ethylbenzene, and *m/p*-xylene are aromatic group, can be considered as an efficient indicator of pollution from road traffic. Toluene was found in high concentrations, followed by *m/p*-xylene, benzene, and ethylbenzene which being lower than the BTEX species concentrations near a road side at the Pathumwan junction in Bangkok [33]. Toluene was the dominant species followed by *m/p*-xylene, benzene, and ethylbenzene as similar to the cases of Iran [34], Ramsis and Haram, Greater in Cairo [35], and in Texas [36]. Ethylbenzene concentration near zero because most concentration data of each VOC species is 0 μg m^{-3}. This is lower than the detection limit because the ambient air sampling was conducted 30 m from ground level on the rooftop, similar to the case at roadside in Bangkok [37]. Toluene had higher concentration than the others due to its stability and its estimated lifetime of 2 days, where it reacts with OH radicals 5 times [38]. Xylene and ethylbenzene are emitted by the same major sources, but they decay at different rates from OH-oxidation in the atmosphere [39].

The BTEX species ratio such as benzene/ toluene (B/T), toluene/benzene (T/B), and *m/p*-xylene/benzene (*m/p*-X/B) are appropriate to use to identify the source of the individual BTEX species. These ratios are indicators that can elucidate traffic emissions and the role of photochemical reactions caused by varying decay rates in the atmosphere under the influence of solar radiation and are indicative of the photochemical age of an air mass. Average BTEX species ratios at roadside in monitoring site are provided in Table 2. B/T ratio is indicative of the photochemical age of an air mass and transport of BTEX species. The lifetime of toluene (1.9 days) was shorter than benzene (9.4 days), resulting in an easier photochemical reaction and rapid subsequent decay of toluene during transport. High B/T ratios reflect a longer transport of vehicular emissions from surrounding cities or result from older air masses due to their differences in lifetime [40].

Table 1 Average (mean±SD in µg m^{-3}) and range of concentrations (µg m^{-3}) for VOCs 29 species from 16th February to 15th March in 2018

VOCs species	Weekday	Weekend
Toluene	3.32±2.29 (0.00–12.79)	3.11±2.93 (0.00–14.69)
Propane	2.70±2.11 (0.00–13.70)	2.88±2.05 (0.59–11.88)
CCl_4	2.16±2.75 (0.00–22.71)	1.43±1.71 (0.00–7.55)
m/p-xylenes	0.58±0.40 (0.00–2.43)	0.59±0.38 (0.00–2.24)
Ethylene	0.37±0.19 (0.00–3.23)	0.38±0.12 (0.21–0.81)
Ethane	0.29±0.24 (0.00–4.02)	0.34±0.17 (0.08–1.04)
Isoprene	0.26±0.28 (0.00–1.79)	0.27±0.32 (0.00–1.53)
n-hexane	0.15±0.16 (0.00–1.25)	0.14±0.15 (0.00–0.62)
1,2,3-TMB	0.11±0.13 (0.00–0.92)	0.10±0.12 (0.00–0.50)
1,2,4-TMB	0.11±0.07 (0.00–0.49)	0.11±0.07 (0.00–0.35)
Benzene	0.09±0.19 (0.00–1.18)	0.11±0.19 (0.00–0.68)
trans-2-pentene	0.06±0.07 (0.00–0.70)	004±0.06 (0.00–0.36)
n-butane	0.05±0.21 (0.00–1.38)	0.12±0.31 (0.00–1.66)
1,3,5-TMB	0.05±0.05 (0.00–0.32)	0.05±0.05 (0.00–0.22)
Styrene	0.03±0.04 (0.00–0.47)	0.03±0.04 (0.00–0.17)
n-pentane	0.02±0.04 (0.00–0.26)	0.03±0.05 (0.00–0.22)
trans-2-butene	0.01±0.13 (0.00–2.07)	nd
o-xylene	0.01±0.03 (0.00–0.52)	0.01±0.02 (0.00–0.08)
cis-2-pentene	0.01±0.06 (0.00–0.88)	nd
cis-2-butene	nd	0.02±0.09 (0.00–0.59)
Acetylene	nd	nd
1-pentene	nd	nd
1-butene	nd	nd
i-butane	nd	nd
Ethylbenzene	nd	nd
1-3-butadiene	nd	nd
n-octane	nd	nd
i-pentane	nd	nd
Dichloromethane	nd	0.01±0.05 (0.00–0.59)

Remark: nd = not detectable, lower than detection limit of each substance

Table 2 Ratio values of BTEX species at monitoring site in Bangkok, Thailand

BTEX ratio	B/T	T/B	*m/p*-X/B
Weekday	0.03±0.01	42.56±13.74	7.31±2.09
Weekend	0.04±0.01	28.00±7.08	5.21±1.03

B/T ratio > 1 reflects a source dominated by coal and biofuel burning, whereas B/T < 0.6 reflects vehicle emission dominated source [41]. The B/T ratio range in weekday (weekend) of 0.03±0.01 (0.04±0.01) at monitoring site was low and indicates that toluene was emitted near the source and associated mainly with vehicular emission. The reciprocal of the B/T ratio is the T/B ratio. T/B ratio was the highest near the pollution source. The T/B ratio > 10 indicates that toluene and benzene was emitted from industrialized city. The T/B ratio < 10 indicates that toluene and benzene was emitted from a non-industrialized city where the main pollution source was traffic exhaust [42]. T/B ratios << 1 indicates that toluene and benzene were emitted from diesel exhaust or emitted from vehicular sources. The T/B ratios range in weekday (weekend) of 42.56±13.74 (28.00±7.08) at the monitoring site showed that toluene and benzene was emitted from industrial areas. The T/B ratios is high due to range of toluene concentration (0.00–12.79 ppb) higher than range of benzene (0.00–1.18 ppb). In another emission profile study, the T/B ratio was 0.61 and 1.61 for diesel and gasoline exhausts, respectively [43], and 1.56 in urban Tianjin in China [44]. m/p-X/B ratios indicated the possibility of air mass transported [45]. Low m/p-X/B ratios (0.6 to 2.7) imply aging of the air mass and photochemical reactions were active [46]. The m/p-X/B ratio in weekday (weekend) of 7.31±2.09 (5.21±1.03) at the monitoring site indicated that BTEX was emitted from further away with longer transport from the source and a corresponding greater photochemical degradation of the isomers m/p-xylene compared to benzene.

4) Ozone formation potentials (OFPs)

The OFP assessed the VOC species involved in O_3 formation and was calculated based on the maximum incremental reactivity coefficients (MIR) and the average of each VOCs [47]. The OFP of each VOC was calculated using Eq. 1.

$$\mathrm{OFPi} = \mathrm{MIRi} \times \mathrm{VOCi} \qquad \text{(Eq. 1)}$$

where MIR is the maximum incremental reactivity of VOCs species i (gm O_3 per gm VOCs), was obtained from paper prepared for the California Air Resources Board Contract 07-339 [48]; and VOCi ($\mu g\ m^{-3}$) is the mass concentration of each VOC specie (denote as i).

The OFP values of the 10 species were calculated for the sampling period. Table 3 showed the top ten VOC species with the highest MIR values due to the MIR values is different where when calculating the OFP values is depended this value as well. The *m/p*-xylenes had the highest OFP value, followed by isoprene, and 1,2,3-TMB [49]. Milt et al. studied about the influence of adding 10% biofuel in gasoline and diesel of all vehicular fuels in Bangkok, Thailand found out that it has caused ozone formation [50]. The main contributor to the OFP was from transportation. While in china's urban areas, ethylene, isoprene, *m/p*-xylenes, and toluene showed higher photochemical reaction reactivity and contributed most to the OFPs [51]. The other OFP values study, toluene and m/p-xylenes were the most important contributors to ozone formation among aromatic hydrocarbons in urban area in India [22], while toluene, isoprene and *m/p*-xylenes were in Taipei, Taiwan [52]. The high O_3 levels is due to the rapid O_3 production in the presence of high VOCs concentration [53]. The patterns of OFP, TVOCs, and NO_x on weekday and weekend were quite similar, this is, two peaks during the morning and evening rush hours. The correlation coefficient between OFP and TVOCs is 0.9.

Table 3 OFP values of top ten MIR in Bangkok, Thailand

VOCs species	MIR	OFP (weekday)	OFP (weekend)
Trans-2-butene	15.16	0.15	0.00
Cis-2-butene	14.24	0.00	0.30
1-3-butadiene	12.61	0.00	0.00
1,2,3-TMB[a]	11.97	1.33	1.20
1,3,5-TMB[b]	11.76	0.55	0.54
Isoprene	10.61	2.74	2.82
Trans-2-pentene	10.56	0.60	0.38
Cis-2-pentene	10.38	0.07	0.00
m/p-xylene	9.75	5.66	5.73
1-butene	9.73	0.00	0.00

Remark: [a]1,2,3-trimethylbenzene; [b]1,3,5-trimethylbenzene

5) Backward trajectory

The backward trajectory calculated by the web version of the HYSPLIT model, developed by the National Oceanic and Atmospheric Administration (NOAA), is used in this study. The Trajectory analysis could be useful for studying for the potential regional sources of O_3 but it cannot be used to pinpoint the exact origin of the air masses [10]. This study calculated 48 hours back-trajectories arriving at 100, 500, and 1000 m above ground level at 1500 UTC on 16th February 2018 (Figure 5). This day had higher concentration of O_3 (102.78 ppb) than other days during the sampling period. This result found that the air flows arriving at 100 and 500 m was transported from the Gulf of Thailand as passes through Samutprakan Province, at 1000 m was transported from the north of Thailand as passes through Chachoengsao Province. This study found that the ethylene concentration of 0.37±0.19 µg m^{-3} (fifth level) is the major species for identifying as shipping emission [54] and the ethylene industry [55]. Theapiriyakit et al. studied on the ozone level in Thailand, found that in the dry season, the ozone level in Samutprakarn Province was higher than in that of Bangkok, as Samutprakarn is a suburban area with different city topography with less atmospheric smog episodes has less severe traffic problem but has many more industrial plants emitting different species of VOCs [12]. Prabamroong et al. found that the high O_3 values (40 ppb) in Rayong, Thailand come from south winds because of local emission, lower atmospheric ventilation, influences of O_3 transportation, and its precursors [56]. The VOCs gas with high concentration does not only sourcing from the roadside therefore a backward trajectory is required. South winds, maritime and the summer southerly Gulf winds, are suggested to be the attributed to the sea breeze effects cause by initiated by differential heating of the ground as the land progressively warms above oceanic temperatures in Thailand's dry season. The concentrations have been brought down to the surface by sea breeze (according to wind breeze) has transported back to Bangkok under the photolytic cycle with added precursors. This process can also be explain with the VOC concentrations emitted from industrial area in the Samutprakan Province [20].

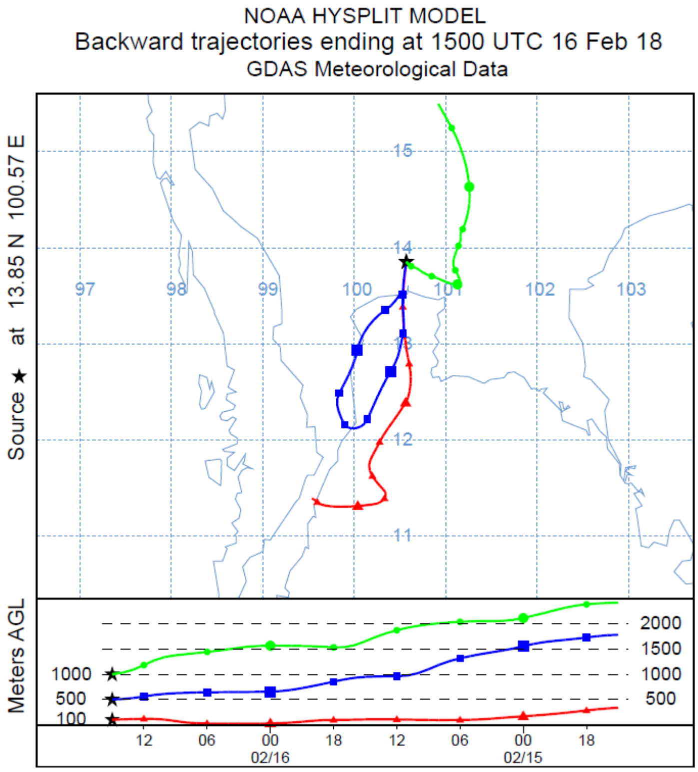


Figure 5 Back trajectory from HYSPLIT at the altitude of 100 m (red line), 500 m (blue line), and 1,000 m (green line) on 16th February 2018.

Conclusions

Meteorological measurements and gas concentrations in summer in urban areas indicated that the wind direction influence on gas concentrations. Due to the wet season having very low concentration VOCs due to wet deposition resulting in a low impact towards the environment. With this, the necessity of finding the VOCs concentration during the period might not be necessary, however in future studies the gap for the measurement during the wet season should be accounted for. The dominant VOCs species were toluene, propane, and CCl_4. The ratios of BTEX species indicated that benzene, toluene, ethylbenzene, and *m/p*-xylene emitted from road traffic and industrial area. High VOCs concentration in monitoring site was influenced by the sea breeze brought the pollutants back. Sea breeze effects initiated by differential heating of the ground as the land progressively warms above oceanic temperatures in the Thai dry season. The concentrations that have been brought down to the surface by sea breeze (according to wind breeze) has transported back to Bangkok under the photolytic cycle with added precursors. This process can also explain the VOC concentrations emitted from industrial area in the Samut Prakan. However, with different direction the results may be different with different wind direction but a new study will be dependent to the source. The results indicated that the control of VOCs emission can reduce O_3 concentration. This study suggests that anthropogenic VOCs in Bangkok emitted from vehicular source from local sources. In the same way as emitted from industrial area in the remote atmosphere of south Bangkok influenced sea breeze. Through this conclusion the best method of controlling the VOCs emission would be through controlling the sources of VOCs production. These findings will be useful for informing relevant policy to control this regional pollution. The consideration must should be put on reducing traffic and industry in order to reduce ambient ozone levels.

Acknowledgments

This study was financially supported by the Monitoring of Microclimate and Air Pollutants in Thailand Project that is supported the finance by Hydro Informatics Institute (HII). Authors would also like to thank the Faculty of Environmental Science, Kasetsart University, Bangkhen Campus, Bangkok for the support of this work.

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