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Carbon and Trace Element Compositions of Total Suspended Particles (TSP) and Nanoparticles ($PM_{0.1}$) in Ambient Air of Southern Thailand and Characterization of Their Sources

Muanfun Inerb [1], Worradorn Phairuang [2,3,*], Phakphum Paluang [3], Mitsuhiko Hata [2], Masami Furuuchi [1,2] and Prasit Wangpakapattanawong [4]

[1] Faculty of Environmental Management, Prince of Songkla University, Hat Yai 90112, Thailand; 6010920027@psu.ac.th (M.I.); mfuruch@staff.kanazawa-u.ac.jp (M.F.)
[2] Faculty of Geoscience and Civil Engineering, Institute of Science and Engineering, Kanazawa University, Kanazawa 920-1192, Japan; hata@se.kanazawa-u.ac.jp
[3] Department of Geography, Faculty of Social Sciences, Chiang Mai University, Chiang Mai 50200, Thailand; phakphum_p@cmu.ac.th
[4] Department of Biology and Environmental Science Research Center, Faculty of Science, Chiang Mai University, Chiang Mai 50200, Thailand; prasit.wang@cmu.ac.th
* Correspondence: worradorn.ph@cmu.ac.th

Abstract: The concentration of total suspended particles (TSP) and nanoparticles ($PM_{0.1}$) over Hat Yai city, Songkhla province, southern Thailand was measured in 2019. Organic carbon (OC) and elemental carbon (EC) were evaluated by carbon aerosol analyzer (IMPROVE-TOR) method. Thirteen trace elements including Al, Ba, K, Cu, Cr, Fe, Mg, Mn, Na, Ni, Ti, Pb, and Zn were evaluated by ICP-OES. Annual average TSP and $PM_{0.1}$ mass concentrations were determined to be 58.3 ± 7.8 and 10.4 ± 1.2 $\mu g/m^3$, respectively. The highest levels of PM occurred in the wet season with the corresponding values for the dry seasons being lower. The averaged OC/EC ratio ranged from 3.8–4.2 (TSP) and 2.5–2.7 ($PM_{0.1}$). The char to soot ratios were constantly less than 1.0 for both TSP and $PM_{0.1}$, indicating that land transportation is the main emission source. A principal component analysis (PCA) revealed that road transportation, industry, and biomass burning are the key sources of these particles. However, PM arising from Indonesian peatland fires causes an increase in the carbon and trace element concentrations in southern Thailand. The findings make useful information for air quality management and strategies for controlling this problem, based on a source apportionment analysis.

Keywords: air quality management; biomass burning; carbon; PCA; $PM_{0.1}$; trace elements

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1. Introduction

Airborne particulate matter (PM), which is thought to be strongly associated with adverse human health effects [1,2] and global warming [3,4] has recently become a subject of great interest. Studies of PM in Thailand and Asian countries are currently based on measurements of PM_{10} and $PM_{2.5}$ through ground-based monitoring and mathematical modeling [5–11]. However, data on the abundance, sources, monthly distribution together with seasonal variations in the concentrations of carbon and elements in the nano-size range ($PM_{0.1}$; particles of diameter ≤ 0.1 μm or 100 nm) and total suspended particles (TSP) remain scarce in developing countries, including Thailand and its neighboring countries [12–14].

Particle-bound total carbon (TC) can be separated into two types, i.e., organic carbon (OC) and elemental carbon (EC) or black carbon (BC) [15]. The term BC is loosely equivalent to EC liable on the analytical method being used [16,17]. The distribution of carbon portions, which basically represent the chemical, optical, and physical appearances of carbon material in PM, varies with time and location [18–20]. Knowledge of carbon

composition in atmospheric PM is also vital for identifying emission sources and control measures for PM and carbon-containing materials, the two main causes of air pollution and global warming [21,22].

In addition, particle-bound trace elements have important effects regarding human health. The toxic elements increase both carcinogenic and non-carcinogenic risk assessments in humans [23–25]. Although they represent a small share of the total mass concentration of PM, trace elements are of particular concern because that have a health risk to humans [23,26,27]. Concerning elements, Mn, Al, and Fe are characteristically found in the crust earth, whereas Cu, Cr, Pb, and Zn are resulting from anthropogenic sources [5,13]. Inhaling toxic elements including Cd, Zn, Cr, Pb, and Ni could be harmful to human health, causing various diseases, particularly cancer [28–30].

To the best of the authors' knowledge, only a few limited studies of the distribution of $PM_{0.1}$ and carbon material in Thailand are reported [12,16]. Previous studies reported only data on OC/EC in the upper part of Thailand and did not report information on the contribution and source apportionment for the $PM_{0.1}$ fraction. Moreover, information concerning particle-bound trace elements in the $PM_{0.1}$ fraction is still lacking in Thailand. Receptor modeling, e.g., principal component analyses (PCA) has been used to identify the emission sources of chemical species in the PM_{10} and $PM_{2.5}$ fractions in Thailand [7,24], although PCA continues to be lacking in $PM_{0.1}$ fraction in Thailand.

Like other cities in Thailand and other Southeast Asian countries, the economy of Hat Yai has grown rapidly in recent years. The city of Hat Yai also suffers from air pollution. Thus, the objectives of this study were (1) to investigate the characteristics of atmospheric OC, EC, and metals in the $PM_{0.1}$ and TSP fractions, (2) to determine the emission sources by PCA, and (3) to identify the possible sources of this pollution in Hat Yai city, the economic capital of the Songkhla province. These relationships would be valuable for identifying potential emission sources in the area and could lead to better air quality management leading to a sustainable city and society.

2. Methodology

2.1. Sampling Site

Hat Yai is an economic city in the southern part of Thailand, covering an area of approximately 852.8 square kilometers and with a population of around 0.6 million. The key economic activities of Hat Yai are related to farming production, i.e., palm oil and para-rubber [31,32]. $PM_{0.1}$ and TSP samples were collected on the top floor of an 8-floor building, which is part of the Faculty of Engineering, Prince of Songkla University (PSU; 7°00′21.8″ N 100°30′08.6″ E). The site is located approximately 30 m above ground level, and is not expected to be influenced by ground-level activities. The monitoring station is located at a distance of approximately 4 km from the commercial area, where household and transportation are not dense. Figure 1 shows the monitoring site in this study. During the monitoring periods, detailed weather information was collected, including relative humidity, rainfall, temperature, pressure, and wind speed was also carefully measured, as shown in Table 1. The weather in southern Thailand can be separated into two seasons depending on the monsoons, namely, a dry season from January to April, and a wet season (or monsoon season) from May to December [33].

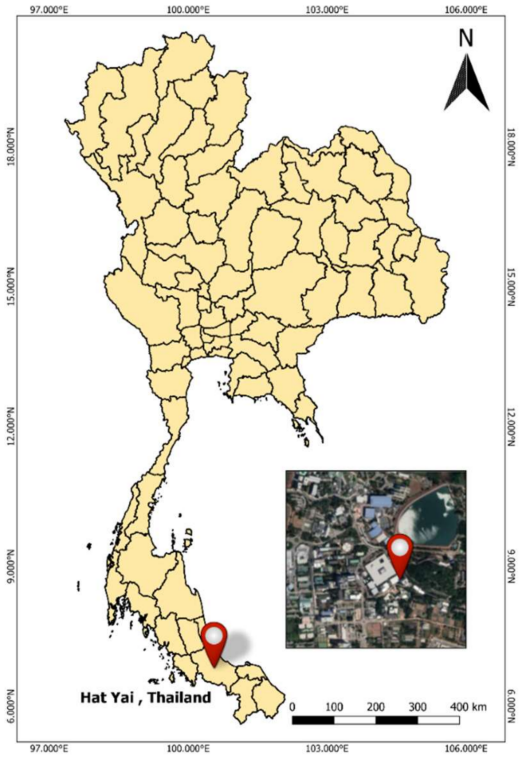

Figure 1. Location of sampling site, in Hat Yai, Songkhla, southern Thailand; Prince of Songkla University (PSU).

Table 1. Meteorological parameters during different sampling times.

Sampling Period	Temperature (°C)	RH (%)	Wind Speed (m/s)	Pressure (hPa)	Rainfall (mm)
Jan–April, 2019	24.7–28.3 (26.8)	65–82 (74)	0.98–1.84 (1.40)	913–1009 (977)	28.6–215.8 (117.9)
May–Aug, 2019	27.0–28.3 (27.9)	77-81 (78)	0.75–1.26 (0.99)	975–1008 (1000)	75.4–245.2 (153.2)

2.2. Cascade Sampler for $PM_{0.1}$ and High-Volume Sampler for TSP

For the air sampling, $PM_{0.1}$ samples were collected on quartz fiber filters (QFF) (2500 QAT-UP, Pall Corp., pure quartz, size 55 mm, New York, NY, USA) using a $PM_{0.1}$ cascade sampler that operated at a flow rate of 40 L/min. Samples were collected for periods of 48 hrs. from January to August 2019. Samples were collected for two consecutive days in each week of each month. A total of 32 $PM_{0.1}$ samples were collected and examined in this study. Moreover, TSP was collected with a portable high-volume sampler (Shibata, Japan). The high-volume sampler contained an air inlet at 500 L/min (2500 QAT-UP, Pall Corp., pure quartz, size 110 mm, New York, NY, USA). To confirm the mass concentration, non-repeated ANOVA was performed for determining the mass concentrations of TSP and $PM_{0.1}$ evaluated at the sampling site, and significant differences ($\rho < 0.05$) were found between seasons. The annual averaged TSP and $PM_{0.1}$ concentrations were computed by dividing the mass of all the monthly averaged TSP and $PM_{0.1}$ by the volume of the air they were taken from.

For TSP sampling, 24-h samples were collected from January to August 2019. Samples were collected four times per month, in parallel with $PM_{0.1}$ samples. A total of 32 TSP samples were collected in this study. QFFs were weighed twice before and after sampling. Before weighing, QFFs were pre-baked in an electric furnace at 350 °C for 1 h to eliminate possible carbon impurities [19,20]. QFFs were balanced for 48 h in a $PM_{2.5}$ chamber with

a controlled relative humidity of 35 ± 5% as well as a temperature of 21.5 ± 1.5 °C [18]. Travel blank filters were also arranged to decrease the effect of any contamination, i.e., the adsorption of volatile organic components during sampling and filter transportation. All filters that were used were stored at a temperature below −20 °C in a freezer until chemical analysis [30].

2.3. Carbon Analysis Method

Carbon compositions in the PM were determined using a Carbon Aerosol Analyzer (Model 5L, Sunset Laboratory, Tigard, OR, USA) at Kanazawa University, Japan and the measures followed the Interagency Monitoring of Protected Visual Environments-Thermal/Optical Reflectance (IMPROVE-TOR method) [34]. QFF sample parts were punched using a 15 mm × 10 mm rectangular cutter before the carbon analysis. Briefly, four OC fractions OC1, OC2, OC3, and OC4 at temperature 120 °C, 250 °C, 450 °C, and 550 °C were analyzed in a 100% helium (He) atmosphere, while the EC fractions EC1, EC2, and EC3 at temperature 550 °C, 700 °C and 800 °C were measured in O_2 (2%) and He (98%) carrier gas. Additionally, the pyrolyzed organic carbon fraction (PyC) was determined at 550 °C between the split time of EC and OC [16,17]. OC and EC are defined respectively as follows: OC = OC1 + OC2 + OC3 + OC4 + PyC as well as EC = EC1 + EC2 + EC3-PyC.

Moreover, Char-EC refers to EC1-PyC and Soot-EC is EC2 + EC3 [15]. In the following studies, char, soot, and char/soot ratios were used as indices of the influence of biomass combustion and fossil fuel together with OC, EC, and OC/EC ratios [19]. The quality assurance (QA) and quality control (QC) were calibrated with blank filters and a reference standard. The calibration standard for the carbon analysis was TC, as analyzed by sucrose ($C_{12}H_{22}O_{11}$) (196-00015, Wako Pure Chemical Industries, Ltd., Osaka, Japan). Minimum detection limits (MDLs) were determined by blank filters. The averaged MDL values of OC and EC in $PM_{0.1}$ (n = 3) in blank filters were 0.2 and 0.2 μg/m^3, as well as those in TSP (n = 3) were 0.2 and 0.1 μg/m^3, respectively. The blank filters month was subtracted from the samples for correction of the carbon species in each month.

2.4. Trace Elements Analysis

All samples were determined for 13 trace elements including Al, Ba, Cu, Na, Ni, K, Cr, Mg, Ti, Fe, Mn, Zn, and Pb. The trace element concentrations were measured by an inductively coupled plasma optical emission spectrometer (ICP-OES; PerkinElmer, AVIO 500) at Prince of Songkla University, Thailand. Briefly, one half of the QFF was cut into small pieces and immersed in 2 mL of 65% HNO_3^- for 1 h, in a water bath at a temperature of 95 °C. Then, the 0.5 mL of 30% H_2O_2 was added for 30 min [30]. The samples were allowed to cool to room temperature, filtered through Whatman No.1 paper, and then diluted to obtain a 10 mL solution [23]. The procedure for the extraction and analysis of trace elements followed the US-EPA method 6010D (SW-846) and the US-EPA method IO-3.1 (compendium of methods for the determination of inorganic compounds in ambient air). Several sampling filters (n = 5) were tested by dividing into two parts, first, the part was extracted normally, for the second part after extraction, the filter residue was extracted again and compared with a blank filter. Quality assurance (QA) and quality control (QC) were confirmed by digesting and analyzing the standards and blanks in common with each sample batch. The quantification was compared by a standard curve (R^2 > 0.99) [23,30].

2.5. Source Apportionment Analysis

The $PM_{0.1}$ and TSP as well as their carbon and trace elements concentration were conducted by using the OriginPro 8.6 program and SPSS (version 22). All PCA were used to categorize possible patterns in the data [35]. The findings show the existence of a correlation between $PM_{0.1}$ and TSP-bound carbon as well as trace elements and other factors (sources). To identify the specific sources, PCA was run using monthly data. The Kaiser–Meyer–Olkin's (KMO) test was performed and KMO value > 0.6 was obtained, which suggested that all data were suitable for PCA [36]. In each chemical species, greater

than 80% of the samples found a signal-to-noise ratio (S/N) of unity and were carefully chosen as variables in the analysis [30].

2.6. Hot Spots and Backward Trajectories

Hotspots or active fires from open burning were derived from the satellite imagery, namely, moderate resolution imaging spectroradiometer (MODIS). The resolution of the hot spots is represented at a 1 km^2 resolution for each spot. This study analyzes the five-day isentropic back-trajectories of air masses to classify the most likely biomass burning source regions influencing the measured PMs at the selection sites in Hat Yai, Thailand. Five days were likely to be sufficient time for most trajectories to pass through the possible source regions in SEA. Back-trajectories were produced using the Hybrid Single-Particle Lagrangian Integrated Trajectory model version 4 (HYSPLIT-4) [37]. It was accessed and run through the NOAA website (https://www.ready.noaa.gov/HYSPLIT.php; accessed on 9 March 2022). As shown in an earlier report, the ending height from 500 m AGL is calculated liable on the reference of the backward trajectory in Thailand [38].

3. Results and Discussion

3.1. $PM_{0.1}$ and TSP Mass Concentrations

Figure 2 displays the $PM_{0.1}$ and TSP mass concentrations for the sampling period. The averaged $PM_{0.1}$ and TSP mass concentrations were 10.4 ± 1.2 and 58.3 ± 7.8 μg/m^3, respectively. These TSP mass concentrations are lower than the TSP standard value from the WHO and Thailand guidelines (100 and 330 μg/m^3, respectively) representing that the sources of PM emission were lower around the sampling site. Nevertheless, there is no standard for ambient $PM_{0.1}$ in Thailand and other countries. The wet season has a higher mass concentration of $PM_{0.1}$ and TSP than the dry season. The highest $PM_{0.1}$ and TSP were found in August, as 12.7 ± 2.0 and 73.5 ± 18.4 μg/m^3, respectively. The level of $PM_{0.1}$ ranges from 14.5 to 20.7% that of TSP. The high mass concentration during the wet season is related to transboundary pollution from other countries. Particulate pollution during the haze period over southern Thailand was affected by peat-land fires in Indonesia [19,39]. High levels of PM pollution transport affect the air quality in southern Thailand almost annually during June–August [19]. In 2019, the high level of $PM_{0.1}$ and TSP found in August at the sampling site indicates that long-range atmospheric transport accounts for an increased level of particulate matter at this time.

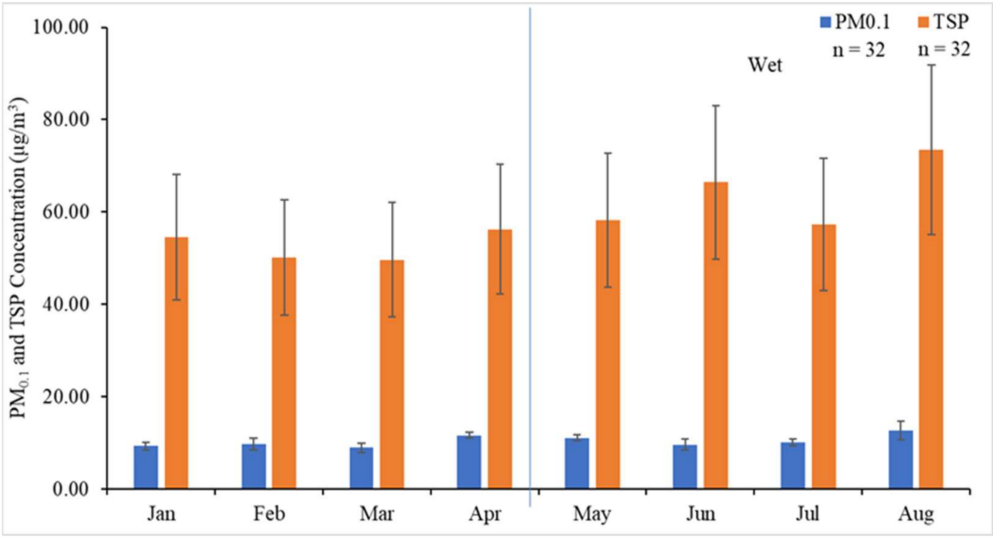

Figure 2. Average mass concentrations of $PM_{0.1}$ and TSP in Hat Yai, Songkhla, Thailand during dry and wet seasons, 2019.

The averaged $PM_{0.1}$ level of 10.4 ± 1.2 μg/m^3 was lower than the corresponding values for Bangkok, and Chiang Mai, Thailand, Hanoi, Vietnam, and North Sumatra, Indonesia [16,17,20]. However, the $PM_{0.1}$ levels were roughly four times higher than those in other urban areas in Asia (Table 2). The mass concentration in Kanazawa, Japan is 4.7, and 2.6–5.4 μg/m^3 in other cities in Japan [40,41]. In other Asian cities, Chen et al. (2010) [42] described that the $PM_{0.1}$ in an urban-traffic area in Taiwan was 1.4 ± 0.6 μg/m^3. However, Ding et al. (2017) [43] reported high levels of $PM_{0.1}$ in a municipal area of Shanghai, China. In Kolkata, India, the $PM_{0.1}$ level reaches levels as high as 8.8 ± 0.6 μg/m^3 during episodes of biomass fires [44]. In the United Kingdom (UK), the mass concentration of $PM_{0.1}$ based on the estimated PM_{10} fraction for each emission source are as follows: waste incineration (4%), industrial combustion (7%), energy combustion in industries (8%), industrial off-road mobile machinery (9%), agriculture (9%), non-road transportations (14%), and production process (15%) [21]. Moreover, the $PM_{0.1}$ mass concentration is very low in western countries. Venecek et al. (2019) [45] studied the county levels of $PM_{0.1}$ across the USA and found that $PM_{0.1}$ surpassed 2 μg/m^3 during a smog period. However, the annual average mass concentration of ambient $PM_{0.1}$ is very small (around 1 μg/m^3). Particle number concentration (PNC) is widely used to measure $PM_{0.1}$ due to a small fraction in many cities [46]. In south part of Thailand, there was a fluctuation, probably related to local emission sources, especially, wood biomass burning in ago-industries [31].

Table 2. Mass concentration of the $PM_{0.1}$ fraction at different locations in Asia (μg/m^3).

Location	Site Description	Concentration	References
Hat Yai, Thailand	Mixed	10.4 ± 1.2	This study
Bangkok, Thailand	Urban-traffic	14.8 ± 2.0	[16]
Chiang Mai, Thailand	Suburban	25.2 ± 4.7	[16]
Hanoi, Vietnam	Urban-traffic	6.0 ± 2.7	[17]
North Sumatra, Indonesia	Urban-traffic	16.8 ± 4.0	[20]
North Sumatra, Indonesia	Rural	7.1 ± 2.5	[20]
Kanazawa, Japan	Mixed	4.7	[40]
Kanazawa, Japan	Mixed	2.6 ± 1.2	[41]
Suzu, Japan	Rural	2.8 ± 0.9	[41]
Toyama, Japan	Urban	5.4 ± 1.6	[41]
Hsinchu, Taiwan	Urban-traffic	2.2 ± 0.6	[42]
Shanghai, China	Urban	13.4	[43]
Kolkata, India	Urban	8.8 ± 2.6	[44]

3.2. Carbon Components in $PM_{0.1}$ and TSP

The seasonal and total averaged carbon species occurrence in $PM_{0.1}$ and TSP from January to August 2019, including OC, EC, Char-EC, Soot-EC, and TC concentrations, OC/EC, and Char-EC/Soot-EC ratio are listed in Table 2. The wet season showed a higher carbon content than the dry season both for $PM_{0.1}$ and TSP. The averages for Char-EC in the $PM_{0.1}$ were high in the wet season compared to the dry season. It is interesting that Soot-EC in the TSP was quite similar in the wet season, although a difference in $PM_{0.1}$ was observed. Almost every year in the wet season in southern Thailand, a southeast-Asian haze with elevated PM rises from Sumatra and the Kalimantan islands, Indonesia and often blankets southern Thailand during June–September [7,47]. Tham et al. (2019) [48] reported that during a $PM_{2.5}$ episode from peat-land fires on Sumatra Island, a large amount of carbon species, i.e., PyC and Soot-EC was released into the atmosphere.

3.3. OC/EC and Char-EC/Soot-EC Ratios

The ratio of OC/EC can be used to identify sources of emitted carbonaceous aerosols. OC/EC ratios from diesel engines, coal burning, and biomass fires are different. Biomass fires have higher OC/EC ratios, while fossil fuel burning results in lower ratios [12,44]. Pongpiachan et al. (2014) [49] described a ratio of OC/EC for biomass burning of ~7–8, while Allen et al. (2001) [50] and Pio et al. (2011) [51] reported a value of ~1–2 from fossil

fuel burning. Table 3 displays the annual ratios of OC/EC in $PM_{0.1}$ and TSP in Hat Yai city, which have relatively higher values in the wet season (2.6 ± 0.7 and 4.2 ± 1.5 for $PM_{0.1}$ and TSP, respectively).

Table 3. Average seasonal and total concentrations of OC, EC, Char-EC, Soot-EC, TC (μg/m^3) and OC/EC, Char-EC/Soot-EC ratio in Hat Yai.

Size	Season	OC	EC	Char-EC	Soot-EC	TC	OC/EC (−)	Char-EC/Soot-EC (−)
PM0.1	Dry	1.6 ± 0.2	0.7 ± 0.1	0.1 ± 0.1	0.5 ± 0.1	2.3 ± 0.4	2.5 ± 0.5	0.3 ± 0.2
	Wet	4.9 ± 0.9	1.8 ± 0.5	0.4 ± 0.1	1.4 ± 0.1	6.7 ± 0.3	2.7 ± 0.7	0.3 ± 0.2
	Total	3.3 ± 0.6	1.2 ± 0.4	0.3 ± 0.1	0.9 ± 0.1	4.5 ± 0.4	2.6 ± 0.6	0.3 ± 0.2
TSP	Dry	5.0 ± 1.2	1.4 ± 0.4	0.4 ± 0.2	1.1 ± 0.3	6.4 ± 2.9	3.8 ± 1.6	0.3 ± 0.1
	Wet	5.5 ± 3.6	1.5 ± 0.9	0.3 ± 0.2	1.1 ± 0.3	7.0 ± 4.2	4.2 ± 1.5	0.2 ± 0.1
	Total	5.3 ± 4.6	1.4 ± 1.0	0.3 ± 0.2	1.1 ± 0.3	6.7 ± 5.4	4.0 ± 1.3	0.3 ± 0.2

The ratios of OC/EC reported in this result are in general agreement with the results described in other studies [32,40]. The ratios of OC/EC for $PM_{0.1}$ in this study are higher than those from Taiwan, which ranged from 0.2 to 1.7 [40,52], but lower than the values for Hanoi, Vietnam, which ranged from 3.8 to 5.9 [46]. In this study, the ratios of OC/EC in southern Thailand were nearly constant, which suggests that the same emission sources might contribute to OC and EC. However, the OC/EC ratios in TSP were in the range of 1.9–6.3 indicating the influence of more varying emission sources than in $PM_{0.1}$ (OC/EC, 1.6–3.5). Thumanu et al. (2009) [32] discussed the OC/EC ratios in Songkhla province in a study of the sources of PM_{10}. The measured OC/EC ratios were as follows: 1.2–1.3 (road traffic), 1.6–2.3 (industrial sources), and 3.9–4.2 (biomass fires). These results show that the origin of the higher OC/EC ratios in $PM_{0.1}$ and TSP could be a variety of sources, i.e., the industrial sector and biomass burning that contributes considerably to the carbon composition in the ambient air above southern Thailand. OC, which dominates especially in $PM_{0.1}$, may be largely attributed to biomass burning. Conversely, the ratio of OC/EC depends on three main causes for correctly classifying the emission source. The three issues are primary emission sources, secondary organic aerosols (SOA), and wet scavenging [53].

Unlike the OC/EC ratio, the Char-EC/Soot-EC ratio is distinct for different primary sources and can be used to identify the source at the origin. Only two factors can affect the char/soot ratio: the wet deposition level and primary emission source. A higher ratio of char/soot is indicative of biomass fires associated with the influence of Char-EC to the total EC content; while ratios smaller than 1.0 indicate that soot from the motor exhaust is a major contributor to the total EC content [15]. The char/soot ratio in the $PM_{0.1}$ fraction in the present study was constantly smaller than 1.0, 0.3 ± 0.2, and in the case of TSP, the value was 0.3 ± 0.2. The char/soot ratio is an index of fossil fuel combustion (0.3~0.5 for diesel soot) [16,19,20]. This suggests that fossil fuels from local transportation exert an important influence on the $PM_{0.1}$ and TSP fractions. No seasonal difference was found for the total EC. The ratios of OC/EC and char/soot were also different from the size distribution [21]. Data on other size-fractionated particulate matter and carbon components are vital for future studies directed at investigating the development of carbonaceous aerosols in more detail.

3.4. Distribution of Metals

The total concentration of thirteen elements in $PM_{0.1}$ and TSP was 161.5 ± 20.7 and 910.6 ± 157.4 ng/m^3, respectively (Table 4). The highest mass concentration of element species in $PM_{0.1}$, was found to be for K, followed by Na > Mg > Al > Fe > Zn, as well as small portions (approximately 1 ng/m^3) of Cu, Ba, Ti, Pb, Ni, Mn and Cr. The high levels of K, Na, Mg, Al, and Fe could be related to crustal elements [54]. Instead, the high Zn could have resulted from anthropogenic activities, for example, industrial activities and road transportation [25,55]. The $PM_{0.1}$-bound trace elements in the wet season were higher than in the dry season. The element K, a tracer for biomass fires [56] is also two times higher

during the wet season. Na, Al, and Fe are often produced from non-road transportation sources, particularly the re-suspension of surface dust into the atmosphere [27,57]. As described by Nghiem et al., (2020) [26], to investigate the distribution of the elemental composition in Hanoi, Vietnam, the total mass concentration of trace elements in the $PM_{0.1}$ fraction accounted for 1.1% ± 0.9% of the total elements, with the most abundant elements being Na, Al, K, Mg, Zn, and Fe. Moreover, Adachi and Buseck (2010) [58] found that 50% of metallic $PM_{0.1}$ contains more than one element and that Ni, Fe, Cr, Ti, and Zn are frequently originated in $PM_{0.1}$ particles containing more than one element.

Table 4. Concentration of trace elements in Hat Yai, southern Thailand (ng/m^3).

Species	$PM_{0.1}$			TSP		
	Dry	Wet	Annual	Dry	Wet	Annual
Al	13.1 ± 8.8	19.1 ± 6.4	16.1 ± 8.1	33.6 ± 16.2	54.6 ± 32.6	44.1 ± 27.4
Ba	0.5 ± 0.3	1.2 ± 0.8	0.8 ± 0.7	0.8 ± 0.4	1.6 ± 0.5	1.2 ± 0.6
Cr	0.04 ± 0.01	0.09 ± 0.05	0.07 ± 0.41	0.13 ± 0.06	0.20 ± 0.11	0.16 ± 0.10
Cu	2.12 ± 0.66	1.16 ± 0.91	1.64 ± 0.91	2.22 ± 0.61	1.99 ± 0.67	2.11 ± 0.64
Fe	11.0 ± 11.9	15.3 ± 8.3	13.1 ± 10.2	56.0 ± 35.2	93.2 ± 40.7	74.6 ± 41.8
K	32.2 ± 22.3	55.2 ± 19.6	43.7 ± 23.5	204.2 ± 185.8	422.3 ± 213.1	313.3 ± 225.1
Mg	23.4 ± 17.3	44.4 ± 24.6	33.9 ± 23.5	131.2 ± 56.5	91.5 ± 48.4	111.4 ± 55.3
Mn	1.26 ± 0.18	2.30 ± 1.65	1.78 ± 1.26	2.25 ± 1.40	4.78 ± 2.16	3.52 ± 2.20
Na	39.7 ± 38.9	43.6 ± 30.2	41.6 ± 33.7	540.2 ± 335.2	151.7 ± 93.3	346.0 ± 311.9
Ni	0.20 ± 0.09	0.31 ± 0.09	0.26 ± 0.10	0.29 ± 0.11	0.34 ± 0.22	0.32 ± 0.17
Pb	0.11 ± 0.11	0.99 ± 1.25	0.55 ± 0.97	1.35 ± 1.42	1.20 ± 0.83	1.27 ± 1.14
Ti	0.71 ± 0.17	0.61 ± 0.10	0.66 ± 0.14	1.36 ± 0.72	2.38 ± 1.59	1.87 ± 1.31
Zn	5.10 ± 1.49	9.12 ± 3.39	7.11 ± 3.27	9.80 ± 5.06	11.94 ± 4.48	10.87 ± 4.80
Total	129.5 ± 18.5	193.5 ± 22.5	161.5 ± 20.7	983.4 ± 173.5	837.8 ± 131.1	910.6 ± 157.5

The highest mass concentration of trace elements in TSP was for Na, followed by K > Mg > Fe > Al > Zn and a small portion (less than 10 ng/m^3) was found for Mn, Cu, Ba, Ti, Pb, Ni, and Cr (Table 3). It is also interesting that in the $PM_{0.1}$ fraction, K that represents a tracer for biomass fires increases during the wet season. Pollution Control Department (PCD), Thailand reported the Emission Inventory (EI) in the Songkhla province, they found that industries (85%), road traffic (10%), and others (5% including household and various types of transportation) are the main TSP emission sources in the Songkhla province [59].

3.5. Source Apportionment of $PM_{0.1}$ and TSP Bound Carbon and Metals

3.5.1. $PM_{0.1}$ Source Apportionment

In Table 5, PCA shows the factors for each season. To interpret the results for each season, four main factors were extracted for the dry season and four for the rainy season.

In the dry season, the PCA analysis suggested that four main clusters account for up to 86.2% of the cumulative variance. The first factor contains Al, K, Mg, Na, Cu, Mn, Fe, and Zn which reveals that this group of metals may have a common source. The total 38.3% of the $PM_{0.1}$ in the dry season could be the result of human-caused activities, particularly diesel and gasoline engines [25,60]. Miller et al. (2007) [61] described a high density of elemental nanoparticles in 30–300 nm diesel exhaust particles, including Cu, Fe, Zn, and Mg. Diesel engines are major contributors of traffic-related $PM_{0.1}$ in the ambient air [58].

The second factor (26.8%) is related to Ba, K, Mg, Na, TC, OC, and EC which may have a common origin. Biomass combustion and crustal dust sources produce particles in an ultrafine size range [62]. Factor 3 includes Cr and Pb and accounts for around 11.9% of the total. These elements indicate that combustion can be considered to be essential. The last factor is deepened on Cu and Ni. It was reported that Ni and Cu primarily originate from vehicular sources from both combustion and non-combustion sources [63].

Table 5. Principal components in the $PM_{0.1}$ fraction by season.

Species	Dry Season (Jan–Apr)				Wet Season (May–Aug)			
	PC1	**PC2**	**PC3**	**PC4**	**PC1**	**PC2**	**PC3**	**PC4**
Al	0.87				0.63			
Ba		0.82			−0.41	0.52	0.48	0.53
Cr		0.47	0.80			−0.69	0.33	0.51
Cu	0.60		0.37	0.61	0.42		0.77	−0.35
Fe	0.89				0.69	−0.57		
K	0.50	0.60	−0.42		0.92			
Mg	0.57	0.57		−0.42	0.80		0.45	
Mn	0.92	0.22			−0.45	0.52	0.39	0.33
Na	0.71	0.64			0.94		0.30	
Ni	−0.36	−0.43		0.65	0.34	−0.59	−0.61	
Pb	0.37	−0.36	0.64	−0.38	0.62	0.63		0.40
Ti	0.48	−0.69	0.41	−0.34	0.56	−0.48		0.59
Zn	0.72	−0.31			0.87		−0.36	0.18
TC	−0.61	0.69			0.42	0.90		
OC	−0.52	0.76			0.36	0.92		
EC	−0.76	0.51			0.60	0.68		
Initial eigenvalue	6.13	4.28	1.897	1.483	6.09	4.58	1.94	1.45
% Variance	38.31	26.75	11.85	9.26	38.07	28.60	12.10	9.03
% Cumulative Variance	38.31	65.06	76.92	86.19	38.07	66.67	78.77	87.80

During the wet season, the PCA analysis revealed that four main factors account for up to 87.8% of the cumulative variance. PC1 included Al, Na, K, Fe, Mg, Pb, Zn, Ti, and EC that accounted for 38.1% of the total variance. This factor is suggestive of mixed sources from road traffic, with diesel particles (Fe, Mg, Na, Pb, Zn, and EC) and road dust (Al, Na, and Ti) together with contributions from the industrial sector (Fe, Pb, and Zn) [27,64]. PC2 included Ba, Pb, TC, OC, and EC that accounted for 28.6%. This is similar to PC2 during the dry season, representing common sources from biomass fires and crustal dust sources which release nano-size range particles [65–67]. PC3, which was loaded with Cu, suggests that contributions from non-exhaust traffic sources, i.e., brakes and tire wear [54] accounted for 14.0%. PC4 is comprised of Pb, Cr, and Ti that accounted for around 9.0% indicating that Ba, Cr, and Ti are derived from construction dust [54].

3.5.2. TSP Source Apportionment

As shown in Table 6, PCA revealed the factors that are important for each season. To interpret the results obtained from the dry and wet seasons, the dry season was extracted, accounting for 4 main factors and the wet season accounted for 3 factors.

In the dry season, 4 PC factors account for 83.2% of the total variance. PC1 accounts for 38.5% and is derived from mixed sources, namely, traffic emissions, the industrial sector, and road soil dust [64]. It represents most of the metals including Ba, K, Pb, Mn, Ti, and Zn. In PC2, accounting for 20.6%, Al, Cr, Mn, Na, and Ti represent markers for crustal elements. In PC3, accounting for 15.3%, TC, OC, and EC appear to arise from biomass combustion. PC4, accounting for 8.7%, is associated with Pb and EC from vehicle emissions [66].

During the wet season, PCA revealed that 3 main factors account for up to 86.9% of cumulative variance. PC1, accounting for 57.2% of the variance, includes all of the metals except Cu (K, Mn, Mg, Al, Ti, Fe, Na, and Ni); this cluster represents mixed sources including road and soil dust [13], as well as diesel exhaust (Mn, Mg, Al, Zn, and Fe) [27]. A considerable amount of road dust is also released by the mechanical abrasion of the road pavement, which contains high levels of Fe, Mg, and Al [67]. PC2 has in height loading factor (more than 0.8) of TC and OC as tracers of biomass burning, which contribute around 20.5% [7]. PC3 contributes 9.2% from vehicular emission with a high loading of Pb and EC [13].

Table 6. Principal components in the TSP fraction by season.

Species	Dry Season (Jan–Apr)				Wet Season (May–Aug)		
	PC1	**PC2**	**PC3**	**PC4**	**PC1**	**PC2**	**PC3**
Al	0.38	0.78			0.85	0.46	
Ba	0.86				0.91		
Cr	0.46	0.69		−0.36	0.96		
Cu	0.33	0.30	−0.55		0.49	−0.67	0.40
Fe	0.90				0.95		0.02
K	0.88	−0.33			0.70	−0.50	−0.31
Mg	−0.35	0.84		0.32	0.95		
Mn	0.97				0.83		−0.32
Na	−0.57	0.72		0.31	0.82	0.34	
Ni	0.42	0.40			0.93		
Pb	0.62			0.51	0.68		0.50
Ti	0.75	0.54			0.94		−0.08
Zn	0.88				0.66	−0.39	0.29
TC			0.93			0.91	0.28
OC			0.89			0.90	−0.06
EC			0.46	0.74	0.35	0.45	0.69
Initial eigenvalue	6.16	3.30	2.46	1.40	9.15	3.29	1.47
% Variance	38.52	20.63	15.35	8.75	57.18	20.56	9.20
% Cumulative Variance	38.52	59.15	74.50	83.25	57.18	77.74	86.94

Factor loads < 0.3 are left in the blanks and factor loads > 0.5 are presented in bold.

3.6. Possible Local and Long-Range Transport of PM

The HYSPLIT-4 model was used to study the potential long-term transport of particle-bound carbon and trace elements at the monitoring site [19]. To detect the influence of long-range transportation of PM-bound chemicals, high levels were considered for this analysis. The transport times were five days (26–31 August 2019) during the wet season. Figure 3 demonstrates the backward trajectories during high PM concentrations for the above-mentioned times. As shown in Figure 3, the 120-h air mass originated from the northern part of Sumatra Island, Indonesia, where peat-land fires were reported by researchers [68]. Air masses moved from southeast Thailand which contributed to high TC and OC concentrations detected at the study site.

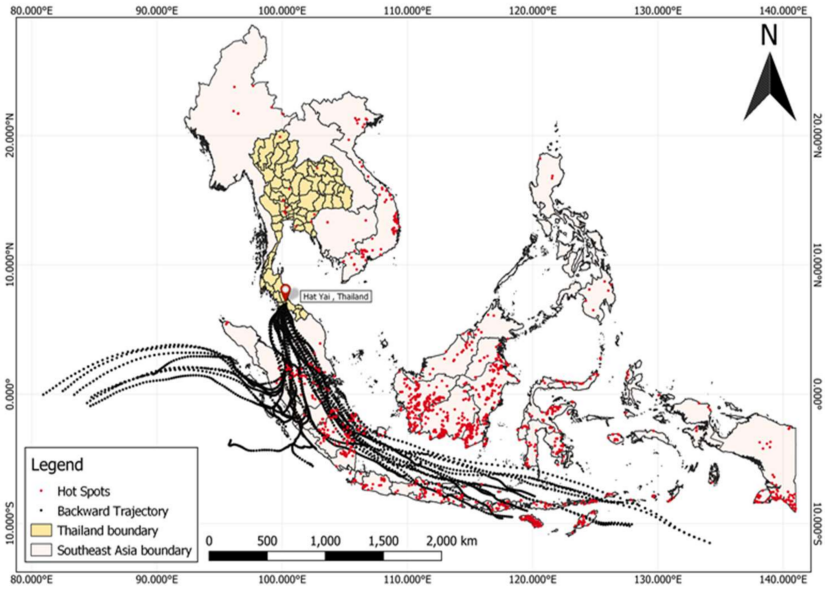

Figure 3. Distribution of hot spots and air mass trajectories during the high episode (August 2019).

4. Conclusions

In this study, particle-bound OC and EC, and Char-EC and Soot-EC as well as trace elements in the $PM_{0.1}$ and TSP fractions were investigated from January–August 2019 in southern Thailand. The possible emission sources of $PM_{0.1}$ and TSP based on analyses for carbon and metals were evaluated. The averaged $PM_{0.1}$ and TSP mass concentrations were found to be 10.4 ± 1.2 and 58.3 ± 7.8 $\mu g/m^3$, respectively. The highest concentrations of PM occurred in the wet season with the corresponding values for the dry season being lower. The OC to EC ratio for $PM_{0.1}$ and TSP was somewhat similar between the seasons, i.e., 2.5–2.7 ($PM_{0.1}$) and 3.8–4.2 (TSP). The char to soot ratio was consistently less than 1 for both the $PM_{0.1}$ and TSP fractions, indicating that land transportation appears to be the main emission source. PCA showed that road transportation (combustion and non-combustion), the industrial sector, and biomass burning are the main sources. These findings emphasize the importance of focusing emission control strategies on particles in southern Thailand, as well as elsewhere. However, cross-border pollution over countries in this area could also be a significant factor that merits future study in a more detailed investigation.

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