

Original Article

Formation potentiality and source apportionment analysis of secondary organic aerosol in urban and suburban area, Thailand

Lalidaporn Punya¹, Sarawut Thepanondh^{1, 2*}, Suphaphat Kwonpongsagoon^{1, 2},
Wanna Laowagul³, Nuttakit Sukjit¹, and Pattaranan Hanma¹

¹ Department of Sanitary Engineering, Faculty of Public Health,
Mahidol University, Ratchathewi, Bangkok, 10400 Thailand

² Center of Excellence on Environmental Health and Toxicology (EHT), Ratchathewi, Bangkok, 10400 Thailand

³ Environmental Research and Training Center, Department of Environmental Quality Promotion,
Ministry of Natural Resources and Environment, Khlong Luang, Pathum Thani, 12120 Thailand

Received: 5 April 2021; Revised: 11 June 2021; Accepted: 28 June 2021

Abstract

Ambient volatile organic compounds (VOCs) concentrations measured in Bangkok and its suburb in Thailand were estimated for their secondary organic aerosol formation potential (SOAP) to elucidate the major compounds contributing to the formation of secondary fine particulates. VOCs were collected to evacuated canisters and analyzed by Gas Chromatography - Mass Spectrophotometer (GC-MS) in accordance with US-EPA TO15. Measured data from January 2013 to December 2019 were used in this study. The results indicate that the top five species of VOCs contributing to SOAP were toluene (66-54%), m-, p-xylene (11-7%), ethylbenzene (9-8%), benzene (12-5%), o-xylene (5-4%) and styrene (9-4%), in rank order. These VOCs accounted for more than 90 % of total SOAP. Positive Matrix Factorization (PMF) was then used to identify the potential emission sources of these compounds. The results clearly indicated a high contribution from mobile sources at roadside monitoring stations. As for the suburban area, most of the total VOC concentrations were from mobile source emissions, chemical use in the households, and open burning. The results of this study highlight the necessity to control these emission groups to reduce the formation potentiality of secondary organic aerosols and PM_{2.5} pollution.

Keywords: volatile organic compound, secondary organic aerosol formation, positive matrix factorization, receptor oriented model, air pollution

1. Introduction

Secondary organic aerosol (SOA) pollution has become a major environmental concern in many cities in Thailand (Dieu *et al.*, 2019; Narita *et al.*, 2019). Its effects include direct health impacts as well as indirect environmental impacts, such as decreasing crops and ambient visibility (Mozaffar, Zhang, Fan, Cao, & Lin, 2020). SOA is the product of photochemical reactions between volatile organic

compounds (VOCs) and other primary air pollutants, such as sulfur dioxide and oxides of nitrogen, induced by sunlight. Besides SOA, others products from the photochemical oxidation of these chemicals include tropospheric ozone, peroxyacyl nitrates, and aldehydes (Leuchner & Rappengluck, 2010; McCarthy, Akiliu, Brown, & Lyder, 2013). Depending on locality, the VOCs can be emitted from anthropogenic or natural sources (Su *et al.*, 2019). It is well recognized that mobile sources, industrial processes, biomass burning, and emissions from chemicals used in households, are the dominant anthropogenic sources of VOCs.

Several studies have indicated the crucial role of VOCs in the formation of SOA (Barsanti, Kroll, & Thornton,

*Corresponding author

Email address: sarawut.the@mahidol.ac.th

2017; Hallquist *et al.*, 2009; Rajeev, Rajput, Singh, & Gupta, 2018). The source contribution studies by Yang *et al.* (2019) and Wang *et al.* (2020) reported that industrial processes are the major VOC sources contributing to ozone formation potential (OFP) and secondary organic aerosol potential (SOAP). Characteristics of VOCs were also assessed in other studies finding that industrial sources were important contributors of precursors to OFP and SOAP (Li *et al.*, 2020; Liu *et al.*, 2020). The VOC species with greatest contributions to SOA formation were toluene (57%), m,p-xylene (16%), benzene (10%), and ethylbenzene (7%), accounting for about 90% of the total SOA formation at western Taiwan (Huang & Hsieh, 2020). The contribution of VOCs to SOA in different seasons in Beijing were investigated. SOA was highest in winter at 2885.1 $\mu\text{g}/\text{m}^3$. Aromatics were the main contributors to SOAP, accounting for ~98.2% of the total SOAP (Li *et al.*, 2020).

The aim of this study was to estimate individual VOC species' reactivity and their contributions to the SOA formation, and to identify VOC sources and their contributions to SOA formation in urban and suburban area in Thailand. Intensive measured VOC data from January 2013 to December 2019 were used in this analysis to reveal the major VOCs contributing to the formation of secondary organic aerosols, and their potential emission sources through the source apportionment analysis. The novelty and the major application of this study is describing the formation of secondary organic aerosols and fine particulates, while currently only information on primary particulates is used to manage the $\text{PM}_{2.5}$ problem.

2. Methodology

2.1 Information about the monitoring sites

Ambient VOCs were measured in Bangkok, the capital city of Thailand. The roadside areas at Chulalongkorn hospital and Dindeang were selected to represent the major emission sources of VOCs in Bangkok. Measured data at Pathum Thani province and at Chiang Mai province represented the suburban areas, and were also used to evaluate the characteristics and profiles of VOCs from land use changes. All four stations are air quality monitoring stations of the Pollution Control Department (<http://air4thai.pcd.go.th/webV2/region.php?region=1>). Characteristics and spatial locations of the VOC monitoring stations are shown in Table 1 and Figure 1.

Chulalongkorn hospital and Dindaeng are classified as roadside monitoring stations. These stations are in the central business district of Bangkok. Heavy traffic affects in these areas. The traffic volume in early morning rush hour (7:00-9:00 am) was about 123,337 vehicles per hour. These locations are surrounded by commercial buildings, low-rise and high-rise residential buildings, small restaurants, and mobile meat grills. ERTC is a rural monitoring station located in Pathum Thani province. The traffic volume in this area is significantly below those in Bangkok. This site is considered not to be directly influenced by heavy traffic on roads. As for the City Hall of Chiang Mai in Chiang Mai province, monitoring campaign was conducted at the area close to the city hall. Low traffic volume was observed at this site. However, this monitoring site is surrounded by forest, with a

stadium, low-rise buildings far from the downtown area, and thus it experiences no large effects from traffic and building factors. Therefore, it is expected that the open burning, including forest fires and agricultural burns, could be potential emission sources influencing profile and concentrations of VOCs at this suburban site.

2.2 VOCs measurement

Ambient VOCs concentration data used in this study were measured by the Thai Pollution Control Department. Measured data in Bangkok, Pathum Thani and Chiang Mai provinces from January 2013 to December 2019 were used in the analyses. Sampling of VOCs were conducted based on US EPA TO15. Evacuated 6 L Silonite TM coated stainless steel vessels were used to collect the samples. Prior to sampling, the canisters were pre-cleaned with high-purity nitrogen and evacuated to 0.05 mmHg. At the sampling sites VOCs were pushed into the vacuum canisters by the external atmospheric pressure. A constant flow rate of $5.5 \times 10^{-8} \text{ m}^3/\text{s}$ from full vacuum to within about 7 kPa or less below ambient pressure was maintained by the flow controller attached to each canister, to allow 24 hours of uniform sampling. Collected samples were then pressurized to $14.2 \times 10^3 \text{ kg}/\text{m}^2$ by humidified nitrogen to prevent contamination from later on entering the sample canister. At the laboratory, VOCs were analyzed and speciated for their compositions by gas chromatography - mass spectrophotometry (GC-MS).

For quality assurance and quality control (QA/QC) of the data, measuring instruments using standard gas were each time using calibration curves, relative standard deviation (RSD), and method detection limit (MDL). The acceptance criteria of the correlation coefficients from the calibration curves were above 0.99. The relative standard deviation values were less than 20%, which were used in the judgment of the calibration. The method detection limit for the 44 VOC species ranged from 0.002 to 0.05 ($\mu\text{g}/\text{m}^3$), as shown in Table 2. Details of VOC sampling and analysis are presented elsewhere (Laowagul, Garivait, Limpaseni, & Yoshizumi, 2008; Saikomol, Thepanondh, & Laowagul, 2019; Thepanondh, Varoonphan, Sarutichart, & Makkasap, 2011). Totally, there were 44 compounds analyzed in this study. Sampling was conducted monthly from January 2013 to December 2019.

2.3 Secondary organic aerosol formation

Formation of secondary organic aerosols in this study was calculated using average concentration and secondary organic aerosol potential (SOAP) of the individual VOC species based on an equation developed by Derwent *et al.* (2010).

SOAP of each VOC species was calculated by multiplying its SOAP conversion factor by the measured concentration of that individual VOC species. Total SOAP was estimated as the sum of individual SOAP contributions from all the VOC species (Niu, Mo, Shao, Lu, & Xie, 2016), as shown in Equation (1).

$$\text{SOAP} = \sum_{i=1}^n C_i \times \text{SOAP}_i \quad (1)$$

Table 1. Characteristics of VOCs monitoring sites

Monitoring station	Region	Province	Area	Location (Lat/Lon)
Dindeang	Central	Bangkok	Roadside	13.762517N/100.5502E
Chulalongkorn hospital	Central	Bangkok	Roadside	13.729852N/100.536501E
ERTC	Central	Pathum Thani	Suburban	14.045808N/100.714087E
City Hall Chiang Mai	Northern	Chiang Mai	Suburban	18.840633N/98.96963E

Table 2. Method detection limits of VOCs

Compound	MDL (ug/m ³)	Compound	MDL (ug/m ³)
1,1,1-Trichloroethane	0.008	Chlorobenzene	0.004
1,1,2,2-Tetrachloroethane	0.02	Chloroethane	0.002
1,1,2-Trichloroethane	0.02	Chloroform	0.01
1,1-Dichloroethane	0.006	Chloromethane	0.003
1,1-Dichloroethylene	0.004	cis-1,2-Dichloroethylene	0.004
1,2,4-Trichlorobenzene	0.05	cis-1,3-Dichloropropene	0.008
1,2,4-Trimethylbenzene	0.009	Dichloromethane	0.006
1,2-Dibromoethane	0.01	Ethylbenzene	0.005
1,2-Dichlorobenzene	0.01	Freon 11	0.009
1,2-Dichloroethane	0.01	Freon 113	0.01
1,2-Dichloropropane	0.01	Freon 114	0.01
1,3,5-Trimethylbenzene	0.01	Freon 12	0.009
1,3-Butadiene	0.003	Hexachloro-1,3-butadiene	0.05
1,3-Dichlorobenzene	0.01	m- & p-Xylene	0.01
1,4-Dichlorobenzene	0.01	o-Xylene	0.005
1-Ethyl-4-methylbenzene	0.006	Styrene	0.007
3-Chloropropene	0.003	Tetrachloroethylene	0.01
Acrylonitrile	0.008	Toluene	0.02
Benzene	0.005	trans-1,3-Dichloropropene	0.006
Benzyl Chloride	0.004	Trichloroethylene	0.007
Bromomethane	0.006	Vinyl chloride	0.003
Carbon Tetrachloride	0.008		

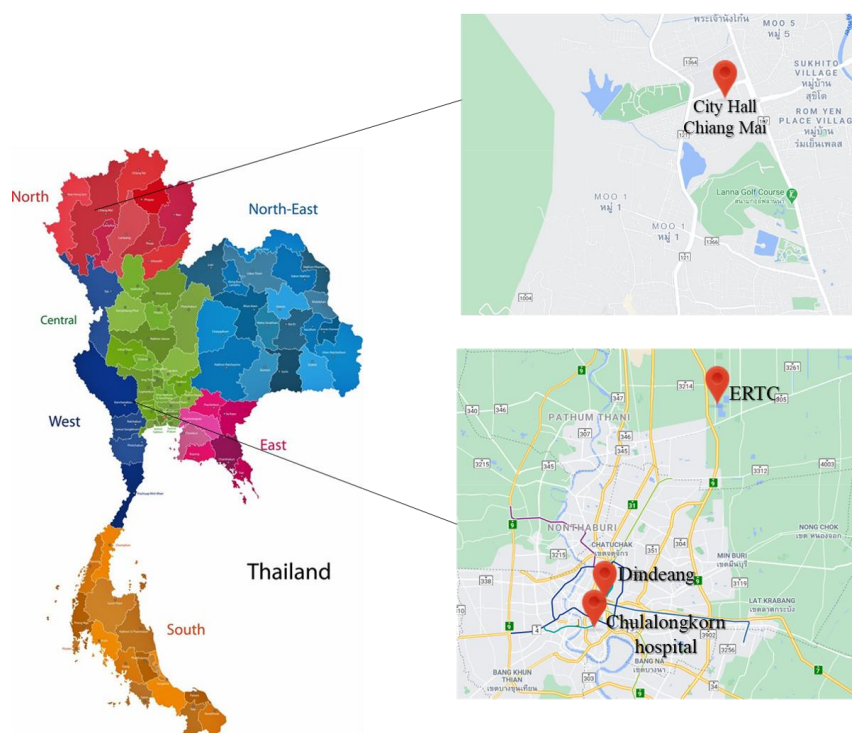


Figure 1. The location of study area and sampling sites

where C_i ($\mu\text{g}/\text{m}^3$) is mass concentration of species i and SOAP_i is the SOA formation potential (conversion factor) of species i .

2.4 PMF model

Positive matrix factorization (PMF) model (version 5.0.14) was applied in the source contribution analysis of this study. It is a multivariate factor analysis tool developed by the US.EPA that decomposes a matrix of speciated sample data into two matrices for factor contributions and factor profiles (US.EPA, 2014). The extent and magnitude of source types are represented as measured source profile. Details of its principles are described in greater detail elsewhere (Jindamane, Thepanondh, Aggapongpisit, & Suktawee, 2020; Paatero, 1997; Paatero & Tapper, 1994).

The application of PMF depends on the estimated uncertainties in each measured data, providing guidelines for estimating the input uncertainties. In this study, the USEPA equation approach was used to estimate the uncertainty associated with each data point and the missing concentration values were replaced by the geometric mean of the species (Kim, Brown, Hafner, & Hopke, 2005). However, species compounds with more than 25% missing values (including values reported as lower than the method detection limit MDL) were rejected from PMF analysis (Saeaw & Thepanondh, 2015).

The uncertainty of VOC concentration was calculated using Equations. (2)-(3). If VOC concentration is greater than MDL, Equation (2) is used, while if VOC concentration is less than or equal MDL, Equation (3) is used.

$$Unc. = \sqrt{(\text{Error Fraction} \times \text{Concentration})^2 + (0.5 \times \text{MDL})^2} \quad (2)$$

$$Unc. = \frac{5}{6} \times \text{Concentration} \quad (3)$$

The error fraction can be 5-20% and it was set at 10% in this study. MDL is the method detection limit.

3. Results and Discussion

3.1 Estimation of secondary organic aerosol formation

Formation of secondary organic aerosols was estimated for each monitoring station. Results are summarized in Table 3. The top-5 VOCs with the highest contributions to SOA formation were evaluated for each monitoring site. It

was found that these top-5 compounds contributed more than 90% of the total SOAP at every site. The total SOAP ranged within 1160.5-5526.2 $\mu\text{g-SOA}/\text{m}^3$ with the highest value in Dindaeng monitoring station (roadside area) at 5526.2 $\mu\text{g-SOA}/\text{m}^3$ and the lowest value in City Hall Chiang Mai (suburban area) at 1160.5 $\mu\text{g-SOA}/\text{m}^3$. The different SOA potentials between these two sites were mainly affected by the measured concentrations of toluene. High concentration of toluene was observed in the roadside (36 $\mu\text{g}/\text{m}^3$) while a much lower concentration was detected in the suburban area (6 $\mu\text{g}/\text{m}^3$).

Toluene, benzene, ethylbenzene and m,p-xylene were the major components at all four monitoring stations. Among these, the most abundant SOAP species was toluene, accounting for more than 50% of total SOAP. We further conducted the source apportionment analysis of these compounds to elucidate the potential major sources of the secondary organic aerosols formed by volatile organic compounds, by using PMF as presented in section 3.2.

3.2 PMF analysis

After pretreatment of data, among the 44 of measured VOCs for each sample, 34 compounds were used in the PMF analysis. Results are presented in terms of source apportionment and source contribution from individual VOC species to SOAP.

3.2.1 Source apportionment

PMF analysis revealed that there were five factors (groups) according to presence of signature compounds in each emission category. Possible VOC emission sources of each factor were determined by using relationship between VOCs and their percentage of contribution in the same group. An example of source profiles of the five factors as well as their contribution percentages in Chiang Mai is shown in Figure 2. Details of source profile at each monitoring station are presented in Figure 3-5.

Factors 1 and 4 were identified as the emissions of chemicals used in households. Factor 1 was characterized by high percentages of trichloroethylene, 1,1-dichloroethylene and cis-1,2-dichloroethylene. Factor 4 was characterized by high percentages of 1,4-dichlorobenzene, 1,1,2,2-tetrachloro ethane and 1,2-dibromoethane, propane, ethylene, ethane, and benzyl chloride. As reported by Kim *et al.* (2005) and Ling, Guo, Cheng, & Yu (2011), these compounds are important indicators of emissions from chemical use in households.

Table 3. Top 5 VOC species with the highest contributions to SOA formation

Monitoring station	SOAP _i from top 5 VOCs species ($\mu\text{g-SOA}/\text{m}^3$)					Percentage of top 5 VOCs to SOAP (%)
Chulalongkorn hospital (total SOAP = 5424.3)	Toluene (3580.7)	m,p-Xylene (467.5)	Ethylbenzene (407.5)	Benzene (295.5)	Styrene (228.6)	91.8
Dindaeng (total SOAP = 5526.2)	Toluene (3276.3)	m,p-Xylene 622.9	Ethylbenzene (434.3)	Benzene (392.3)	o-Xylene (297.5)	90.9
ERTC (total SOAP = 1974.7)	Toluene (1167.2)	Ethylbenzene (192.6)	Styrene (184.6)	m,p-Xylene (150.4)	Benzene (132.8)	92.5
City Hall Chiang Mai (total SOAP = 1160.5)	Toluene (627.6)	Benzene (146.8)	m,p-Xylene (114.5)	Ethylbenzene (90.3)	o-Xylene (61.5)	90.0

Remark: SOAP expressed in $\mu\text{g-SOA}/\text{m}^3$

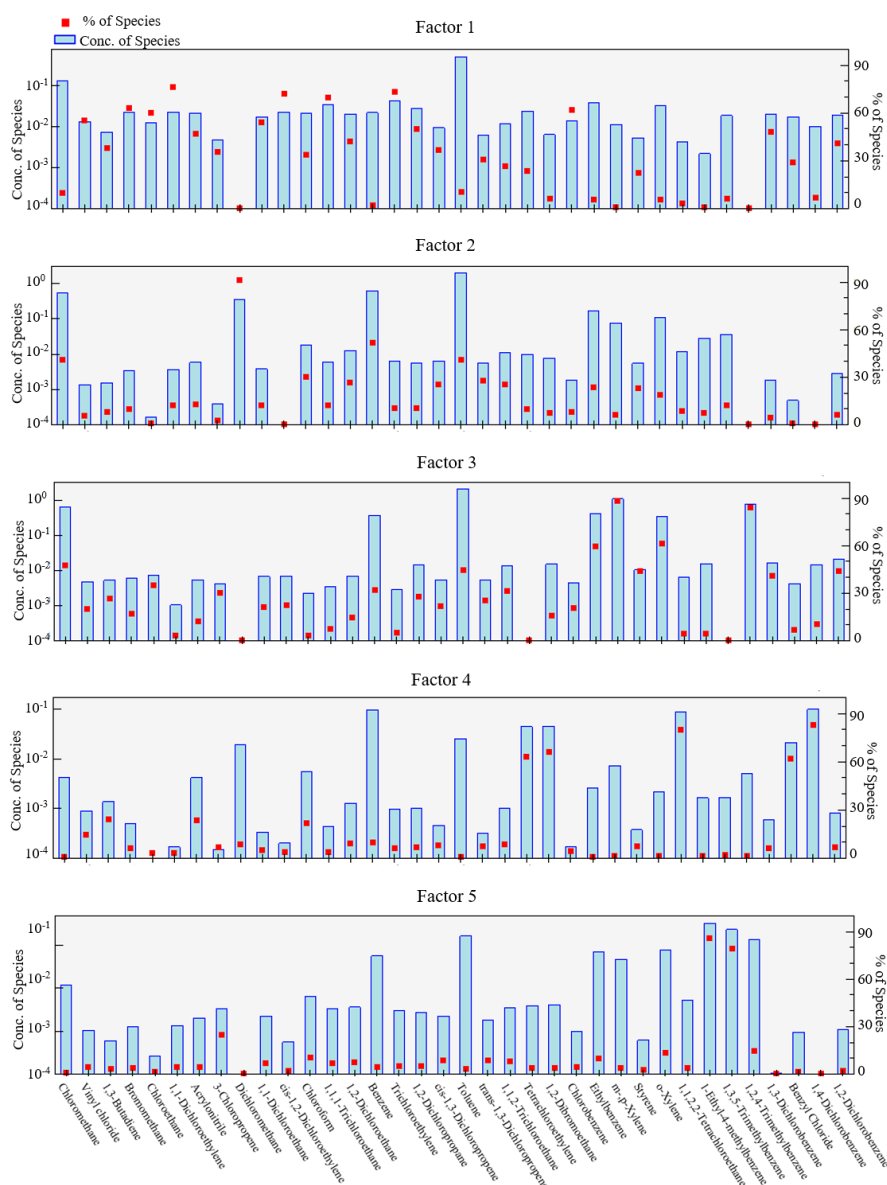


Figure 3. VOCs source profiles in Chulalongkorn hospital

Factor 2 was characterized by high percentages of toluene, benzene, o-xylene, ethylbenzene and chloromethane. Liu *et al.* (2008) reported that chloromethane was a typical/signature of VOCs released from biomass burning, while ethylene, ethane and the BTEX group were also typical VOCs emitted from this emission source. Therefore, this factor was classified as the influence from open burning activity.

High percentages of toluene and other aromatics such as benzene, m-, p-xylene, o-xylene, ethylbenzene and 1,2,4-trimethylbenzene were characterized in factor 3. Previous studies have reported that these VOC species are a signature of mobile source emissions (Geng *et al.*, 2010; Muezzinoglu, Odabasi, & Onat, 2001). Likewise, factor 5 was mainly contributed by 1-ethyl-4-methylbenzene, 1,3,5-trimethylbenzene and 1,2,4-trimethylbenzene, in which the trimethylbenzenes are key components of diesel vehicle

exhausts (Demir & Saral, 2013; Xue *et al.*, 2017). Therefore, factors 3 and 5 were primarily attributed to mobile sources.

Contributions of each emission source to total VOCs concentrations at the four monitoring stations of this study are summarized in Figure 6. At Bangkok (Chulalongkorn hospital and Dindaeng), the major emissions sources contributing to total VOC concentrations were mobile sources (more than 50%) and about 47.9% and 43.5% of total VOCs were contributed from chemical use in the households at Chulalongkorn hospital and Dindaeng, respectively. This indicates the ability of PMF analysis in this study to distinguish the major emission groups in each area. Chemical use in household sources provides many groups of VOCs, which indicates the importance of further study of individual species in order to analyze the characteristics and composition of chemical use in households (Ling *et al.*, 2011). At ERTC, contributions to total VOCs were 33.6%, 33.8% and 32.4%

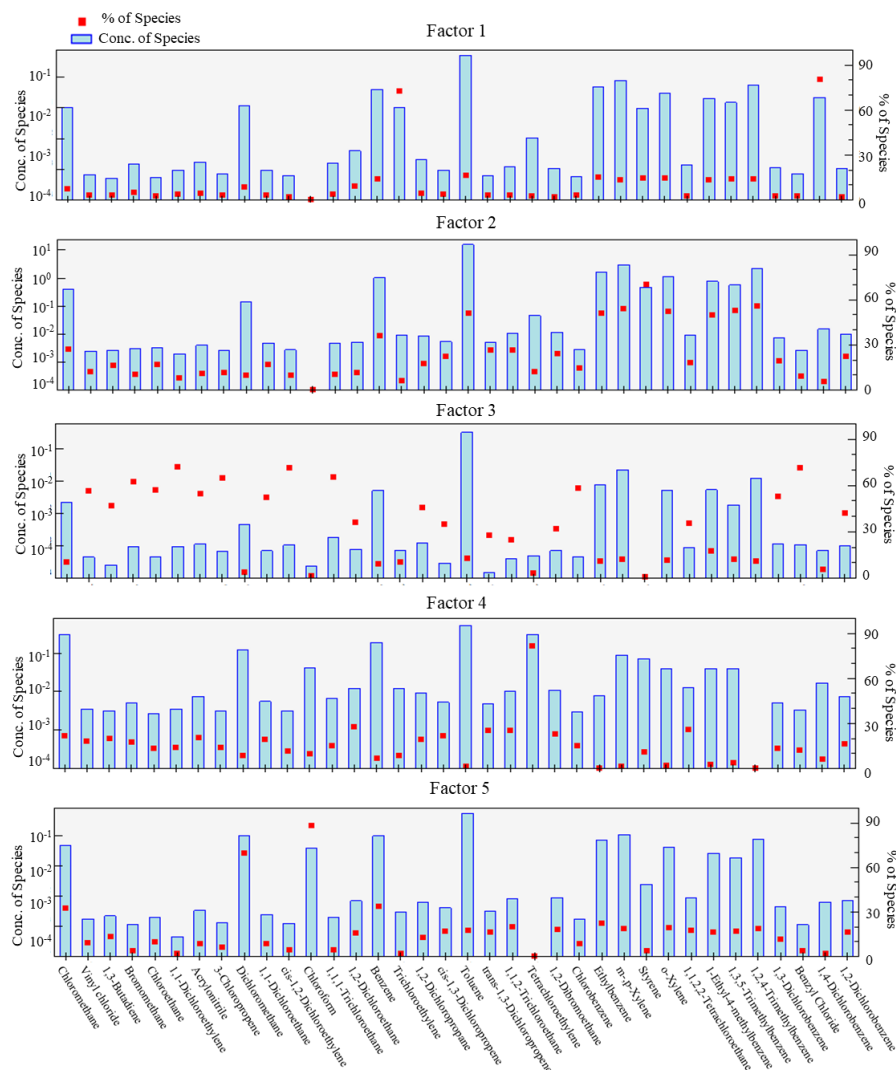


Figure 4. VOCs source profiles in Dindeang

from mobile sources, chemical use in households, and open burning, respectively. At City Hall Chiang Mai, about 54.5%, 14.4% and 31.1% of total VOCs were assigned to mobile sources, chemical use in households, and open burning, respectively.

3.2.2 Source contributions to individual VOC species for SOAP

Figure 7 shows the top 5 VOC species most contributing to SOAP and their corresponding source contributions at each monitoring station in this study. At Chulalongkorn hospital, toluene, m,p-xylene, ethylbenzene, benzene, and styrene were the top 5 VOC species in the formation of secondary organic aerosols, accounting for 66.1%, 8.6%, 7.5%, 5.4%, and 4.1%, respectively. PMF model revealed that the contributions from mobile sources to these compounds were about 51.0%, 54.0%, 51.4%, 36.2%, and 70.1%, respectively. Chemical use in households contributed about 49.9%, 45.9%, 48.5%, 63.7%, and 29.9% to toluene, m,p-xylene, ethylbenzene, benzene, and styrene,

respectively.

The top 5 VOC species for SOAP at Dindaeng were toluene (59.3%), m,p-xylene (11.3%), ethylbenzene (7.8%), benzene (7.1%), and o-xylene (5.4%). The source contributions of these species were quite similar to those in Chulalongkorn hospital. At ERTC, toluene, ethylbenzene, styrene, m,p-xylene and benzene were the top 5 VOCs species, accounting for 59.1%, 9.7%, 9.3%, 7.6%, and 6.7%, respectively, of the total SOAP. They were mainly emitted from mobile sources, chemical use in households, and open burning; except for styrene, which was mostly contributed from mobile sources and chemical use in households. Toluene was also the largest SOAP contributor in the City Hall Chiang Mai, followed by benzene, m/p-xylene, ethylbenzene, and o-xylene. They constituted 54.1%, 12.6%, 9.8%, 7.7%, and 5.3% of the total SOAP. The source contributions of toluene benzene, m/p-xylene, ethylbenzene, and o-xylene in this monitoring station were similar with those at the ERTC. Comparison of findings in this study with other related studies is summarized in Table 4.

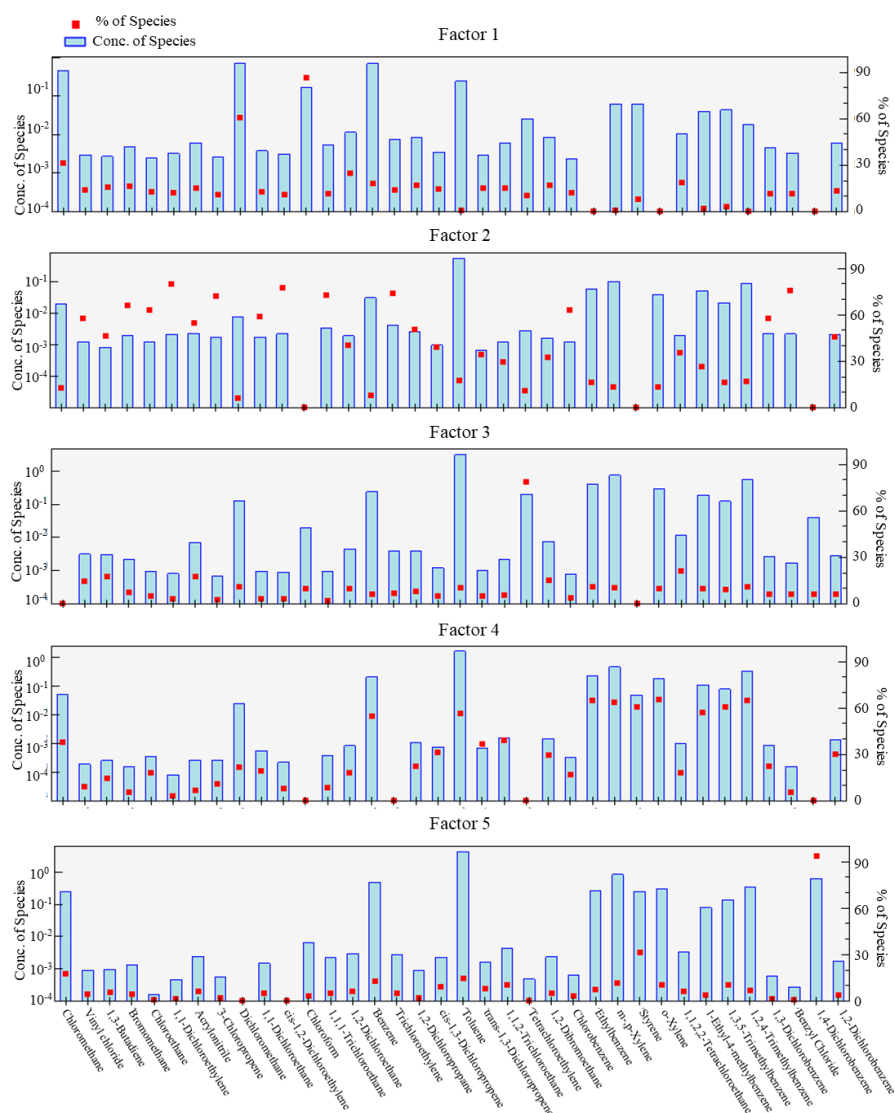


Figure 5. VOCs source profiles in ERTC

Table 4. Comparison of SOAP in different national and international cities

City, Country	Area	Top VOC species	% of SOAP	Source	Reference
Bangkok, Thailand	urban	toluene	54-75	mobile source	This study
Pathum Thani, Thailand	suburban	toluene	54.1	open burning	This study
Chiang Mai, Thailand	suburban	toluene	59.1	mobile source	This study
Western Taiwan	urban and industrial	toluene	-	industrial emissions	Huang <i>et al.</i> (2020)
Nanjing, China	suburban	toluene	-	traffic emissions	Mozaffar <i>et al.</i> (2020)
Calgary, Canada	urban	toluene	70	oil and gas production	Xiong, Zhou, Xing, and Du, (2020)
Beijing–Tianjin–Hebei, China	urban	toluene	43.1	on-road vehicles	Wu and Xei (2018)
Yangtze River Delta, China	urban	toluene	47.8	petrochemical industries	Wu and Xei (2018)
Pearl River Delta, China	urban	toluene	39.8	surface coating	Wu and Xei (2018)
Sichuan–Chongqing, China	urban	toluene	44.6	on- road vehicles	Wu and Xei (2018)

In summary, the top 5 VOCs constituted more than 90% of SOAP in each monitoring station. Toluene, m,p-xylene, o-xylene, ethylbenzene, benzene, and styrene were the key species. Mobile sources and chemical use in households

were generally the major sources of VOCs contributing to formation of secondary organic aerosols. Higher contribution of open burning to the formation of secondary organic aerosols was found in the suburban area.

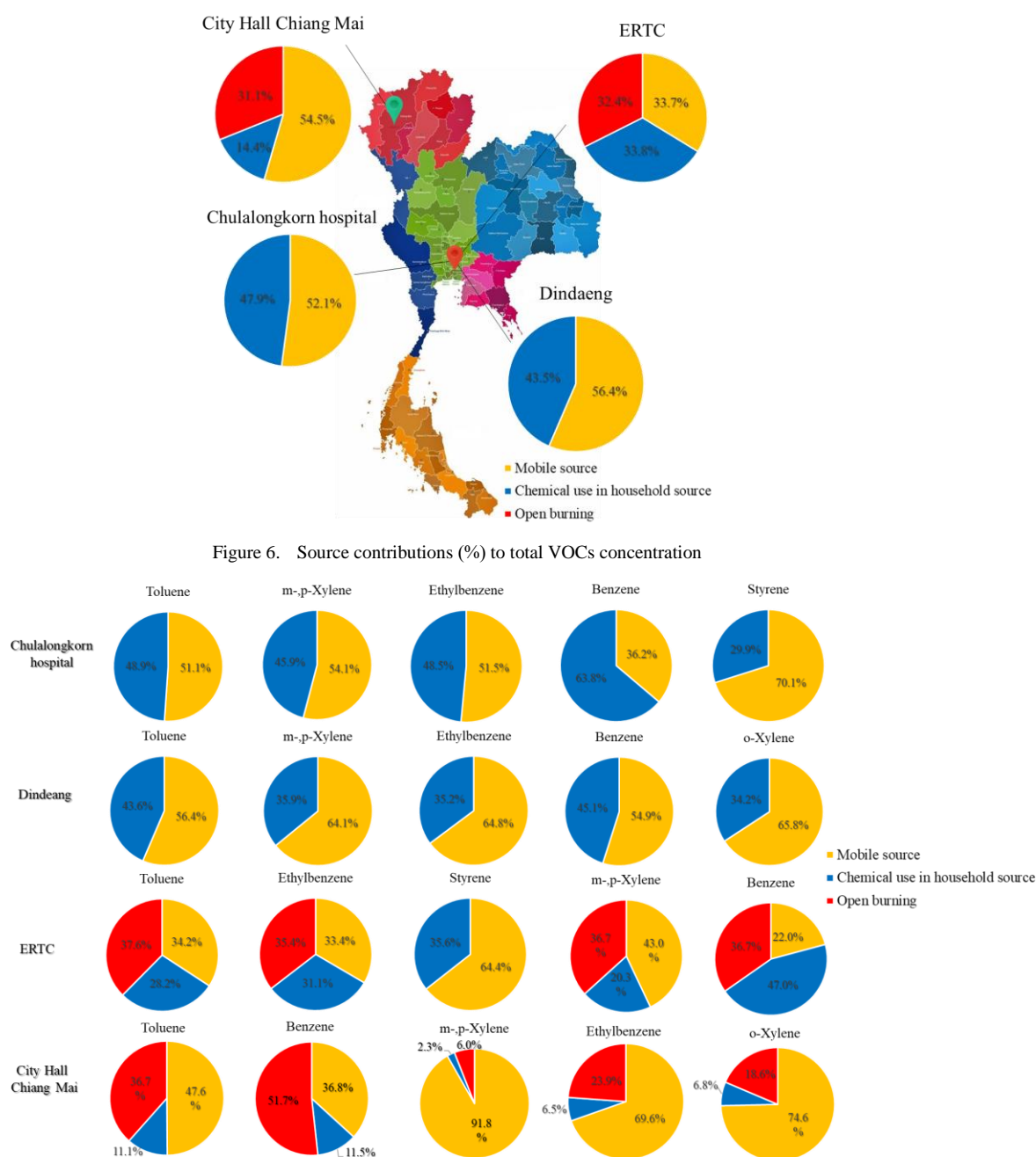


Figure 7. Source contributions to top five species affecting SOAP in the four monitoring stations

4. Conclusions

In this study, 44 VOCs ambient concentrations were measured in urban and rural (suburban) areas in Thailand and were comprehensively analyze for their potential to form secondary organic aerosols. Long term monitoring data from January 2013 to December 2019 were evaluated to identify the major VOC species contributing to the formation of fine particulates. Results indicated that the top 5 major components contributing to the formation processes were toluene, m-p-xylene, o-xylene, ethylbenzene, benzene and

styrene, in decreasing rank order. Source apportionment analysis was further performed to identify the potential emission sources of those compounds using the US.EPA PMF model. Results revealed that mobile sources were dominant accounting for more than 50 % of these top 5 VOC concentrations in Bangkok (roadside area). Another emission source contributing to VOC concentrations was chemical use in households. As for the rural/suburban areas (Pathum Thani province and Chiang Mai province), besides the contributions from mobile sources and chemical use in households, we also found significant influence of emissions from open/biomass

burning; in the target VOCs concentrations more than 30% of the total. Findings and methodology introduced in our study demonstrate the necessity in understanding the key VOCs attributed for the formation of secondary organic aerosols, as well as their potential emission sources, to effectively manage and control their emissions and mitigate the fine particulate pollution.

5. Acknowledgements

The authors would like to thank the Pollution Control Department of Thailand (PCD) for providing measured VOCs and monitoring station data used in this study. This study was partially supported for publication by the Faculty of Public Health, Mahidol University, Thailand.

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