



Long-range Transboundary Atmospheric Transport of Polycyclic Aromatic Hydrocarbons, Carbonaceous Compositions, and Water-soluble Ionic Species in Southern Thailand

Chomsri Choochuay¹, Siwatt Pongpiachan^{2*}, Danai Tipmanee³, Woranuch Deelaman¹, Oramas Suttinun¹, Qiyuan Wang⁴, Li Xing⁴, Guohui Li⁴, Yongming Han⁴, Jittree Palakun⁵, Saran Poshyachinda⁶, Suparerk Aukkaravittayapun⁶, Vanisa Surapipith⁶, Junji Cao⁴

¹ Faculty of Environmental Management, Prince of Songkla University Hat-Yai Campus, Songkla 90112, Thailand

² NIDA Center for Research & Development of Disaster Prevention & Management, School of Social and Environmental Development, National Institute of Development Administration (NIDA), Bangkok 10240, Thailand

³ Faculty of Technology and Environment, Prince of Songkla University, Phuket 83120, Thailand

⁴ SKLLQG and Key Lab of Aerosol Chemistry & Physics, Institute of Earth Environment, Chinese Academy of Sciences (IEECAS), Xi'an 710061, China

⁵ Faculty of Education, Valaya Alongkorn Rajabhat University under the Royal Patronage (VRU), Pathumthani 13180, Thailand

⁶ National Astronomical Research Institute of Thailand (Public Organization), Chiang-Mai 50180, Thailand

ABSTRACT

This study investigated atmospheric particulate matter (PM) with an aerodynamic diameter of $< 2.5 \mu\text{m}$ ($\text{PM}_{2.5}$) observed at the Prince of Songkla University (Phuket Campus) in southern Thailand. All samples ($n = 75$) were collected using MiniVol™ portable air samplers from March 2017 to February 2018. Carbonaceous aerosol compositions, i.e., organic carbon (OC) and elemental carbon (EC), water-soluble ionic species (WSIS), and polycyclic aromatic hydrocarbons (PAHs) in the $\text{PM}_{2.5}$ samples were identified and quantified. We found that the average $\text{PM}_{2.5}$ concentration was $42.26 \pm 13.45 \mu\text{g m}^{-3}$, while the average concentrations of OC and EC were 3.05 ± 1.70 and $0.63 \pm 0.58 \mu\text{g m}^{-3}$, respectively. The OC/EC ratio was in the range of 2.69–16.9 (mean: 6.05 ± 2.70), and the average concentration of 10 selected ions was $6.91 \pm 3.54 \mu\text{g m}^{-3}$. The average concentration of SO_4^{2-} was the highest throughout the entire study period ($2.33 \pm 1.73 \mu\text{g m}^{-3}$); the average contribution of SO_4^{2-} to the major ionic components was 34%. Surprisingly, the average concentrations of NO_3^- and NH_4^+ were relatively low. The mean ratio of $[\text{NO}_3^-]/[\text{SO}_4^{2-}]$ was 0.33 ± 0.24 . Strong positive correlation was found between K^+ and both OC and EC ($r = 0.90$ and $r = 0.93$, respectively). It is also precious to highlight that biomass burning (BB) is the major source of OC, EC and K^+ , which multiple studies have confirmed that the role of K^+ as a biomass marker. Results showed that BB episodes might play a major role in producing the observed high levels of OC. The relatively high abundance of both B[g,h,i]P and Ind suggests that motor vehicles, petroleum/oil combustion, and industrial waste burning are the primary emission sources of PAHs in the ambient air of Phuket. Interestingly, principal component analysis (PCA) indicated that vehicular exhausts are the main source of carbonaceous aerosol compositions found in the ambient air of Phuket, whereas the contributions of biomass burning, diesel emissions, sea salt aerosols and industrial emissions were also important.

Keywords: $\text{PM}_{2.5}$; PAHs; Carbonaceous compositions; Water soluble ionic species; Biomass burning.

INTRODUCTION

Although air pollution is primarily an urban phenomenon, it is an important problem globally. In population centres

such as Thailand, large quantities of fuel are consumed in various economic sectors, for e.g., industry (Gocht *et al.*, 2001; Vicente and Alves, 2018; Salma *et al.*, 2020), transportation (Silva, 2005; Zhang *et al.*, 2014; Lin *et al.*, 2019), and electricity generation (Dung, 1996; Chen *et al.*, 2020). Combustion of fossil fuels such as coal and petroleum is responsible for causing the majority of air pollution (Sookkai *et al.*, 2000; Vicente and Alves, 2018; Salma *et al.*, 2020). Air pollution in the form of dust, especially particulate matter (PM) with an aerodynamic diameter of $< 2.5 \mu\text{m}$ ($\text{PM}_{2.5}$), is

* Corresponding author.

Tel.: 00 66 2 727 3113; Fax: 00 66 2 732 0276

E-mail address: pongpiapun@gmail.com

among the most dangerous. This is because it can affect the human respiratory system (Wheeler *et al.*, 2006; Doiron *et al.*, 2019; Lelieveld *et al.*, 2019; Nhung *et al.*, 2019), exacerbating conditions such as bronchitis, influenza, pneumonia, emphysema, and asthma, especially in children, the elderly, and people with underlying cardiopulmonary/respiratory diseases (Jinsart *et al.*, 2002; Cohen *et al.*, 2017; Lelieveld *et al.*, 2019).

Carbonaceous aerosols have been studied thoroughly over recent decades because they can affect human health, ecosystems, and the climate system (Shih *et al.*, 2008; Chen *et al.*, 2017; Pani *et al.*, 2018). Another major concern is that they are persistent organic pollutants that can remain in the environment for long periods (Jones and Voogt, 1999; Dachs and Eisenreich, 2000; Al-Mulali *et al.*, 2015; Bakirtas and Akpolat, 2018). Several studies have investigated the presence of carcinogenic and/or mutagenic substances in the atmosphere, derived via gas-particle partitioning, e.g., polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls, the origin of which is incomplete combustion attributable to both natural and anthropogenic sources (Zhang *et al.*, 2014; Achten and Andersson, 2015; Wincent *et al.*, 2016; Bocchi *et al.*, 2017; Idowu *et al.*, 2019). These substances, which are classified as semi-volatile compounds, can be released as solid material or vapour that can adhere to the surface of other particles (Smith and Harrison, 1998; Jones and Voogt, 1999; Dachs and Eisenreich, 2000; Schummer *et al.*, 2010; Lawal, 2017). Thus, they can spread from their source via many media, posing a danger to human health and the ecosystem. Therefore, measurement of the concentration of these carbonaceous aerosols is highly important.

Over the past few decades, BB and traffic emissions have been extensively evaluated in the northern and central parts of Thailand its release large amounts of particulate matter, including, OC-EC, WSIS and PAHs that increased environmental pollution (Chaiyo *et al.*, 2011, 2013; Duangkaew *et al.*, 2013; Pongpiachan, 2013; Pongpiachan *et al.*, 2013; Tsai *et al.*, 2013; Chaiyo and Garivait, 2014; Pongpiachan *et al.*, 2014a, b; Janta and Chantara, 2017; Pongpiachan *et al.*, 2017; Pani *et al.*, 2018; Thepnuan *et al.*, 2019; Choochuay *et al.*, 2020). In Thailand, information on PAHs, carbonaceous compositions, i.e., organic carbon (OC) and elemental carbon (EC), and water-soluble ionic species (WSIS) in the ambient air of southern parts of the country is rare. Previous study of carbonaceous aerosols in the coastal city of Hat-Yai (southern Thailand) found that aged marine aerosols from long-range transportation and/or particles from biomass burning (BB) made a major contribution to the carbonaceous aerosols measured at the top of a building in the study area (Pongpiachan *et al.*, 2009, 2013). Therefore, this study selected an observation site at the Prince of Songkla University (Phuket Campus) in southern Thailand to investigate atmospheric PM_{2.5}. Phuket is the largest island in Thailand. It is located in the south and encircled by the Andaman Sea. It has long slender shape with north-south orientation. In addition, Phuket has several other large and small satellite islands. Approximately 70% of the land area is mountainous, while the remaining 30% comprises plains. The climate of Phuket is warm and moist throughout the year.

The first unambiguous evidence that the air pollution seen frequently in fine atmospheric particles is caused by human activities became available several decades ago. Comprehension of the composition and major sources of carbonaceous aerosols is important for improving air quality. Therefore, the objective of this study was to determine the characteristics of OC, EC, WSIS, and PAHs in the PM_{2.5} samples obtained at the study site. The analysis focused primarily on the following: (i) characterization of the chemical compounds detected in the PM_{2.5} samples, (ii) statistical analysis of the chemical composition and its relation to source identification, and (iii) statistical source apportionment of the chemical composition, including OC, EC, WSIS, and PAHs.

MATERIALS AND METHODS

Air Quality Observatory Sites

The aerosol sampling campaign was undertaken at Building 6 of the Prince of Songkla University (Phuket Campus) in Thailand (Fig. 1). Phuket, the largest island in Thailand, is in the south and surrounded by the Andaman Sea. The main island has long slender shape with north-south orientation and it has several other large and small satellite islands. Around 70% of the land area is mountainous, while the remaining 30% comprises plains. The climate of Phuket is warm and moist throughout the year. The MiniVol™ air samplers were installed on the rooftop of Building 6 (4th Floor): 7.89318°N, 98.35209°E (GPS coordinates: 7°53'35.5"N, 98°21'07.5"E). The monitoring campaign was conducted from March 2017 to February 2018.

Samples of PM_{2.5} ($n = 75$) were obtained using MiniVol™ portable air samplers (Airmetrics, USA) with 47-mm quartz filters and a flow rate of 5 L min⁻¹. All samples were collected over 72-h periods. All PM_{2.5} samples were stored carefully in individual petri slide dishes and refrigerated to retain their chemical composition until required for further analysis. The quartz-fibre filter samples were divided into two segments. One of the filters was analyzed for OC-EC, and the other one was analyzed for PAHs and WSIS.

Chemical Analysis

Carbonaceous Aerosol Analyses: Organic Carbon (OC) and Elemental Carbon (EC)

The measurements of carbonaceous aerosol compositions including calibration and quality assurance/quality control (QA/QC) processes were performed at the laboratory of the Institute of Earth Environment, Chinese Academy of Science (Xian, China). The protocols adopted were the same as reported previous by Chow *et al.* (2007). Normally, the OC content was considered as the sum of individual OC fractions (i.e., OC1 + OC2 + OC3 + OC4) and the EC content was considered as the sum of individual EC fractions (i.e., EC1 + EC2 + EC3 + OP), based on the IMPROVE_A thermal optical reflectance protocol (Fung *et al.*, 2002; Chow *et al.*, 2007).

Carbonate carbon was determined through assessment of CO₂ acidification from organic samples prior to the normal carbon analysis procedure. Seven temperatures were used for different fractions. The temperature protocol was applied

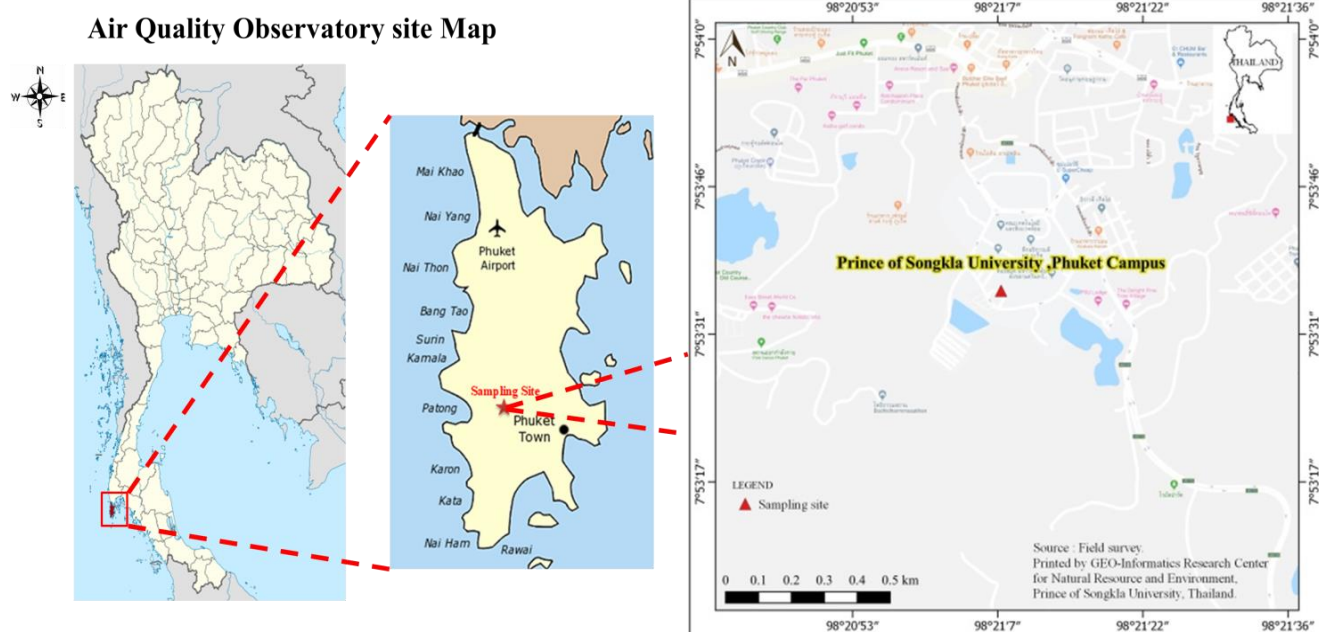


Fig. 1. Location of the sampling site used in this study.

to separate OC and EC in a process similar to the thermal optical reflectance and thermal optical transmittance pyrolysis correction. This protocol produces evaluations of total OC, total EC, and total carbon (TC), monitored by both reflectance and transmittance. For the QA/QC procedures that have been described elsewhere (Cao *et al.*, 2003), the instrument was calibrated daily with known quantities of methane. Replicate analyses were performed for each group for 10 samples and the relative deviation of the replicate analyses was < 5% for TC and < 10% for both OC and EC.

Water-soluble Ionic Species (WSIS)

The concentrations of WSIS included five cations (i.e., Na^+ , NH_4^+ , K^+ , Mg^{2+} , and Ca^{2+}) and five anions (i.e., Cl^- , F^- , NO_2^- , NO_3^- , and SO_4^{2-}). An ion chromatograph with a separation column was used for the extraction from all $\text{PM}_{2.5}$ samples. The QA/QC procedure for this analysis required all glassware to undergo ultrasonic cleaning and oven drying at 450°C for approximately 6 h. All solvents used in the analysis procedure were pesticide residue grade (Wang *et al.*, 2005).

Polycyclic Aromatic Hydrocarbons (PAHs)

The concentrations of PAHs in the $\text{PM}_{2.5}$ samples were measured using in-injection port thermal desorption coupled with gas chromatography/mass spectrometry, which quantified the concentration of 19 PAHs as non-polar organic compounds. This analytical procedure is similar to the alternative method of traditional solvent extraction followed by gas chromatography/mass spectrometry analysis. The analytical procedures have been described in previous studies (Ho and Yu, 2004).

Statistical Analysis

This study used the SPSS System for Windows Version 22 to produce descriptive statistics (minimum, maximum, mean, and standard deviation) of the measured concentrations

of PAHs, carbonaceous compositions, and WSIS. We also used PCA for identification of source appointment.

RESULTS AND DISCUSSION

Concentrations of Total Carbon (TC), Organic Carbon (OC), and Elemental Carbon (EC)

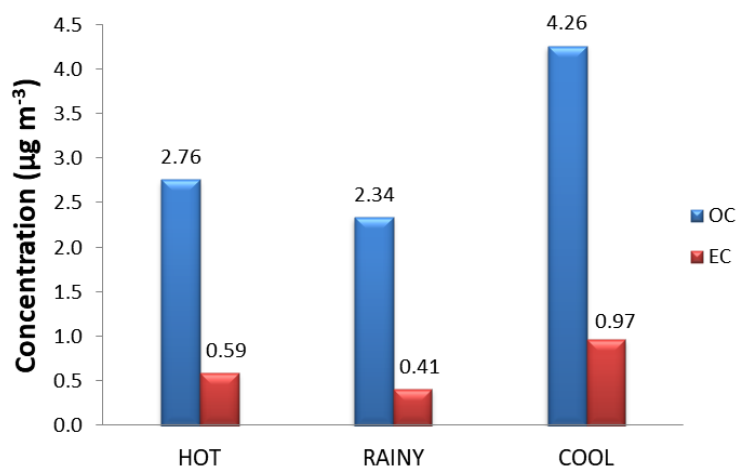
The average concentrations of each carbon fraction for OC, EC, TC, and $\text{PM}_{2.5}$ in the samples from Phuket are presented in Table 1, and the concentrations of OC and EC in each individual sample are shown in Fig. 2.

Seinfeld and Pandis (2006) reported that the highest carbonaceous fraction of fine atmospheric PM is OC at 70–80%, followed by EC and inorganic carbon at 5%. The average concentrations of carbonaceous chemical components found in our samples are listed in Table 1. It can be seen that of the OC fractions, OC3 was the highest, followed in descending order by OC4, OC2, and OC1. For the EC fractions, EC1 was the highest, followed by EC2 and EC3. In characterizing the chemical composition of aerosols in northern Indochina in March and April 2010, Chuang *et al.* (2013) found OC3 to be a reasonable tracer of BB, whereas OC2 is known as a tracer of both coal combustion (Chow *et al.*, 2004) and vehicular exhausts (Cheng *et al.*, 2015).

In observations of ambient air throughout an entire year in Phuket, the OC fraction was found to be the major component because it is released directly into the ambient air following incomplete combustion of organic compounds (Jimenez *et al.*, 2008). It can be emitted directly from various sources such as industrial processes and natural occurrences, e.g., BB (primary OC) or it can be formed from gas-particle partitioning in the air (secondary OC: SOC). It is well known that OC can have substantial impact on human health (Mauderly and Chow, 2008). Conversely, the EC fraction was found to be much lower than the OC fraction.

Table 1. Concentrations of OC, EC, TC, and PM_{2.5} samples from Phuket, Thailand.

Species	Minimum ($\mu\text{g m}^{-3}$)	Maximum ($\mu\text{g m}^{-3}$)	Mean \pm SD ($n = 75$)
OC1	0.00	0.22	0.04 ± 0.05
OC2	0.22	1.56	0.53 ± 0.23
OC3	0.73	5.34	1.57 ± 0.75
OC4	0.12	3.37	0.71 ± 0.62
OC	1.08	10.90	3.05 ± 1.70
EC1	0.10	3.74	0.70 ± 0.64
EC2	0.00	0.28	0.13 ± 0.06
EC3	0.00	0.00	0.00 ± 0.00
EC	0.09	3.90	0.63 ± 0.58
TC	1.17	14.80	3.67 ± 2.25
PM _{2.5}	20.07	91.02	42.26 ± 13.45

**Fig. 2.** Average concentration of seasonal variation of OC and EC in PM_{2.5} samples collected in Phuket.

As the chemical structure of EC is similar to that of impure graphite, it appears reasonable to assume that vehicular exhausts are a major source of EC. Consequently, the most important sources of EC are fossil fuel combustion and/or BB (Gelencsér, 2004).

The mean values of OC and EC in the PM_{2.5} samples of this study were 3.05 ± 1.70 and $0.63 \pm 0.58 \mu\text{g m}^{-3}$, respectively. These values are much smaller in comparison with those from other areas. However, the average mean concentrations of OC and EC determined in this study are similar to those reported in autumn and winter in Cape Hedo, Okinawa (Kunwar and Kawamura, 2014). Generally, EC is released from any combustion source and it is usually used as a tracer of primary OC (Turpin and Huntzicker, 1995). Hence, the relationship between OC and EC can be used to estimate the source of carbonaceous particles. The relationship between OC and EC in the PM_{2.5} samples obtained in Phuket in this study is illustrated in Fig. 3. The large R^2 values (0.86) have been found in this study indicated that the impact of local primary sources (traffic and biomass burning) have a big role in Phuket's atmosphere.

OC/EC Ratios and Secondary Organic Carbon (SOC)

Contributions

OC/EC Ratios

Carbonaceous compounds represent a significant fraction

of atmospheric aerosols, accounting for 20–35% of PM₁₀ and 20–45% of PM_{2.5} (Yttri *et al.*, 2007; Putaud *et al.*, 2010). The OC/EC ratio is applied frequently to explain the emission sources of carbonaceous aerosol compounds (Han *et al.*, 2007, 2009; Wu *et al.*, 2019; Xing *et al.*, 2020). In our study, the OC/EC ratios determined in this study were in the range of 2.69–16.9 with an annual mean value of 6.05 ± 2.70 . The season averaged OC/EC ratios are 4.94 (hot), 6.84 (rainy), and 5.70 (cool) (Fig. 4).

The measurement of atmospheric PM_{2.5}-bound carbonaceous aerosol composition widely studied in Thailand, especially in the northern and the central part of Thailand. In this study, the annual mean OC/EC ratios value is 1.1 times lower than the value reported from Chiang-Mai, Thailand (Choochuay *et al.*, 2020). Most of the time previous studies the availability of data from southern Thailand is limited. The chemical characteristics of carbonaceous aerosols and PAHs of PM₁₀ in the city of Hat-Yai in southern Thailand have been studied by Pongpiachan *et al.* (2014a). Their study suggested that the persistence of OC/EC ratios could have been attributable from BB, vehicular, industrial emissions, and/or long-range transportation and agricultural waste burning aerosols. The OC/EC ratio can be used to estimate the primary sources of pollution. Several studies on carbonaceous PM in different parts of the world have reported that high OC/EC ratios are related to SOC (Chow *et al.*, 1993; Turpin

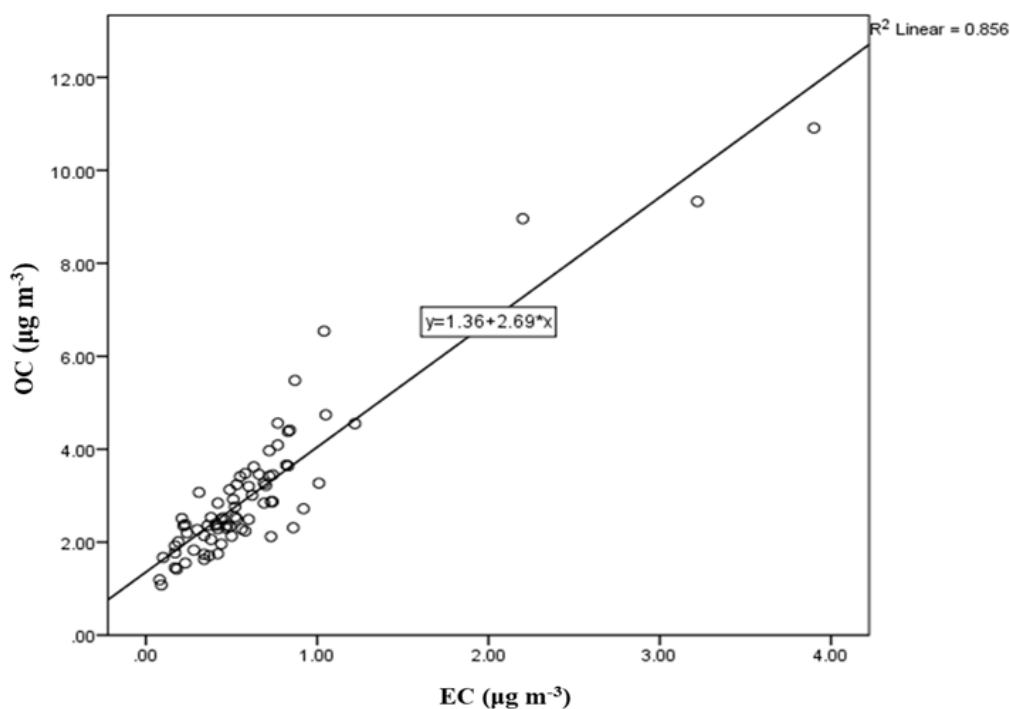


Fig. 3. EC vs. OC correlation in the PM_{2.5} samples collected in Phuket during March 2017 to February 2018.

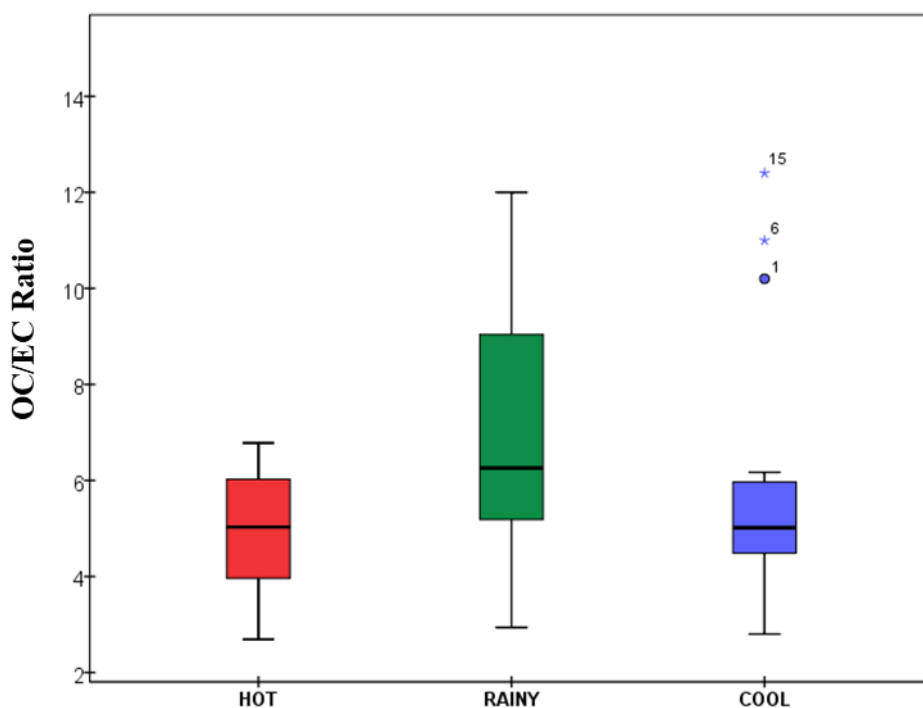


Fig. 4. The OC/EC ratios obtained in Phuket during March 2017 to February 2018.

and Huntzicker, 1995). Carbonaceous aerosols with OC/EC values > 2 can be considered to contain significant quantities of SOC, which larger OC/EC values are also attributed to (i) biogenic emissions, (ii) BB aerosols (Wu *et al.*, 2019; Kalita *et al.*, 2020; Kaskaoutis *et al.*, 2020). In this study, the range of OC/EC ratios was 2.69–16.9 (mean: 6.05 ± 2.70). However, a high value of the OC/EC ratio (12) was reported

by Cao *et al.* (2005) in aerosols derived from residential coal combustion. The result from recent study in Southeast Asia regions (SEA) reported that the Biomass burning and biogenic emissions were significantly larger compared to other regions in south Asia (Kalita *et al.*, 2020). However, the concentrations of carbonaceous compounds vary inter-regionally in relation to local emissions and weather (Heald *et al.*, 2008).

Estimation of Secondary Organic Carbon (SOC)

The SOC contribution can be estimated by measuring OC and EC concentrations and an appropriately selected primary OC to EC ratio. Many studies have used a widely accepted EC tracer method to measure SOC. Using this method, the contribution of SOC can be calculated based on the minimum values of OC/EC ratios, where EC is used as a measure of primary OC (Castro *et al.*, 1999). In this study, SOC was estimated using the following equation:

$$\text{SOC} = \text{OC}_{\text{total}} - \text{EC} \times (\text{OC/EC})_{\text{pri}} \quad (1)$$

where OC_{total} represents the total OC and $(\text{OC/EC})_{\text{pri}}$ is the mean of the three lowest OC/EC ratios.

The mean of the three lowest OC/EC ratios (2.79) was applied in this study to estimate the SOC content of the $\text{PM}_{2.5}$ samples. Based on this technique, it was determined that the annual mean value of SOC was $1.30 \pm 1.63 \mu\text{g m}^{-3}$ and the highest value was $2.82 \mu\text{g m}^{-3}$. The percentage contribution of SOC to OC_{total} was 42.6% in this study. This value is 1.4 times lower than the value (59.2%) detected in Okinawa, Japan (Kunwar and Kawamura, 2014) and 1.5 times lower than both the value (67.8%) reported for Hat-Yai, Thailand (Pongpiachan *et al.*, 2014a) and the value (65%) found in Claremont, USA (Na *et al.*, 2004). Conversely, our value is 2.5 times higher than that observed in Birmingham, United Kingdom (Castro *et al.*, 1999). Our result is close to that found by Li *et al.* (2009) in their study conducted at a coastal site (37.7%), and similar to values observed in Kaohsiung in Taiwan (40%) by Lin and Tai (2001). The application of diagnostic binary ratios of OC/EC and estimations of secondary organic carbon (SOC) in this study highlighted that the enhanced impacts of incomplete combustion emissions, such as motor vehicle exhaust, fuel burning, and biomass burning, which can be remained in the atmosphere for several days (Wu *et al.*, 2019; Kaskaoutis *et al.*, 2020).

Atmospheric Concentrations of Water-soluble Ionic Species (WSIS) and PAHs in $\text{PM}_{2.5}$

Given that Phuket is the largest island in Thailand, it was considered important to examine the impact of marine aerosols on the characterization of carbonaceous compositions. The chemical characteristics of WSIS have been studied

thoroughly in different areas of the world. Several studies have reported that SO_4^{2-} and Cl^- are the main contributors to WSIS found in marine aerosols, whereas NH_4^+ and K^+ are the main contributors to WSIS in aerosols attributable to BB (Kocaka *et al.*, 2007; Park and Cho, 2011).

The individual and average concentrations of 10 selected ions (SO_4^{2-} , Na^+ , Ca^{2+} , Cl^- , NO_3^- , NO_2^- , NH_4^+ , K^+ , Mg^{2+} and F^-) considered in this study are presented in Table 2.

Several previous studies have used diagnostic ratios to analyse the sources of marine aerosols and non-marine aerosols or non-sea-salt for WSIS (Karthikeyan and Balasubramanian, 2006). Previous work has reported that the sources of K^+ , SO_4^{2-} , and Ca^{2+} are not solely from marine aerosols (Wang and Shooter, 2001). Therefore, the contribution of each of these ions from non-sea-salt sources was calculated using the following equations (Hedge *et al.*, 2007; George *et al.*, 2008; Behrooz *et al.*, 2017):

$$\text{nss-SO}_4^{2-} = (\text{SO}_4^{2-}) - 0.2516 * (\text{Na}^+) \quad (2)$$

$$\text{nss-Ca}^{2+} = (\text{Ca}^{2+}) - 0.0385 * (\text{Na}^+) \quad (3)$$

$$\text{nss-K}^+ = (\text{K}^+) - 0.037 * (\text{Na}^+) \quad (4)$$

*Note, nss-SO_4^{2-} , nss-Ca^{2+} , and nss-K^+ can be used in the formulas above, assuming that marine aerosols are the same as sea-salt in terms of chemical composition. Meanwhile, Na^+ has been used as a marker for marine aerosols, by assuming that whole Na^+ comes from the marine source. (George *et al.*, 2008; Behrooz *et al.*, 2017).

Based on the OC/EC ratios in this study, long-range atmospheric transport of BB plumes from nearby countries could represent one source. In this region, BB is a widespread activity and it is known that PM is transported from Indonesia (Southeast Asia) into southern Thailand (Phairuang *et al.*, 2020). Moreover, strong correlation ($r = 0.94$) was found between nss-K^+ and OC, which was found related to long-range atmospheric transport and the influence of BB on organic aerosols during the cool period.

Previous study reported that SO_4^{2-} , K^+ , and NH_4^+ are the major fractions in the form of secondary inorganic aerosols and biomass burning. Moreover, WSIS of NH_4^+ , K^+ , Ca^{2+} , Na^+ were extracted from the $\text{PM}_{2.5}$ ambient air samples, which Na^+ , NH_4^+ , and Cl^- are mainly originated from aged

Table 2. Concentrations of water-soluble ionic species (WSIS) observed in the $\text{PM}_{2.5}$ samples from Phuket, Thailand.

Ionic Species	Mean ($\mu\text{g m}^{-3}$)	Min. ($\mu\text{g m}^{-3}$)	Max. ($\mu\text{g m}^{-3}$)	% mass of total ion content
F^-	0.09 ± 0.01	0.07	0.12	1
Cl^-	0.53 ± 0.28	0.32	2.47	8
NO_2^-	0.30 ± 0.19	0.00	0.73	4
NO_3^-	0.53 ± 0.21	0.00	1.62	8
SO_4^{2-}	2.33 ± 1.73	0.33	9.21	34
Na^+	1.47 ± 0.39	0.65	3.04	21
NH_4^+	0.29 ± 0.32	0.00	2.38	4
K^+	0.28 ± 0.24	0.00	1.58	4
Mg^{2+}	0.13 ± 0.03	0.06	0.24	2
Ca^{2+}	0.96 ± 0.14	0.66	1.37	14
Total	6.91 ± 3.54	-	-	-

sea salt and mixed industrial, whilst Mg^{2+} and Ca^{2+} are generally made from mineral dust (Dahari *et al.*, 2019). In this study, the average SO_4^{2-} concentration was the highest throughout the entire study period for all season (Fig. 5).

As previously mentioned, the most dominant species in this study were SO_4^{2-} , Na^+ and Ca^{2+} which mainly contributed from secondary inorganic aerosols, biomass burning, sea salt and mixed industrial for the ambient air in Phuket (Dahari *et al.*, 2019). The statistics showed that there were no obvious differences on F^- , Cl^- , NO_2^- and NO_3^- in all seasons, while SO_4^{2-} , Na^+ , Ca^{2+} , NH_4^+ , K^+ and Mg^{2+} were obvious differences between rainy and cool ($p > 0.05$).

For the classification, $[NO_3^-]/[SO_4^{2-}]$ ratios were applied carefully to identify the incidence of stationary sources (e.g., boilers industries, power plants, etc.) and mobile sources (e.g., vehicular exhausts) of nitrogen and sulphur. They are generally formed via atmospheric reactions of their gaseous phase, e.g., NO_x and SO_2 . Normally, SO_2 is released via coal combustion, whilst NO_x results from any type of combustion, e.g., coal power plants and vehicular emissions in aerosols (Liu *et al.*, 2011; Mkoma *et al.*, 2014 Javid *et al.*, 2015; Park *et al.*, 2015; Deng *et al.*, 2016; Huang *et al.*, 2016). A high $[NO_3^-]/[SO_4^{2-}]$ ratio (1.06) was found in a region with high levels of vehicular emissions (Li *et al.*, 2009). The mean $[NO_3^-]/[SO_4^{2-}]$ ratio found during the annual was 0.33 ± 0.24 , while the season averaged ratios were 0.22 (hot), 0.31 (rainy), and 0.17 (cool). It is lower than that found in other

areas in summer in China, e.g., Beijing (0.83), Tianjin (0.71), and Shijiazhuang (0.56) (Dao *et al.*, 2014). Hence, this result means that the local sources from vehicular emissions (tracers for NO_3^-) are limited and the ratio decreases, as sulfate has more regional sources. The high temperatures in Phuket modulate particulate nitrate into the gaseous phase, which reduces the $[NO_3^-]/[SO_4^{2-}]$ ratio (Cuccia *et al.*, 2013; Titos *et al.*, 2014; Dumka *et al.*, 2017). However, the ratio of 0.3–0.5 found in this study is also lower than that usually found in China because of the widespread use of sulphur-containing coal by the Chinese (Yao *et al.*, 2002).

The ions SO_4^{2-} and NH_4^+ are secondary ions that have a complex reaction in that NH_4^+ responds rapidly with SO_4^{2-} to the constant form of ammonium salts (Lai *et al.*, 2007; Li *et al.*, 2012; Wang *et al.*, 2013). Generally, SO_4^{2-} is influenced by anthropogenic sources in industrial areas. The concentration of SO_4^{2-} was significantly higher than that of Na^+ and Cl^- , whereas nss- SO_4^{2-} was the primary species for acid replacement (Zhang *et al.*, 2010). Similar to other ions with anthropogenic sources (e.g., NO_3^-), the correlation with those of nss- SO_4^{2-} was reasonable (Zhang *et al.*, 2010).

In general, Na^+ and Cl^- are the sea salt ions that form the largest fractions in marine aerosols. In this study, the highest concentrations of Na^+ and Cl^- were 1.47 ± 0.39 and $0.53 \pm 0.28 \mu g m^{-3}$, which accounted for 21.0% and 8.0% of the total ionic species, respectively. For marine aerosols, Zhang *et al.* (2010) reported that sea salt aerosols (i.e., NaCl) can

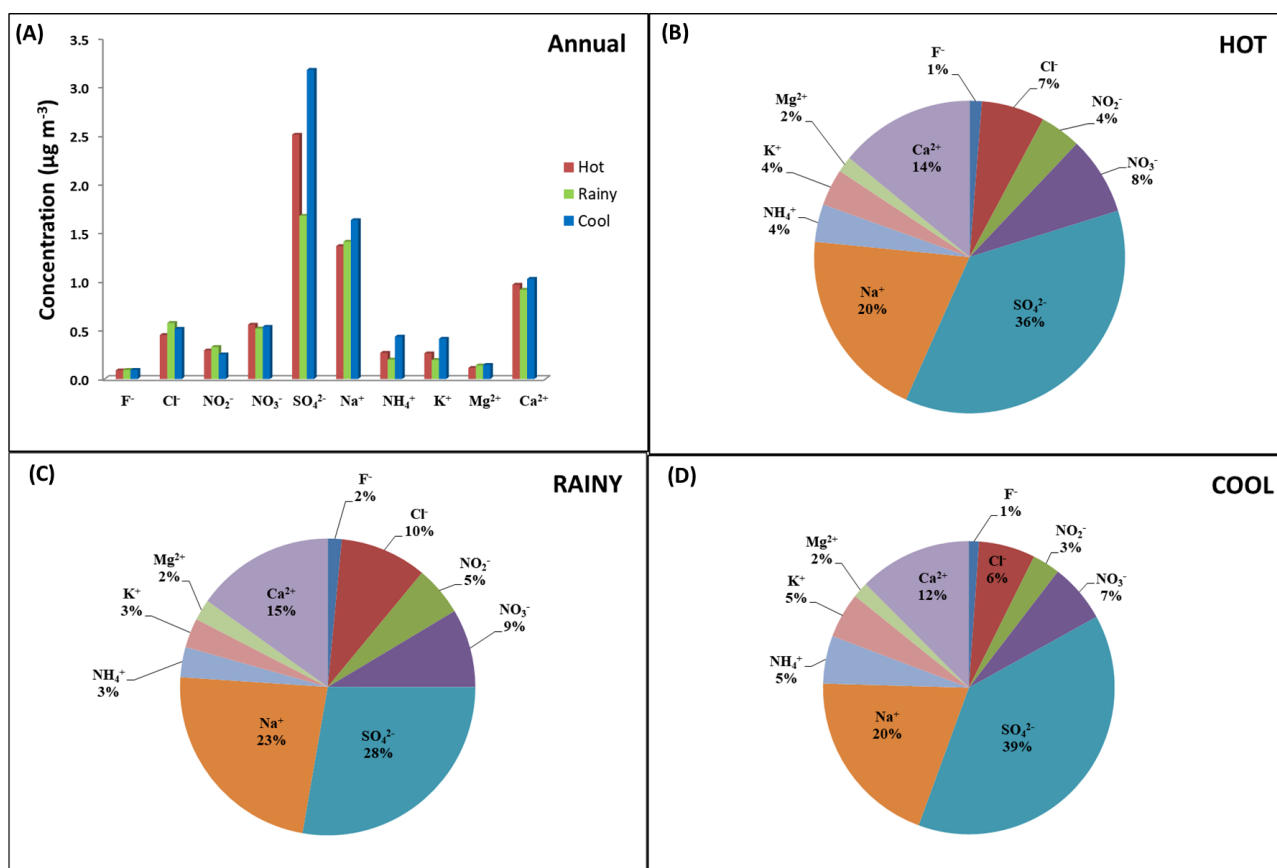


Fig. 5. (A) Annual concentration of individual WSIS, Percentage contributions of individual WSIS in (B) hot, (C) rainy and (D) cool season collected from Phuket.

emit HCl via exchange with sulphuric acid and nitric acid, which results in a shortage of Cl^- relative to Na^+ . The annual average equivalent ratios of Cl^-/Na^+ in the aerosols from Phuket were 3.4 and 3.2 times lower than those on Yongxing Island and those of seawater, respectively (Table 3). In our study, we find that there is chloride depletion is the result of the interaction of sea salt with acidic species, nitrate, sulfate followed by the losing of Cl^- in term of HCl gas (Stogiannidis and Laane, 2015). Moreover, the ratio of Mg^{2+} to Na^+ was 0.09, which is 2.3 and 2.4 times lower in comparison with the values from Yongxing Island and seawater, suggesting that the ratio of $\text{Mg}^{2+}/\text{Na}^+$ (Mg^{2+} loss) in $\text{PM}_{2.5}$ samples maybe due to the leach of magnesium chloride (MgCl_2), which is a component of bittern in sea salt.

Correlations of Chemical Composition of $\text{PM}_{2.5}$ and its Relation to Source Identification

Some ions in carbonaceous aerosol composition such as K^+ , SO_4^{2-} , and Ca^{2+} have multiple sources, e.g., ocean and land surfaces. Additionally, nss-SO_4^{2-} in the atmosphere can be derived from various sources. It originates from the combustion of fossil fuels such as coal, oil, and natural gas (Cuccia *et al.*, 2013; Kunwar and Kawamura, 2014; Titos *et al.*, 2014; Dumka *et al.*, 2017). In Phuket, we found the highest concentrations of carbonaceous aerosols found in OC and SO_4^{2-} were 3.05 and 2.33 $\mu\text{g m}^{-3}$, respectively. Several previous studies reported that SO_4^{2-} and Cl^- are the principal species of WSIS normally found in marine aerosols, whereas K^+ and NH_4^+ are the primary species associated with BB and agricultural waste burning (Matsumoto *et al.*, 1998; Kocaka *et al.*, 2007; Park and Cho, 2011; Pongpiachan *et al.*, 2014a). The correlations of OC, EC, and WSIS shown in Table 4. The results showed strong correlation between K^+ and both OC ($r = 0.89$) and EC ($r = 0.93$). It is well known that K^+ is a marker of BB (Kundu *et al.*, 2010), whereas EC is a marker of incomplete combustion of biomass and/or fossil fuel. We also found strong correlation between nss-K^+ and both EC ($r = 0.93$) and OC ($r = 0.89$); therefore, BB episodes might also play a major role in generating the higher OC concentrations. Previous analysis of satellite imagery revealed evidence of frequent BB episodes in southern Thailand, e.g., in preparation for agriculture, agricultural produce burning, and forest fires.

Among the ions measured in this study, NH_4^+ was

strongly correlated with K^+ ($r = 0.81$). It is assumed that one effect of BB was significant enrichment in $\text{PM}_{2.5}$. Previous studies related that fertilizer use as well as agriculture waste and related domestic activities are sources of gaseous ammonia emissions (Thepanondh *et al.*, 2005).

Concentrations of Polycyclic Aromatic Hydrocarbons (PAHs)

The concentrations of PAHs are summarized in Table 5. The total concentration of all 19 PAHs was $0.3780 \pm 0.3480 \text{ ng m}^{-3}$. The values determined in this study are lower than those measured in other areas of Thailand such as Chiang-Mai and Bangkok, which are known as heavily polluted areas (Pongpiachan, 2013; Pongpiachan *et al.*, 2014b).

Several previous studies have investigated the environmental cycle of PAHs in different environmental situations in Thailand (Pongpiachan, 2013; Pongpiachan *et al.*, 2014b, 2015). In northern Thailand, BB, forest fires, and agricultural waste burning during winter emit large quantities of PM into the atmosphere, especially ultra-fine particles that include $\text{PM}_{2.5}$ -bound PAHs (Vadrevu *et al.*, 2015, 2019). In central Thailand, vehicular emissions represent a major contributor to atmospheric PM. However, in southern Thailand, especially Phuket, the limited availability of PAH data makes it difficult to identify the sources of the pollution emitted into the atmosphere.

The concentrations of the individual PAHs in the $\text{PM}_{2.5}$ samples obtained in Phuket during March 2017 to February 2018 decreased in the following order: B[g,h,i]P > Ind > Phe > B[a]A > Cor > B[b]F > B[k]F > B[a]P > B[e]P > Ace > D[a,h]A > Fluo > Fl > Pyr > D[a,e]P > Chry > Ant > Per > B[a]F. Of the 16 priority PAHs identified by the United States Environmental Protection Agency, 9 are emitted via combustion processes such as those involving coal, diesel, and petroleum. Ravindra *et al.* (2008) reported that Flu, Pry, B[a]A, Chry, B[b]F, B[k]F, B[a]P, B[g,h,i]P, and Ind are combustion PAHs. The ratios of the concentrations of these combustion PAHs have been analysed in many studies to identify the sources of the PAHs in aerosols (Manoli *et al.*, 2004). In this study, high abundances of B[g,h,i]P and Ind were detected, indicating that motor vehicles, petroleum/oil combustion, and industrial waste burning are emission sources of the PAHs found in the ambient air of Phuket (Zhou *et al.*, 1999; Ravindra *et al.*, 2008).

Table 3. Comparison of equivalent ratios of ionic species in aerosols observed in Phuket, on Yongxing Island (Xiao *et al.*, 2017), and those in seawater (Keene *et al.*, 1986).

Ion ratios	Phuket Island	Yongxing Island	Seawater
Cl^-/Na^+	0.37	1.25	1.17
$\text{Mg}^{2+}/\text{Na}^+$	0.09	0.21	0.22
K^+/Na^+	0.19	0.048	0.021
$\text{Ca}^{2+}/\text{Na}^+$	0.68	0.62	0.044
$\text{SO}_4^{2-}/\text{Na}^+$	1.54	0.66	0.12
$\text{nss-SO}_4^{2-}/\text{Na}^+$	1.33	0.54	-
$\text{NO}_3^-/\text{Na}^+$	0.37	0.18	-
$\text{NH}_4^+/\text{Na}^+$	0.20	0.022	-
$\text{NO}_3^-/\text{nss-SO}_4^{2-}$	0.27	0.34	-
$\text{NH}_4^+/\text{nss-Ca}^{2+}$	0.32	0.038	-

Table 4. Pearson correlation analysis of OC, EC, and WSIS in PM_{2.5} samples obtained from Phuket during March 2017 to February 2018.

	F ⁻	Cl ⁻	NO ₂ ⁻	NO ₃ ⁻	SO ₄ ²⁻	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺	nss-SO ₄ ²⁻	nss-Ca ²⁺	nss-K ⁺	OC	EC
F ⁻	1														
Cl ⁻	0.13	1													
NO ₂ ⁻	0.24*	0.00	1												
NO ₃ ⁻	0.17	0.15	0.52**	1											
SO ₄ ²⁻	-0.16	-0.07	-0.02	0.55**	1										
Na ⁺	-0.01	0.58**	-0.05	0.40**	0.61**	1									
NH ₄ ⁺	0.07	-0.12	0.20	0.67**	0.73**	0.27*	1								
K ⁺	-0.05	-0.08	0.14	0.50**	0.79**	0.39**	0.81**	1							
Mg ²⁺	0.13	0.33**	0.03	0.47**	0.46**	0.71**	0.37**	0.32**	1						
Ca ²⁺	0.00	0.15	0.02	0.34**	0.72**	0.60**	0.52**	0.65**	0.50**	1					
nss-SO ₄ ²⁻	-0.16	-0.11	-0.01	0.55**	0.99**	0.58**	0.74**	0.79**	0.44**	0.71**	1				
nss-Ca ²⁺	0.00	0.10	0.03	0.32**	0.69**	0.53**	0.53**	0.65**	0.45**	0.99**	0.68**	1			
nss-K ⁺	-0.05	-0.12	0.14	0.49**	0.77**	0.34**	0.82**	0.99**	0.28*	0.63**	0.78**	0.63**	1		
OC	-0.07	-0.12	0.06	0.41**	0.73**	0.39**	0.72**	0.89**	0.28*	0.59**	0.73**	0.58**	0.89**	1	
EC	-0.04	-0.15	0.10	0.46**	0.77**	0.36**	0.78**	0.93**	0.25*	0.58**	0.78**	0.58**	0.93**	0.93**	1

t-test is < 0.01 for the correlation where r is > 0.70.

Table 5. Summary of PAH concentrations in Phuket, Thailand.

PAH (ng m ⁻³)	Mean	SD	Min.	Max.
Ace	0.0140	0.0096	0.0015	0.0507
Fl	0.0112	0.0079	0.0082	0.0270
Phe	0.0409	0.0411	0.0261	0.0711
Ant	0.0067	0.0045	0.0059	0.0160
Fluo	0.0120	0.0120	0.0070	0.0221
Pyr	0.0110	0.0127	0.0062	0.0160
B[a]A	0.0340	0.0163	0.0206	0.0581
Chry	0.0067	0.0070	0.0028	0.0099
B[b]F	0.0239	0.0220	0.0228	0.0464
B[k]F	0.0238	0.0229	0.0086	0.0340
B[a]F	0.0032	0.0030	0.0025	0.0057
B[e]P	0.0144	0.0134	0.0056	0.0169
B[a]P	0.0174	0.0190	0.0072	0.0224
Per	0.0048	0.0058	0.0029	0.0061
Ind	0.0507	0.0500	0.0359	0.0652
B[g,h,i]P	0.0575	0.0590	0.0348	0.0709
D[a,h]A	0.0133	0.0118	0.0243	0.0032
Cor	0.0239	0.0208	0.0185	0.0356
D[a,e]P	0.0085	0.0091	0.0069	0.0129
ΣPAHs*	0.3780	0.3480	-	-

*ΣPAHs is the sum of Ace, Fl, Phe, Ant, Fluo, Pyr, B[a]A, Chry, B[b]F, B[k]F, B[a]F, B[e]P, B[a]P, Per, Ind, B[g,h,i]P, D[a,h]A, Cor, and D[a,e]P

Principal Component Analysis (PCA)

We used PCA to identify potential sources of the carbonaceous and WSIS compositions of the PM_{2.5} samples. The PCA method is a multivariate procedure that links multivariate data reduction by transforming the data into rectangular components. Hence, PCA reduces multidimensional data into smaller dimensions (Wold *et al.*, 1987). In this section, source identification coupled with quantitative source apportionment of targeted chemical species is considered using PCA.

In this study, the concentrations of OC, EC, WSIS, and 19 individual PAHs from 75 samples were collected as active

variables. The majority of the variance (82.8%) of the scaled data was explained by five eigenvectors/principal components (PCs) (Table 6). The first PC (PC1) accounts for 55.5% of the total variance, while the second PC (PC2) explains 10.9% of the total variance, followed by PC3–PC5 that describe 10.6%, 5.2%, and 3.7% of the total variance, respectively.

In accounting for 55.5% of the total variance, PC1 showed high loading of B[g,h,i]P, Cor, Ind, B[e]P, B[b]F, D[a,h]A, B[a]F, Pyr, B[a]P, B[k]F, Fluo, Chry, D[a,e]P, and Ant with corresponding correlation coefficients of 0.946, 0.938, 0.936, 0.893, 0.886, 0.874, 0.852, 0.850, 0.849, 0.835, 0.795, 0.774, 0.761, and 0.623, respectively. Anthropogenic activity

Table 6. Rotated component matrix^a of carbonaceous compounds, WSIS, and 19 individual PAHs in the PM_{2.5} samples from Phuket, Thailand.

Compositions	Principal Component (PC)				
	PC1	PC2	PC3	PC4	PC5
TC	0.423	0.854	0.139	0.033	−0.005
OC	0.441	0.824	0.157	0.041	−0.023
EC	0.345	0.895	0.080	0.007	0.050
F [−]	−0.225	−0.002	−0.038	0.052	0.618
Cl [−]	−0.050	−0.218	−0.015	0.810	0.060
NO ₂ [−]	0.229	−0.050	−0.014	−0.055	0.774
NO ₃ [−]	0.191	0.387	0.215	0.284	0.694
SO ₄ ^{2−}	0.315	0.754	0.198	0.297	−0.020
Na ⁺	0.155	0.308	0.101	0.883	−0.043
NH ₄ ⁺	0.312	0.734	0.265	0.022	0.345
K ⁺	0.424	0.845	0.127	0.090	0.077
Mg ²⁺	−0.013	0.299	0.154	0.718	0.211
Ca ²⁺	0.130	0.625	0.291	0.452	−0.071
Ace	0.259	0.092	0.838	0.053	0.011
Fl	0.119	0.486	0.663	0.017	0.287
Phe	0.408	0.478	0.614	0.075	0.212
Ant	0.623	0.336	0.595	0.079	0.069
Fluo	0.795	0.465	0.324	0.030	0.041
Pyr	0.850	0.436	0.218	0.014	0.046
B[a]A	0.257	0.187	0.780	0.178	−0.127
Chry	0.774	0.571	0.202	0.015	0.026
B[b]F	0.886	0.322	0.171	−0.013	0.014
B[k]F	0.835	0.379	0.176	0.018	−0.021
B[a]F	0.852	0.290	0.233	0.007	0.114
B[e]P	0.893	0.328	0.231	0.032	0.008
B[a]P	0.849	0.399	0.244	0.061	0.015
Per	0.373	−0.021	0.069	0.000	−0.208
Ind	0.936	0.255	0.133	0.047	−0.005
B[g,h,i]P	0.946	0.200	0.149	0.050	−0.005
D[a,h]A	0.874	0.109	0.069	0.057	0.031
Cor	0.938	0.163	0.090	0.037	0.018
D[a,e]P	0.761	0.095	0.045	0.099	−0.046
Variance [%]	55.5	10.9	6.1	5.2	5.1
Estimated source	Vehicular Exhausts	Biomass Burning	Diesel Emissions	Sea Salt Aerosols	Industrial Emissions

^a Rotation Method: Varimax with Kaiser Normalization.

Bold: loading > 0.5

is concentrated in urban areas; therefore, these positive loadings in PC1 could be attributed to anthropogenic activities involving combustion of coal, diesel, and petroleum. In particular, the high levels of molecular 4–6 ring PAHs found in PC1 could be related to vehicular exhausts (Miguel and Pereira, 1989; Harrison *et al.*, 1996) and/or gasoline vehicles (Schauer *et al.*, 2002; Teixeira *et al.*, 2013).

Significant correlations of EC, TC, K⁺, OC, SO₄^{2−}, NH₄⁺, Ca²⁺, and Chry were found in PC2 with correlation coefficients of 0.895, 0.854, 0.845, 0.824, 0.754, 0.734, 0.625, and 0.571, respectively, accounting for 10.9% of the total variance. This PC is believed to be the biomass burning source of carbonaceous compositions. Due to OC, EC and K⁺ are generated from biomass burning, BB emissions contain a significant amount of WSIS, such as NH₄⁺ and K⁺ (Lee *et al.*, 2016; Pani *et al.*, 2018). Moreover, OC and EC can be

related to biomass burning as well (Mkoma *et al.*, 2013).

As illustrated in Table 6, PC3 represented 6.1% of the total variance. Several studies reported that Phe and Ant could be used as geochemical tracers of PM released from diesel engine exhausts and coal combustion (Fang *et al.*, 2006). Findings of a previous study that analysed air samples collected at Singapore suggested that PAH congeners with two and three rings were higher in concentration while the levels of the PAHs of higher molecular weight, four to six rings, are less. The difference in the concentration trends may be a result of the distinctive depletion rates of individual PAHs related differences in fuel characteristics (See. *et al.*, 2006). In this study, Phe exhibited the highest atmospheric concentrations with an average value of 0.041 ± 0.041 μg m^{−3}.

PC4 represented 5.2% of the total variance. The comparatively high loadings of Na⁺ ($r = 0.883$), Cl[−] ($r =$

0.810), and Mg^{2+} ($r = 0.718$) underline the importance of marine aerosols, which explanation is in good compliance with previous studies conducted in Auckland and Brisbane, underlined that Cl^- as a chemical tracer of maritime aerosols (Chan *et al.*, 1997; Wang *et al.*, 2005).

PC5 represented 5.1% of the total variance, with a high loading factor for NO_2^- , NO_3^- and F^- . Over recent decades, numerous studies have underlined the importance of industrial activities as one of the major sources of particulate F^- in the urban atmosphere (Lovelock, 1971; Haidouti *et al.*, 1993; Mukherjee *et al.*, 2003). For instance, hydrofluoric acid is used widely in the manufacture of chemicals and plastics and in laundries (WHO, 2000). The relatively low percentage contribution of industrial emissions was found in reasonable accord with the fact that the factories in Phuket account for only 0.31% of total number of factories in Thailand, based on a statistical survey conducted by the Department of Industrial Works of the Ministry of Industry in 2019. Consequently, it appears plausible that “industrial emissions” represented by PC5 account for only 5.1% of the total variance.

CONCLUSIONS

This study investigated the carbonaceous aerosol compositions (OC, EC, WSIS, and PAHs) of $\text{PM}_{2.5}$ samples obtained in Phuket. The average $\text{PM}_{2.5}$ concentration was 1.7 times higher than the USEPA standard. The application of diagnostic binary ratios of OC/EC and estimations of secondary organic carbon (SOC) in this study highlighted that the enhanced impacts of incomplete combustion emissions, such as motor vehicle exhaust, fuel burning, and biomass burning. Strong correlation ($r = 0.80$) was found between nss-K^+ and OC, which was also shown to be affected significantly by long-range atmospheric transport of organic aerosols associated with BB. In this study, the concentration of individual PAHs relatively high abundances of B[g,h,i]P and Ind were detected, indicating that motor vehicles, petroleum/oil combustion, and industrial waste burning are emission sources of the PAHs found in the ambient air of Phuket.

Source identification of the chemical species by PCA revealed that five sources of carbonaceous composition observed in the $\text{PM}_{2.5}$ samples explained 82.8% of the total variance. The highlight showed that vehicular exhausts, BB, diesel emissions, sea salt aerosols, and industrial emissions accounted for 55.5%, 10.9%, 6.1%, 5.20%, and 5.1% of the total variance, respectively. Interestingly, the PCA result showed vehicular exhausts as the main source. However, the contributions of both marine aerosols and BB to SOC also played a major role. Overall, 17.2% of the variance could not be attributed to the five primary local and/or regional sources; this proportion was considered to originate from other combustion activities such as incinerators, incense burning, and cooking.

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