

Seasonal Source Apportionment of Volatile Organic Compounds in Bangkok Ambient Air

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ABSTRACT: Volatile organic compounds (VOCs) are emitted from various sources, both anthropogenic and biogenic. They are important precursors in photochemical reactions, and the formation of secondary aerosols. Therefore in controlling ozone concentration, sources of precursors need to be defined. Although ozone precursors are both NO_x and VOCs, the study from Pollution Control Department concluded that ozone episodes in Bangkok are VOC-limiting. However, uncertainties in emission inventory are greater for VOCs than the criteria pollutants such as TSP, SO_2 and NO_x . The diversity of VOCs sources includes vehicles, industry, solvent-containing products and biological processes. An estimation of volatilization is very difficult due to uncertainties in estimating solvent use, temperature and manner of applications. In this study the U.S. EPA receptor model, CMB7 was used to complement the emission inventory by identifying contribution from various sources to ambient VOC concentration. The receptor model methodology requires investigation of concentration of VOCs at receptor (ambient), and composition of VOCs at sources, which are then input to a statistical model. Ambient air samplings took place at four PCD air monitoring stations: (1) Din Daeng, in central of Bangkok with heavy traffic, (2) Rajabhat University at Chandrakasem, a commercial and residential area north of Bangkok, (3) Rajabhat University at Ban Somdej Chao Praya, a commercial and residential area south of Bangkok, (4) Ratburana Post Office, an industrial area south of Bangkok. The air samplings were conducted for eight months during July 2003 to February 2004 covering the two prevailing wind directions in Thailand, the southwest and northeast monsoon seasons. The air samples were collected in the morning between 8:00 am – 12:00 pm every 6 days at each station. These air samples were collected on sorbent tubes, Chromosorp 106, by pump and analyzed by the thermal desorption-gas chromatography/mass spectrometer, which speciated the samples into 18 VOC species. VOC emission source profiles included in the study were exhaust gases from tailpipes of gasoline vehicles, exhaust gases from tailpipes of diesel vehicles, gasoline vapors, flue gas from fuel oil boilers, vapors of solvent-based paints and liquid thinners, smoke from burning biomass, smoke from food barbequing on charcoal stoves, and air samples from municipal waste disposal sites. The results from CMB receptor modeling ($R^2 = 0.95-1.00$) showed that during the southwest monsoon season the average contribution to the ambient VOC concentration for all stations were from the exhaust gas from tailpipes of gasoline vehicles 21%, the exhaust gas from tailpipes of diesel vehicles 5%, the vapor of gasoline 12%, flue gas from fuel oil boilers 22%, the vapor of solvent-based paint and thinner 8%, smoke from biomass burning 19%, smoke from food barbequing 2%, air samples from municipal waste disposal 4% and unexplained sources 7%. During the northeast monsoon season the average contribution to the ambient VOC concentration for all stations were from the exhaust gas from tailpipes of gasoline vehicles 50%, the exhaust gas from tailpipes of diesel vehicles 6%, the vapor of gasoline 12%, flue gas from fuel oil boilers 2%, the vapor of solvent-based paint and thinner 3%, smoke from food barbequing 5%, air samples from municipal waste disposal 12% and unexplained sources 10%.

KEYWORDS: source apportionment, VOCs, Bangkok ambient air, receptor model.

INTRODUCTION

Bangkok is the capital city of Thailand, located in the central part of the country. The location is at latitude $13^\circ 45'$ north and longitude $100^\circ 28'$ east. The total area is 1,568.737 sq. km. The registry record shows Bangkok population close to seven millions, but perhaps

there are about 10 millions of daytime population. Bangkok is influenced by two seasonal monsoons, namely the southwest monsoon and the northeast monsoon, as well as the land-sea breeze. Bangkok city has a large central business district surrounded by mixed areas of government offices, commercial and residential buildings. There is a large industrial area

south of Bangkok in Samut Prakan province.

Volatile organic compounds (VOCs) are important precursors in photochemical reactions and the formation of secondary aerosols. As precursors in photochemical reactions, they cause elevated ozone levels in urban areas. Pollution Control Department (PCD) reported that the maximum 1-hr ozone concentration in Bangkok from 1996-2003 frequently exceeded the National Ambient Air Quality Standard^{1,2}. PCD study concluded that ozone episodes in Bangkok are VOC-limiting³.

VOCs are emitted from various sources, both anthropogenic and biogenic. In urban areas, VOCs are mainly from anthropogenic sources such as vehicle tailpipes emitting gases from both the complete and incomplete combustion of gasoline, evaporation of gasoline from engines and tanks, solvents used in industry as well as in household products⁴. An estimation by the PCD in 1997 showed that 95% of VOCs was from vehicles and 4.7 % from evaporation at gas stations³. The data excluded solvent usages, which scatter over the whole area of Bangkok. Data on solvent use was not available in the PCD emission inventory in 1997. VOC emission database of Bangkok in 2000 showed that around 60 % of VOCs was from vehicles and around 40 % from area sources such as refueling and usage of solvent-containing products⁵.

In controlling ozone concentration, sources of precursors need to be identified. However, uncertainties in emission inventory are greater for VOCs than the criteria air pollutants such as TSP, SO₂, and NO₂⁶. The diversity of VOCs sources include vehicles, industry, solvent-containing products, and biological processes. An estimation of volatilization is very difficult due to uncertainties in estimating solvent usage, temperature, and manner of application.

In this study the U.S. EPA receptor model, CMB7, was used to complement the emission inventory by identifying contribution from various sources contribution to ambient VOC concentrations.

MATERIALS AND METHODS

To find out the contribution of VOCs from various sources, this study is divided into three parts as follows: measurement of VOC concentrations at the receptors (ambient air), determination of VOC compositions from emission sources, and receptor modeling using the U.S. EPA's CMB7 model⁷.

VOC concentrations at receptors and composition of VOCs from emission sources are used as input data in the receptor model. The target VOC species analyzed in this study refer to 54 VOC species which are mostly found in urban air and are important to ozone formation⁸. Due to the limitation of instruments and

availability of mixed standard gases, only 18 VOC species were quantified (Table 1). The methodology satisfies the receptor model requirement that the number of fitting species must be greater than the number of emission sources concerned.

Receptor Model

Table 1. List of VOC species quantified in this study.

Item	Name	Item	Name	Item	Name
1	1-Pentene	7	3-Methyl pentane	13	Toluene
2	n-Pentane	8	n-Hexane	14	n-Octane
3	trans-2-Pentene	9	Cyclohexane	15	m/p-Xylene
4	Isoprene	10	Benzene	16	n-Nonane
5	2-Methyl pentane	11	Isooctane	17	1,3,5-Trimethyl benzene
6	Cyclopentane	12	n-Heptane	18	n-Decane

Receptor model is a statistical model used to identify contribution from sources to receptors. Source contribution is calculated by weighted least-square multiple regression analysis. The equation is shown below⁷:

$$C_i = F_{i1} S_1 + F_{i2} S_2 + \dots + F_{ij} S_j + \dots + F_{iJ} S_J + E \quad i = 1 \dots I, j = 1 \dots J$$

Where:	C_i	=	Concentration of VOC species i measured at a receptor site
	F_{ij}	=	Fraction of VOC species i in emission from source j
	S_j	=	Estimation of the contribution of source j
	I	=	Number of VOC species (I e•J)
	J	=	Number of source types
	E	=	Uncertainty

S_j are calculated by the model; after that, the model uses S_j to calculate parameters which show the goodness of fit on modeling calculation such as R², Chi-square, TSTAT, and the percent ratio of the sum of the model-calculated source contribution estimates to the measured mass concentration.

The closer R² is to one, the better the source contribution estimates explain the measured concentration. TSTAT is the ratio of the source contribution estimate to the standard deviation of the calculated source contribution. TSTAT values of more than 2 is accepted. Chi-square is the weighted sum of the square of the differences between the calculated and measured VOC species concentration. The acceptable range of the chi-square is less than 2. Percent

mass is the percent ratio of the sum of the model-calculated source contribution estimates to the measured mass concentration. The acceptable range of the percent mass is between 80-120 %.

Although VOCs are reactive species, a conclusion

of many studies showed that transportation time of 3-5 hours from sources to receptors did not affect source apportionment estimation by using chemical mass balance^{6,9-12}. All 18 VOC species in Table 1 have lifetime of more than 3 hours¹³.

VOC Concentrations at Receptors (Ambient Air)

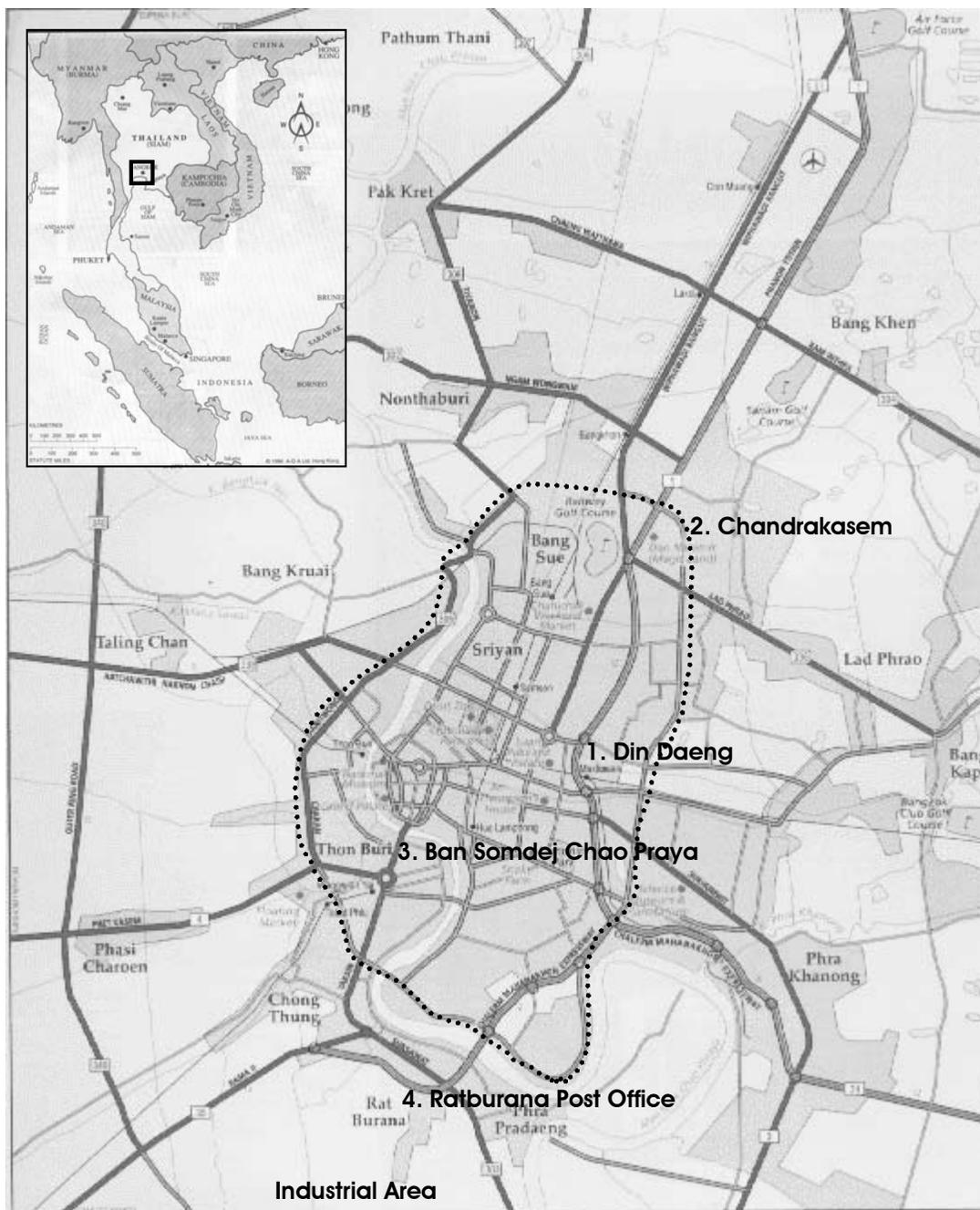


Fig 1. Location of air sampling Sites
 ••••• Central Business District.

Prevailing wind directions:
 ↗ SW monsoon March-October
 ↙ NE monsoon November - February

Sampling Locations

Ambient air sampling took place at four PCD air monitoring stations (Figure 1) as follows: (1) Din Daeng, a central of Bangkok with heavy traffic, (2) Rajabhat University at Chandrakasem, a commercial and residential area north of Bangkok, (3) Rajabhat University at Ban Somdej Chao Praya, a commercial and residential area south of Bangkok, and (4) Ratburana Post Office, an industrial area south of Bangkok.

Sampling Period

The air samplings were carried out during July 2003 to February 2004 for a period of 8 months. This period covered both seasons in Thailand, namely the southwest and northeast monsoon seasons. The sampling equipments were placed on the roof of the PCD monitoring stations with the sample intakes about three meters above the ground. The air samples were collected every 6 days at each station in order to cover each day of a week and collected in the morning between 8:00 am – 12:00 pm in order to get fresh emissions⁴.

Sampling and Analysis Method¹⁴

A procedure of ambient air sampling is as follows.

Ambient air samples were withdrawn by a pump and adsorbed on sorbent tubes, ¼ inch O.D., 3.5 inch long, packed with 250 mg of chromosorb 106 (Supelco).

Air collection flow rate was designed at 10 ml/min¹⁴. The actual flow rate was 11.0 ml/min. ± 0.9 ml/min.

Sampling time was four hours. When the sampling was finished, the sorbent tube was capped with brass Swagelok, wrapped with aluminium foil and put in a plastic bag, which was kept in an ice box and transported to the laboratory, and the samples were stored in a solvent-free refrigerator. The analysis work was carried out within one week after the sampling.

Sorbent tubes for air sampling were cleaned each time before usage at 240 °C for 90 minutes.

Air samples were analyzed by the thermal desorption-GC/MS at Environmental Research and Training Center, Pathum Thani, Thailand.

The Thermal Desorber Unit (TDU) was from Unity, Markes International Limited. The TDU temperature program used was as follows: Purge for 5 minutes, and desorb at 190 °C for 20 minutes to cold trap -10 °C for 15 minutes and immediately heat to 310 °C to GC column. The GC/MS-QP5000 Column: was a Capillary column SPB™ 624, 60 m. long, 0.32 mm. i.d., thickness 1.8 µm. from Shimadzu. Mass spectrometry analysis used an Electron Impact, Quadrupole mass spectrometer.

The GC temperature program was as follows: start at 35 °C and hold for 5 minutes, ramp up at 3 °C/min

to 160 °C, ramp up at 20 °C/min to 200 °C, and hold for 30 minutes. The solvent cut time was 5 minutes, and the scanned mass range was from 35 - 200 amu.

The mixed standard gases were provided by courtesy of Dr. Ian Weeks, Atmospheric Research Division, CSIRO, Australia.

Composition of VOCs from Emission Sources (Source Profiles)

The study utilized nine VOC emission sources including exhaust gases of gasoline vehicles, exhaust gases of diesel vehicles, gasoline vapors, flue gas of fuel oil boilers, vapor of solvent-based paints and thinners, smoke of biomass burning, smoke of food barbequing on charcoal stove, and air samples from municipal waste disposal.

Four emission sources including exhaust gases of gasoline vehicles, exhaust gases of diesel vehicles, flue gas of fuel oil boilers and smoke of biomass burning, are the existing source profiles conducted by the authors in 2000¹⁵. The additional sources were identified in this study.

Vapor of gasoline source profile was analyzed from the five popular brands in Thailand. The gasoline samples were purchased and analyzed for the composition of VOC species in their vapor by modified cold trap GC/MS (Shimadzu QP 5050A, GC Column: CP-sil PONA CB fused Silica WCOT, 100 m long, 0.25 mm i.d., 0.5 mm, MS: Electron Impact, Mass spectrometer) at National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Japan.

Vapor of solvent-based paint source profile was analyzed from the five highest selling solvent-based paint brands. The solvent-based paints were purchased and analyzed for the composition of VOC species in their vapor using the same procedures as vapor of gasoline samples.

Five brands of thinner from auto-body repair shops were obtained and analyzed for the composition of VOC species. The thinner samples were analyzed by direct injection to GC/MS at National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Japan.

Air samples from food barbequing on charcoal stove from three food vendors at various places in Bangkok were analyzed. These samples were collected by sorbent tubes and analyzed by the thermal desorption GC/MS using the same procedure as the ambient air samples.

Air samples from municipal waste disposal were collected by sorbent tubes and analyzed by the thermal desorption GC/MS using the same procedure as the ambient air samples.

RESULTS AND DISCUSSION

VOC concentrations at receptors

Ambient air samples were collected every six days at four stations during July 2003 to February 2004. Seventy-five ambient air samples were analyzed and results obtained from sixty-eight samples. The results of three replications of mixed standard gas analysis

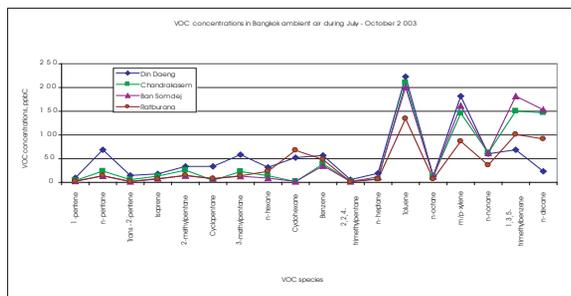


Fig 2. VOC concentrations at each station during the southwest monsoon season.

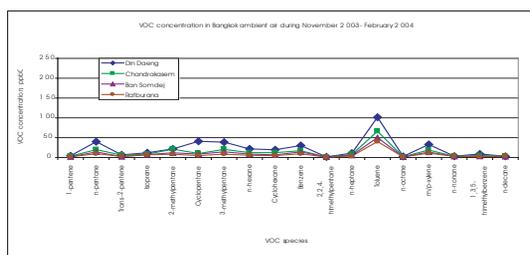


Fig 3. VOC concentrations at each station during the northeast monsoon season.

showed RSD around 8.2%. The VOC thermal desorption extraction efficiency was $95.0 \pm 7.6\%$.

The ambient VOC concentrations in Bangkok was found to be different between the southwest and the northeast monsoon seasons. At all four stations in Bangkok (Figures 2 and 3), the average VOC concentrations during the southwest monsoon season were higher than the concentrations during the northeast monsoon season. This was likely to be caused by the difference in atmospheric conditions. During the southwest monsoon, the prevalent weather conditions were of cloud and rain. In contrast, during the northeast monsoon the weather conditions were clear with strong sunshine. The report on ozone episodes showed that these mostly occurred in the dry season, November to March⁴. Ozone episodes were caused by strong sunshine promoting photochemical reactions leading to high ozone concentrations but probably resulting in a depletion of ozone precursor substances.

Source Profiles

In this study, five new emission source profiles relevant to Thailand were determined: vapor of gasoline (VG), vapor of solvent-based paints (VP), thinners (LT), smoke from food barbecuing on charcoal stoves (BBQ), and air samples from municipal waste disposal (MW). The four existing emission source profiles employed in the study including exhaust gases from gasoline vehicles (GV), exhaust gases from diesel vehicles (DV), flue gas from fuel oil boilers (FB) and smoke from biomass burning (BB)¹⁵. Table 2 shows the fraction of VOCs

Table 2. Emission source profiles showing fraction of various VOC species.

VOC species	Fraction								
	GV	VG	DV	FB	BB	BBQ	LT	VP	MW
1- Pentene	0.0068	0.0125	0.0855	0.0014	0.0163	0.0930	0.0000	0.0000	0.0000
n-Pentane	0.0746	0.1892	0.0348	0.0079	0.0214	0.1028	0.0000	0.0000	0.0184
trans-2-Pentene	0.0169	0.0200	0.0155	0.0043	0.0117	0.0164	0.0000	0.0000	0.0000
Isoprene	0.0016	0.0000	0.0003	0.0000	0.0487	0.0151	0.0000	0.0000	0.0019
2-Methylpentane	0.0832	0.2275	0.0995	0.0470	0.0457	0.0089	0.0000	0.0000	0.0032
Cyclopentane	0.0032	0.0000	0.0027	0.0003	0.0008	0.0828	0.0000	0.0000	0.0000
3-Methylpentane	0.0572	0.1299	0.0347	0.0682	0.0095	0.0068	0.0000	0.0000	0.0049
n-Hexane	0.0572	0.1274	0.0347	0.0616	0.0157	0.0848	0.0000	0.0000	0.0149
Cyclohexane	0.0516	0.0234	0.0385	0.0312	0.0118	0.0359	0.0000	0.0000	0.0425
Benzene	0.1093	0.0544	0.2677	0.0464	0.3949	0.1901	0.0000	0.0000	0.0000
2,2,4-Trimethylpentane	0.0000	0.0038	0.0000	0.0086	0.0000	0.0298	0.0000	0.0000	0.0259
n-Heptane	0.0280	0.0299	0.0176	0.0343	0.0172	0.0985	0.0000	0.0000	0.0332
Toluene	0.3103	0.1731	0.1939	0.3454	0.3108	0.1033	0.9669	0.0635	0.7400
n-Octane	0.0119	0.0000	0.0148	0.0278	0.0044	0.0934	0.0000	0.2455	0.0256
m/p-Xylene	0.1841	0.0088	0.0629	0.2201	0.0583	0.0000	0.0230	0.2964	0.0644
n-Nonane	0.0052	0.0000	0.0245	0.0272	0.0049	0.0757	0.0000	0.2306	0.0146
1,3,5-Trimethylbenzene	0.0268	0.0000	0.0252	0.0433	0.0202	0.0000	0.0101	0.0000	0.0031
n-Decane	0.0033	0.0000	0.0598	0.0250	0.0076	0.0433	0.0000	0.1640	0.0075
Total	1								

Note: GV, DV, FB, and BB source profiles were taken from Limpaseni et al¹⁵, other source profiles were from this study.

from these nine emission sources.

Table 3. Average Source Contribution of VOCs in Bangkok Ambient Air (ppbc).

Sources	Average Source Contribution, ppbc	
	SW monsoon season	NE monsoon season
Gasoline vehicles	159.4	123.0
Diesel vehicles	38.0	14.8
Fuel oil boilers	167.0	4.9
Vapor of gasoline	91.1	29.5
Vapor of paint and thinner	60.7	7.4
Biomass burning	144.2	-
Food barbequing	15.2	12.3
Municipal waste	30.4	29.5
Unexplained	53.1	24.6
Total	759.1	246.0

Table 4. Average Source Contribution of VOCs in Bangkok Ambient Air (percent).

Sources	Average Source Contribution, %		
	SW monsoon season	NE monsoon season	Average
Gasoline vehicles	21	50	36
Diesel vehicles	5	6	6
Fuel oil boilers	22	2	12
Vapor of gasoline	12	12	12
Vapor of paint and thinner	8	3	5
Biomass burning	19	-	10
Food barbequing	2	5	3
Municipal waste	4	12	8
Unexplained	7	10	8
Total %	100	100	100

Receptor Model

By choosing only the sampling dates during which the wind direction was entirely from the southwest and the northeast wind directions, only a small number of samples were suitable for use. To avoid misrepresentation, source contribution of VOCs in Bangkok ambient air was examined for all wind directions. The data from all sixty-eight samples were used for modeling. There were forty-five samples showing the percent mass between 80-120%, which were accepted. Although the other samples showed acceptable R^2 between 0.91-0.99, and Chi-square between 1-2, the percent mass of those samples were below 80% or above 120%; thus twenty-three samples were discarded.

The results from CMB receptor modeling are shown in Tables 3 and 4. The average source apportionment of VOCs from the four stations during the southwest monsoon season showed that the largest three emission sources were fuel oil boilers (22%), gasoline vehicles

(21%), and biomass burning (19%). The different situation occurred during the northeast monsoon season when gasoline vehicles became the most dominant source at 50% and fuel oil boilers accounted for only 2%. The reason for this is that all stations were downwind from an industrial area during the southwest monsoon season but upwind from it during the northeast monsoon season and thus had little impact from the industrial sources. On the other hand, biomass burning had no contribution during the northeast monsoon season (November to February). Although gasoline vehicles contributed a little less in term of concentration in the northeast monsoon season (Table 3), it had a much larger contribution in relative term since the other two sources contributed much less.

Taking the average of both seasons, the results show that the source contribution from the exhaust gas of both gasoline vehicles and diesel vehicles was 42%. The source contribution from fuel oil boilers was 12%. The remaining source contributions were from fugitive sources and area sources, such as vapor of gasoline, vapor of solvent-based paints and thinners, biomass burning, food barbequing and municipal waste disposal for a total of 38%. It may be concluded that the vehicles and area sources are of equal importance at around 40%, while the remaining sources are fuel oil boilers and unexplained sources.

The source contribution of VOCs from this study

Table 5. Comparing source contribution between emission inventories and the receptor model.

Emission sources	Source contribution, %			
	Emission inventory in 1997 ³	Emission inventory in 2000 ⁵	Receptor model March 2003 ¹⁶	Receptor model This study
Line source:				
traffic	95.1	60	33-88	—
Gasoline vehicles	—	—	—	36
Diesel vehicles	—	—	—	6
Point source:				
boiler	0.2	0	0-46	12
Area source:				
Vapor of gasoline	4.7	3	0	12
Vapor of paint and thinner	—	37	0	5
Biomass burning	—	—	12-61	10
Food barbequing	—	—	—	3
Municipal waste	—	—	—	8
Unexplained sources	—	—	—	8

Note: Classification of emission sources referred to the original emission inventory^{3,5}
 — Not applicable

was compared to the emission inventories of Bangkok in 1997 and 2000 and the result of CMB modeling on one day in March 2000 (Table 5). It is clear that important sources of VOCs not covered by the emission inventories are area and fugitive sources identified by source apportionment undertaken in this study. The source apportionment study can complement the emission inventory and provide a more comprehensive coverage of source types which are difficult to inventory.

CONCLUSIONS

The results from source apportionment provide additional information to the emission inventory. While vehicles and vapor of gasoline are confirmed as the major sources of VOCs in Bangkok, the source apportionment discovered other important sources including vapor of paint and thinners, biomass burning, food barbecuing and municipal waste disposal. The source apportionment also shows seasonal variation, pointing to fuel oil boilers and biomass burning as two of the most important sources of VOC emission during the southwest monsoon season.

The results indicate that for effective control of VOCs, which are ozone precursors, the targets need to include VOC fugitive sources such as vapor of gasoline, vapor of paint and thinners, biomass burning, food barbecuing and municipal waste disposal, these fugitive sources are difficult to inventory and thus until now have not been included in mitigation measures. The relative importance of each source type also varies according to season and must be considered in developing mitigation measures.

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