Final Report

Seasonal variation of carbonaceous aerosols in the size-classified particulate matter down to PM0.1 in Songkhla Province, Thailand

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Seasonal variation of carbonaceous aerosols in the size-classified particulate matter down to PM0.1 in Songkhla Province, Thailand

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Prince of Songkla University

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บทคัดย่อ

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ชื่อโครงการ: ความแปรผันตามฤดูกาลขององค์ประกอบคาร์บอนในฝุ่นแยกขนาดเล็กลงถึงระดับอนุภาคนาโน (PM0.1) ในจังหวัดสงขลา ประเทศไทย

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บทคัดย่อ:

โครงการวิจัยนี้ได้ศึกษาองค์ประกอบคาร์บอนชนิดคาร์บอนอินทรีย์ (OC) และคาร์บอนธาตุ (EC) ในฝุ่นแยกขนาดเล็กลงถึงระดับอนุภาคนาโน (PM0.1) ในจังหวัดสงขลา ประเทศไทย โดยเก็บตัวอย่างฝุ่นด้วยเครื่องเก็บตัวอย่างฝุ่นอนุภาคนาโนในบรรยากาศเป็นระยะเวลาต่อเนื่อง 1 ปี ระหว่างเดือนมกราคมถึงธันวาคม 2561 ในอำเภอหาดใหญ่ จังหวัดสงขลา ทำการวิเคราะห์องค์ประกอบ OC และ EC ด้วยวิธีความร้อนและแสง (thermal-optical method) โดยเครื่องวิเคราะห์คาร์บอน (OC-EC Analyzer) เพื่อพิจารณาสัดส่วนขององค์ประกอบคาร์บอนในฝุ่นขนาดต่าง ๆ เพื่อศึกษาแหล่งที่มาของมวลอนุภาค จากผลการศึกษาพบว่าค่าเฉลี่ยรายเดือนของฝุ่นรวมมีค่าความเข้มข้นอยู่ระหว่าง 52.61 ถึง 104.68 ไมโครกรัมต่อลูกบาศก์เมตร โดยพบความเข้มข้นฝุ่นสูงสุดในฝุ่นขนาด 0.5-1.0 ไมโครเมตร ความเข้มข้นคาร์บอนอินทรีย์มีค่าสูงกว่าคาร์บอนธาตุในทุกขนาดฝุ่นและอัตราส่วนคาร์บอนอินทรีย์ต่อคาร์บอนธาตุ (OC/EC) มีค่าสูงเนื่องจากได้รับคาร์บอนอินทรีย์หลักมาจากการเผาชีวมวลในพื้นที่ หรือการเคลื่อนที่ระยะไกลจากต่างประเทศ รวมถึงคาร์บอนอินทรีย์ทุติยภูมิ สัดส่วนคาร์บอนทุติยภูมิมีค่าสูงสุดในฤดูก่อนมรสุม ตามด้วยฤดูมรสมและฤดูแล้งตามลำดับ องค์ประกอบคาร์บอนในฝุ่นขนาดต่าง ๆ ในพื้นที่ภาคใต้ ได้รับอิทธิพลมาจากกิจกรรมในพื้นที่เป็นหลัก ยกเว้นช่วงฤดูก่อนมรสุม การเคลื่อนที่ระยะไกลจากแหล่งกำเนิดไฟป่าบนเกาะสุมาตรา ประเทศอินโดนีเซีย ทำให้เพิ่มระดับคาร์บอนในฝุ่นในบรรยากาศของจังหวัดสงขลาให้สูงขึ้น ทั้งฝุ่นละเอียด (PM0.5-1.0 และ PM1.0-2.5) และฝุ่นหยาบ (PM2.5-10 และ PM>10) นอกจากนี้ยังพบว่าเฉพาะคาร์บอนอินทรีย์ในอนุภาคนาโน (PM<0.1) เพิ่มระดับสูงขึ้นเนื่องจากแหล่งกำเนิดไฟป่าขนาดใหญ่พัดพาเข้ามาในพื้นที่ การศึกษานี้จะเป็นประโยชน์ต่อการจัดการคุณภาพอากาศอันเนื่องมาจากฝุ่นละอองขนาดต่าง ๆ ในพื้นที่ภาคใต้ของประเทศไทย

คำสำคัญ: การเผาชีวมวล องค์ประกอบคาร์บอน มวลอากาศ อนุภาคนาโน การเคลื่อนย้ายระยะไกล

Abstract

Project Code: MRG6180077

Project Title: Seasonal variation of carbonaceous aerosols in the size-classified particulate matter down to PM0.1 in Songkhla Province, Thailand

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Abstract

In this study, particle-bound organic carbon (OC) and elemental carbon (EC) as well as char-EC and soot-EC in size-fractionated particulate matter (PM) down to ultrafine (PM0.1) particles were collected in Hat Yai city, Songkhla province, southern Thailand. The status and possible emission source based on carbon components were evaluated for one year (January - December 2018). The OC and EC were evaluated by thermal/optical reflectance (IMPROVE_TOR) method. Monthly and seasonal total suspended particles (TSP) mass concentrations in Hat Yai ranged from 52.61 to 104.68 μg/m^3. The results displayed that the highest concentrations of PM arose in the pre-monsoon season with the corresponding values in the dry and monsoon seasons being lower. The largest size of PM levels was PM0.5-1.0. The OC originates from primary emission sources and the formation by photochemical activities in the atmosphere. The concentrations of OC were higher than the concentrations of EC in all seasons. High OC/EC ratios might be attributed to OC-rich sources of emissions (i.e., biomass burning) or long-range transportation and secondary organic carbon (SOC) formation. The SOC concentrations contributed high in pre-monsoon followed by monsoon and dry, respectively. The main influence of characteristics of size-specific PM appears to be local sources, except during the pre-monsoon season, where the long-range transport of particles from Indonesian peatland fires causes an increase in the carbon concentration in southern Thailand. The transport plumes from Indonesian peatland fires may increase the OC and EC concentrations both in fine (PM0.5-1.0 and PM1.0-2.5) and coarse (PM2.5-10 and PM>10) particles. The OC fractions in ultrafine (PM<0.1) particles were also significantly affected by the Indonesian peatland fires during pre-monsoon season. This result indicates the importance of focusing emission control strategies on different PM particle sizes in southern Thailand.

Keywords: Biomass burning; Carbon component; Air mass; Nanoparticles; Long-range transport

Executive Summary

1. Introduction to the research problem and its significance

Recently, considerable attention to air pollution has been paid to ambient particulate matter (PM), which is believed to be strongly associated with global warming and public human health consequences. PMs are categorized into three modes: coarse mode (diameter ranged from 2.5 to 10 μm), fine mode (predominantly in accumulation mode, diameter ranged from 0 to 2.5 μm), and ultrafine mode (nucleation mode, diameter smaller than 0.1 μm, or 100 nm). The coarse mode particles are mainly generated from the attrition process, i.e., road and soil dust re-suspension, volcanic eruptions, and sea spray. On the other hand, the fine and ultrafine modes evolve from the combustion process, including biomass burning, motor vehicle, and coal combustion, and photochemical processes in the atmosphere. In the past decade, it has been reported that the finer particles (particularly, ultrafine (PM0.1) particles) pose a higher risk to human health consequences. The ambient PMs are associated with increased morbidity and mortality in humans. There is significant evidence that the PMs have detrimental impacts on the respiratory, circular, cardiovascular, and nervous systems. For the last decades in Thailand, the transportation of PMs in the atmosphere is important around 100-200 km (multi-provincial scale). Moreover, Southeast Asia (SEA) has been a source of PM pollutants affecting countries within the region and also outside the region. The transport plume of Indonesian forest fires affected air quality in Singapore, Malaysia, Brunei, and southern Thailand. Additionally, the recent study suggested that the PM pollutants from open biomass burning transport from northern SEA to East Asia, including southern China and Taiwan, and during the dry season.

The most considerable fraction of atmospheric particulate matter (PM) is carbon material with various physical and chemical characteristics, which contains around 20-50% mass concentration of PMs. The particle-bound total carbon (TC) can be separated into two types, including organic carbon (OC) and elemental carbon (EC) or black carbon (BC). BC and EC can be used interchangeably, depending on the analytical technique. BC is mainly emitted by high-temperature combustion processes (diesel and gasoline exhaust, coal burning, and biomass combustion). In contrast, OC is the light-scattering material, mainly generated from biomass and petroleum combustion and/or secondary chemical process in the atmosphere. The Intergovernmental Panel on Climate Change (IPCC) expected that EC led a global direct radiative forcing of around +0.2 Wm^{-2}, whereas OC was approximately the same magnitude with the opposite sign. Consequently, the primary emissions of EC present exclusive global warming potential and influence of the hydrological cycle. Primary pollutants, including NOx, EC, and OC, will involve an atmospheric photochemical activity to produce secondary organic aerosols (SOA) and ozone (O_3), creating an even more complicated effect.

Knowledge of carbon composition in the aerosol is also vital to distinguish emission sources and control measures for both PM and carbon components, the two main causes of global warming and air pollution. Most of the PM observed in Thailand, and Southeast Asia is presently based on PM10 measurement and a limited extent to PM2.5 observation via ground monitoring and satellite remote sensing. The impact of regional and global warming is very high uncertainty due to carbonaceous aerosols emitted into the atmosphere. This is because the distribution of carbon fractions varies in each time and location, which fundamentally contributed to the chemical, physical and optical characteristics of carbon components in PMs. Accordingly, size-specific characteristics of carbon material in PMs are vital to assess their radiative effects on global warming.

Hat Yai, the economic capital of Songkhla province, is the largest city in southern Thailand. Previous studies show that the main airborne pollutants are caused by biomass burning in rubber industries and transportation emissions. In southern Thailand, agro-industries based on the leading agricultural crop mostly use fuelwood from a para rubber tree. Ribbed Smoke Sheet rubbers (RSS) one of the main industries that always use this fuelwood in the production process. These results suggested that the environmental problem due to dangerous air pollutants, i.e., polycyclic aromatic hydrocarbons (PAHs) and smoke dust, is serious in southern Thailand. According to my best knowledge, there is only one study under the carbon components in the size-fractionated particulate matter down to PM0.1, or nanoparticles in the upper part of Thailand, collected at Chiang Mai and Bangkok. Overall, the main objective of this study is to investigate the seasonal variation of carbon components, including OC and EC in size-classified PM down to PM0.1 in the atmospheric environment collected in Hat Yai city, Songkhla province, southern Thailand. The data from this project will provide knowledge of the temporal variation of size-specific and compositions in an urban area, which leads to a critical global warming problem and negative health consequences in developing countries.

2. Literature review

2.1 Carbonaceous Aerosol

The main source of carbonaceous aerosols (CA) is biomass, coal, and fossil combustion. Biomass burning is the largest contributor to black carbon (BC) and organic carbon (OC) in the atmosphere (Bond et al., 2004, 2013). The term, BC which is loosely equivalent to elemental carbon (EC), is used in climate models, which often refer to light-absorbing materials. At the same time, EC is preferred in fields relating to aerosol chemistry (Watson et al., 2005). Different sources of CA, e.g., diesel soot, coal burning, and biomass combustion, have different OC/EC ratios (Han et al., 2009; Guo, 2016).

Table 1 Source identification with OC/EC and Char-EC/Soot-EC ratios

Sources	OC/EC	Char-EC/Soot-EC
Diesel exhaust	<1.0 (Allen et al., 2001)	1.0-2.0 (Chow et al., 2004)
Gasoline exhaust	2.0-2.4 (Liu et al., 2006)	1.0-2.0 (Chow et al., 2004)
Biomass combustion	7.0-8.0 (Zhang et al., 2007)	2.0-5.0 (Chen et al., 2007)
Wood combustion	16.8-40.0 (Schauer et al., 2001)	N/A
Residential coal combustion	2.5-10.5 (Chen et al., 2006)	1.5-3.0 (Cao et al., 2005)
Residential cooking produced	32.9-81.9 (He et al., 2004)	2.0-6.0 (Chow et al., 2004)

The higher ratio results from biomass burning, while coal combustion and motor vehicle resulting in a lower ratio of OC/EC (Cao et al., 2005; Pio et al., 2011; Han et al., 2016). At the same time, the ratio of OC/EC depends on three factors for correctly classifying the emission source. The three factors are included in primary emission sources, deposition rates, and secondary organic aerosols (SOA) (Khan et al., 2016). Further, EC can be separated into char-EC and soot-EC. Char-EC is generated in low-temperature burning processes and contains the source material. Conversely, soot-EC largely emitted from the high-temperature gas phase of the condensation of hydrocarbons (Cao et al., 2005; Han et al., 2007). There is a shortage of data on OC and EC in addition to char-EC and soot-EC ratios in size-specific PM. Unlike the OC/EC ratios, Char/Soot ratios are distinct for different origin sources and can be used to identify the source at the origin. Only two factors that can interrupt the char/soot ratio are the primary emission source and the deposition rate. A higher ratio of char/soot indicates the dominance of biomass combustion associated with char contributions to total EC contents. In contrast, ratios smaller than 2.0 shows that soot from fossil fuel is a large contributor to the total EC concentrations (Han et al., 2016) (Table 1).

Source apportionments of air pollutants comprising BC and OC have been studied based on the source-receptor techniques (Castro et al., 1999; Favez et al., 2010). Source-receptor methods like the positive matrix factorization (PMF) and the chemical mass balance (CMB) have also been used to identify carbonaceous aerosol and to quantify the contribution of each source. Bressi et al. (2014) studied seven sources at urban sites in Paris, France, including ammonium nitrate, ammonium sulfate, metal manufacturing, marine aerosols, fatty oil burning, road transportation, and biomass burning in fine particulate matter (PM2.5). They found that road transportation and biomass combustion are the two main sources of primary organic aerosols, contributing 14% and 12% to PM2.5 mass concentration, respectively.

However, only a few publications have reported the carbonaceous aerosol (OC and EC) pattern in Thailand (Duangkaew et al., 2013; Pongpiachan et al., 2014a; Phairuang et al., 2019a). There have been scarce studies of carbon components, both spatial and temporal

variation in Thailand, particularly of the nano-scale ambient particles related to carbon components. Phairuang et al. (2019a) described that the emission from agricultural activities and forest fires was mainly distributed the carbonaceous aerosol in the upper part of Thailand (Chiang Mai and Bangkok), where the open biomass burning is very intensive. PM0.1, which usually has more diesel emission content, was presented to be very sensitive to the influence off open biomass burning.

2.2 Air Quality in Songkhla province, southern Thailand

Currently, many environmental problems in Thailand are related to air pollution from fossil fuels burning along with biomass burning, which will take one of the most critical environmental studies. Oil palm and para rubber are the leading plantations in the southern part of Thailand (OAE, 2016). Many residues of these plants are used as the main fuel for agro-industries, i.e., rubber sheets and oil palm. Fuelwood burning in the agro-industry process contains particulate smoke and PAHs, which are environmental loads in this area (Choosong et al., 2007). The southern Thailand, para-rubber fuelwood burning in the ribbed smoke sheet rubbers (RSS) factory is the major source of PAHs (Furuuchi et al., 2006; Tekasakul et al., 2008). The wood moisture content and burning period are the main factors to control smoke particles and associated PAHs into the workplace and surrounding areas (Chomanee et al., 2009; Choosong et al., 2010). However, the para-rubber residues are used as fuel in industries, not only in rubber industries but also in several industries in the southern part of Thailand, which use fuelwood boilers. Pongpiachan et al., 2014a-b indicated that the impacts of maritime aerosols, biomass combustion, and possible agricultural residue burning are the critical source of PM10 related to carbon components. Therefore, the physical and chemical characteristics of size-fractionated ambient particles down to PM0.1, or nanoparticles in southern Thailand as a case study of Songkhla province must be investigated for the relationship between the main emission sources and air quality in this area.

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3. Objectives

The main purpose of this project is to have a better understanding of the emission profiles of carbonaceous aerosol in Songkhla, Thailand, and their relationship with the emission source. In order to achieve this goal, these objectives will be followed:

1) To study the physical and chemical characteristics of size-classified ambient particles down to PM0.1, or nanoparticles in Songkhla province, southern Thailand.
2) To investigate the meteorological effects of the physical and chemical characteristics of size-classified ambient particles down to PM0.1, or nanoparticles in Songkhla province, southern Thailand.
3) To investigate the relationship between the emission source and size-classified ambient particles down to PM0.1, or nanoparticles in Songkhla province, southern Thailand.
4) To investigate the fate and transportation size-classified ambient particles down to PM0.1, or nanoparticles in Songkhla province, southern Thailand.

4. Research Methodology

4.1 Sampling location

Hat Yai city is located on Songkhla province in the southern part of Thailand. PM concentrations were collected on the top floor of an 8-floor building, which is part of the Sirindhorn Applied Research Engineering Building, Faculty of Engineering, Prince of Songkla University (PSU; 7°00'21.8"N 100°30'08.6"E). The sampling site is located approximately 4-km away from the business area, where household and traffic are not severe. A total of events such as domestic cooking, local industry, road transportation, and building construction can be considered to be significant local sources of PMs emission into the Songkhla atmosphere. Figure 1. shows the monitoring site in this study.

Figure 1 Location of the sampling site, Prince of Songkla University (PSU), Hat Yai, Songkhla, southern Thailand

During monitoring periods, the weather parameter data, including relative humidity, rainfall, temperature, pressure, and wind speed, were also collected in detail, as shown in Table 2. The weather in the southern part of Thailand can be divided into three seasons, depending on the monsoons in the region, the dry season from January to April, the pre-monsoon season (May to August), and the monsoon season (September to December).

Table 2 Meteorological parameters during different sampling times

Sampling period	Temperature (°C)	RH (%)	Wind speed (Knot)	Pressure (hpa)	Rainfall (mm)
Jan-April, 2018	24.7-28.3 (26.8)	65-82 (74)	1.92-3.59 (2.88)	913-1009 (977)	28.6-215.8 (117.9)
May-Aug, 2018	27.0-28.3 (27.9)	77-81 (78)	1.45-2.45 (1.93)	975-1008 (1000)	75.4-245.2 (153.2)
Oct-Dec, 2018	26.3-27.2 (26.7)	79-84 (82)	1.55-2.62 (2.05)	976-1010 (993)	155.2-369.4(252.9)
All year, 2018	24.7-28.3 (27.1)	65-84 (78)	1.45-3.59 (2.29)	913-1010 (990)	28.6-369.4 (174.6)

4.2 Cascade sampler for size-segregated ambient particles

A cascade air sampler consisting of four impactor stages (10, 2.5, 1.0, 0.5 μm, of cutoff sizes) and one inertial filter stage for the separation of particles less than 0.1 μm, (PM0.1) along with a backup filter for the collection of PM0.1 (Furuuchi et al., 2010) was used at a flow rate of 40 l/min. Quartz fibrous filters (QFF) (Pallflex Tissuquartz 2500QAT-UP; Pall Corp., Japan) of Ø 55 mm. All of the filters were preheated for one hour at 350 °C to eliminate any possible contamination and then conditioned RH at 35 ± 5 % and temperature at 21.5 ± 1.5 ^{o}C, in a weighing chamber (PWS-PM2.5, Tokyo Dylec Corp., Japan) for 48 hrs before and after sampling. Webbed stainless steel fibers (average fiber diameter of 9.8 μm, steel type SUS316; Nippon Seisen, Japan) were located inside the nozzle of the inertial filter consisting of a duralumin cartridge with a 5.25 mm diameter nozzle that was 5.5 mm in length. 48-hr samples were collected from January to December 2018. Samples were collected for 6 consecutive days (2 days per sample) in the second week of each month. A total of 34 sample sets were collected in this study. After the air sampling, the samples were kept in a refrigerator at -20 ^{o}C until chemical analysis. A blank filter was also prepared to check for possible contamination during filter transportation and the sampling period.

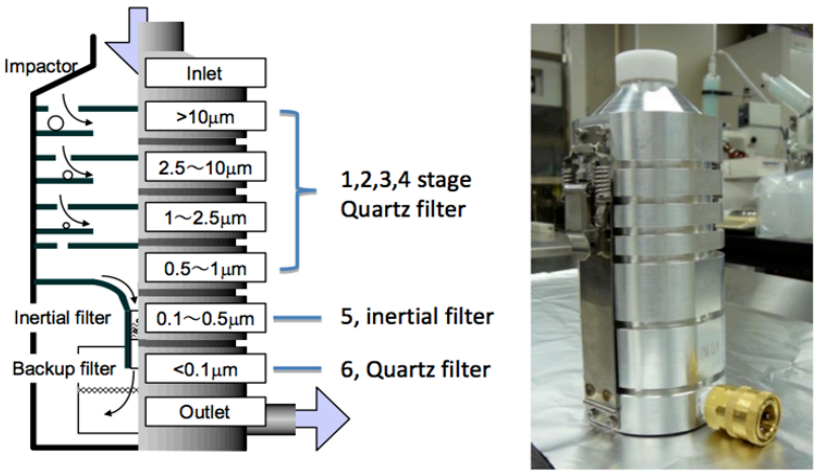


Figure 2 Cascade sampler for size-segregated ambient particles (Nano-Sampler)

4.3 Chemical composition analysis

Particle-bound carbonaceous components as organic carbon (OC) and elemental carbon (EC) were evaluated using a Sunset Laboratory Carbon Aerosol Analyzer with the IMPROVE_TOR protocol (Chow et al., 2004, Watson et al., 2005). An area of 1.5 cm^2 (1.5 cm x 1 cm) squarely punched from each filter sample was analyzed for four OC fractions (OC1, OC2, OC3, OC4 at 120 ^oC, 250 ^oC, 450 ^oC 550 ^oC, respectively) in 100% Helium (He) while the EC fractions (EC1, EC2, and EC3 at 550 ^oC, 700 ^oC, 800 ^oC, respectively) were analyzed in a 2% O_2/98% He atmosphere. The data were corrected for the charring of organic materials (OPC) using a reflectance laser. OC and EC are defined respectively as follows:

OC = OC1+OC2+OC3+OC4+OPC (1)

EC = EC1+EC2+EC3-OPC (2)

The char-EC/soot-EC ratio, where char-EC is defined as EC1-OPC and soot-EC as EC2 + EC3 (Han et al., 2007, 2009), was also evaluated. Quality assurance and quality control (QA/QC) for carbon analysis were regulated against a reference standard and blank filters. The calibration of the carbon analysis was based on total carbon (TC), as analyzed against sucrose ($C_{12}H_{22}O_{11}$) (196-00015, Sucrose, Wako Pure Chemical Industries, Ltd., Japan) as a reference chemical. The minimum detection limit (MDL) was evaluated as 0.2 μg g/cm^2 and 0.1 μg/cm^2, respectively, for OC and EC based on the measured travel-blank filters (n = 3). The carbon concentration from the blank filters was subtracted from the samples.

4.4 Hot Spots and Backward Trajectories

The hotspots from open biomass burning were identified from satellite remote sensing imagery at a 1 km^2 resolution and were provided by the Moderate Resolution Imaging Spectrora Diometer (MODIS) onboard NASA's Aqua and Terra satellites. (The MODIS hotspot data are available from http://earthdata.nasa.gov/data/near-real-time-data/firms). Backward trajectories were used to examine the transport and migration of PM from distant areas. Back trajectory analysis was conducted by the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) transport model. The backward trajectories were used to examine the transport and migration of PMs from distant areas. The 72-hr backward trajectories of air parcel arriving at the sampling site in Hat Yai, Songkhla province at 500 meters from the average ground level (AGL), were evaluated. The Global Data Assimilation System (GDAS) resolution 0.5 degree, from the NOAA website (https://ready.arl.noaa.gov/gdas1.php), was used to obtain the meteorological data. As noted in a previous report (Kim Oanh and Leelasakultum, 2011), the ending height from 500 m AGL calculated depending on the reference of the backward trajectory in Thailand.

5. Results and Discussion

5.1 Mass concentrations of size-fractionated particulate matter

Table 3 Monthly variation of size-fractionated particles mass concentration in Songkhla, southern Thailand

Size	Jan	Feb	Mar	Apr	May	Jun
< 0.1	10.03 ± 1.54	11.31 ± 1.29	8.61 ± 0.29	8.68 ± 3.48	9.35 ± 0.66	11.76 ± 1.18
0.1 - 0.5	2.46 ± 0.54	2.44 ± 0.84	1.29 ± 1.05	1.48 ± 0.73	3.81 ± 0.26	5.61 ± 1.21
0.5 - 1.0	11.09 ± 1.08	15.01 ± 1.96	11.54 ± 1.28	12.68 ± 3.95	12.84 ± 0.46	18.58 ± 1.22
1.0 - 2.5	11.54 ± 0.85	12.95 ± 4.88	11.25 ± 1.73	11.93 ± 2.64	10.88 ± 2.12	15.48 ± 0.48
2.5 - 10	13.36 ± 5.46	15.56 ± 3.02	14.97 ± 5.09	13.48 ± 3.12	16.58 ± 1.32	16.56 ± 5.73
>10	13.11 ± 2.86	12.14 ± 0.41	11.14 ± 2.65	11.53 ± 3.35	11.40 ± 1.36	18.81 ± 5.85
Size	Jul	Aug	Sep	Oct	Nov	Dec
< 0.1	11.08 ± 0.95	16.04 ± 2.51	7.41 ± 0.93	9.08 ± 0.28	8.93 ± 0.13	9.73 ± 0.13
0.1 - 0.5	4.93 ± 1.53	6.81 ± 1.78	3.83 ± 1.22	1.71 ± 0.09	1.72 ± 0.13	2.77 ± 0.18
0.5 - 1.0	15.69 ± 3.03	22.09 ± 3.24	11.68 ± 2.10	11.07 ± 1.06	10.91 ± 1.45	10.72 ± 0.16
1.0 - 2.5	13.85 ± 1.65	17.69 ± 1.19	10.42 ± 1.05	9.53 ± 0.64	9.19 ± 0.37	9.53 ±1.87
2.5 - 10	20.85 ± 3.35	25.59 ± 2.41	14.89 ± 1.08	13.03 ± 2.33	11.84 ± 1.55	14.92 ± 1.04
>10	14.24 ± 1.74	16.48 ± 0.60	9.84 ± 1.00	11.05 ± 1.80	10.02 ± 0.23	9.82 ± 0.57

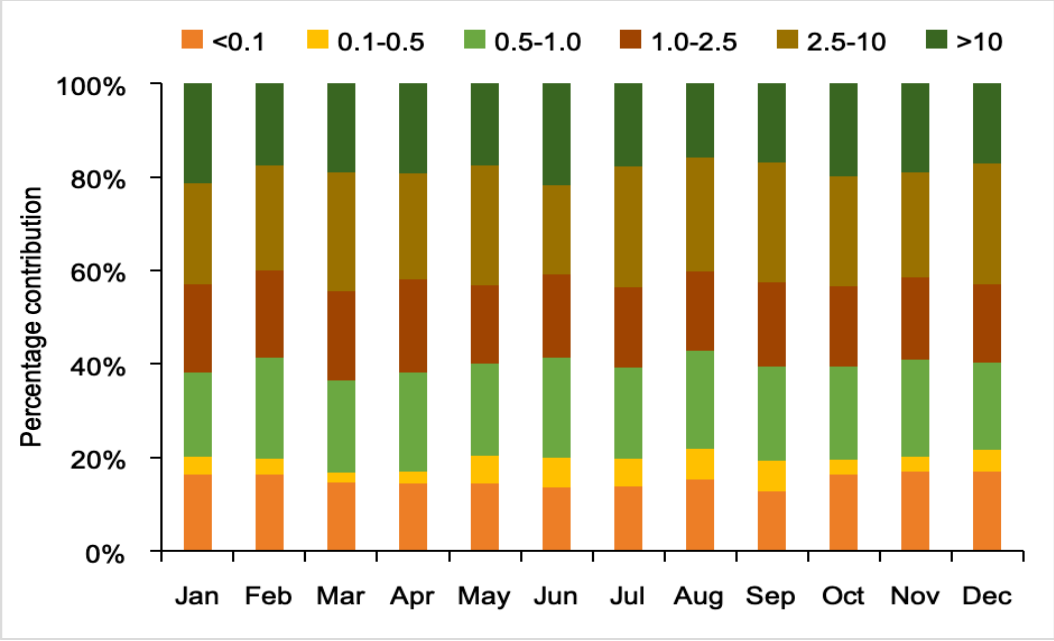


Figure 3 Percentage contribution of individual size fractions to TSP load in different month

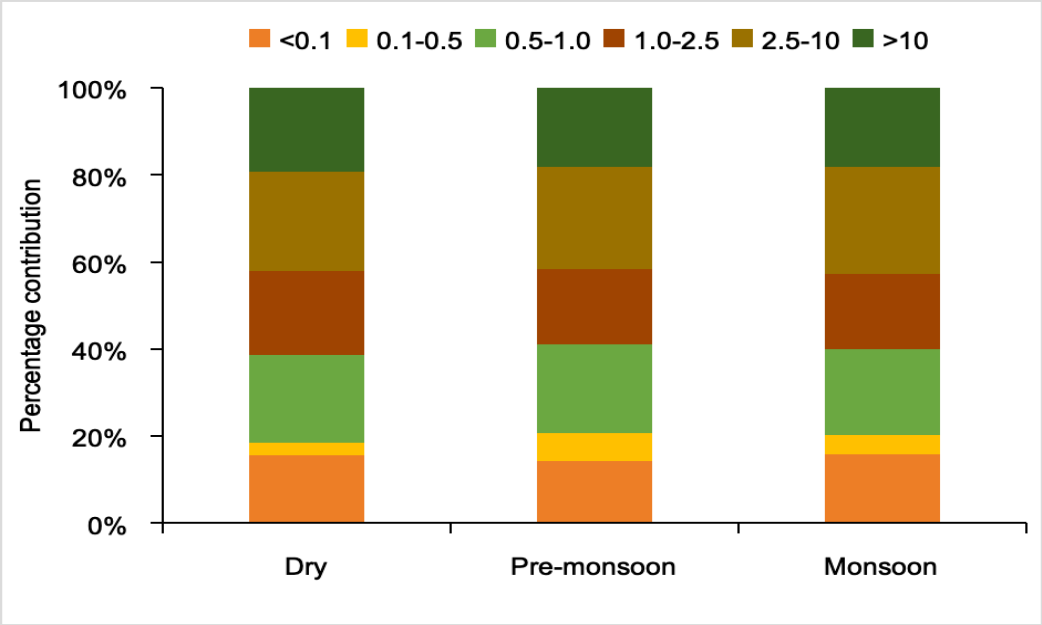


Figure 4 Percentage contribution of individual size fractions to TSP load in different season

Table 3. displays the size distribution of PM in Hat Yai, Songkhla province, southern Thailand. The highest mass concentration of PM is particle size 2.5-10 μm, followed by PM0.5-1.0. TSP concentrations lower than the TSP standard value from the WHO and Thailand guidelines (100 and 330 μg/m^3, respectively) except during haze episodes in August. On the other hand, PM10 and PM2.5 exceeded the NAAQSs of PM10 (50 μg/m^3) and PM2.5 (25 μg/m^3). All of these values exceeded the suggested values by the World Health Organization (WHO), 60, 20, 10 μg/m^3, respectively. However, there is no standard for ambient PM0.1 in Thailand and other countries. In each month, the august has the highest PM0.1 mass concentration to be 16.04 ± 2.51 μg/m^3 followed by the June to be 11.76 ± 1.18 μg/m^3 and the lowest PM0.1 mass concentration in the rainy season especially, September to be 7.41 ± 0.93 μg/m^3, which starting monsoon season in southern Thailand.

Figures 3 and 4 show the percentage contribution of size-fractionated particles in each month and season. The PM2.5-10 contribution to TSP was the highest (24%), followed by PM0.5-1.0 (20%) on an annual average basis. PM0.1 contributed around 17%, whereas PM0.1-0.5 is the lowest percentage of contribution (5%). The seasonal variation of the percentage of size-specific particles in southern Thailand is almost constant (Figure 4). In southern Thailand, there was a fluctuation of PM mass concentration, probably related to local emission sources, especially wood biomass burning in agro-industries (Pongpiachan et al., 2014; Phairuang et al., 2019b). Although, there might be transboundary of smoke aerosol during large-scale forest fires. Extreme event pf pre-monsoon season (June-August), Indonesian smoke haze is transported to lower southern Thailand. (Pentamwa and Kin Oanh, 2008; Chisholm et al., 2016). During such haze episodes, massive numbers of PMs are

transported from one country to others depending on the wind direction and meteorological condition.

5.2 Mass concentrations of OC, EC, Char-EC, Soot-EC, and TC

Table 4 Annual average concentrations of OC, EC, Char-EC, Soot-EC, T, C and mass (ug/m^3) in Songkhla

Size	OC	EC	Char-EC	Soot-EC	TC	PM
$PM_{<0.1}$	0.68 ± 0.60	0.23 ± 0.14	0.06 ± 0.04	0.17 ± 0.12	0.90 ± 0.82	10.17 ± 2.23
$PM_{0.5\text{-}1.0}$	0.93 ± 0.37	0.28 ± 0.22	0.14 ± 0.09	0.15 ± 0.15	1.22 ± 0.53	13.66 ± 3.57
$PM_{1.0\text{-}2.5}$	0.45 ± 0.40	0.12 ± 0.08	0.07 ± 0.05	0.05 ± 0.03	0.57 ± 0.45	12.02 ± 2.58
$PM_{2.5\text{-}10}$	0.62 ± 0.35	0.18 ± 0.12	0.11 ± 0.08	0.06 ± 0.04	0.79 ± 0.45	15.97 ± 3.80
$PM_{>10}$	0.18 ± 0.16	0.06 ± 0.06	0.03 ± 0.13	0.03 ± 0.03	0.23 ± 0.21	12.47 ± 2.79

The OC, EC, Cher-EC, Soot-EC, TC (OC+EC), and particle mass concentrations for each particle size are shown in Table 4. The annual averages in each carbon species for size-fractionated PM were collected from January to December 2018 at the PSU monitoring site. The TC was highest in particle size 0.5-1.0 μm followed by PM<0.1, PM2.5-10, PM1.0-2.5 and PM>10, respectively. Moreover, OC and EC are also high in finer particles, especially PM0.5-1.5 to be 0.93 ± 0.37 and 0.28 ± 0.22 μg/m^3, respectively. This result is similar to size-fractionated particles from other parts of Thailand (Chiang Mai and Bangkok) (Phairuang et al., 2019a). In Hat Yai, PM<0.1 and PM2.5-10 were also dominant in the OC and EC concentration. OC levels were 0.68 ± 0.60 and 0.62 ± 0.35 μg/m^3, respectively, while those for EC were 0.23 ± 0.14 and 0.18 ± 0.12 μg/m^3, respectively. Additionally, the annual averages of char-EC as well as soot-EC concentrations were highest in the PM0.5-1.0 range to be 0.14 ± 0.09 and 0.15 ± 0.15 μg/m^3, respectively.

Table 5 Seasonal average of OC and EC concentrations (ug/m^3) and OC/EC ratio in Songkhla

D = Dry season P = Pre-monsoon season M = Monsoon

Size	OC (ug/m^3)			EC (ug/m^3)			OC/EC (-)		
	D	P	M	D	P	M	D	P	M
PM<0.1	0.44	1.22	0.42	0.18	0.34	0.14	2.75	3.00	3.15
PM0.5-1	0.35	1.90	0.47	0.31	0.36	0.17	1.12	5.47	2.77
PM1-2.5	0.25	0.80	0.26	0.11	0.29	0.28	2.77	4.53	4.05
PM2.5-10	0.30	0.92	0.63	0.09	0.27	0.16	3.87	3.67	5.35
PM>10	0.08	0.31	0.14	0.02	0.11	0.04	3.98	2.85	4.57

Table 6 Seasonal average of Char-EC and Soot-EC concentrations (ug/m^3) and Char-EC/Soot-EC ratio in Songkhla D = Dry season P = Pre-monsoon season M = Monsoon

Size	Char-EC (ug/m^3)			Soot-EC (ug/m^3)			Char-EC/Soot-EC (-)		
	D	P	M	D	P	M	D	P	M
PM<0.1	0.05	0.08	0.04	0.14	0.25	0.10	0.37	0.35	0.34
PM0.5-1	0.14	0.17	0.09	0.16	0.19	0.08	0.82	1.47	1.15
PM1-2.5	0.06	0.11	0.04	0.05	0.07	0.04	1.41	1.59	1.16
PM2.5-10	0.06	0.18	0.10	0.03	0.09	0.06	1.98	2.18	1.89
PM>10	0.01	0.05	0.02	0.01	0.06	0.02	1.27	0.92	1.07

The ratios of OE/EC can be used to identify specific sources of carbonaceous aerosols. Ratios for biomass combustion, coal burning, for diesel engines are all different. Particles derived from biomass combustion have a higher ratio, while the OC/EC ratios for oil and fossil combustion results are lower (Guo, 2016). The ratio of OC to EC for biomass combustion is higher (~3-7) (Pongpiachan et al., 2014; Thuy et al., 2018) and that from fossil fuel is lower (<1) (Allen et al., 2001; Pio et al., 2011). Characteristic of emission sources of carbon fractions include diesel exhaust (OC/EC ~ 0.06-0.8) (Na et al., 2004; Dallman et al., 2014), biomass combustion (OC/EC ~ 3.9-4.2) (Thumanu et al., 2009) and long-range transport/aged aerosol (OC/EC ~ 12) (Saarikoski et al., 2008).

The OC and EC concentrations during pre-monsoon season are the highest concentration in every particle size (Table 5). The ratios of OC/EC reported in this study were in general agreement with previously reported findings. The ratios of OC/EC for ambient PM0.1 in this study (2.75, 3.00 and 3.15 for dry, pre-monsoon and monsoon, respectively) are lower than those from ambient PM0.1 in Chiang Mai during a haze episode (3.49), Bangkok (3.37) (Phairuang et al., 2019a). Additionally, this result is also lower the corresponding values for Hanoi, Vietnam that ranged from 3.79-5.68 (Thuy et al., 2018), but were higher than those from Taiwan that ranged from 0.21-1.71 (Chen et al., 2010; Zhu et al., 2010). The OC/EC ratios in Hat Yai showed different particle size distributions. The OC/EC ratio from particle size range 0.5-10 μm is also higher than others, especially pre-monsoon season. However, the OC/EC ratios in every size range were higher than 1 and represent a mixture of emission sources.

Moreover, Thumanu et al. (2009) reported on OC/EC ratios in PM10 from various sources that were obtained in Songkhla province, Thailand. The measured OC/EC ratios in that study were as follows; 1.2-1.3 (traffic), 1.6-2.3 (industry), and 3.9-4.2 (biomass burning). These outcomes indicate that industrial and biomass burning sources account for much of the carbon in the higher OC/EC ratios in the ambient air above Hat Yai, southern Thailand. OC dominates,

especially in PM0.1 and this may be attributed mainly to biomass burning. Conversely, the ratio of OC/EC depends on three factors for correctly classifying the emission source. The three factors include primary emission sources, the deposition rate, and secondary organic aerosols (SOA) (Cachier et al., 1996; Hallquist et al., 2009).

Unlike the OC/EC ratio, the char-EC/soot-EC ratio is different from each source, and it is frequently possible to identify the sources (Duangkaew et al., 2013; Guo, 2015). Only two factors can interrupt the char/soot ratio, namely the primary emission source and the deposition rate. A higher ratio of char/soot (generally >2) is indicative of biomass combustion and char contributes to the total EC levels; whereas char/soot < 1.0 suggest that emission from diesel engines is a significant contributor to the total EC concentrations (Han et al., 2016). The char/soot ratio in PM0.1 fraction in the present study was nearly constant, at around 0.35. The char/soot ratio serves as an index of fossil fuel combustion (<1.0) (Thuy et al., 2018; Phairuang et al., 2019a). The findings suggested that fossil fuel used in local transportation and diesel exhaust are essential factors in the atmospheric nanoparticles. However, the ratio of char/soot in the PM2.5-10 fraction was increased to 2.18 during pre-monsoon season and was also high in fine and coarse mode particles. The influence of biomass combustion is an important source for the increased Char-EC levels and represents the biomass burning emission source (Kumar and Attri, 2016). The OC/EC and char/soot ratios were also different from the size distribution (Guo, 2015). Moreover, other size-segregated PMs and chemical components are essential in terms of future, more detailed investigations in the further development of source apportionment studies.

5.3 Estimation of Secondary Organic Carbon (SOC)

A breakdown between primary and secondary sources of OC cannot be directly obtained from a carbon analyzer. EC is a good tracer of primary OC (POC) since EC and POC are predominantly emitted from the same emission sources. Based on the EC tracer method (Turpin and Huntzicker, 1995), the expected POC is commonly estimated by analysis of a subset of data collected at a time and place when POC emissions are dominant. The EC-tracer method can be used to calculate the POC based on the following equation: (Lim and Turpin, 2002)

$$POC = (OC/EC)_{min} \times (EC) \quad (1)$$

$$SOC = OC_{tot} - POC \quad (2)$$

where POC is the primary organic carbon, SOC refers to secondary organic carbon, $(OC/EC)_{min}$ is the value of the lowest OC/EC ratio in each season, OC_{tot} is the Total OC. In this study, the carbon distributions for each species were estimated for each season, and the contribution of POC, SOC, and EC relative to TC, (TC = EC + OC, OC = POC + SOC) are presented in Figure 5-9.

The averaged proportion of SOC showed that the contribution of PM0.1 to be 14%, 36%, 30% in the dry, pre-monsoon, and monsoon seasons, respectively (Figure 5). In Thailand, secondary atmospheric PM0.1 has not been studied so far. This is the first semi-direct attempt to estimate the SOC in PM0.1 in each season. The averaged SOC in each season in southern Thailand was lower than the corresponding values for Hanoi, Vietnam. Thuy et al. (2018) reported that SOC smaller particles were dominant compared to larger particles; the SOC in PM0.1 contributes up to 42.7% of the OC level in Hanoi, Vietnam. Instead, the percentage of SOC in the fine particle (PM0.5-1.0) is distributed from 23%, 56%, 45% in the dry, pre-monsoon, and monsoon seasons, respectively (Figure 6). In the same manner, the SOC in PM1.0-2.5 contributed around 35%, 59% and 40% in the dry, pre-monsoon, and monsoon seasons, respectively (Figure 7). The SOC contribution increased in fine particles (PM0.5-1.0 and PM1.0-2.5) during the pre-monsoon season due to meteorological conditions during this time of the year. During the pre-monsoon season, the stable atmospheric conditions may intensify atmospheric oxidation, and the low temperatures could increase the condensation of volatile secondary organic component (VOC) (Hallquist et al., 2009).

As shown in Figure 8, the highest SOC in the coarser particle (PM2.5-10) came from the monsoon season, followed by pre-monsoon and dry, respectively (42%, 30%, and 20%). On the other hand, the PM>10, the highest SOC came from dry season followed by pre-monsoon and monsoon, respectively (55%, 3,1% and 28%) (Figure 9). The higher SOC/OC is indicative of the important contribution of secondary formation of particles thin the southern Thailand atmosphere. Accordingly, this issue will be the subject of a future study related to the role of increased levels of polluted air.

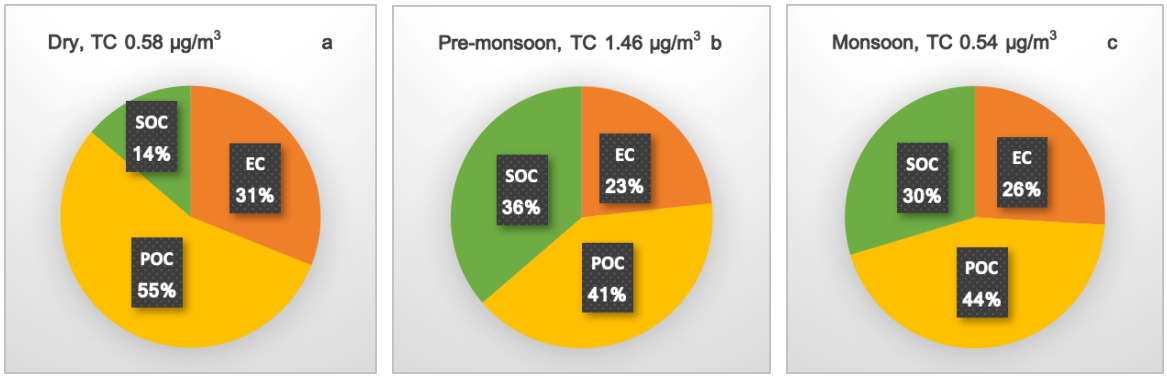


Figure 5 Carbon distribution of each species for three seasons in PM0.1
EC: elemental carbon POC: primary organic carbon SOC: secondary organic carbon

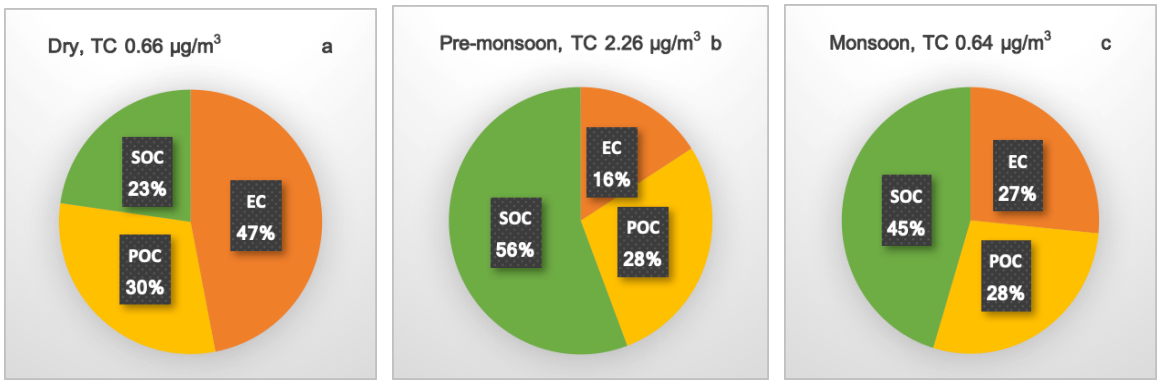


Figure 6 Carbon distribution of each species for three seasons in PM0.5-1.0

EC: elemental carbon POC: primary organic carbon SOC: secondary organic carbon

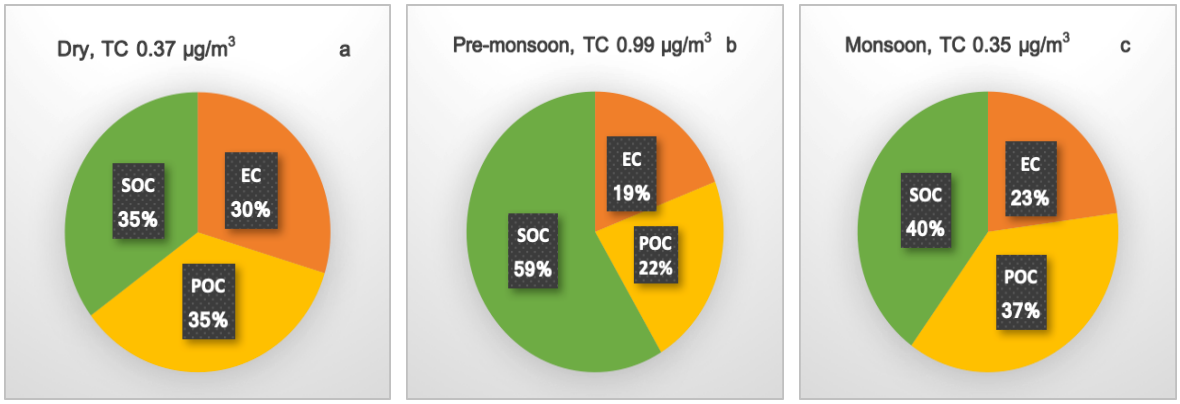


Figure 7 Carbon distribution of each species for three seasons in PM1.0-2.5

EC: elemental carbon POC: primary organic carbon SOC: secondary organic carbon

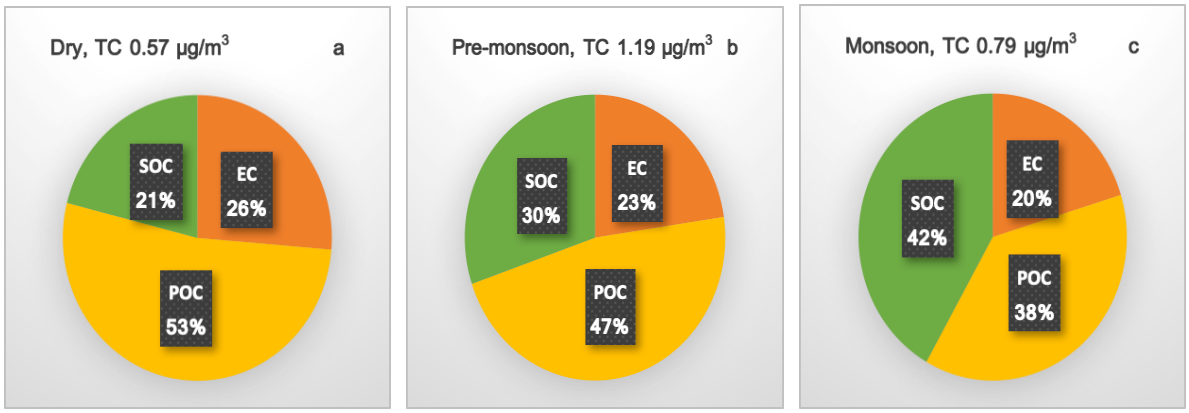


Figure 8 Carbon distribution of each species for three seasons in PM2.5-10

EC: elemental carbon POC: primary organic carbon SOC: secondary organic carbon

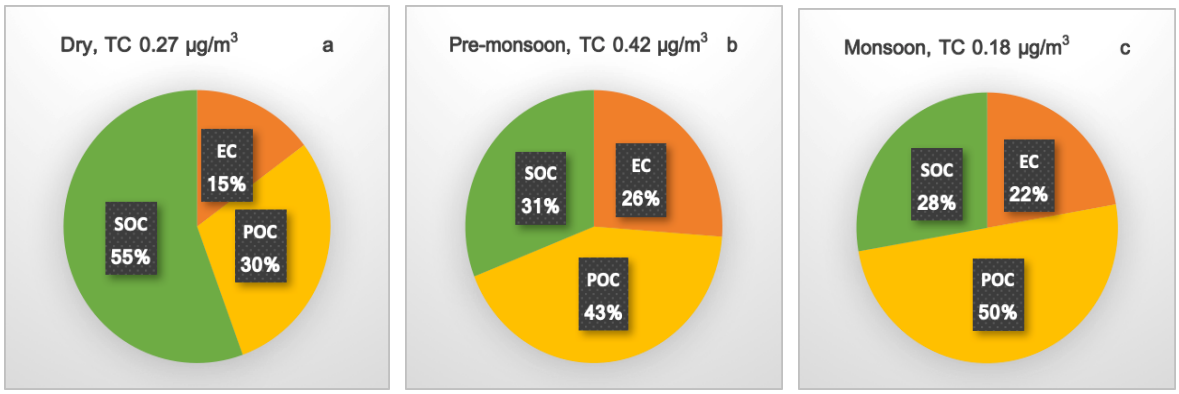
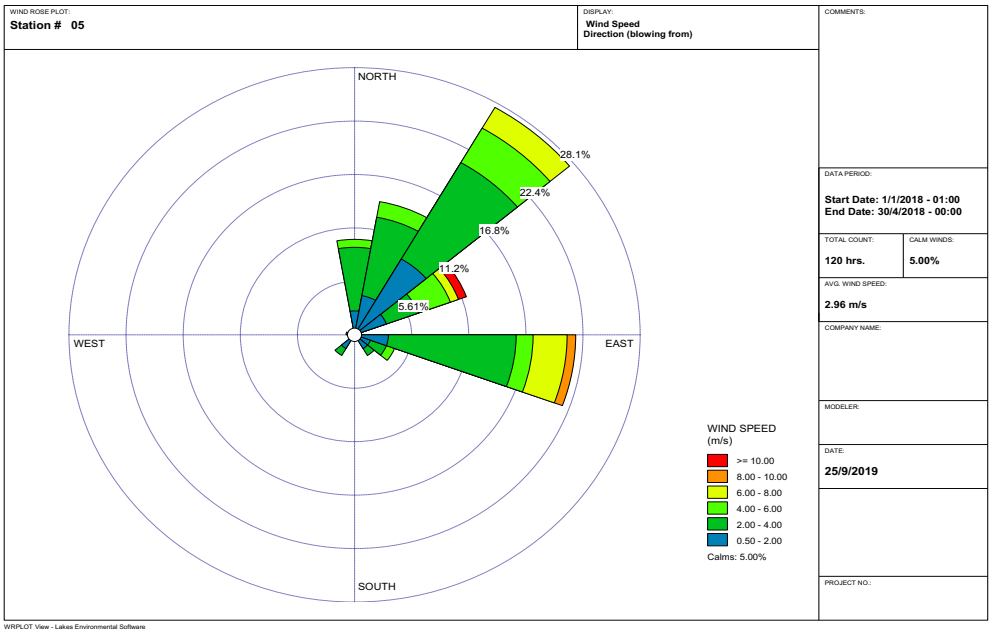


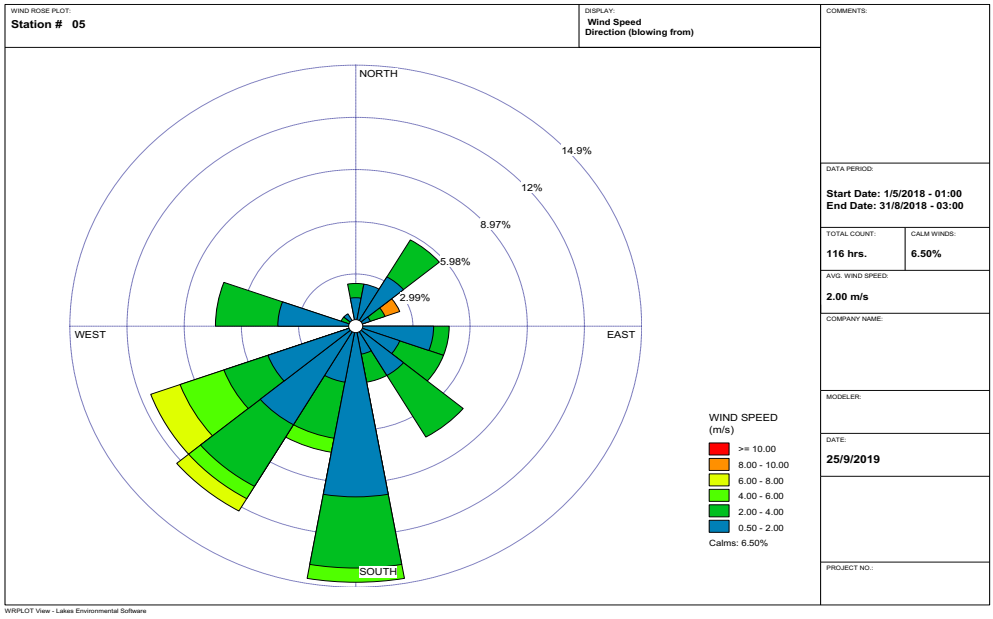
Figure 9 Carbon distribution of each species for three seasons in PM>10
EC: elemental carbon POC: primary organic carbon SOC: secondary organic carbon

5.5 Possible local and long-range transport of carbon materials

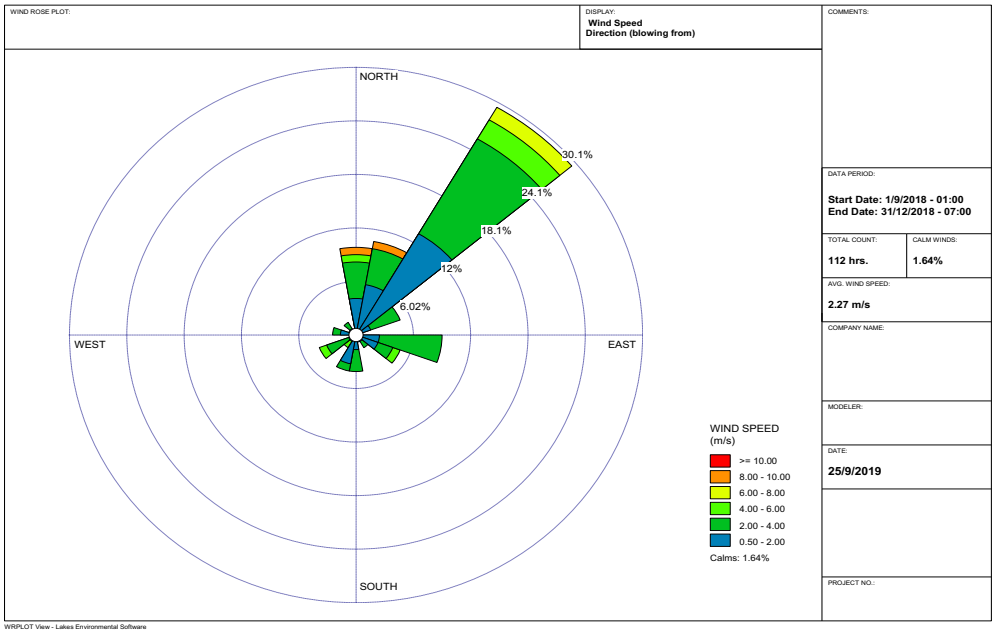
Figure 10 provides information on the wind speed and wind direction at the monitoring site in Hat Yai city, southern Thailand. The difference in the pattern among the three seasons affected both the mass and carbon composition in PMs. In the dry season, the prevailing wind direction was east and north-east (Figure 10a). On the other hand, in the pre-monsoon, the ubiquitous air direction was from the south and south-west direction (Figure 10b). In the monsoon season, most of the wind was from a north-easterly direction (Figure 10c) and was under the influence of the north-east monsoon, in which clean air masses from the Pacific Ocean brought a low OC and EC level to Hat Yai city in Thailand.



(a) Dry



(b) Pre-monsoon



(c) Monsoon

Figure 10 Differences in wind speed and wind direction in the three seasons (a) Dry, (b) Pre-monsoon and (c) Monsoon

Particle-bound carbon, with a lifetime of around one week, can migrate for long distances in the atmosphere (Cape et al., 2012). In this study, high levels of OC and EC were observed on a few sampling days during the pre-monsoon season (16-18 August 2018) for PM size distribution. These high concentrations could be either contributed from local sources and from transboundary over countries through the long-range transport of PM pollutants.

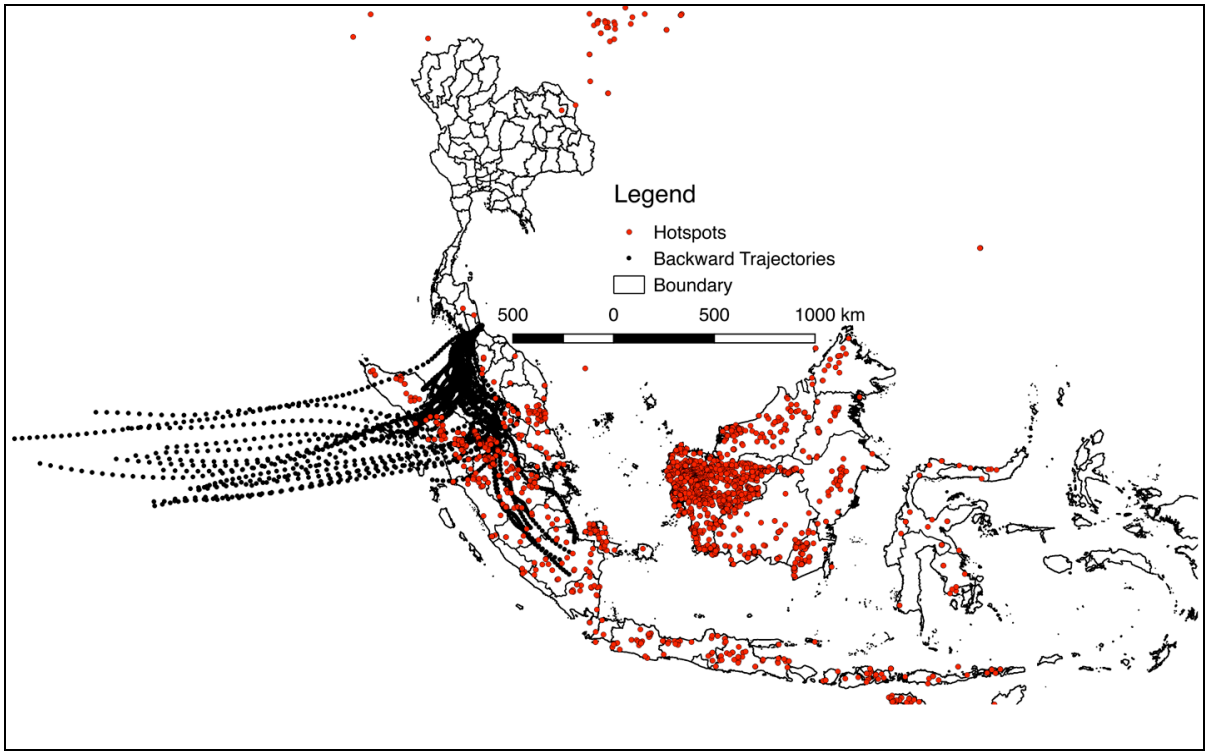


Figure 11 Distribution of hotspots and air mass trajectories during the high episode (14-16 August 2018)

The HYSPLIT-4 model was used to investigate the potential long-range transport of particle-bound OC and EC arriving at the sampling site. To examine the contribution of the long-range transport of OC and EC, high levels were considered for this analysis. There were 72-hrs (14-16, August 2018) during the pre-monsoon season. Figure 11. shows the backward trajectories during high carbon material levels for the dates mentioned above. As shown in Fig. 5., all of the 72-hr air mass originated from the central part of Sumatra island in Indonesia, where peat-land fires have been reported by several researchers (Heil and Golddammer, 2001; Vadrevu et al., 2014; Tham et al., 2019). An air mass that moves from southeast Thailand, which contributes to the high particle-bound carbon levels, was observed at the study site.

6. CONCLUSIONS

In this study, size-fractionated aerosol samples were collected in Hat Yai in southern Thailand during 2018. The particle-bound OC and EC, as well as char-EC and soot-EC, were investigated for one year (January - December 2018). The status and possible emission source of the ambient base on carbon composition were evaluated. The highest concentrations of PMs in the Hat Yai ambient air occurred in the pre-monsoon season, with the corresponding values in the dry and monsoon seasons being lower. The OC derives from primary emission sources and the formation of SOC by photochemical activities in the atmosphere. The concentrations of OC were higher than the concentrations of EC in all seasons. The char/soot ratio was consistently less than 1 for PM0.1, indicating that transportation as local emission sources

appear to be the main emission sources of PM0.1 in Hat Yai, Songkhla province, southern Thailand. High OC/EC ratios might be attributed to OC-rich sources of emission, (i.e., biomass burning) or to long-range transportation and secondary organic aerosol formation. These findings emphasize the importance of focusing emission control strategies on different particle sizes in southern Thailand, as well as elsewhere. However, transboundary migration over countries in this area could also be a key that merits future study in a more detailed investigation.

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7. Output and Appendix

7.1 International Journal Publication

- Phairuang, W., Inerb, M., Furuuchi, M., Hata, M., Tekasakul, S., & Tekasakul, P. (2020). Size-fractionated carbonaceous aerosols down to PM0.1 in southern Thailand: Local and long-range transport effects. *Environmental Pollution*, *260*, 114031.
- Phairuang, W., Suwattiga, P., Chetiyanukornkul, T., Hongtieab, S., Limpaseni, W., Ikemori, F., Hata, M. & Furuuchi, M. (2019). The influence of the open burning of agricultural biomass and forest fires in Thailand on the carbonaceous components in size-fractionated particles. *Environmental Pollution*, *247*, 238-247.

7.2 International conference

- Worradorn Phairuang, Perapong Tekasakul, Surajit Tekasakul, Mitsuhiko Hata and Masami Furuuchi. (2019) Characteristics of particulate matter down to PM0.1 in Southern Thailand. 11th Asian Aerosol Conference (AAC) in Hong Kong SAR, China, 27-30 May 2019.
- Worradorn Phairuang, Surapa Hongtieab; Mitsuhiko Hata; Atsushi Matsuki; Masami Furuuchi; Kazuhiko Sekiguchi, et al. (2018). Ambient Nano-Aerosol in East Asian Cities based on East Asia Nanoparticle Monitoring Network (EA-Nanonet). 22nd ETH Conference on Combustion Generated Nanoparticles. ETH-Zurich, Zurich, Switzerland, 18-21 June 2018.

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Size-fractionated carbonaceous aerosols down to $PM_{0.1}$ in southern Thailand: Local and long-range transport effects☆

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ABSTRACT

In this study, size-fractionated particulate matters (PM) down to ultrafine ($PM_{0.1}$) particles were collected using a cascade air sampler with a $PM_{0.1}$ stage, in Hat Yai city, Songkhla province, southern Thailand during the year 2018. The particle-bound carbonaceous aerosols (CA) as elemental carbon (EC) and organic carbon (OC) were quantified with the thermal/optical reflectance method following the IMPROVE_TOR protocol. The concentrations of different temperature carbon fractions (OC1-OC4, EC1-EC3 and PyO) in the size-fractionated PM were evaluated to discern OC and EC correlations as well as those between char-EC and soot-EC. The results showed that biomass burning, motor vehicle, and secondary organic aerosols (SOC) all contributed to the size-fractionated PM. The OC/EC ratios ranged from 2.90 to 4.30 over the year, with the ratios of $PM_{2.5\text{-}10}$ being the highest, except during the open biomass burning period. The concentration of CA was found to increase during the pre-monsoon season and had its peak value in the $PM_{0.5\text{-}1.0}$ fraction. The long-range transport of PMs from Indonesia, southwest of Thailand toward southern Thailand became more obvious during the pre-monsoon season. Transported plumes from biomass burning in Indonesia may increase the concentration of OC and EC both in the fine ($PM_{0.5\text{-}1.0}$ and $PM_{1.0\text{-}2.5}$) and coarse ($PM_{2.5\text{-}10}$ and $PM_{>10}$) fractions. The OC fraction in $PM_{0.1}$ was also shown to be significantly affected by the transported plumes during the pre-monsoon season. Good OC and EC correlations (R^2 = 0.824–0.915) in the fine particle fractions indicated that they had common sources such as fossil fuel combustion. However, the lower and moderate correlations (R^2 = 0.093–0.678) among the coarser particles suggesting that they have a more complex pattern of emission sources during the dry and monsoon seasons. This indicates the importance of focusing emission control strategies on different PM particle sizes in southern Thailand.

1. Introduction

Carbonaceous material is one of the major components of atmospheric air-borne particles and occupies around 20–50% of particle mass concentration (Gu et al., 2010; Contini et al., 2018). The particulate total carbon (TC) is divided into two main fractions, organic carbon (OC) and elemental carbon (EC), the latter of which is loosely equivalent to black carbon (BC) and soot, and BC or EC are used interchangeably depending on the method of analysis (Lavanchy et al., 1999; Watson et al., 2005). The atmospheric photochemical process of producing secondary organic aerosols (SOA) involves primary pollutants, including SO_2, NO_x, and carbonaceous aerosols, creating a complicated effect in the atmosphere (Hallquist et al., 2009; Irei et al., 2016). Epidemiological studies have linked these pollutants to respiratory and cardiovascular health impacts, and it has been estimated that one hundred of every ten thousand premature deaths are caused by BC each year

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0269-7491/

(Badarinath et al., 2007; Fullerton et al., 2008; WHO, 2012).

The principal source of particle-bound carbonaceous aerosols is fossil-fuel and biomass combustion. In a global-scale emission inventory (EI), the highest emissions of OC and EC result from open biomass burning (savannah fires, crop residue burning and forest fires), followed by indoor solid-biomass as well as fossil-fuel combustion (Bond et al., 2004, 2013). The OC/EC ratio can identify the sources of carbonaceous material in the atmosphere (Briggs and Long, 2016; Kumar et al., 2018). Coal combustion, domestic cooking, biomass burning and the combustion of gasoline and diesel in vehicle engines cause differences in the ratio of OC/EC. A higher ratio of OC to EC results from biomass combustion, whereas petroleum combustion causes a lower OC/EC ratio (Pio et al., 2011; Kumar and Attri, 2016). Nevertheless, the ratio of OC/EC is dependent on three factors, the emission sources, deposition rate, and SOA (Guo, 2016; Khan et al., 2016). Further, EC can be divided into char-EC and soot-EC. Char is mainly generated in low combustion processes and contains the primary source material. Conversely, soot is mainly derived from the hot-gas temperature of hydrocarbon condensation (Han et al., 2007; Kim et al., 2011). The char/soot ratio differs according to the primary emission source and can also be used to identify the original source (Han et al., 2010).

The particulate matter (PM) contents observed in Thailand and South-East Asian countries are presently based on the levels of $PM_{2.5}$ and PM_{10} as well as, to a lesser extent, the measurement of PM_1 based on ground monitoring and satellite remote sensing (Ee-Ling et al., 2015; Tsay et al., 2016; Phairuang et al., 2017; Vejpongsa et al., 2017). However, only a small number of previous reports have described and discussed the monthly- and seasonally-based carbonaceous aerosol patterns in Thailand (Duangkaew et al., 2013; Pongpiachan et al., 2014b; Phairuang et al., 2019a). Additionally, there have been few studies of carbon components based on size-fractionated PMs in Thailand and neighboring countries, particularly, down to the ultrafine size range (<100 nm) of ambient particles. Phairuang et al. (2019a), reported that open biomass burning in central and northern Thailand dominated the carbon matter in the ambient air. In particular, the $PM_{0.1}$ content, which is largely derived from motor vehicle emissions, was shown to be at a level which strongly agreed with open burning in the upper part of Thailand being the major factor in air quality.

The study reported was conducted through the East-Asia Nanoparticle Monitoring Network (EA-NanoNet), which was established to observe the sources and the physical, chemical and optical characteristics of nanoparticles ($PM_{0.1}$) in the East and South-East Asian atmosphere i.e., Japan, Taiwan, Cambodia, Vietnam, Malaysia and Thailand (Hata et al., 2015). The main motivation of this study was to investigate the monthly and seasonal variation of carbonaceous aerosols including EC and OC in size-fractionated PM down to $PM_{0.1}$ in the ambient air collected in Songkhla province, southern Thailand. Further, the Hybrid Single-Particle Lagrangian Integrated Trajectory Model, version 4 (HYSPLIT4) was used to investigate the possible migration of air-borne pollutants associated with high carbon concentrations in southern Thailand. The data from this study will provide knowledge of the temporal variation in the size distribution of carbon components, which are a critical factor in global warming and affect human health, in the ambient air of an urban area in a developing country.

2. Materials and methodology

2.1. Study area

The sampling site was located in Hat Yai, Songkhla province on the top (eighth) floor of the Sirindhorn Applied Research Engineering Building, Faculty of Engineering, Prince of Songkla University (PSU; 7°00′21.8″ N, 100°30′08.6″ E). The high elevation of the sampler allowed for a better mixing of Hat Yai background atmosphere because the sampler was some distance from main street-level emission sources. The height is also relevant to occupants of high-rise building in the area in terms of small-particle exposure. PSU is located around 4 km from the busy city center, in an area where traffic is less severe and there are fewer households. The main activities which contribute to PM in Hat Yai are domestic cooking, fuel combustion by factories, road transportation and building construction. Hat Yai is the largest city in southern Thailand with an area of 852.79 km^2 and a population of approximately 0.5 million. The main industries of Hat Yai relate to agricultural products including para rubber, and marine products. In common with other cities in Thailand and South-East Asian countries, Hat Yai suffers from air pollution associated with rapid regional economic development. According to the EI for Songkhla province conducted in 2009, the total suspended particles (TSP) of emissions were mainly from factories (85%) followed by motor vehicles (10%) with others (households, ships and other modes of transportation, 5%) also contributing (Pollution Control Department (PCD), 2009). The sampling site is shown in Fig. 1. Moreover, a study of the carbon components in PM_{10} by Pongpiachan et al. (2014a) suggested that local activities such as night-time tourism activities affect the air-borne carbonaceous content at ground level. However, the main impacts on carbon matter in PM_{10} in Hat Yai are from biomass combustion, crop residue burning and maritime aerosols. The weather parameters during the monitoring period, consisting of temperature, rainfall, relative humidity, wind speed and direction, are detailed in Supplementary Table S1. The climate in the southern part of Thailand may be divided into three seasons which depend on the monsoon, the dry season (January–April), the pre-monsoon season (May–August) and the monsoon season (September–December) (Supplementary Table S2). Because of its geographical location in proximity to the ocean, Hat Yai experiences a tropical climate, characterized by relatively uniform high temperatures (24.7–32.5 °C), relative humidity (65–84%), rainfall (2096 mm) and low monthly average wind speeds (1.4–3.5 m/s) throughout the year. Further, as reported by the Thailand Meteorological Department (TMD, 2015), there have been frequent tropical cyclones passing across Thailand during the 65 years between 1951 and 2015.

2.2. Cascade sampler for size-classification of particulate matter

The air sampling at the PSU site was conducted using a $PM_{0.1}$ cascade sampler with four impactor stages (>10, 10-2.5, 2.5–1.0, 1.0–0.5 μm), an inertial filter (IF) stage (0.5–0.1 μm) and a backup filter (<0.1 μm) located downstream of IF. The nano-sampler was operated with the air inlet at 40 l/min (Furuuchi et al., 2010). Each set of filter samples covered a period of 48 h and sets were collected, three times per week in second week of each month in the year 2018. Quartz fiber filters (QFF) (Pall, 2500 QAT-UP, diameter 55 mm) were used for all stages except the IF stage. The filters were preheated for 1 h (350 °C) to remove any possible contamination and were then kept at a controlled temperature of 21.5 ± 1.5 °C and a relative humidity of 35 ± 5% for 48 h before and after weighing in a weighing chamber (PWS-PM2.5, Tokyo Dylec Corp., Japan). Blank filters were prepared to confirm the influence of any contamination from the adsorption of gaseous carbon components during sampling and filter transportation. As reported by Kuwabara et al. (2016), nano-samplers of the type used in this study have around 30% gaseous OC which penetrates through the backup filter for $PM_{0.1}$ particles. However, the artifacts due to the

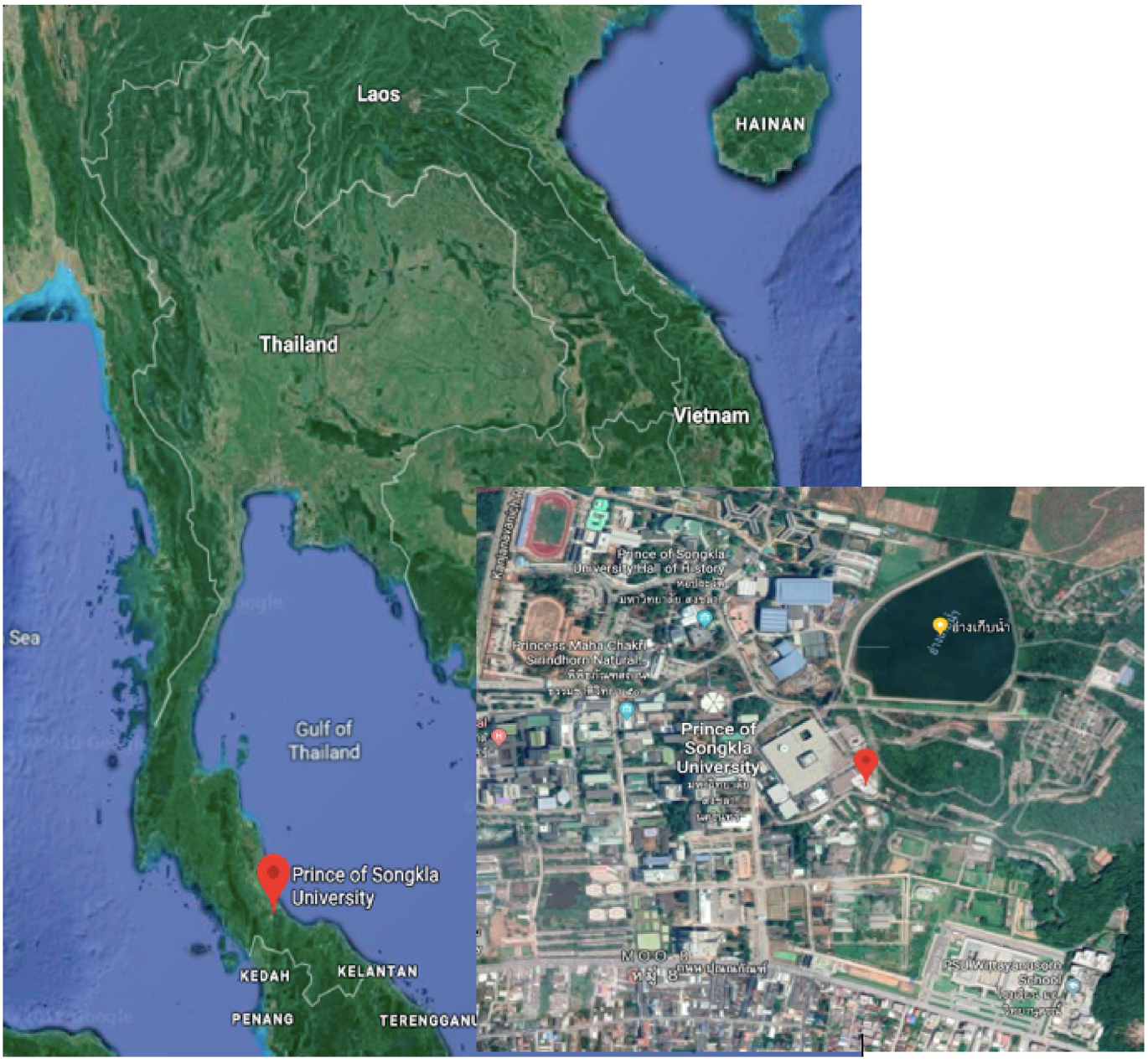

Fig. 1. Location of sampling site Prince of Songkla University (PSU), Hat Yai, Songkhla, southern Thailand.

evaporation of semi-volatiles should be much smaller than those of conventional types of nanoparticle samplers, e.g., low-pressure impactor and Nano-MOUDI (Hering et al., 1979; Furuuchi et al., 2010; MSP Cooperation, 2018). Other chemical substances in the nano-sampler were not evaluated although they could also be an important factor in air pollution which merits future study in a more detailed investigation into the further development of nano-sampler equipment.

2.3. Thermal/optical carbon analysis

The carbonaceous aerosols collected on the QFF were analyzed by estimating the CO_2 acidification from the organic samples on a 1.5 cm^2 section of the QFF that was punched out with a 1.5 cm × 1.0 cm rectangular cutter. The samples were analyzed with an EC-OC analyzer (Sunset Instruments Inc., Model 5L, USA) based on the IMPROVE (Interagency Monitoring of Protected Visual Environments) Thermal/Optical Reflectance (TOR) protocol (Chow et al., 2004; Watson et al., 2005), which has been widely used to classify carbonaceous material in the atmosphere (Han et al., 2007; Shi et al., 2016). The spot number of particle deposition on each punched sample for PM sizes, >10, 10-2.5, 2.5–1.0 and 1.0–0.5 μm was set at 1, 2, 4 and 8, respectively. The $PM_{0.1}$ samples were punched-out using the same cutter from a backup filter. As noted in a previous report (Phairuang et al., 2019a), the IMPROVE_TOR protocol yields equivalent OC/EC splits on filters. The different carbon analyses were confirmed as being within an error of less than 5% of TC and 10% of EC and OC. The distinctive particle size on the 1.5 cm^2 samples from each stage were analyzed using IMPROVE_TOR, without any preparation. Since as previously noted, the IMPROVE_TOR method cannot be applied to PM collected on an IF stage ($PM_{0.5-0.1}$), or from webbed stainless-steel fibers in a nozzle,

the IF samples were not subjected to carbon analysis with the exception of evaluating the TC on the travel blanks.

For the above analysis, four OC fractions (OC1, OC2, OC3, OC4) were analyzed in non-oxidizing Helium (He) at air temperatures of 120, 250, 450 and 550 °C, respectively. Additionally, the final fraction of the OC was evaluated as pyrolyzed organic carbon (PyC), which is carbon which has evolved in the oxidizing atmosphere in 2% O_2/98% He. Moreover, three EC fractions (EC1, EC2 and EC3) were measured in an oxidizing atmosphere at 550, 700 and 800 °C, respectively (Chow et al., 2004; Watson et al., 2005). In the following, OC and EC are defined respectively as OC1+OC2+OC3+OC4+PyC and EC1+EC2+EC3-PyC, in which EC can be divided into char-EC, defined as EC1-PyC, and soot-EC, defined as EC2+EC3 (Han et al., 2007, 2009). Further, char-EC, soot-EC and the char/soot ratio were used to identify the impact of fossil-fuel and biomass combustion along with OC, EC and the OC/EC ratio (Cao et al., 2005; Guo, 2015; Shi et al., 2016).

2.4. Hotspots and Backward trajectories

The geographic locations of active fires or open burning were detected by Moderate Resolution Imaging Spectroradiometer (MODIS) satellite remote sensing imagery (accessible from http://earthdata.nasa.gov/data/near-real-time-data/firms). The active fires were located based on a 1 km × 1 km area within each image. In order to investigate the impact of the transportation of carbonaceous aerosols by air-parcel movement from neighboring areas and their long-range transport, 72-h air mass back trajectories arriving at the monitoring site in Hat Yai, 500 m above the average ground level (AGL) were evaluated using a model based on Hybrid Single-Particle Lagrangian Integrated Trajectory, version 4 (HYSPLIT4) (ALR, 2019). The meteorological data was derived from the Global Data Assimilation System (GDAS), resolution 0.5°, in the NOAA website (https://ready.arl.noaa.gov/gdas1.php). To examine the magnitude of the effect of air-parcel movement HYSPLIT4 models were created at three different heights above ground level (30, 50, and 500 m). The results showed that the trajectory pathways calculated at the PSU site, for the same ending times at the three different levels were not significantly different (Supplementary Fig. S1). Accordingly, the low friction from the Earth's surface at low levels does not appear to significantly affect the resulting backward trajectory pathways in Hat Yai city. Therefore, the data collected at 500 m above the ground can be taken to represent the backward trajectory for the lower part of the planetary boundary layer in this area, and the final height of 500 m above ground level was adopted following previous reports on backward trajectories in Thailand (Pentamwa and Kim Oanh, 2008; Kim Oanh and Leelasakultum, 2011).

3. Results and discussion

3.1. Mass concentrations of size-fractionated aerosols

The average monthly mass concentrations of size-segregated PM aerosols in Hat Yai during all three seasons of 2018 are shown in Fig. 2. The annual average TSP concentrations did not exceed the annual average limits of the National Ambient Air Quality Standards (NAAQSs) in Thailand (100 μg/m^3). The monthly TSP concentrations in Hat Yai ranged from 52.61 to 104.68 μg/m^3. On the other hand, PM_{10} and $PM_{2.5}$ exceeded the NAAQSs of PM_{10} (50 μg/m^3) and $PM_{2.5}$ (25 μg/m^3). All of these values exceeded the values suggested by the World Health Organization (WHO), which are 60, 20, 10 μg/m^3, respectively. It is interesting to note that the highest concentrations of PM in Songkhla's ambient air occur in the pre-monsoon season with those in the dry and monsoon seasons being lower. The high concentration of particles in that period is mainly due to transboundary air pollutants from other countries (Pentamwa and Kin Oanh, 2008). Almost every year in June–August, the lower southern part of Thailand is affected by regional smoke haze in South-East Asia (Quah and Varkkey, 2013; Velasco and Rastan, 2015). The seasonal variations of concentration in size fractionated PM down to PM0.1 can be explained as being due to the combined impact of weather conditions. During the monsoon season in southern Thailand, which usually prevails from October to December, clean air masses coming from the ocean are able to dilute the ambient PM. Moreover, the seasonally high rainfall is able to dilute the PM concentration via the wet-deposition mechanism. Hence, the carbon material in PM in Hat

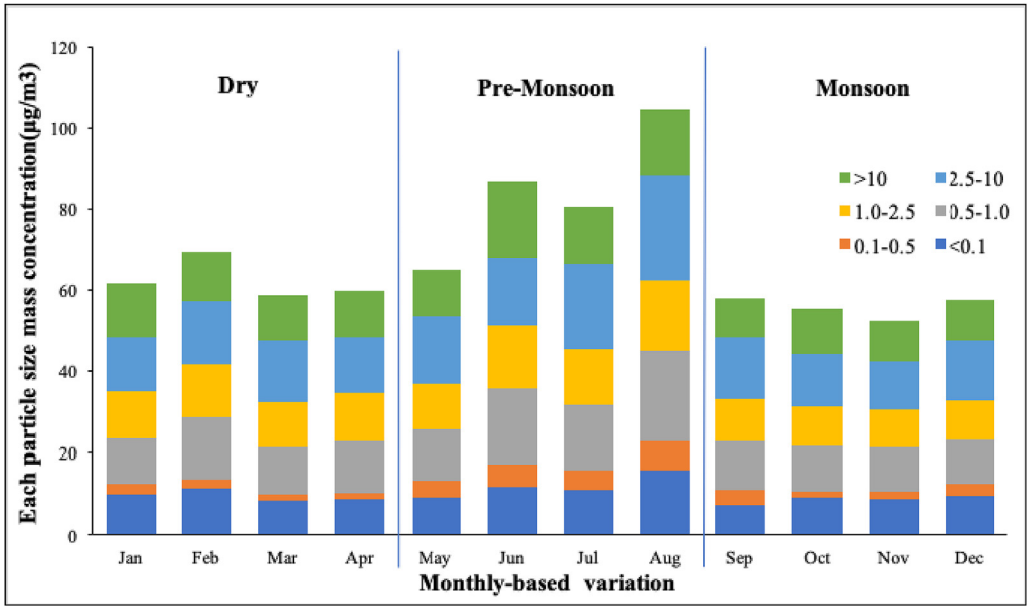

Fig. 2. Monthly and Seasonal variation of size-fractionated particles' mass concentration in Songkhla, southern Thailand.

Table 1
Annual average concentrations of OC, EC, Char-EC, Soot-EC, TC and mass (μg/m^3) in Songkhla.

	OC	EC	Char-EC	Soot-EC	TC	PM
$PM_{0.1}$	0.68 ± 0.60	0.23 ± 0.14	0.06 ± 0.04	0.17 ± 0.12	0.90 ± 0.82	10.17 ± 2.23
$PM_{0.5\text{-}1.0}$	0.93 ± 0.37	0.28 ± 0.22	0.14 ± 0.09	0.15 ± 0.15	1.22 ± 0.53	13.66 ± 3.57
$PM_{1.0\text{-}2.5}$	0.45 ± 0.40	0.12 ± 0.08	0.07 ± 0.05	0.05 ± 0.03	0.57 ± 0.45	12.02 ± 2.58
$PM_{2.5\text{-}10}$	0.62 ± 0.35	0.18 ± 0.12	0.11 ± 0.08	0.06 ± 0.04	0.79 ± 0.45	15.97 ± 3.80
$PM_{>10}$	0.18 ± 0.16	0.06 ± 0.06	0.03 ± 0.13	0.03 ± 0.03	0.23 ± 0.21	12.47 ± 2.79

Yai is very limited during the monsoon season. However, during the dry and pre-monsoon seasons there are higher PM concentrations, which can also be linked to particular weather conditions and high emission sources, i.e., low precipitation and long-range transport from distant areas.

3.2. Mass concentrations of OC, EC, Char-EC, Soot-EC, and TC

The annual average carbon species consisting of OC, EC, Char-EC, Soot-EC and TC concentrations from January to December, 2018 at the PSU sampling site are listed in Table 1. The highest OC and EC concentrations were found in the finer-size particle range, 0.5–1.0 μm ($PM_{0.5\text{-}1.0}$) with values of 0.93 ± 0.37 and 0.28 ± 0.22 μg/m^3, respectively. This finding agrees with those for size-fractionated particles from other sites further north in Thailand (Bangkok and Chiang Mai) (Phairuang et al., 2019a). In Songkhla, $PM_{0.1}$ nanoparticles as well as coarser sized particles ($PM_{2.5\text{-}10}$) were also dominant in the OC and EC concentrations. The average OC concentrations were 0.68 ± 0.60 and 0.62 ± 0.35 μg/m^3, respectively, while those for EC were 0.23 ± 0.14 and 0.18 ± 0.12 μg/m^3, respectively. The $PM_{0.5\text{-}1.0}$ particles formed the largest proportion of the TC at the PSU location. Moreover, the annual averages of char-EC as well as soot-EC concentrations were highest in the $PM_{0.5\text{-}1.0}$ range, with values of 0.14 ± 0.09 and 0.15 ± 0.15 μg/m^3, respectively.

3.3. Size distribution of OC and EC fractions

Based on the IMPROVE_TOR method, eight carbon fractions were isolated comprising OC1, OC2, OC3 OC4, PyC and EC1, EC2, EC3 (Chow et al., 2004, 2010). These subcomponents have different emission sources (Cao et al., 2005; Kim et al., 2011) and the eight carbon fractions were investigated in order to identify and quantify the sources of the carbonaceous aerosols (Han et al., 2010; Zhu et al., 2010). OC1 and EC1 represent the biomass burning contribution. OC2 might originate from secondary organic carbon (SOC) as well as the combustion of coal and the burning of biomass (Chow et al., 2004). OC3 is generally emitted from gasoline and LPG exhaust (Cao et al., 2006) while OC4 is derived from road dust. Further, PyC might be associated with the water-soluble organic carbon (WSOC) content, which represents incomplete combustion during the cooking of meat and biomass burning (Cao et al., 2005). In addition, EC1-PyC and OC3 are good indicators of biomass burning sources nearby (Chuang et al., 2013), and EC2 and EC3 are good indicators of hot-gas evaporation from motor vehicles.

Fig. 3 shows the eight carbon fractions in the size-fractionated particles as percentages across the three seasons in southern Thailand. The different emission sources of PM can be indicated by the OC and EC subcomponents. The OC is typically optically scattered while EC mainly consists of light absorbing particles. EC is therefore detected by optical-thermal analysis whereas PyO usually absorbs near visible light, which also represents the optical absorbing properties of the atmosphere.

In the $PM_{0.1}$ particles, the percentages of the eight fractions were ordered: OC3 > OC2 > EC1 > EC2 > OC4 > PyO > EC3 > OC1. The concentrations of OC3 and OC2 were higher than those of the others indicating that the most abundant sources were gasoline-burning vehicles and the burning of biomass, as well as SOA formation in the atmosphere (Cao et al., 2006; Lee et al., 2006). The EC1 concentration was also high especially during the pre-monsoon season, which indicates the contribution of nanoparticles from biomass combustion. This finding was similar to that of Tham et al. (2019), who collected $PM_{2.5}$ near a peat forest burning site at Jambi city in Sumatra, which displayed dominant OC3 in the $PM_{2.5}$ samples due to the fresh smoke derived from the peat-land-fire emissions. The OC1 and OC2 mostly consist of volatile organic

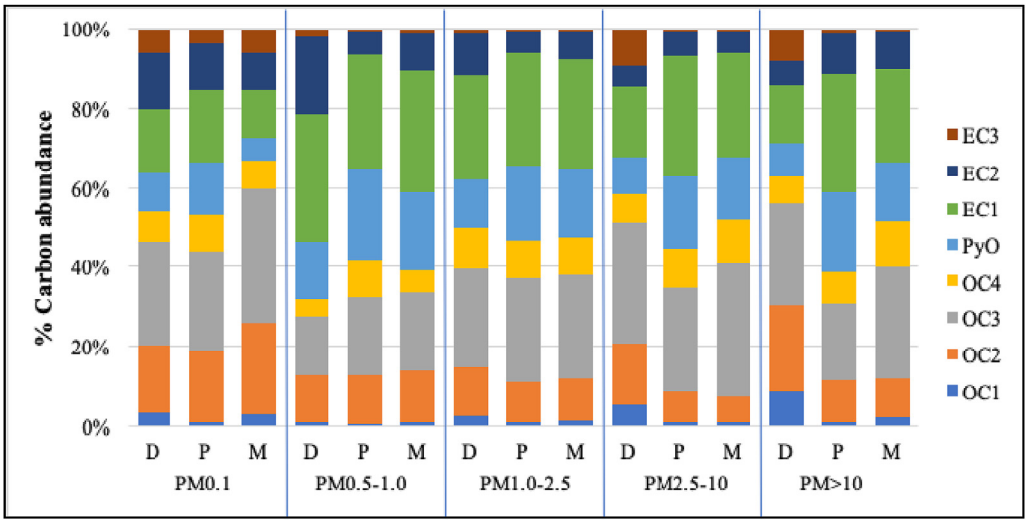

Fig. 3. The percentage of eight carbon fractions in size-fractionated particles.

substances that can quickly change during long-rang transport (Chuang et al. 2013), The level of EC2+EC3 (soot-EC) was high all year round and represents the abundance of carbon-emissions from motor vehicles.

The distribution of the fine particles was dissimilar from that of the ultrafine particles, and the order of the percentages was: EC1 > OC3 > PyO > OC2 > EC2 > OC4 > EC3 > OC1. The EC1 fraction was the highest carbon species in $PM_{0.5\text{-}1.0}$ and $PM_{1.0\text{-}2.5}$ (fine size-range) representing the contribution of biomass burning. Previous studies have also found that $PM_{0.5\text{-}1.0}$ from biomass combustion is dominant (Hata et al., 2014; Chomanee et al., 2018; Phairuang et al., 2019a). Nevertheless, OC3 and PyO were the second and third most abundant amongst the fine particles, which are most likely derived from incomplete combustion in meat cooking and biomass burning as well as from gasoline and LPG exhaust.

Among the coarse particles, the percentage of OC3 was highest in all three seasons and represents local sources from gasoline and LPG exhaust, However, in the pre-monsoon season the levels of EC1 and PyO were also high due to biomass burning and other sources of coarser carbon particles in the air. It is notable that the EC3 percentage provided the lowest EC contribution in each particle-size range and very little EC was detected at a hot gas high-temperature (800 °C) in any of the samples.

Table 2
Seasonal concentrations (μg/m^3) of carbon indices in $PM_{0.1}$ collected over Songkhla, southern Thailand.

Species	Dry	Pre-Monsoon	Monsoon
OC	0.44	1.12	0.42
EC	0.18	0.33	0.14
TC	0.62	1.45	0.56
OC/EC	2.44	3.29	3.00
EC/TC	0.29	0.22	0.25
POC [a]	0.32 (72%)	0.59 (53%)	0.24 (57%)
SOC [b]	0.08 (28%)	0.53 (47%)	0.16 (43%)
Char-EC [c]	0.05	0.08	0.04
Soot-EC [d]	0.14	0.26	0.10
Char-EC/Soot-EC	0.36	0.31	0.40

*Indicates the % contributions of POC and SOC to organic carbon (OC) in brackets.
[a] $POC = (OC/EC)_{min}$ x (EC).
[b] $SOC = (OC)_{tot} - POC$.
[c] Char-EC = EC1 - OP.
[d] Soot-EC = EC2 + EC.

3.4. Carbon characteristics of $PM_{0.1}$ particles

Table 2 displays the seasonal carbon concentrations in $PM_{0.1}$ collected in Hat Yai. The concentrations of OC and EC in the $PM_{0.1}$ samples indicate the seasonal changes in each species of carbon, with the pre-monsoon concentration of carbon being higher than those in the dry and monsoon seasons. The OC/EC ratios can be used to identify different emission sources such as fossil fuels, biomass burning and secondary formation (Pongpiachan et al., 2014a; Rajput et al., 2014). The EC proportion in the OC/EC ratios is elevated by biomass combustion and by secondary organic carbon (SOC) formation. In this study, the OC/EC ratio in $PM_{0.1}$ was found to vary between 2.44 and 3.29 (Table 2). In previous studies, the OC/EC ratios for motor exhaust, coal combustion, residential wood combustion, and forest fires have been found to be 1.4, 2.7, 4.3 and 15.7, respectively (Watson et al., 2001). Additionally, Lee et al. (2006), reported OC/EC ratios of around 0.6 in Hong Kong due to motor vehicle fuel combustion. Similarly, Zhu et al. (2010) reported that the OC/EC ratios in $PM_{0.1}$ and $PM_{2.5}$ were 0.68 and 1.26, respectively. Thumanu et al. (2009) found that the ratios of OC/EC in PM_{10} collected from sources in Songkhla province were 1.2–1.3 (due to transportation), 1.6–2.3 (due to industrial activity) and 3.9–4.2 (due to biomass burning). In this study, the higher OC/EC ratio suggests that the biomass combustion and industrial sector sources contribute much of the carbon material in the atmosphere above southern Thailand. Table 3 shows the annual average OC/EC and Char-EC/Soot-EC ratios in Thai cities reported in the literature. The OC is generally derived from primary emission sources and photochemical reaction processes. However, the breakdown between primary and secondary sources of OC cannot be obtained from carbon analysis. Lim and Turpin (2002) used the EC tracer method to calculate the primary organic carbon (POC) as follows:

$$POC = (OC/EC)_{min} \times (EC)$$

Table 3
Comparison of OC/EC, Char-EC/Soot-EC ratio in the present study with those reported in the literature in Thailand.

City	Period	Site Description	Method	Particle size	OC/EC	Char-EC/Soot-EC	Reference
Songkhla	January–December 2018	Suburban	IMPROVE_TOR	$PM_{0.1}$	2.91	0.36	This Study
				$PM_{0.5\text{-}1.0}$	3.06	1.15	
				$PM_{1.0\text{-}2.5}$	3.78	1.40	
				$PM_{2.5\text{-}10}$	4.30	2.02	
				$PM_{>10}$	3.80	1.09	
Chiang Mai	Dry season, March 2015	Suburban	IMPROVE_TOR	$PM_{0.1}$	3.49	2.73	Phairuang et al. (2019a)
				$PM_{0.5\text{-}1.0}$	3.42	9.07	
				$PM_{1.0\text{-}2.5}$	3.00	5.43	
				$PM_{2.5\text{-}10}$	2.41	3.82	
				$PM_{>10}$	2.90	2.54	
Chiang Mai	Dry season, February-April 2016	Suburban	IMPROVE_TOR	$PM_{2.5}$	3.6–11.0	/	Thepnuan et al. (2019)
Bangkok	Dry season, January 2015	Urban	IMPROVE_TOR	$PM_{0.1}$	3.37	1.07	Phairuang et al. (2019a)
				$PM_{0.5\text{-}1.0}$	5.87	4.58	
				$PM_{1.0\text{-}2.5}$	3.63	6.96	
				$PM_{2.5\text{-}10}$	2.37	2.44	
				$PM_{>10}$	3.22	0.84	
Bangkok	Dry Season, December 2011–February 2012	Urban	IMPROVE_TOR	$PM_{2.5}$	4.52	1.7	Duangkaew et al. (2013)
Bangkok	February-December, 2007	Urban, traffic	IMPROVE_TOR	PM_{10}	0.99	/	Pongpiachan et al. (2014b)
Songkhla	Pre-Monsoon, August 2007	Background	IMPROVE_TOR	PM_{10}	2.48–3.43	/	Thumanu et al. (2009)
	Pre-Monsoon, August 2007	Traffic			1.29–1.37		
	Pre-Monsoon, August 2007	Industry			1.68–2.33		
	Monsoon, November 2007	Biomass			3.93–4.22		

$$SOC = OC_{tot} - POC$$

The lowest OC/EC ratios were used to calculate the POC in different seasons. The amount of SOC is an important contributor to the OC amount throughout the year but as can be seen in Table 2. the percentage shows considerable seasonal variation. The lowest SOC contribution is during the dry season as compared to the pre-monsoon and monsoon seasons.

3.5. Correlations between levels of different carbon particles

The characteristic substances in EC do not change significantly in the ambient air. Accordingly, EC is a poor indicator of primary human source emissions, although the correlation between OC and EC has often been used as a means of estimating the primary OC (Guo, 2015). Moreover, the level of OC particles can be estimated based on the correlation between OC and EC. The OC/EC correlation provides some indication of the origin of carbonaceous aerosols. In many sampling locations, OC as well as EC show good correlations, which indicate that they have common sources, i.e., biomass or fossil-fuel burning (Zhang et al., 2011; He et al., 2015; Guo, 2016; Wang et al., 2017). Fig. 4 shows the OC and EC correlations in the size-fractionated particles. The atmospheric PMs smaller than 0.1 μm display high correlations in the dry and monsoon seasons ($R^2 = 0.915$ and 0.885, respectively) suggesting that they come from the same emission sources. However, the OC and EC correlation decreased during the pre-monsoon season ($R^2 = 0.632$), suggesting a more complex pattern of emission sources during this period. In contrast, the relationship between OC and EC among fine particles showed moderate correlations in all three seasons while for $PM_{0.5\text{-}1.0}$ particles, there was a good correlation in the dry season, suggesting that the OC and EC were from similar emission sources. However, the correlations in the monsoon and pre-monsoon seasons were only moderate suggesting an increased contribution from other sources. Moreover, the $PM_{1.0\text{-}2.5}$, $PM_{2.5\text{-}10}$ and $PM_{>10}$ particles showed lower correlations between OC and EC, suggesting that more complex emission sources are involved in those particle ranges.

The different fractions of PMs in Hat Yai were plotted to elucidate the relationship between EC and char-EC, together with EC and soot-EC. Generally, char is derived from biomass burning at lower temperatures and remains as the primary material with a diameter ranging from 1 to 100 μm (Masiello, 2004). However, soot is largely emitted at high temperatures in the hot-gas from motor vehicles and forest fires and is composed of submicron particles (Han et al., 2010). As presented in Fig. 5. the char-EC and soot-EC correlation with EC was strongest in the fine and coarse particle distributions ($R^2 = 0.931$–0.943), indicating that char dominated the total EC in the coarser range, whereas the finer particles ($PM_{0.1}$ and $PM_{0.5\text{-}1.0}$) showed lower correlations ($R^2 = 0.541$ and 0.617, respectively). However, soot-EC showed a high correspondence with EC in all size ranges ($R^2 = 0.770$–0.946). In general, soot is comprised of finer particles (<1 μm) which can continue to be suspended in the atmosphere for over a month and are thus more difficult to remove (Ogren and Charlson, 1983).

The char/soot ratio has often been used to identify primary sources. Chow et al. (2004) reported that a lower ratio (<1) indicating motor vehicle exhaust, a ratio of 1.31 indicated coal combustion and one of 22.6 indicated biomass combustion. In this study, the char/soot ratios varied according to the size of the particles. The nanoparticles showed a lower char/soot ratio (0.36), suggesting a greater contribution from motor vehicle exhaust. On the other hand, for larger particle sizes the char/soot ratio was higher than 1 and at its maximum for the $PM_{0.5\text{-}1.0}$ particles (2.02). The particle sizes in Hat Yai's atmosphere were therefore found to result from the mixing of many sources. Char reflects local sources whereas soot reveals regional sources, thus a lower char/soot ratio may indicate the influence of longer lasting carbonaceous aerosols. As recently reported by Tham et al. (2019), the influence of peat-fire smoke on urban $PM_{2.5}$ affects the PM carbonaceous profile, which tends to show more abundant soot-EC than char-EC during haze episodes originating in peat-burning (flaming mode) from Riau, Sumatra. The char to soot ratio from peat fires is lower than that originating in other more locally situated biomass burnings. In is interesting that as shown in the EI for Songkhla province, PM in this area is mainly derived from industrial processes, which account for up to 85% of TSP. The industries in Songkhla and the southern part of Thailand are mainly related to agricultural products or so called, agro-industries. Para rubber and oil palm trees are the main commercial crops grown in southern Thailand (OAE, 2016) and the residues from these plants are used as the main fuel in many agro-industry-related factories such as those producing palm oil and rubber products. The burning of wood as a fuel in factories releases sub-micron PM and PAHs, which are environmental pollutants in this area (Choosong et al., 2010; Chomanee et al., 2018). Moreover, para-rubber wood is used as a fuel, not only in the rubber-goods industry, but also in other factories in southern Thailand which use fuel-wood boilers. Pongpiachan et al. (2014a) suggested that biomass combustion, and possibly the burning of crop residues and maritime aerosols are the main source of carbon-related particles in PM_{10}. However, it is undeniable that the char and soot contribution to TSP in this area is mainly derived from wood combustion.

3.6. Air mass trajectories, hotspots and wind direction

The high concentrations of PM and their high carbon content could be either contributed by local sources or from distant sources due to the long-range transport of pollutants. The HYSPLIT4 model was used to determine the likelihood of long-range transport of OC and EC from distant sources at the monitoring site. In order to study the contribution of the long-range transport of carbon, a 3-day period (14–16 August) during the pre-monsoon season, during which there were high levels of OC and EC, was analyzed. Fig. 6 displays the 3-day backward trajectories during the period of high OC and EC concentrations on the above-mentioned dates. As shown in Fig. 6, all the 72-h backward trajectories during these days originated from Sumatra in Indonesia, where high intensity forest fires have been reported by several researchers (Heil and Goldammer, 2001; Heil et al., 2007; Vadrevu et al., 2014; Velasco and Rastan, 2015; Nuthammachot et al., 2019) between June and August, due to open burning and a stable atmospheric boundary layer. The air mass parcel then moved towards the northeast which contributed to the high carbonaceous aerosol levels observed at the PSU site. It is worth noting that the atmospheric lifetime of BC particles in ambient air is around 7 days (Cape et al., 2012), and the BC from the Sumatra forest fires would have been able to migrate long distances across other countries in the region. Therefore, it is most likely that during August 2018 the high OC and EC noted in all particle size ranges were a result of substantial emissions from forest fires on Sumatra.

On the other hand, the air mass direction during the monsoon season in November and December was from the opposite direction and represented a local source of clean ocean air. Supplementary Fig. S2 specifies information on the wind speed and wind direction at the monitoring site in Hat Yai city, southern Thailand. The difference in the pattern among the three seasons had an effect on both the mass and carbon composition in the PMs. In the dry season (January-April, 2018), the prevailing wind direction was east and north-east (Fig. S2a) whereas pre-monsoon (May-August, 2018),

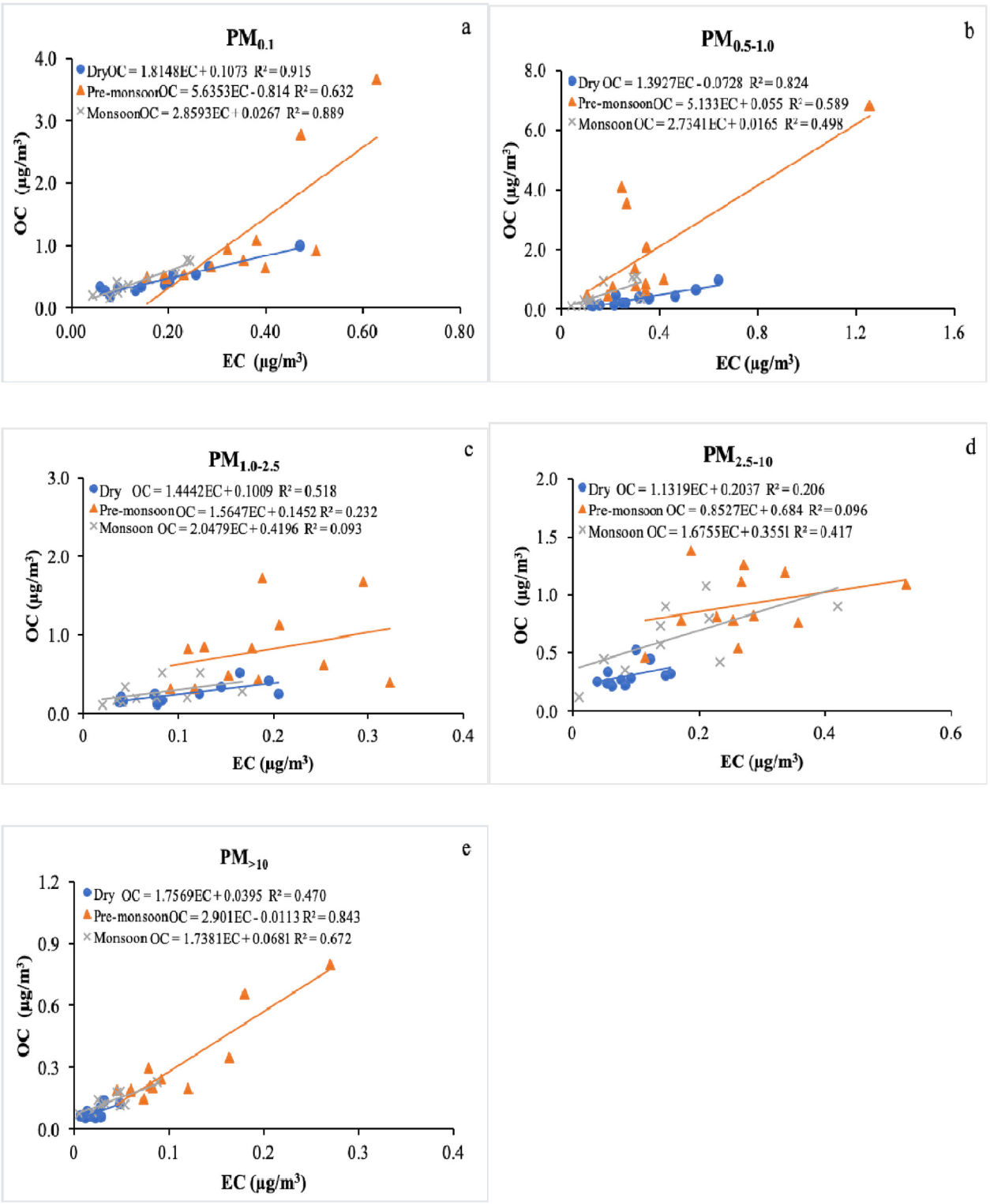

Fig. 4. Correlation between OC and EC in size-fractionated aerosol samples during different seasons at the PSU site.

the dominant air direction was from the south and south-west (Fig. S2b). In the monsoon season (September-October, 2018), most of the wind was from a north-easterly direction (Fig. S2c), and was under the influence of the north-east monsoon, in which clean air masses from the Pacific Ocean brought a low OC and EC fraction to Hat Yai city. The high OC and EC in $PM_{0.5\text{-}1.0}$, which represented the major carbon component in the PM, therefore came substantially from emissions from the combustion of wood as fuel in agro-industry related factories in the southern part of Thailand as well as in the wood-fired boilers of other types of factories. These boilers were an important source of carbonaceous aerosols during that period with their level tending to vary depending on the peak season of production of fuel-wood from the felling of para-rubber trees in the area (Tekasakul et al., 2008; Choosong et al., 2010;

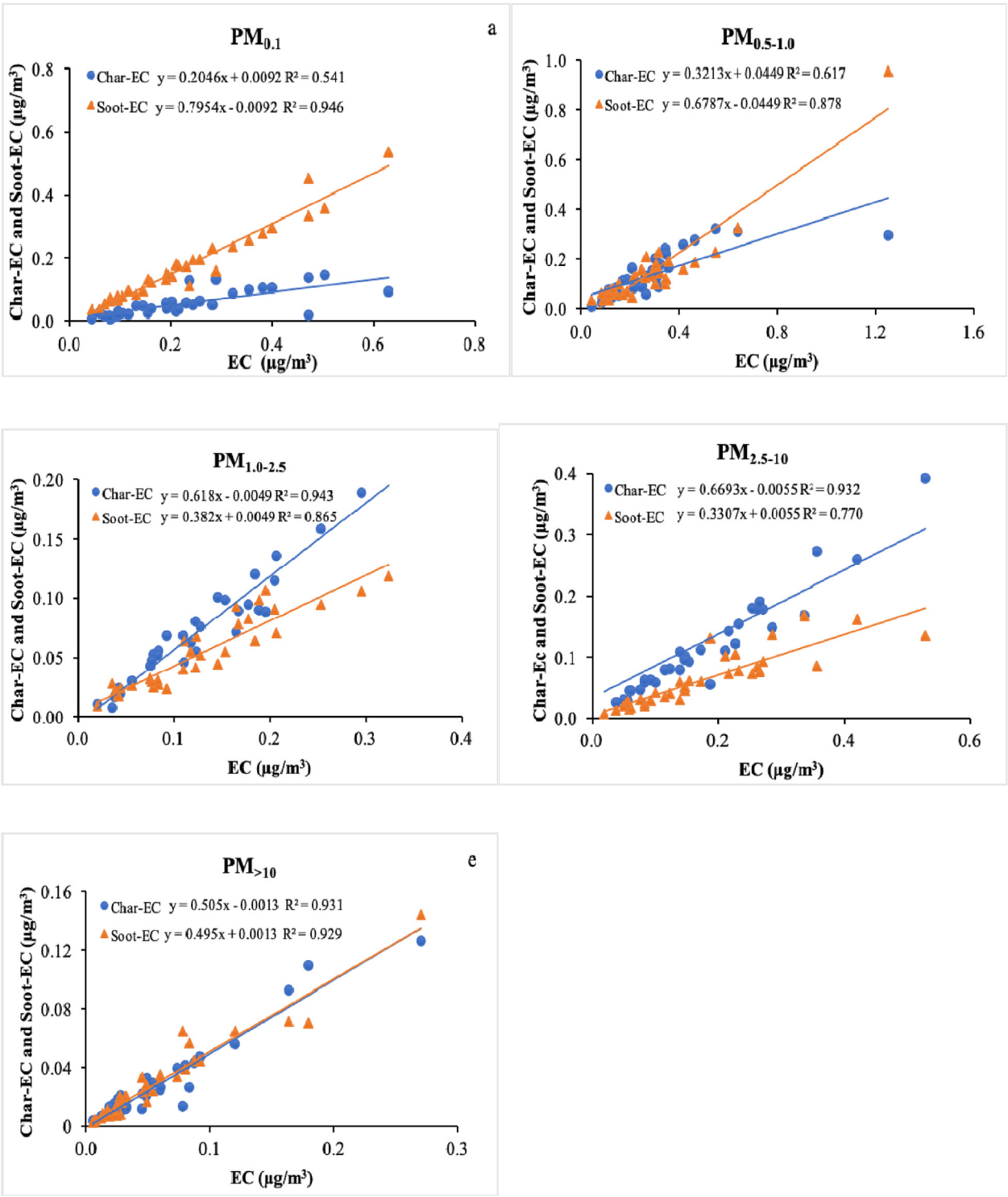

Fig. 5. Correlations between EC and Char-EC, and EC and Soot = EC at the PSU site.

Chomanee et al., 2018; Phairuang et al., 2019b).

4. Conclusion

This study was conducted to characterize the pollution from size-fractionated carbonaceous PM down to $PM_{0.1}$ in Hat Yai, Songkhla province, southern Thailand during three seasons in 2018. The carbon components among the PM were measured with an OC-EC thermal/optical carbon analyze.

(1) Monthly and seasonal TSP mass concentrations in Hat Yai ranged from 52.61 to 104.68 µg/m^3. The highest fraction of PM concentrations was $PM_{0.5\text{-}1.0}$. Moreover, the highest concentrations of PM in Songkhla's ambient air occurred in the pre-monsoon season with those in the dry and monsoon seasons being lower.

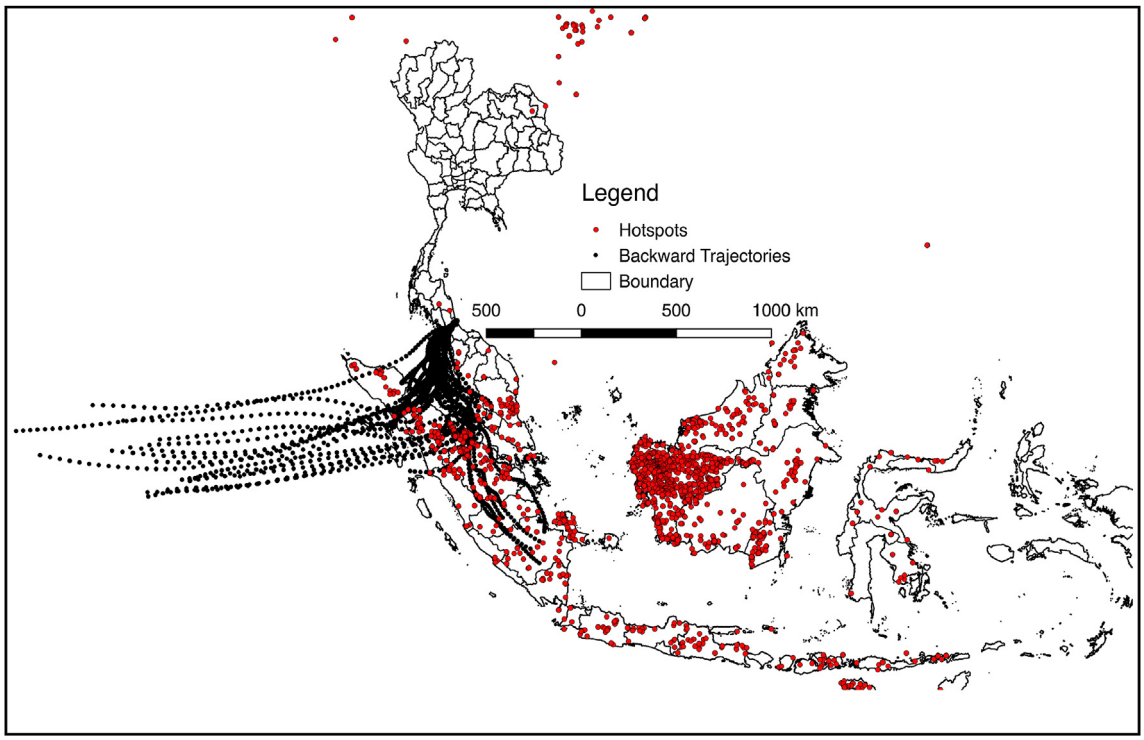

Fig. 6. Hotspots and Backward trajectories during the sampling period at PSU over high PMs episode, 14–16 August 2018.

(2) The concentrations of OC were higher than EC in every size range. High OC/EC ratios might be attributed to OC-rich source emissions (i.e. biomass burning) or to long-range transportation and secondary organic aerosol formation.
(3) Good correlations between the OC and EC levels in smaller particle size ranges indicated that they had common dominant sources, such as emissions from the combustion of fossil fuels, whereas the lower and moderate correlations in the coarser particle size range suggested a more complex pattern of emission sources, and this was especially notable in the dry and monsoon seasons.
(4) Transported plumes from Indonesian forest fires may have increased the OC and EC concentrations both in fine mode ($PM_{0.5\text{-}1.0}$ and $PM_{1.0\text{-}2.5}$) as well as in the coarse mode ($PM_{2.5\text{-}10}$ and $PM_{>10}$) fractions. The OC fraction in $PM_{0.1}$ was also shown to be significantly affected by the transported plumes during the pre-monsoon season.
(5) These findings emphasize the importance of focusing emission control strategies on different particle size ranges in the PM of the ambient air in southern Thailand. Moreover, based on the results of this study, the local and regional government departments with responsibility for maintaining air quality should make appropriate plans to control PM which contributes to the critical global warming problem and also has adverse impacts on public health.

Declaration of competing interest

The authors declare that they have no competing interests.

CRediT authorship contribution statement

Worradorn Phairuang: Conceptualization, Writing - original draft, Funding acquisition. **Muanfun Inerb:** Visualization. **Masami Furuuchi:** Conceptualization. **Mitsuhiko Hata:** Investigation. **Surajit Tekasakul:** Visualization. **Perapong Tekasakul:** Writing - review & editing.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2020.114031.

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The influence of the open burning of agricultural biomass and forest fires in Thailand on the carbonaceous components in size-fractionated particles☆

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ABSTRACT

Size-segregated ambient particles down to particles smaller than 0.1 μm ($PM_{0.1}$) were collected during the year 2014–2015 using cascade air samplers with a $PM_{0.1}$ stage, at two cities in Thailand, Bangkok and Chiang Mai. Their characteristics and seasonal behavior were evaluated based on the thermal/optical reflectance (IMPROVE_TOR) method. Diagnostic indices for their emission sources and the black carbon (BC) concentration were assessed using an aethalometer and related to the monthly emission inventory (EI) of particle-bound BC and organic carbon (OC) in order to investigate the contribution of agricultural activities and forest fires as well as agro-industries in Thailand. Monthly provincial EIs were evaluated based on the number of agricultural crops produced corresponding to field residue burning and the use of residues as fuel in agro-industries, and also on the number of hot spots from satellite images corresponding to the areas burned by forest fires. The ratio of char-EC/soot-EC describing the relative influence of biomass combustion to diesel emission was found to be in agreement with the EI of BC from biomass burning in the size range <1 μm. This was especially true for $PM_{0.1}$, which usually tends to be indicative of diesel exhaust particles, and was shown to be very sensitive to the EI of biomass burning. In Chiang Mai, the northern part of Thailand, the forest fires located upwind of the monitoring site were found to be the largest contributor while the carbon behavior at the site in Bangkok was better accounted for by the EI of provinces in central Thailand including Bangkok and its surrounding provinces, where the burning of crop residues and the cultivation of sugarcane for sugar production are significant factors. This suggests that the influence of transportation of polluted air masses is important on a multi-provincial scale (100–200 km) in Thailand.

1. Introduction

Carbon is one of the main components of atmospheric particulate matter (PM), which occupies up to 20–60% of fine particle concentration (He et al., 2004; Gu et al., 2010). Carbon species in PM include organic carbon (OC) as well as elemental or black carbon (EC or BC, the terms are often used interchangeably depended on the methodical technique (Chow et al., 2010)). BC consists of light-

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absorbing carbon components, while OC is comprised of the light-scattering carbon components (Venkatachari et al., 2006). OC is obtained from primary sources or chemical reactions relating to gaseous organic precursors, whereas EC is generated exclusively from primary incomplete combustion (Turpin and Huntzicker, 1995). The major source of carbon in aerosols is carbon-containing fuel combustion including the fossil and biomass. The largest source of EC emission is biomass burning. Bond et al. (2004) reported data on the global emission of OC and BC noting that the highest emissions were from open biomass burning followed by the indoor combustion of biomass and fossil energy. Open biomass burning (in forests, savannas, as well as of crop residues) accounted for 74% of total global OC emissions. Furthermore, high EC emissions of around 40% of the global total came from open biomass burning.

The carbon in PM plays a major role in global climate and human health (Jacobson, 2001; Pöschl, 2005). The health risks from OC, BC, and $PM_{2.5}$ are significantly associated with cardiovascular mortality and daily mortality (Klemm et al., 2004; Ostro et al., 2006). Carbonaceous aerosols have directly influenced the radiative forcing of the Earth's atmosphere, which has increased global warming in the last decade (Ramana et al., 2010). Additionally, carbonaceous particles indirectly act as cloud condensation nuclei (CCN), so influencing the albedo and lifetime of clouds (Leaitch et al., 2010).

Thailand and neighboring countries in Southeast Asia have agriculture-based economies and generate a large total of agricultural residues, which are burnt in the field and also burnt in agro-industry. Additionally, emissions from forest fires make a considerable contribution to the volume of carbonaceous aerosols in Thailand (Chaiyo et al., 2013; Chaiyo and Garivait, 2014). However, only a few publications have reported as well as reviewed the behavior of carbon component in aerosols in Thailand (Duangkaew et al., 2011; Pongpiachan et al., 2013; 2014). There has been a lack of studies of carbonaceous aerosols in size-segregated PM in Thailand, especially in urban and suburban areas. Moreover, the $PM_{0.1}$ ambient particle related to carbon components has not so far been studied in Thailand and neighboring countries.

The study reported in this paper was managed by the East Asia Nanoparticle Monitoring Network (EA-Nanonet). The EA-Nanonet was founded to monitor the source, characteristic as well as chemical composition of nanoparticle emissions along with the migration of PMs in many countries in East and South-East Asia including Japan, South Korea, China, Vietnam, Singapore, Cambadia Combodia and Thailand (Hata et al., 2015). In this report, the influence of biomass burning on air quality in Thailand was investigated, focusing particularly on particle-bound carbon. Size-segregated ambient particles down to $PM_{0.1}$ were collected throughout a period of one year in two cities in Thailand, Bangkok and Chiang Mai and the characteristics and seasonal behavior of the carbonaceous components and their diagnostic indices for emission sources were considered in relation to the monthly emission inventory (EI) of particle-bound OC and BC corresponding to agricultural activities and forest fires as well as agro-industries in Thailand. Following the procedure reported by Phairuang et al. (2017), monthly provincial EIs were evaluated based on the amount of agricultural crop production corresponding to pre-harvest burning, residue field burning and residue usage as a fuel in agro-industries, and also on the number of hot spots detected by satellite (Moderate Resolution Imaging Spectroradiometer; MODIS) on board a NASA satellite by which data relating to the area burned by forest fires was collected (Junpen et al., 2013). The EIs of BC and OC in provinces surrounding the monitoring sites were considered in relation to the concentration of carbonaceous components in the form of EC and OC evaluated by a thermal/optical carbon analysis as well as diagnostic indices such as char-EC to soot-EC, denoting the relative influence of biomass burning to diesel emission. In order to establish possible influences between provinces, the number of provinces selected was also considered taking into account similarities in agricultural activity between provinces and the possible migration of air pollutants, which were estimated by backward air mass trajectory analysis.

2. Methodology

2.1. Sampling locations

2.1.1. Bangkok

The sampling site in Bangkok was located on the roof top of a 10-story building, which belongs to the Faculty of Applied Science, King Mongkut's University of Technology North Bangkok (KMUTNB) (13.82° N, 100.51° E) (Fig. 1). The sampling site is located in the northern suburban area of Bangkok. There are heavily used major roads to the west, south, and east of the site although the area to the north of the site could be described as predominantly agricultural. Because of the elevation of the site (~40 m), a well-mixed atmosphere with minimal local influences could be expected. The surroundings of this site could be categorized as on the periphery of the Bangkok Metropolitan Region (BMR). As proxies for the mobile emissions and residential background in BMR, data from two air-quality monitoring (AQM) stations in BMR were used, one in a suburban area (Bang Kruai Nonthaburi (BKN), 13.82° N 100.49° E) and one in an urban area (Dindang Housing Authority (DHA), 13.46° N 100.32° E). Chuersuwan et al. (2008) reported particulate matter (PM) in BMR including $PM_{2.5}$ and PM_{10} in the years 2002–2003 and described that the main sources of $PM_{2.5}$ and PM_{10} were vehicles as well as biomass burning, where biomass burning contributed 6–41% to $PM_{2.5}$ and 28–36% to PM_{10} depending on the sites. Moreover, recent research on the source of $PM_{2.5}$ in BMR during the period 2015–2017 by Kim Oanh (2017), reported that during the dry period there were higher relative contributions from biomass burning (35% in Bangkok and 38% at the Asian Institute of Technology; AIT) compared to the wet season (24.6% in Bangkok and 24.9% at AIT).

2.1.2. Chiang Mai

The sampling site in Chiang Mai was located in an open corridor on the fifth floor of the Department of Biology, Faculty of Science, Chiang Mai University (CMU) (18.48° N, 98.57° E) and data from two AQM stations in Chiang Mai, one in a suburban location (CM1, 18.83° N 98.97° E) and one in an urban location (CM2, 18.78° N 98.99° E), were also collected. (Fig. 1). CMU is located in the outskirts of Chiang Mai municipality so that the sampling site is located around 5 km away from the city downtown, where traffic and biomass burning are not severe. According to the emission inventory for Chiang Mai municipality estimated by the Pollution Control Department (PCD), the total particulate emissions were mostly divided between local sources or forest fires (89%), the burning of solid waste (5.4%) and the burning of crop residues (2.3%) while longer-range sources accounted for only 2.5% (Kim Oanh and Leelasakultum, 2011).

2.2. Air sampling and carbon monitoring

2.2.1. Cascade sampler for nanoparticles

For the field sampling, cascade air samplers for nanoparticles developed by Furuuchi et al. (2010) were used both at the KMUTNB and CMU sites, both of which have been previously used as continuous monitoring sites for ambient nanoparticles in EA-Nanonet (Hata et al., 2015). The sampler used consisted of four impactor stages (including >10, 2.5–10, 1–2.5, 0.5–1 μm) as well as

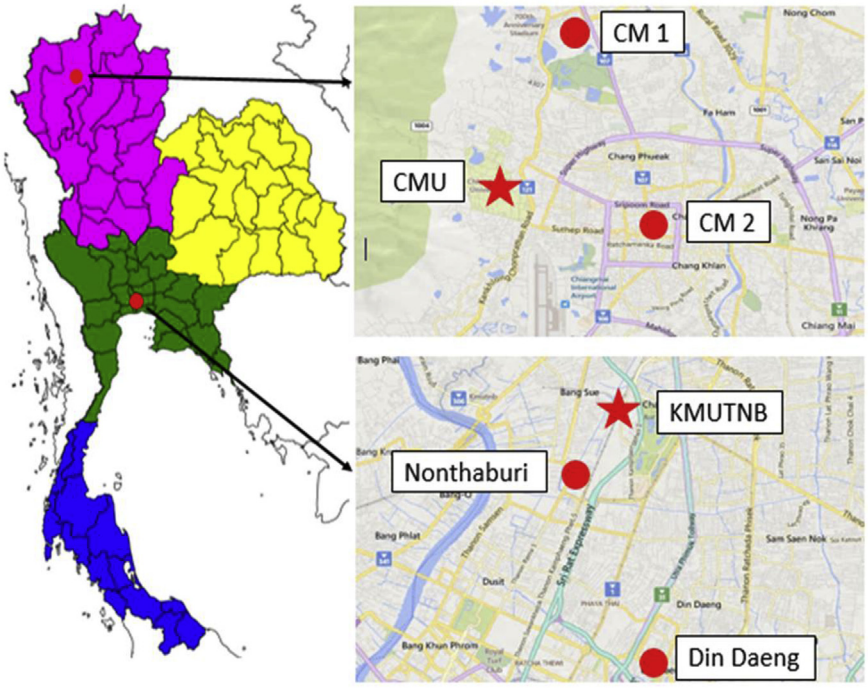


Fig. 1. Location of sampling sites at Chiang Mai and Bangkok and geographical area divided into 4 regions.

a spacial inertial technology filter (0.1–0.5 μm) along with a backup filter downstream of the inertial filter (<0.1 μm). This sampler was operated with 40 L/min of flow rate, which is large enough to obtain samples for the carbon analysis from 24 h of sampling at all the sites. The sampler has the advantage of a low pressure drop (5–10 kPa) for $PM_{0.1}$ separation. Artifacts due to the evaporation of semi-volatiles, therefore, should be much smaller than those of conventional samplers, for example the low-pressure impactor (LPI) and Nano-MOUDI which have pressure drops of 60–80 kPa (Hering et al., 1978; Hering et al., 1979; MSP Cooperation, 2018) although approximately 30% gaseous OC was reported to penetrate through a backup filter for $PM_{0.1}$ (Kuwabara et al., 2016).

Taking into account the difference in the particulate concentrations and the manpower available, at the KMUTNB site, particle samples were collected every 6 days (5 times/month) for 24 h while at the CMU site, samples were collected every 10 days (3 times/month) for 24 h. Quartz fibrous filters (QFF) (ϕ 55 mm, Pallflex, 2500QAT- UP) pre-baked for sampling to minimize possible contamination was used at high temperature 350 °C for 1 h. Since the webbed stainless steel (SUS) fibers in the inertial filter could have been damaged by baking, a SUS fiber mat was used without baking after confirming its background carbon level to be lower than the detection limit of carbons on a total carbon basis. The filters were kept at a controlled temperature (21.5 ± 1.5 °C) as well as relative humidity (35 ± 5% RH) in a weighing chamber for 48 h and the filter weight (readability to 1 μg) was measured using a microbalance installed in the weighing chamber before and after sampling (PWS-PM2.5, Tokyo Dylec Corp., Japan). After weighing, the filters were saved in a sealed polyethylene (PE) bag to avoid the adsorption of any chemicals from the outside air. The filter was also enclosed with aluminum foil to avoid any contamination by direct contact with the PE bag's surface, which cannot be neglected for some samples with small amouts of particles. Travel blank filters were prepared to check the adsorption of gas-phase carbons or any contamination during sampling and sample restoration-transportation. The used filters were kept at below −25 °C in a refrigerator until being analyzed at all times except during the mailing procedure.

2.2.2. Black carbon (BC) monitor

The BC concentration was monitored online using an aethalometer (AethLabs, microAeth®Model AE51), which evaluates the amount of BC in particles deposited on a glass fiber filter based on the transmitted light (880 nm) rate of change in absorption light, because of the continuous collection of aerosols. The aethalometer was installed at the same site in KMUTNB and operated for 24 h along with the cascade air sampler every 6 days.

2.3. Analysis of carbonaceous components

The carbon components in PM collected on the QFFs were investigated using a Sunset Laboratory Carbon Aerosol Analyzer, following the Interagency Monitoring of Protected Visual Environments_Thermal/Optical Reflectance (IMPROVE_TOR) protocol (Chow et al., 2004: Watson et al., 2005), in which the OC was described as the total of the OC fractions, including OC1, OC2, OC3 and OC4 at temperature 120, 250, 450 and 550 °C, respectively, in a non-oxidizing helium (He) air, while the EC was taken as the total of the EC fractions, including EC1, EC2, and EC3 at temperature 550, 700, 800 °C, respectively. in a 2% O_2 and 98% He air, but excluded the pyrolyzed carbon fraction (POC). Thus, the EC fraction refers to EC1+EC2+EC3-POC. EC can be divided into Char-EC and Soot-EC based on the auto-split time setting, where Char-EC is defined as EC1 minus POC as well as Soot-EC as EC2 plus EC3 (Chow et al., 2004; Han et al., 2007, 2010). As a reference for the analysis, sucrose ($C_{12}H_{22}O_{11}$) was used before each analysis to confirm the analysis reliablity based on the total carbon (TC). Han et al. (2010) studied that char-EC can be taken to indicate the influence of coal or biomass burning. The char-EC/soot-EC ratio is a portion of the effect of biomass burning (Shi et al., 2016). Soot-EC, char-EC and char-EC/soot-EC ratio were therefore evaluated and used as indices to indicate the influences of biomass burning and emissions from

traffic along with common carbon indices relating to OC, EC and the OC/EC ratio.

Filter samples for the carbon analysis were prepared by cutting 15 mm × 10 mm sections from the original circular filter using a rectangular cutter. The number of spots on each punched sample was set at 1, 2, 4 and 8 for >10, 10–2.5, 2.5–1 and 1–0.5 μm, respectively. $PM_{0.1}$ samples collected uniformly on a backup filter were punched-out using the same cutter. The mass concentrations of carbon for each size fraction were calculated from the carbon mass on a punched-out sample along with the total number of spots and the air sampling flow rate. As pointed out in previous reported (Jaffrezo et al., 2005), the TOR protocol correction yields equivalent OC/EC splits on filters. The concentrations of the different size-fractions on the 1.5 cm^2 samples from each stage of the total aerosol sampling were analyzed using TOR, without any preparation, from the impactor aerosol sampling utilized for the calculation of the atmospheric concentrations. The repeatability of the analysis was confirmed by analyzing samples punched-out from the same filter. The reliability of the analysis of the spot samples was also confirmed by comparing the soot-EC and char-EC with the EC of samples at different concentrations of each size fraction. The correlation factor, R^2 for linear regressions between those parameters are listed in Supplement Table S1. As described in the following section, there were very good correlations for each size-fraction both in Bangkok and Chiang Mai regardless of the month when sampling occurred. Travel blank filters were also analyzed following the same procedure described above. Since the IMPROVE_TOR protocol cannot be applied to particles collected on an inertial filter, or, webbed stainless steel fibers in a small nozzle, carbon analysis was not conducted for the inertial filter samples with the exception of testing for the total carbon on the travel blanks.

2.4. Carbon emission inventory

Monthly emission inventories of carbons emitted from biomass burning were estimated for each province in Thailand following a previously reported procedure, which calculates the amount of pollutants emitted based on the mass and emission factors of agricultural crop residues burned in fields, forest fires and biomass burning as a fuel in agro-industries (Phairuang et al., 2017). The total of a pollutant released from biomass burning can be estimated from the following equation (Giglio et al., 2006)

$$E = M \times EF \quad (1)$$

where E (g) refers to emitted mass of a pollutant from fuel-burning, M (kg) is the fuel consumption and EF (g $kg^{-1}_{dry\ mass}$) is the emission factor. The *EFs* related to the carbonaceous components, BC and OC shown in Table 1 were used in this study. Since, most data are country specific the values reported for Thailand in previous studies were used (Andreae and Merlet, 2001; Cao et al., 2008; Kanokkanjana, 2010; Kim Oanh et al., 2011; Chaiyo and Garivait, 2014) However, there have only been a few reports relating to *EFs* on EC, for instance on residential stoves in China (Li et al., 2009; Shen et al., 2010) while *EFs* on BC and OC are the only parameters available in Thailand at present, and both BC and OC are forms of solid carbon originate commonly in the fine portion of atmospheric particulate matters. To establish the relationships between the carbon inventory and the carbons monitored, e.g, EC, it is better to use *EFs* for EC since there is a difference between the methods used for measuring EC and BC. EC is usually measured by a thermal or a thermal/optical method whereas BC is measured by an optical method (Watson et al., 2005; Chen et al., 2006). However, bearing in mind the consistency between, e.g., EC and BC especially from the same emission sources (Chow et al., 2010; Janssen et al., 2012). EC and other categories of carbons were established by reference to the emission inventory for BC or OC.

Table 1
Emission Factors of carbon species (Unit: $g/kg_{dry\ mass}$).

Carbon	Rice	Corn	Cassava	Sugar cane	Soy bean	Potato	Forest
OC	2.99[a]	2.79[b]	3.30[c]	1.25[d]	3.30[c]	3.30[c]	5.20[c]
BC	0.77[b]	0.48[b]	0.60[f]	0.71[b]	0.60[f]	0.60[f]	0.77[e]

[a] Kim Oanh et al. (2011) Field burning of rice straw in Thailand.
[b] Kanokkanjana, 2010 Carbonaceous Aerosols from Agricultural Open Burning in Thailand.
[c] Andreae and Merlet (2001) Values are best guess for any combination of crop residue.
[d] Zhang et al. (2013) Biomass burning of agricultural residue in China.
[e] Chaiyo and Garivait (2014) Black carbon emissions from forest fire in Thailand.
[f] Streets et al., 2003 Data were likely obtained from various area sources.

2.4.1. Agricultural residue burning

The total of agricultural residues burned was calculated from Eq. (2) (Streets et al., 2003):

$$M = P \times N \times D \times \beta \times F \quad (2)$$

where M (g) is the amount of crop residue burned in the field, P (kg) is the total crop production, N is the residue to crop ratio, D is the burning efficiency, β is the fraction burned in the field and F is the crop-specific combustion efficiency ratio.

2.4.2. Forest fires

The carbon emission from forest fires was estimated by converting hot spot data to burned area using Eq. (3) (Shrestha et al., 2013):

$$M = A \times B \times C \quad (3)$$

where A (km^2) is the burned area, B ($kg_{biomass}\ km^{-2}$) is the biomass density of a specific forest type in Thailand ($3.76 \times 10^5\ kg_{biomass}\ km^{-2}$) and C is the combustion efficiency, or, 0.79 in typical forest areas in Thailand (Junpen et al., 2011).

2.4.3. Agro-industry

Sugarcane is one of the most significant crops in Thailand and contributes significantly to the bio-economy in 47 out of the total of 77 provinces, in most areas except the south of Thailand (Sriroth et al., 2016). In sugar factories, bagasse, which is the residue obtained after squeezing the juice out of the sugarcane, is used as the fuel for biomass boilers during the sugar period. The operating days for the boilers varies from 120 to 240 days at an average of 140 days. Because of the large amount of biomass fuel consumed in the sugar industry with some emission control, the control of PM emission is estimated as 60% (Phairuang et al., 2017). With regard to other agro-industries, rice mills are also important and rice husk is used as a fuel to generate heat and electricity in agro-processing (Prasara-A and Grant, 2011). However, rice mills in Thailand are mostly small-scale community-level factories located near to paddy fields (Ritthaisong et al., 2014), so that it is difficult to get reliable data for rice mills. The leading crops in the south of Thailand are para-rubber and oil palm and neither are subject to field burning so the emissions from open biomass burning directly related to agricultural activities are not significant. However, the residues of these crops are used as fuel not only in the rubber and palm oil industries, but also in various industries in this area, which use fuel-wood boilers. Since central and northern Thailand were the focus of this study and wood consumption data is not currently

available, the contribution from fuel-wood consumption is not discussed here but will be discussed in a future work.

The consumption of residues from sugarcane bagasse in sugar factories as a fuel can be estimated by the following equation (4):

$$E = W_{fuel/year} \times J/W_{fuel} \times W/J \quad (4)$$

where $W_{fuel/year}$ (Mg) refers the total bagasse consumed as a fuel according to the Office of the Cane and Sugar Board (OCSB, 2016). J/W_{fuel} (7600 kJ/kg) is the average lower heating value (LHV) of bagasse (Jenjariyakosoln et al., 2014). *W/J* is the EFs of the carbon component released from a boiler in a sugar factory according to the Global Atmospheric Pollution Forum manual version 5.0 (GAPF, 2012). The *EFs* depend on the production rate and the emission control facilities. Office of Natural Resource and Environment Policy and Planning (ONEP) reported the Environmental impact assessment (EIA) of the detail the existent emission control equipment used in sugar mills and indicate that all sugar mills use multicyclone to eliminate particulate matter (PM) before PM produced by bagasse burning into the atmosphere, with the PM removal efficiency being estimated at ~60% (ONEP, 2015).

2.5. Inventory to be related to monitored carbon data

The air pollutants monitored at the selected sites arose not only from local or adjacent sources but also from sources located at a distance from the main data collection sites, i.e., from surrounding provinces. Therefore, it is important to discuss to what extent the carbon EI relates to carbonaceous components arising in the immediate environs of the monitored sites as opposed to arising from sources in other provinces. Chuang et al. (2016) studied biomass burning in northern Thailand and noted that there is an aging process which freshly burned biomass aerosols will undergo involving coagulation and condensation, and this will convert their size distribution during 24 h of migration. Source areas should be within 100–200 km to contribute to the PM characterized, especially, nanoparticles or, $PM_{0.1}$. Moreover, the office of agricultural economics (OAE) has classified Thailand into four regions consisting of the north, northeast central and south according to their agricultural crops and food products. Northern Thailand is covered with forest areas in the upper north with paddy fields and sugarcane in the lower north. Northeastern and central Thailand are mainly covered with crops such as rice, sugarcane and cassava, while in the south, two perennial crops, para-rubber and oil palm are dominant (Supplement Fig. S1). For this study, the concentrations and indices of carbonaceous components evaluated at KMUTNB and CMU, respectively, were related to the EIs for the central and northern areas of Thailand. However, at a simpler level, the EIs of the BMR and Chiang Mai province could be considered as being defined by the measurements obtained from the monitoring of the data at the collection sites.

2.6. Air mass trajectory to monitoring location

In order to discuss the influence of the migration of air pollutants by air mass movement from surrounding provinces, the 24-hr backward trajectory of air masses arriving at the monitoring sites in Bangkok and Chiang Mai at 50 m from the average ground level (AGL)were analyzed using the HYbrid Single-Particle Lagrangian Integrated Trajectory Model version 4 (HYSPLIT4) (ALR, 2018). The meteorological data used for the HYSPLIT4 model were obtained from the Global Data Assimilation System (GDAS) resolution 1°, from the NOAA website (https://ready.arl.noaa.gov/gdas1.php). To investigate the magnitude of this effect attempts were made to run the HYSPLIT4 model at different ending heights from the ground (50 m) level to 1000 m above ground (50, 500 and 1000 m). The results show that the trajectory pathways calculated at Bangkok and Chiang Mai have ending times for the three different levels (50, 500 and 1000 m) which are not significantly different (Supplements Figs. S2a and S2b). The emissions from the huge area of crop burning and dense forest fires in the area would encounter only a little friction from the Earth's surface and the low level ending levels might not be significantly affected by any backward trajectory movement. Consequently, the ending level at the ground (50 m) can be used to represent the backward trajectory for the ground level of the global boundary layer.

3. Results and discussion

3.1. Emission inventory of the distribution of carbons in Thailand

The provincial distributions of the annual total emitted amount of OC and BC from biomass combustion in Thailand, estimated for the year 2014 based on the procedure described above are shown in Fig. 2. There were larger emissions in the north-east, the north and central Thailand (except BMR) while the emissions were lower in the south, depending on the crops cultivated in each area, which include rice, sugarcane, cassava, potatoes and soybean (LDD, 2016). The BC emissions from agricultural residue burning and forest fires are more serious in the northern and north-eastern regions and dominate the emissions in those areas. This is consistent with a previous report relating to the spatial and temporal summaries of high-resolution emissions throughout Thailand by Vongmahadlek et al. (2009), which suggested that the main contributors to PM_{10}, OC and BC are biomass burning (40.2%), industrial sectors (31.5%), road transportation (17.1%) and power plants (10.9%). Most crop residues are burned in the field after harvesting or used in agro-

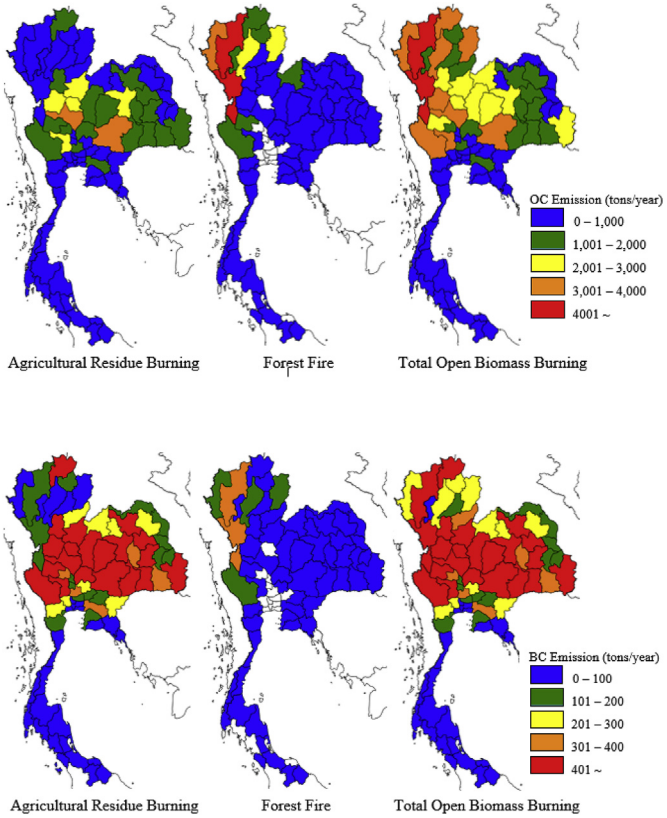


Fig. 2. Distribution of each pollutant by province in Thailand (2014) (a) OC, (b) BC.

industries as fuel although sugarcane leaves are frequently burned in the field before harvesting. As reported by Phairuang et al. (2017), the emission of PM from biomass burning dominates the ambient PM concentration in northern, northeastern and central Thailand, where rice and sugarcane are the main crops and forest fires are a serious problem, especially in the north during the dry season. As previously described, emissions from open biomass burning directly related to agricultural activities are not significant in southern Thailand although emissions from biomass burning used as fuel in industries are an issue which needs to be carefully considered (Chomanee et al., 2009).

As shown in Supplement Fig. 2a and 2b, the monthly OC and BC emissions from agricultural residue burning evaluated for the four different regions, categorized by the OAE (2016). Regardless of the region, peaks appear in the dry season from December to May and the emission decreases in the wet period between June and October. Peaks in November to December can be mainly attributed to sugarcane pre-harvesting burning and those in April and May to the open burning of rice residues, both of which are significant in the northeastern region. As shown in Supplement Figs. S3a and S3b, OC and BC emissions from forest fires are very significant in March, which is attributable mainly to forest fires in the northern region.

3.2. Monitored concentrations of carbonaceous aerosols

The annual average organic carbon (OC), elemental carbon (EC), Char-EC, Soot-EC and Total carbon (TC) concentrations at KMUTNB are listed in Table 2 for the period between August 2014 and July 2015. The annual averages for OC and EC were highest in the size range 0.5–1.0 μm ($PM_{0.5\text{-}1.0}$), with values of 1.93 ± 1.53 and 0.53 ± 0.24 μg/m^3, respectively. In the Bangkok site, $PM_{0.1}$ (or nanoparticle) and $PM_{2.5\text{-}10}$ were the dominant OC and EC and the OC values were 1.49 ± 0.90 and 1.24 ± 0.64 μg/m^3, respectively, whereas those for EC were 0.44 ± 0.17 and 0.48 ± 0.16 μg/m^3, respectively. TC in KMUTNB was maximum in the size range of $PM_{0.5\text{-}1.0}$.

The annual average OC, EC, Char-EC, Soot-EC and TC concentrations in Northern Thailand (Chiang Mai) are shown in Table 3. The annual OC concentration was very high in ultrafine particle ($PM_{0.1}$) at 3.76 ± 2.53 μg/m^3, whereas EC was highest in small particle sizes < 1 μm ($PM_{0.5\text{-}1.0}$) with a value of 1.37 ± 1.14 μg/m^3. TC in particulate matter was higher in smaller particle sizes than in larger sizes. Nanoparticle, or, $PM_{0.1}$ comprises the highest TC mass concentration.

3.3. Correlation between EI of open biomass burning and carbonaceous parameters

The carbon composition ratio in PM can distinguish the release source from all combustion activities (Cao et al., 2005). Fluctuating of OC and EC are produced from different source categories. The ratio of OC/EC in size-classified particles has been generally used to classify emission sources. Nevertheless, the OC/EC ratio often relies on other factors to correctly identify the emission source. The three factors that can disrupt the ration of OC/EC are the primary emission source, secondary organic aerosols (SOA) and the removal rate by wet deposition (Cachier et al., 1996). The correlation between the carbon EI and the amount of carbon species emitted in each size fraction is very important in investigating carbon behavior.

3.3.1. Behavior of Char-EC and Soot-EC

Figs. 3 and 4 show the Char-EC/Soot-EC ratios for the BMR and Chiang Mai, respectively. The EI extrapolated to zero was very similar for $PM_{0.1}$ at ~0.5 in both locations, indicating that $PM_{0.1}$ has the characteristics of diesel soot (Char-EC/Soot-EC < 1) (Allen et al., 2001). In Chiang Mai, the correlations are clear for fine size ranges smaller than 2.5 μm, especially for PM_1 and $PM_{0.1}$. An increase in the Char-EC/Soot-EC ratio is suggestive of more biomass combustion behavior, with the EI showing significant $PM_{0.5\text{-}1}$ where the ratio also has a peak value (~10). This may be related to the dominant size of the PM emitted from biomass burning being in the size range of 0.5–1.0 μm (Hata et al., 2014; Chomanee et al., 2018). The correlation in $PM_{0.1}$ at the Bangkok site is slightly lower but still clear. However, the correlations in other size ranges are rather poor and the Char-EC/Soot-EC ratio fluctuated around 3–4 for particle size 1–10 μm while it was <1 for particle size >10 μm. Such a difference is probably related to the carbon emission sources with the Soot-EC being richer in diesel soot while the Char-EC consists of smoke particles from biomass burning. As can be seen in Supplement Figs. S4a–4e and Figs. S5a–5e, in Bangkok, as compared to Chiang Mai, the Soot-EC had larger concentrations in every size-range of particles throughout the year and they were very high in $PM_{0.1}$ while in Bangkok only $PM_{0.1}$ was sensitive to the influence of biomass combustion.

Pongpiachan et al. (2014) studied the chemical characteristics of the carbonaceous aerosol PM_{10} in Bangkok, and suggested that the low OC/EC ratios in February–December 2007 were indicative of a major contribution from road transportation. However, the site stations in that study were PCD air quality monitoring sites, which can be considered as focusing on diesel engine sources in an urban residential background.

The BC concentration was continuously measured by aethalometer. The average 1-hr BC concentrations in the Bangkok ambient air ranged from 1.59 to 4.19 μg/m^3, with an average of 2.47 μg/m^3. The hourly variation indicated that the highest values occurred between 6 and 9 a.m. with another peak in the evening (Supplement Fig. S6). This result suggests that the behavior of the Soot-EC in relation to BC behavior as measured by aethalometer is closely related to road transportation.

During haze episodes in northern Thailand, including those in Chiang Mai, air pollutants are wildly spread and cover the total area in the dry season (Chantara et al., 2012; Khamkaew et al., 2016). The Particulate matter contributes carbon components in the form of both BC and OC that originate from open biomass burning and this significantly affects the carbon behavior in size-segregated aerosols, and in particular $PM_{0.1}$ particles.

3.4. Air mass trajectories and hotspots

3.4.1. Bangkok

The contribution of carbon from biomass burning can be

Table 2
Annual average concentrations of OC, EC, Char-EC, Soot-EC, TC and mass (ug/m^3) in Bangkok.

	OC	EC	Char-EC	Soot-EC	TC	PM
$PM_{0.1}$	1.49 ± 0.90	0.44 ± 0.17	0.18 ± 0.10	0.25 ± 0.08	1.92 ± 1.04	14.80 ± 1.99
$PM_{0.5\text{-}1.0}$	1.93 ± 1.53	0.53 ± 0.24	0.41 ± 0.20	0.12 ± 0.06	2.46 ± 1.71	21.85 ± 4.62
$PM_{1.0\text{-}2.5}$	1.11 ± 0.75	0.39 ± 0.19	0.30 ± 0.16	0.08 ± 0.04	1.50 ± 0.88	18.76 ± 3.34
$PM_{2.5\text{-}10}$	1.24 ± 0.64	0.48 ± 0.16	0.31 ± 0.11	0.17 ± 0.08	1.72 ± 0.77	27.83 ± 4.43
$PM_{>10}$	0.60 ± 0.36	0.19 ± 0.10	0.09 ± 0.11	0.11 ± 0.06	0.79 ± 0.45	20.11 ± 2.59

Table 3
Annual average concentrations of OC, EC, Char-EC, Soot-EC, TC and mass (ug/m^3) in Chiang Mai.

	OC	EC	Char-EC	Soot-EC	TC	PM
$PM_{0.1}$	3.76 ± 2.53	1.11 ± 1.06	0.66 ± 0.39	0.44 ± 0.26	4.87 ± 3.65	25.21 ± 4.73
$PM_{0.5\text{-}1.0}$	2.33 ± 1.64	1.37 ± 1.14	1.19 ± 1.34	0.18 ± 0.10	3.70 ± 2.94	26.20 ± 4.73
$PM_{1.0\text{-}2.5}$	1.24 ± 0.70	0.53 ± 0.36	0.40 ± 0.33	0.13 ± 0.09	1.77 ± 0.95	20.77 ± 2.84
$PM_{2.5\text{-}10}$	1.12 ± 0.42	0.35 ± 0.16	0.26 ± 0.14	0.09 ± 0.04	1.48 ± 0.53	21.83 ± 1.99
$PM_{>10}$	0.52 ± 0.40	0.11 ± 0.05	0.07 ± 0.03	0.04 ± 0.03	0.63 ± 0.44	18.51 ± 0.96

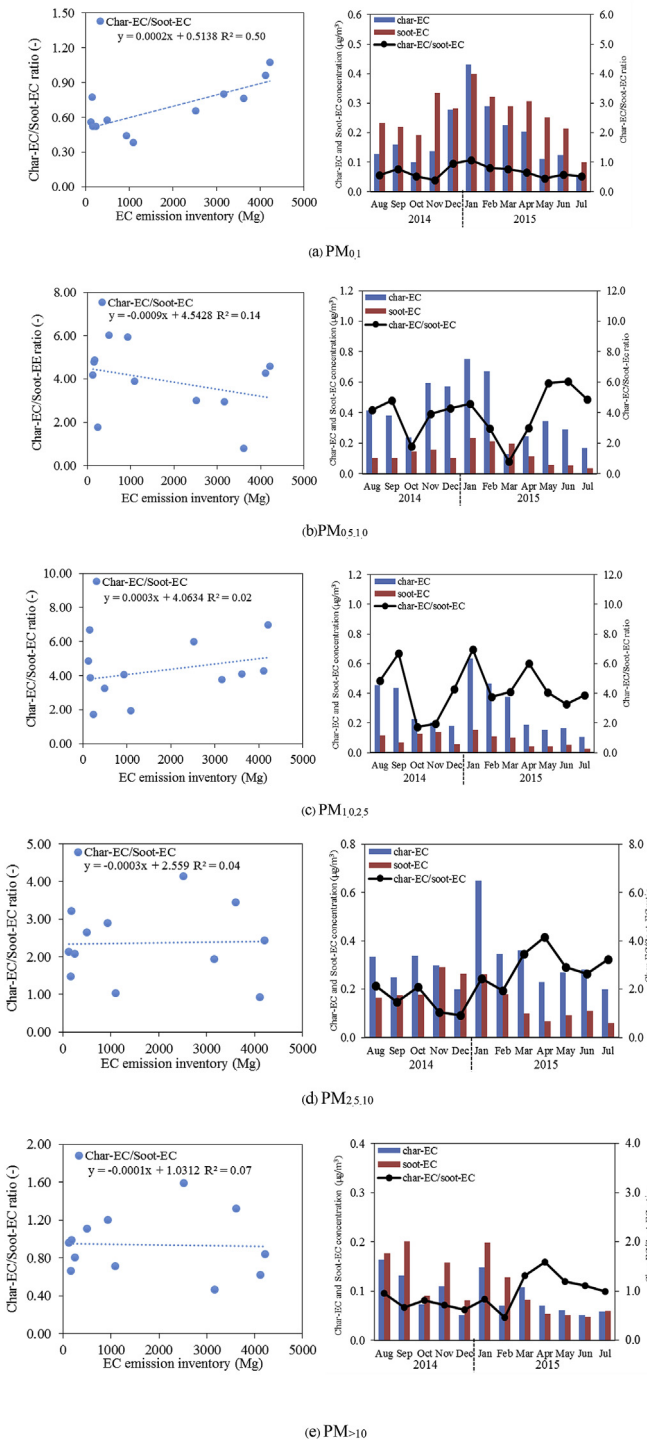


Fig. 3. Char-EC/Soot-EC ratio for source identification in Bangkok.

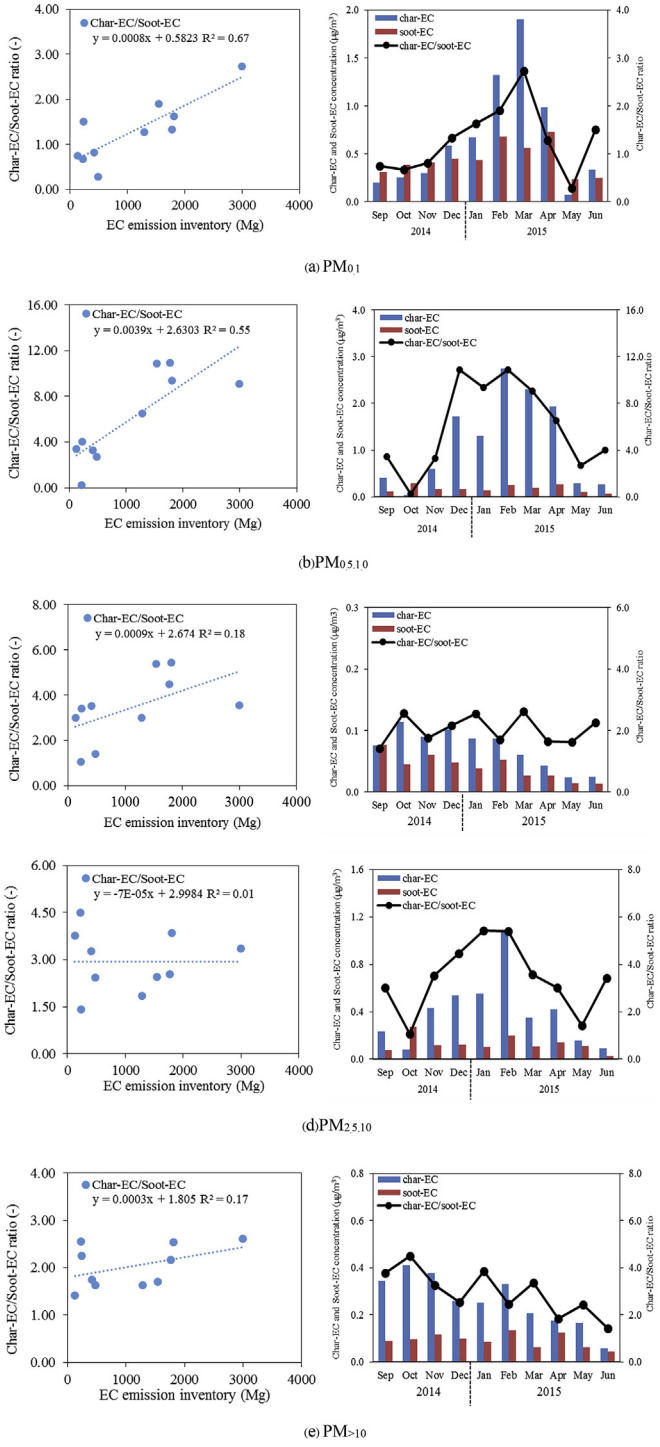
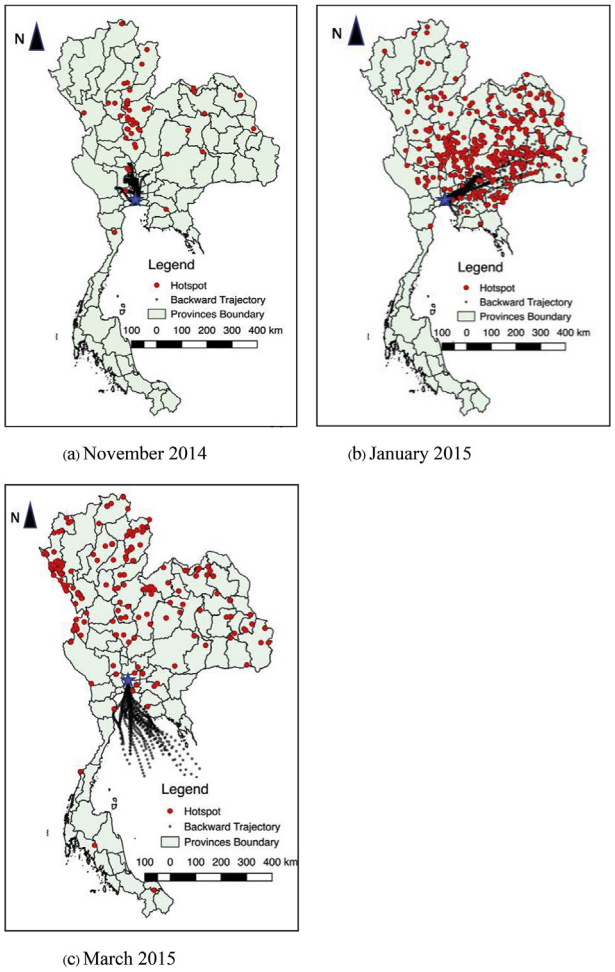


Fig. 4. Char-EC/Soot-EC ratio for source identification in Chiang Mai.

investigated based on backward trajectories. As shown in Fig. 5, air masses from the north direction of Bangkok moved over Bangkok in November 2014 at the start of the dry season and from the northeast of BMR in January 2015. Fig. 5a displays that the air masses came from the huge agricultural area in the Chaopraya river basin where rice is widely cultivated. In the dry season, there is large burning of rice residues after harvesting between November and April (Tipayarom and Kim Oanh, 2007; Kanokkanjana, 2010; Cheewapongphan and Garivait, 2013). Accordingly, an enormous amount of particle smoke is released which travels to BMR during that period. As reported by Kim Oanh et al. (2018), the emission of BC from agricultural residue burning dominates in the central part of Thailand particularly in the Chaopraya river basin where rice and sugarcane are predominantly cultivated. That area is between 100 and 200 km away from Bangkok. The time taken for the air mass transports from the emission source area to reach Bangkok is around 12–24 h. Taking into account that the atmospheric lifetime of BC is around one week (Cape et al., 2012), the BC from open biomass burning may contribute significantly to carbonaceous aerosols in the surrounding areas including Bangkok. Furthermore, Fig. 5b shows that an air mass migrated from the northeast influenced by the northeast monsoon season in Thailand. The air mass movements are from the northeast passed through the northeastern part of Thailand where there is widespread burning of rice and sugarcane residues. However, as Fig. 5c shows, later air mass movements emanated in the south from the gulf of Thailand which brought clean air to Bangkok and this resulted in a slightly lower carbon concentration in March 2015 due to the clean air from the ocean.



(a) November 2014 (b) January 2015

(c) March 2015

Fig. 5. Hotspot and Backward trajectory during the sampling periods at KMUTNB over high episode (a) Novemb. er 2014 (b) January 2015 (c) March 2015.

3.4.2. Chiang Mai

The haze in northern Thailand occurs every year from February to early April. The potentially high impact of open biomass burning was an essential contributor to the PM pollution observed in Chiang Mai at the CMU monitoring station. Backward trajectories were also analyzed during the sampling time to identify potential sources using the HYSPLIT4 model for air masses 50 m above ground level at the CMU station. Fig. 6 show heavy air mass travels from the south and southwest direction. The air movement passed through many hotspot areas on 21 March 2015 (the sampling time). Biomass combustion is a key potentially high contributor to the PM pollution observed at the CMU station. From February to April every year, forest fires often occur in the upper part of Thailand. Therefore, a vast amount of smoke is released and migrates to Chiang Mai during that period.

Furthermore, air pollution from neighboring countries i.e. Myanmar and Laos needs to be measured for an accurate evaluation in northern Thailand. The air pollutants emitted from major biomass burning can migrate from one country to its neighbors and to other regional countries as well. The influence of carbonaceous aerosols from other countries is not discussed here, due to the difficulty of obtaining their emission inventories both of spatial and temporal distributions. The issue of air pollution in Thailand from other countries and pollution from Thailand in other countries constitutes the main area of uncertainty in this study and a future study needs to investigate those topics.

4. Conclusion

In this study, particle-bound carbonaceous components in size-segregated ambient particles down to nanoparticles ($PM_{0.1}$) were related to the EI of particle-bound BC and OC from agricultural and forest fire related biomass burning in Thailand. The results indicate that the carbon emissions related to agricultural activities, for

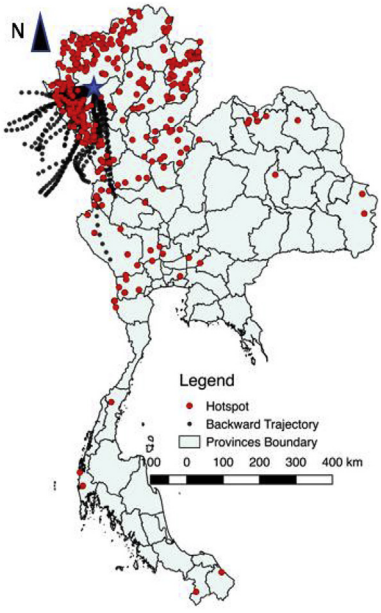


Fig. 6. Hotspot and Backward trajectory during the sampling period at CMU over high episode, 21 March 2015.

example pre-harvest burning, field residue burning, forest fires and the use of field residue as a fuel in agro-industries dominate the ambient carbon concentration, not only in an area where open biomass burning is very intensive (Chiang Mai) but also in a region to which the emissions migrate from surrounding provinces (Bangkok). The Char-EC/Soot-EC diagnostic index of carbonaceous components, which describes the relative influence of biomass burning to diesel emission, was clearly reflected in the EI of BC from agricultural biomass burning, particularly for particles smaller than 1 μm. This was especially true for $PM_{0.1}$, which basically provides evidence of diesel emissions, but was found to be very sensitive to the EI of agricultural biomass burning. In Chiang Mai, the northern part of Thailand, forest fires occurring upwind of the CMU site were found to be the largest contributor while the carbon behavior at the site in Bangkok was better described by the EI for BMR and the surrounding provinces in the central Thailand region, where the open burning of agricultural residues, particularly from the cultivation of sugarcane for sugar production, is significant. This suggests that the influence of the migration or diffusion of polluted air masses is important at a multi-provincial scale (100–200 km) in Thailand although transboundary migration over countries in this area could also be an important factor which merits future study in a more detailed investigation.

Conflicts of interest

There are no conflicts of interest to declare.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2019.01.001.

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