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Chomsri ChooChuay[a], Siwatt Pongpiachan[b], Danai Tipmanee[c], Woranuch Deelaman[a], Natthapong Iadtem[a], Oramas Suttinun[a], Qiyuan Wang[d], Li Xing[d], Guohui Li[d], Yongming Han[d], Muhammad Zaffar Hashmi[e] [ID], Jittree Palakun[f], Saran Poshyachinda[g], Suparerk Aukkaravittayapun[g], Vanisa Surapipith[g], and Junji Cao[d]

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ABSTRACT

$PM_{2.5}$ is widely regarded as a major air pollutant due to its adverse health impacts and intimate relationship with the climate system. This study aims to characterize the chemical components (e.g., organic carbon (OC), elemental carbon (EC), water soluble ionic species (WSIS) and polycyclic aromatic hydrocarbons (PAHs) in $PM_{2.5}$ collected at Doi–Inthanon in Chiang-Mai, Thailand, the highest mountain in Thailand. All samples ($n = 50$) were collected by MiniVolTM portable air samplers from March 2017 to March 2018. In this study we found the average $PM_{2.5}$ concentration was $100 \pm 48.6\ \mu g\ m^{-3}$. The OC/EC ratio was 6.8 ± 3.0, and the decreasing order of the WSIS concentrations was $SO_4^{2-} > Na^+ > Ca^{2+} > NH_4^+ > NO_3^- > K^+ > Cl^- > NO_2^- > Mg^{2+} > F^-$. The total concentrations of nineteen PAHs were defined as 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. The concentration of total PAHs was $2.361 \pm 2.154\ \mu g\ m^{-3}$. Principal component analysis (PCA) highlights the importance of vehicular exhaust, biomass burning, diesel emissions, sea-salt aerosols and volatilization from fertilizers as the five dominant potential sources that accounted for 51.6%, 16.2%, 10.6%, 5.20% and 3.70% of the total variance, respectively. The rest of the 12.7% variance probably is associated with unidentified local and regional sources such as incinerators, joss paper/incense burning, and domestic cooking. Interestingly, the results from the source estimations from the PCA underlined the importance of vehicular exhaust as the major contributor to the $PM_{2.5}$ concentrations in the ambient air of Don-Inthanon, Chiang-Mai province. However, it is crucial to emphasize that the impacts of agricultural waste burning, fossil fuel combustion, coal combustion and forest fires on the variations of OC, EC and WSIS contents were not negligible.

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1. Introduction

Particulate matter (PM) or aerosols are defined as very small diameter solids or liquids that remain suspended in the atmosphere,[1] and they are a significant worldwide environmental issue, are well known as public health and climate hazards[2–9] and cause visibility degradation.[10] Over the past decades, international attention has been paid to fine particles discharged as a consequence of open biomass burning. Agricultural waste burning has usually operated to clean land before the next crop cycle.[11] Biomass burning (BB) is reported almost every year with varying intensities during the dry season in northern Thailand and in neighboring countries, such as Myanmar, Laos, Cambodia and Vietnam, due to agricultural waste burning,[8,9,11] and emits substantial amounts of $PM_{2.5}$ and trace gaseous species into the atmosphere.[8,9,12–15] BB emits substantial amounts of trace gaseous species and PM into the atmosphere and its contribution to carbonaceous aerosols at urban sites has been identified to be significant.

BB emissions contain significant amounts of water-soluble ionic species (WSIS), such as NH_4^+, K^+, and NO_3^-[8,16–19] Chemical analyses of smoke aerosols during forest fires have shown that potassium (K^+) and ammonium ions (NH_4^+) are the dominant cations and that sulfate ions (SO_4^{2-}) are the most abundant anions.[20] It is also crucial to highlight that biomass burning is the major source of OC, EC, NO_3^-, NH_4^+, SO_4^{2-} and K^+.[21] Multiple studies have confirmed the role of K^+ as a biosmoke tracer.[22–25]

In recent years, scientists around the world have paid more attention to carbonaceous particulates because they influence global warming, cloud microphysics,[26,27] global climate change[28,29] and have adverse effects on human health.[30] Moreover, cardiovascular mortality and morbidity rates are associated with increased levels of urban carbonaceous particulate matter.[31] Furthermore, several organic compounds such as PAHs and PCBs are found in OC mixtures, which possess carcinogenic and mutagenic effects.[32] Currently, most studies have highlighted the behavior of particulate PAHs in tropical countries and in northern Thailand.[33–41] Despite countless measurements of PAH compositions in coarse and fine particles around the world, there is inadequate information focusing on the characterization of particulate PAHs in tropical atmospheres. Most studies have targeted estimations of source apportionment.[42–46] Unfortunately, there is limited information associated with $PM_{2.5}$-bound OC, EC, WSIS and PAHs in northern Thailand.

Chiang-Mai is the third-largest province in Thailand, covering an area of 20,107 square kilometers. It is the second-largest province in the country in terms of population and also attracts many millions of visitors each year. Due to its mountainous geographical features, Chiang-Mai has suffered from poor air quality, especially during the dry season, for decades. Previous studies have indicated that biomass burning, vehicular emissions, industrial emissions, and anthropogenic activities are the main contributors for air pollutants.[8,14,47,48] The understanding of atmospheric aerosol compositions and their major sources is undoubtedly essential for reducing ambient PM levels and improving air quality. This study was carried out to evaluate the emission sources and chemical characteristics of the OC, EC, WSIS and PAHs components in $PM_{2.5}$. Overall, the main objectives of this study are to (*i*) characterize the chemical compounds (e.g., OC, EC, OC/EC, WSIS, and PAHs) from $PM_{2.5}$ samples collected in Doi-Inthanon, Chiang-Mai province; (*ii*) statistically analyze the chemical composition of $PM_{2.5}$ and its relationships to source identification; and (*iii*) perform source apportionment of the chemical composition of $PM_{2.5}$ using hierarchical cluster (HCA) and principal component analysis (PCA).

2. Materials and methods

2.1. Sampling site

The air quality observatory site is located at the National Astronomical Research Institute of Thailand (NARIT) at the summit of Doi–Inthanon, Chiang-Mai province (Figure 1). It is
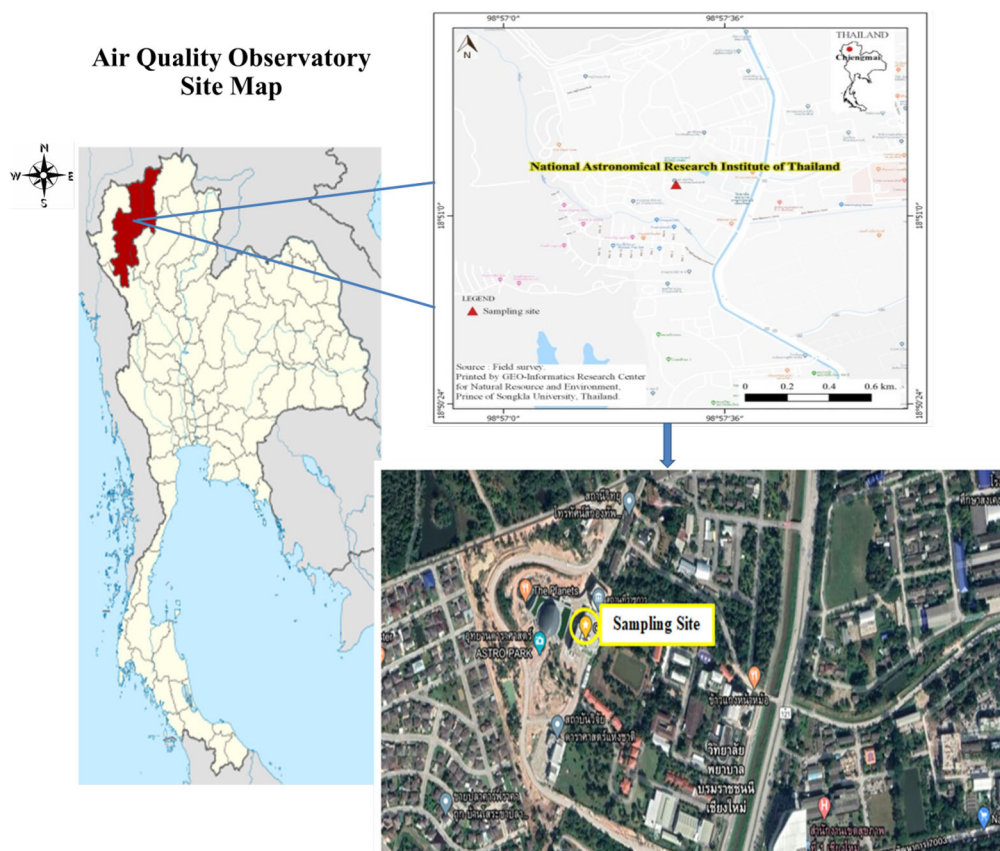
Figure 1. Map of the air quality observatory sites in this study.

important to mention that Doi-Inthanon is the highest mountain in Thailand. The elevations of Doi-Inthanon range between 800 and 2,565 meters. The elevation of Doi-Inthanon means that it is cooler temperature than the surrounding plains. It has high humidity and cold weather all the year. Hence, Doi-Inthanon is one of the most popular destinations in Thailand. It is a popular tourist destination for both foreign and Thai tourists, with a peak of 12,000 visitors visiting on New Year's Day. In addition to a range of tourist facilities on the summit, there is also a Royal Thai Air Force weather radar station at the summit and the Thai National Observatory (TNO) at km44. The main entrance is about 70 km southwest from Chiang-Mai city center. The sampling location is at 18 54'40.5"N and 99 13'01.4"E. Chiang-Mai is the third-largest province in Thailand, covering an area of 20,107 square kilometers. It is the second-largest in the country in terms of population, with 1,746,840 people currently residing in Chiang-Mai. The city is a popular destination among travelers. Chiang-Mai has relatively cool weather throughout the year. There has been an increasing number of articles related to air quality in northern Thailand.[36,49] These can be correlated with the $PM_{2.5}$ levels, which are caused by large-scale 'hot spot' locations. The pollutant sources can be identified, for example, pollutants from forest fires, burning of agriculture waste, and trans-boundary haze pollution.

All samples ($n = 50$) were collected on quartz-fibre filters using MiniVolTM portable air samplers (Airmetrics) through 47 mm filters at flow rates of 5 L min^{-1}. All samples were collected over a period of 72 h, and then, sample collections were halted for 24 h and were then followed by another collection session of 72 h; this pattern was repeated throughout the study period. After the sample collections, the filters were carefully kept in Petri slide dishes that were individually

wrapped in aluminum foil to avoid any loss from photodegradation and were stored in a freezer at 4 °C to maintain their chemical stability until subsequent 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. This monitoring campaign was conducted from March 2017 to March 2018.

2.2. Chemical analysis

The carbonaceous aerosol components, OC and EC, were quantified using a Desert Research Institute (DRI) Model 2001 carbon analyzer (Atmoslytic Inc., Calabasas, CA, USA). The IMPROVE_A thermal/optical reflectance (TOR) protocol was used for the analyses.[50] The thermal/optical carbon analyzer bases its analysis on the oxidation of organic carbon (OC) compounds and of elemental carbon (EC) at different temperatures.

Carbonate carbon is determined by estimating the CO_2 acidification from organic sample that punches before the normal carbon analysis procedure. This protocol uses seven temperature programs for seven different fractions. The temperature protocol is applied for separating OC and EC. It is the same as that for the TOR and TOT burning correction. This protocol includes total OC, total EC, and total carbon, which are monitored by both reflectance (OPR) and transmittance (OPT). This protocol is dependent on the thermal/optical analysis that is applied for quantification of the thermally derived sub-fractions of OC and EC.

Thermal analysis using heat (thermal/optical carbon analyzer) follows the IMPROVE protocol (Interagency Monitoring of Protected Visual Environments) using the thermal/optical reflectance (TOR) method. There are eight types of carbonaceous compounds that are differentiated by their combustion temperatures, namely, OC1, OC2, OC3, OC4, EC1, EC2, EC3 and OP. Total OC was defined as the sum of four OC fractions (OC1-OC4) plus OP, whereas the total EC was technically defined as the sum of three EC fractions (EC1-EC3) minus OP.

In this study, TD-GC/MS was employed for both qualitative and quantitative analyses of PAHs. Injection port thermal desorption (TD), coupled with gas chromatography/mass spectrometry (GC/MS), was applied to quantify the non-polar organic compounds including 19 PAHs in the $PM_{2.5}$ samples. This method acts like an alternative method of traditional solvent extraction followed by GC/MS analysis. The detail of analytical procedures have been reported in previous publications.[51–53] An Agilent 7890 A GC/5975C MS system (Agilent Technology, Inc., Santa Clara, CA, USA) were applied which operated in electron impact (EI) ionization mode. In brief, aliquots of the sample filter (0.1–0.526 cm^2, subjected to the loading) was cut into small pieces and transferred into a TD tube. The sample-loaded tube was placed into the GC injection port at 50 °C. The injection port temperature was high up to 275 °C for desorption in a splitless mode while the GC oven temperature was kept around 30 °C. Separation of organic compounds was performed with a HP-5MS (30 m × 0.250 mm i.d., 0.25 μm film thickness, Agilent Technologies,Inc.) capillary column. The constant flow rate of carrier gas helium (UHP grade, 99.999% purity) was 1.2 mL/min. The MSD was scanned from 50 to 550 amu. The EI voltage was 70 eV and selected ion mode was applied for the quantification.

The concentrations of Water-Soluble Ionic Species (WSIS), five inorganic cations (Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}), four inorganic anions, which included Cl^-, F^-, NO_3^- and SO_4^{2-} in the samples were extracted by ion chromatography (IC, Dionex 600) that consists of a separation column (Dionex Ionpac AS11 for anion and CS12A for cation), a guard column (Dionex Ionpac AG 11 for anion and AG12A for cation) The quality assurance/quality control (QA/QC) procedures for this analysis, all glassware used in sample preparations was washed by an ultrasonic and then took them to a furnace for baked at 450 °C around 6 hours..[54]

Table 1. Summary of the concentrations of PM$_{2.5}$, TC, OC and EC in Doi-Inthanon Chiang-Mai province.

Species	Unit	Mean ± SD ($n = 50$)	Minimum	Maximum
PM$_{2.5}$	μg m^{-3}	100 ± 48.6	30.4	204
TC	μg m^{-3}	9.98 ± 8.00	0.74	32.8
OC	μg m^{-3}	8.38 ± 6.41	0.67	26.8
EC	μg m^{-3}	1.60 ± 1.65	0.07	7.26
OC1	μg m^{-3}	0.10 ± 0.08	0.00	0.30
OC2	μg m^{-3}	1.26 ± 1.02	0.16	4.69
OC3	μg m^{-3}	3.60 ± 2.54	0.40	11.4
OC4	μg m^{-3}	2.57 ± 2.49	0.08	10.5
EC1	μg m^{-3}	2.28 ± 1.95	0.01	7.10
EC2	μg m^{-3}	0.16 ± 0.07	0.05	0.34
EC3	μg m^{-3}	0.02 ± 0.05	0.00	0.24

2.2.1. Quality assurance and control (QA/QC)

The quality assurance/quality control (QA/QC) procedures for this analysis have been described here. All glassware used in sample preparations was washed by an ultrasonic and then took them to a furnace for baked at 450 °C around 6 hours. All solvents that used for extraction and analysis were pesticide residue grade.

For the OC/EC analysis, the instrument was calibrated with known quantities of methane daily. Replicate analyses were done for each group of ten samples. Blank filters were also analyzed and the sample results were corrected for the average of the blank concentrations, which were 1.72 and 0.09 μg m^{-3} for OC and EC, respectively. The detection limits for EC and OC were $<1.0 \mu$g m^{-3}. The relative deviation of replicate analyses was $<5\%$ for TC (total carbon) and $<10\%$ for OC and EC.

For PAHs was quantified using the TD-GC/MS instrumentation. Chrysene-d$_{12}$ (C$_{18}$D$_{12}$) (98%, Sigma-Aldrich, Bellefonte, PA, USA) was added as internal standard (IS) for the PAH. A five-point calibration over a concentration range of 0.5–5.0 ng for each of the target compounds from a standard mixture (Sigma-Aldrich, Bellefonte, PA, USA) was established, and the correlation coefficients (R^2) for linear regressions of the calibration curves were >0.99. The recovery of the TD method was calculated based on the response signal of liquid standards and its known amount spiked into the blank filter. Repeatability and reproducibility were also investigated for this method. For every 10 samples, one replicate analysis was performed; and for every 5 samples, one backup filter was analyzed to check any potential contamination and the results show $< 5\%$ contribution. In addition, ambient samples spiked with known amounts of internal and external standards were analyzed to check the potential interference. All data were corrected for the average value of the blanks.

For WSIS, after collected the sample, all samples were immediately stored in a freezer at -20 °C to avoid the loss of semi-volatiles. For this study, the field blanks were extremely low, normally below or close to the method detection limit. Therefore, the background contamination/interference was insignificant and can thus be ignored. The determination coefficient (R^2) of the calibration curve for each chemical species analyzed is required to be higher than 0.995. At least 10% of the samples were analyzed by spiking with a known amount of metallic and ionic species to determine their recovery percentages.

2.3. Statistical analysis

Hierarchical cluster analysis (HCA) and principal component analysis (PCA) were used for the classification and source apportionments of OC, EC, WSIS and PAHs using the Statistical Package for Social Sciences (SPSS) version 13 software. Data preprocessing consisted first of subjecting all data to a logarithmic transformation. This technique is the most common approach for transforming environmental data, as it is robust for non-normal data distributions.[55] PCA enables

multivariate data reduction by transforming the data into orthogonal components that are linear combinations of the original variables. Therefore, PCA reduces multidimensional data to fewer dimensions. Hence, this method is used to identify trends and clusters within the data. In addition, the correlation coefficients of OC, EC and WSIS are regularly used to identify the relationships between aerosol components[56,57] and are used in this study to observe these relationships and to identify the sources of OC, EC and WSIS.

3. Results & discussion

3.1. Mass concentrations of PM$_{2.5}$ and carbonaceous compounds (OC and EC)

The average mass concentrations of atmospheric PM$_{2.5}$ and its carbonaceous chemical components (OC and EC) in Chiang-Mai were collected from March 2017 to March 2018. The concentrations of PM$_{2.5}$ and the average compositions of each carbon fraction relative to the total carbon (TC), OC and EC levels in Chiang-Mai are summarized in Table 1. The average concentrations of PM$_{2.5}$ are 100 ± 48.6 and the carbonaceous chemical components are 9.98 ± 8.00, 8.38 ± 6.41, 1.60 ± 1.65, 0.10 ± 0.08, 1.26 ± 1.02, 3.60 ± 2.54, 2.57 ± 2.49, 2.28 ± 1.95, 0.16 ± 0.07 and $0.02 \pm 0.05 \,\mu g \, m^{-3}$ for TC, OC, EC, OC1, OC2, OC3, OC4, EC1, EC2 and EC3, respectively.

In Chiang-Mai,[58] presented the concentrations of carbonaceous compounds in PM$_{2.5}$ at Doi Ang Khang (DAK) and at Chiang Mai University (CMU) during the dry season of 2015. The TC mass concentrations were 54.9 ± 15.8 and $56.0 \pm 22.4 \,\mu g \, m^{-3}$ at DAK and at CMU, respectively. The percentage contributions from OC were 87% and 83% at DAK and CMU, respectively. In addition, the percentage contribution of OC obtained from this study was 84%. This result is consistent with previous studies indicating that BB might have been the principal source of carbonaceous substances.[8,14,16] In contrast, the EC fractions were regarded as being relatively lower than the OC fractions. Since EC has a chemical structure similar to impure graphite, it appears reasonable to assume that vehicle exhaust is the major source of EC. As a consequence, the most important sources of EC are fossil fuel combustion and biomass burning.[59]

OC can either be directly emitted into the atmosphere from the incomplete combustion of organic compounds or can form by the condensation of compounds produced by atmospheric photooxidation and polymerization of organic species,[60] including thousands of organic compounds (e.g., aromatic compounds, carboxylic compounds with polar substituents and aliphatic compounds) with a variety of physical and chemical properties. OC may be emitted directly from sources such as industrial processes and by natural occurrences (e.g., primary OC) or can form in the atmosphere due to gas-to-particle conversion of semi- and low-volatility organic compounds (e.g., secondary OC). Quantification of the contributions of primary and secondary organic carbon is quite difficult to determine through direct chemical analysis, since OC contains a complex mixture of many compounds. Particles containing OC might show a significant risk to human health, as well. The carbonaceous compounds or total carbon (TC) in the atmosphere represent one of the main components of the total mass of suspended fine particles. They are composed of organic carbon (OC) with a contribution of 70–80%.[61]

In the case of four OC fractions, OC3 was the most abundant ($3.60 \pm 2.54 \,\mu g \, m^{-3}$), followed by OC4 ($2.57 \pm 2.49 \,\mu g \, m^{-3}$), OC2 ($1.26 \pm 1.02 \,\mu g \, m^{-3}$) and OC1 ($0.10 \pm 0.08 \,\mu g \, m^{-3}$). Additionally, EC1 was the major fraction ($2.28 \pm 1.95 \,\mu g \, m^{-3}$) in the EC. 62) reported that OC3 can be used as a biomass burning tracer adjacent to emission sources, while OC2 is the most abundant fraction of OC released from coal combustion[63] and motor vehicle exhaust.[64] Biomass burning combustion sources are considered to be the greatest contributors to primary carbonaceous particle levels in the atmosphere[2,22,65,66] and many studies have used their fractions for source apportionment of biomass burning.[63,65,67–69]

Table 2. The values of the OC, EC and OC to EC for different emission sources.

Emission source	EC ($\mu g\ m^{-3}$)	OC ($\mu g\ m^{-3}$)	OC/EC ratio	References
Light-duty gasoline vehiclesa	22.6	50.1	2.2	70
Light-duty gasoline vehiclesb	13.5 ± 8.02	30.1 ± 12.3	2.2	71
Heavy-duty diesel vehiclesa	40.5	32.6	0.8	70
Tunnelsc	25.5 ± 4.90	19.2 ± 8.46	0.76	72
Tunnelsb	16.9	16.8	N.A.	
Paved road dustb	1.12 ± 0.30	14.7 ± 2.00	13.1	71
Residential wood combustionb	12.3 ± 4.20	51.3 ± 11.7	4.15	73
Meat charbroilinga	0.0 ± 0.5	33.8 ± 2.0	N.A.	74
Natural gas home appliancesa	6.7	84.9	12.7	70
Forest firesa	3.23 ± 1.80	46.8 ± 15.6	14.51	73
Agriculture Residues	0.69–0.13	3.3	5–8	75
Biomass burning	0.86	2.6	5.1	20
	1.16–7.38	3.43–14.5	4.0-8.0	25
	3.33 ± 0.87	18.62 ± 4.10	5.7	62
	3.5	20.2	6.8	17
	1.2	N.A	9	76
	11.3 ± 6.9	34.1 ± 18.0	3.3	65
	12.3 ± 5.3	61.9 ± 33.2	5.1	65
	11.3 ± 6.9	34.1 ± 18.0	12.3	65
	7.1 ± 2.0	47.8 ± 13.9	6.8 ± 0.6	58
	9.5 ± 4.3	46.6 ± 19.0	5.2 ± 1.3	58
	N.A	N.A	16.7	77
	N.A	N.A	6.7	78
	N.A	N.A	10	79
Fossil fuel combustion	N.A	N.A	1.1	73
	N.A	N.A	4	80
	N.A	N.A	4.1	65
Coal combustion	N.A	N.A	2.7	73
	N.A	N.A	3.0 and 12.0	65
Secondary organic carbon	N.A	N.A	3.3	81
	N.A	N.A	21–33	82
Long-range transport	N.A	N.A	12	81
Traffic	N.A	N.A	0.7	81
Cooking emissions	1.60 ± 0.20	3.37 ± 0.46	4.3–7.7	83

Note: N.A.=not applicable,.
aMass % of fine particle mass,.
bMass concentration.
cmg per vehicle-kilometr.

3.2. The OC/EC ratios, atmospheric concentrations of Water-Soluble ionic species (WSIS) and PAHs in $PM_{2.5}$

3.2.1. Oc/EC ratios

The ratio of OC to EC (OC/EC) is often used to obtain information on emission sources.[28,63,65,67,68] Table 2 shows the values of the OC, EC and OC to EC for different emission sources reported for different emission sources. The average OC/EC ratio from this study was 6.8 ± 3.0

OC/EC ratios can be used to classify the main sources of air pollutants. Some studies have reported that OC/EC ratios greater than two indicate SOC formation.[63,84] Table 2 shows that the OC/EC ratios from both vehicle exhaust and biomass burning range from 4.0 to 16.7,[17,20,62,65,75–79,85–87] the ratios for motor vehicles (diesel and gasoline) range from 0.02 to 4.0,[30,88–93] the ratios for fossil fuel combustion range from 1.1 to 4.1,[65,73,80] the ratios for coal combustion range from 2.7 to 12.0[65,73] the ratios for forest fires show a range of 14.51–16.0,[73] the ratios secondary organic carbon show a range of 3.3–33,[81,82] the ratios for long-range transport show a range of 3.01-12[81,94] and cooking emissions ratios show a range of 4.3–7.7.[83] It is important to emphasize that the PM 2.5-bound OC/EC ratios collected during the dry season at Doi Ang Khang and at Chiang Mai University were 6.8 ± 0.6 and 5.2 ± 1.3, respectively (Figure 2).[58] As a consequence, the OC/EC ratio obtained from this study (e.g., 6.8 ± 3.0) is surprisingly
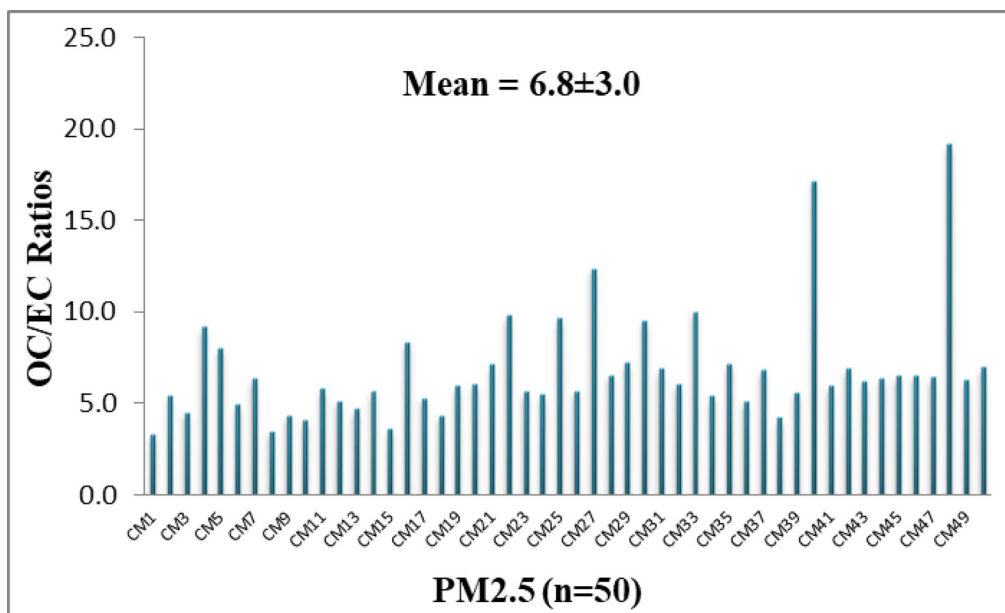
Figure 2. Temporal variations in the OC/EC ratios for 50 samples from Doi-Inthanon Chiang-Mai, Thailand from March 2017 to March 2018.

consistent with the results of Pani et al.[58] Thus, the source of carbonaceous compounds in this region might be from biomass burning, vehicle exhaust, coal combustion, secondary organic carbon, long-range transport and/or cooking emissions.

3.2.2. Secondary organic carbon (SOC) contributions

Carbonaceous aerosols with OC/EC values greater than two can be considered to contain significant quantities of SOC.[84,95] According to Castro et al.,[96] the SOC contribution can be estimated by calculating the minimum values of the OC/EC ratios in all samples. The SOC was computed from equation 1:

$$ SOC \ = \ OCtotal \ - \ EC \ \times \ (OC/EC)min \qquad \text{Equation 1} $$

where SOC is the secondary OC, OC_{total} denotes the total OC, and $(OC/EC)_{min}$ is the average of the three minimum OC/EC ratios. Ji et al.[97] described the formation and influencing factors of SOC, which show that photochemical oxidation and atmospheric temperatures play important roles in the formation of SOCs. SOCs are distributed in a particle phase after the oxidation of volatile organic carbon (VOC) by active radicals in the atmosphere.[98] Chemical reactions involving gas–particle conversion occur during long-distance transport of aerosol particles.[99]

In this study, there was a high correlation between OC-EC and K^+. Normally, OC, EC and K^+ are generated from biomass burning and from in $PM_{2.5}$. It is crucial to note that the high OC/EC ratios (6.8 ± 3.0), coupled with the high SOC contents ($8.12 \pm 0.26 \, \mu g \, m^{-3}$) detected at Don-Inthanon, highlight the dominant effects of biomass/agricultural waste burning in northern Thailand. These findings are consistent with the comparatively high SO_4^{2-}, NH_4^+, K^+ levels observed at Don-Inthanon, Chiang-Mai. Strong positive correlations of OC *vs.* K^+ ($r = 0.95$) and EC *vs.* K^+ ($r = 0.90$) were also detected. BB is another possible means for forming SOC[100] and it has been common in Chiang-Mai to burn biomass for farming preparation.

Table 3. Summary of the concentrations of WSIS in Doi-Inthanon Chiang-Mai province.

Species	Unit	Mean ± SD ($n = 50$)	Minimum	Maximum
F^-	$\mu g\ m^{-3}$	0.10 ± 0.02	0.05	0.15
Cl^-	$\mu g\ m^{-3}$	0.39 ± 0.11	0.18	0.85
NO_2^-	$\mu g\ m^{-3}$	0.30 ± 0.15	0.00	0.64
NO_3^-	$\mu g\ m^{-3}$	0.65 ± 0.30	0.00	1.51
SO_4^{2-}	$\mu g\ m^{-3}$	4.60 ± 3.90	0.18	15.70
Na^+	$\mu g\ m^{-3}$	1.40 ± 0.35	0.45	2.26
NH_4^+	$\mu g\ m^{-3}$	0.99 ± 0.97	0.00	3.98
K^+	$\mu g\ m^{-3}$	0.62 ± 0.50	0.00	1.90
Mg^{2+}	$\mu g\ m^{-3}$	0.11 ± 0.03	0.03	0.17
Ca^{2+}	$\mu g\ m^{-3}$	1.06 ± 0.32	0.27	2.08

3.2.3. Water soluble ionic species (WSIS)

WSIS dominate major portions of atmospheric particles[101,102] and they may be harmful to human health as they can easily enter human lungs and trigger respiratory diseases.[103] For this particular reason, chemical characterizations of WSIS have been conducted worldwide by many researchers using various types of analytical techniques.[104–107] Most of these studies have investigated the major ions, such as NH_4^+, Ca^{2+}, K^+, Na^+, Mg^{2+}, Cl^-, NO_3^-, and SO_4^{2-}. It is well known that BB emissions are responsible for relatively large amounts of WSIS in atmospheric particles.[8,17,62] Since BB releases great amounts of carbonaceous aerosols, coupled with WSIS, into the atmosphere,[18,24,62,65] the assessment of WSIS, OC, and EC can provide valuable information regarding their formation, characteristics, and potential emission sources.

As previously mentioned, the most dominant species in this study were SO_4^{2-}, which mainly converts SO_2 gaseous precursors into particles. In this study, it presents the concentrations of ten WSIS species in $PM_{2.5}$. The results showed that SO_4^{2-}, Na^+, Ca^{2+}, NH_4^+, NO_3^-, K^+, Cl^-, NO_2^-, Mg^{2+} and F^- were the dominant WSIS species, which accounted for 44%, 14%, 11%, 10%, 6%, 6%, 4%, 3%, 1% and 1% of the total mass of ions, respectively (Table 3). Secondary inorganic aerosols (SIA), including SO_4^{2-}, NH_4^+, and NO_3^-, were the major ions found in this study. This finding suggests that SIA are mainly present in fine particles, which is in good agreement with previous studies.[58,108,109] It is also crucial to note that Na^+, Ca^{2+}, Mg^{2+} and F^- can be used as geochemical tracers representing maritime aerosols.[107,110]

For this study, the individual WSIS concentrations of the $PM_{2.5}$ samples in Don-Inthanon, Chiang-Mai during March 2017 to March 2018 were in a decreasing order of $SO_4^{2-}>Na^+>Ca^{2+}>NH_4^+>NO_3^->K^+>Cl^->NO_2^->Mg^{2+}>F^-$. These data indicate that fossil fuel combustion (vehicles and energy production) (e.g., SO_4^{2-}) and long-range atmospheric transport of maritime aerosols (e.g., Na^+) are the two major contributors to WSIS in $PM_{2.5}$. SO_4^{2-}, and NO_3^- and NH_4^+ mainly form in the atmosphere by gas-to-particle conversion from their precursor gases (e.g., SO_2, NO_x and NH_3). Although the major source of NO_x is traffic emissions, in particular, incomplete combustion of fossil fuels from diesel engines and cement kiln manufacturing can be another major source of NO_x emissions.[111,112] and Yao et al.[113] reported that SO_4^{2-} is normally created mainly from fossil fuel combustion by the photochemical formation of SO_2 from traffic and industrial activity. In contrast,[114] indicated that vehicle exhaust and biomass burning were two major sources of SO_4^{2-} in the ambient air of northern Thailand. Chantara et al.[12] conducted atmospheric pollutants in Mae Hia Research Center, Chiang-Mai province over a five-year (2005–2009). During 5 years of sampling the result found that the dominant ion species was SO_4^{2-} (>39% of total ion concentrations). As a consequence, the SO_4^{2-} was found the most dominant ion from this study. It is surprisingly consistent with the results of Chantara et al.[12] However, the elevation of the sampling site from this study was higher than the sampling site that was conducted from Mae Hia Research Center but the result from both study is not different because the surrounding around the sampling site of this study, there is a Royal Thai Air Force weather radar station at the summit and the Thai National Observatory and the important reason

Table 4. Summary of PAHs concentrations in Doi-Inthanon, Chiang-Mai province.

PAHs (µg m^{-3})	Mean	S.D.	Min.	Max.
Ace	0.040	0.042	0.005	0.226
Fl	0.032	0.038	0.008	0.219
Phe	0.125	0.200	0.022	1.039
Ant	0.020	0.022	0.006	0.122
Fluo	0.054	0.046	0.008	0.206
Pyr	0.054	0.048	0.006	0.184
B[a]A	0.123	0.092	0.019	0.470
Chry	0.052	0.049	0.002	0.181
B[b]F	0.177	0.167	0.006	0.733
B[k]F	0.203	0.203	0.001	0.930
B[a]F	0.028	0.026	0.002	0.117
B[e]P	0.096	0.084	0.005	0.381
B[a]P	0.170	0.167	0.009	0.703
Per	0.019	0.017	0.001	0.077
Ind	0.468	0.380	0.004	1.713
B[g,h,i]P	0.398	0.322	0.004	1.403
D[a,h]A	0.104	0.087	0.002	0.405
Cor	0.144	0.108	0.001	0.467
D[a,e]P	0.055	0.056	0.007	0.274
ΣPAHs*	2.361	2.154		

*Note that 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.

is Doi-Inthanon is a popular tourist destination for both foreign and Thai tourists, with a peak of 12,000 visitors visiting on New Year's Day. Consequently, it seems reasonable to conclude that the $PM_{2.5}$-bound SO_4^{2-} obtained in this study could be derived from mixed sources between biomass burning and vehicles and energy production.

3.2.5. Concentrations of PAHs

The concentrations of PAHs are summarized in Table 4 for the concentrations of $\sum$PAHs (e.g., the sum of 19 PAH contents) for the annual averages, the standard deviations (SD) and the ranges for 19 PAHs examined in this study. The concentration of total PAHs was 2.361 ± 2.154 µg m^{-3}. The values in this study were lower than those measured in other areas such as Beijing and Guangzhou, which are often known as heavily polluted areas in China.[115,116]

Over the past few decades, numerous studies concerned with source identifications, quantitative source apportionments, and ecological risk assessments of PAHs in the ambient air of Thailand have conducted intensive investigations.[35,36,39,40] In the northern part of Thailand, agricultural waste and biomass burning during cold periods have released large amounts of particulate matter, especially ultra-fine particles, including $PM_{2.5}$-bound PAHs, into the atmosphere. There are a number of recent studies that use PAHs data to obtain inferences of particulate pollution sources.[34,117–119] Nine of the 16 USEPA priority PAHs are related to combustion processes such as those of coal, petroleum and diesel. These PAHs are known as combustion PAHs (CPAH), namely, Flu, Pry, B[a]A, Chry, B[b]F, B[k]F, B[a]P, B[g,h,i]P and Ind.[120] The ratios of the concentrations of these CPAHs have been used in many studies to identify potential sources of PAHs in aerosols.[121] In this study, relatively high abundances of Ind and B[g,h,i]P were detected, indicating that traffic emissions, petroleum combustion and industrial waste burning can be considered as the emission sources of PAHs for the ambient air of Chiang-Mai.[120,122]

3.2.5.1. Diagnostic ratios. The diagnostic ratios of PAHs have been used as indicators for emission sources, such as biomass and coal combustion, petroleum and mixed sources pollutants.[123] The four ratios of Fluo/(Fluo + Pyr), An/(An + Phe), Ind/(Ind + B[ghi]P) and B[a]A/ (B[a]A + Chry) were used for source identification in this study, Fluo/(Fluo + Pyr) of 0.4–0.5

recommended for gasoline engine, < 0.4 petroleum source and > 0.5 recommended for biomass burning (for example grass and/or wood) and coal combustion. An/(An + Phe) < 0.1 was guided for petroleum source and > 0.1 was guide for combustion source. Ind/(Ind + B[ghi]P) > 0.5 was indicated to biomass burning (such as grass, wood) and coal combustion, a value < 0.2 was related as a petroleum source and 0.2–0.5 related to petroleum combustion (Figure 3). B[a]A/(B[a]A + Chry), 0.35 was recommended for petroleum combustion and mixed sources (such as a combustion of petroleum, biomass burning and coal combustion. Scatter plots of four ratios in PM$_{2.5}$ from Don-Inthanon, Chiang-Mai province are shown in Figure 3. The rang of Fluo/(Fluo + Pyr) ratio was 0.43-0.68, indicated that a source from gasoline engine, biomass burning (for example grass and/or wood) and coal combustion. An/(An + Phe) was 0.10-0.30, indicating a combustion source. Ind/(Ind + B[ghi]P) was 0.46-0.57, guided that a source from petroleum combustion, biomass burning (such as grass, wood) and coal combustion. For B[a]A/(B[a]A + Chry) was more than 0.35, the range was 0.49-0.95, indicating the fraction of petroleum combustion, for example vehicular exhaust and crude oil combustion. Hence, the results of the ratio expose that the main sources of PM$_{2.5}$-bound PAHs can be formed biomass burning, coal combustion and petroleum during the study periods in Chiang-Mai province.

3.2.5.2. Pearson correlation analysis of OC-EC, WSIS and PAHs.
The Pearson correlations of OC-EC, WSIS and PAHs in PM$_{2.5}$ were investigated, the OC concentrations were strongly correlated with EC (r = 0.95). The SO_4^{2-} concentrations were greatly correlated with NH_4^+ (r = 0.91). The K^+ concentrations were positively correlated with OC (r = 0.95) and EC (r = 0.89). The B[k]F concentrations were strongly correlated with B[b]F (r = 0.98). The B[g,h,i]P concentrations were strongly correlated with Ind (r = 0.98). The Phe concentrations were strongly correlated with Ace (r = 0.97) and Ant (r = 0.98).[58] reported that strong correlations were also found between SO_4^{2-} and NH_4^+ (r = 0.95) from studies in Chiang-Mai during the dry period. The result of this study also demonstrated the similar patterns observed in previous investigations conducted in Doi-Inthanon, Chiang-Mai province.

3.3. Source identification and pattern recognition

To identify the potential sources of the OC, EC, WSIS and PAHs, two multivariate statistical analyses are introduced in this section. In section 3.2.4, the relatively high correlation coefficients of K^+ vs. OC and K^+ vs. EC indicate that biomass burning is the main contributor to carbonaceous aerosols. Furthermore, the relatively high abundance of Ind and B[g,h,i]P suggests that motor vehicle exhaust can be considered as one of the major emission sources of PAHs in this region. In this section, source identification, coupled with quantitative source apportionment of targeted chemical species, is investigated by using two multivariate statistical analyses, namely, hierarchical cluster analysis (HCA) and principal component analysis (PCA).

3.3.1. Classification of chemical compounds by hierarchical cluster analysis (HCA)
HCA was performed to identify the homogeneous groups of carbonaceous aerosol components, including OC, EC, WSIS and 19 individual PAHs in PM$_{2.5}$. The hierarchical dendrogram is shown in Fig 4. Three major groups of carbonaceous aerosol components were clearly distinguished. The first group contains F$^-$, Mg^{2+}, Cl$^-$, NO$_2^-$, NO$_3^-$, K$^+$, Na$^+$, Ca^{2+}, NH$_4^+$, EC, SO$_4^{2-}$, TC, OC, B[a]F, Per, Ace, Ant, Fl, Fluo, Pyr, Chry, D[a,e]P, all OCs, EC and WSIS. This indicates that the major sources are mixtures of biomass burning, fossil fuel, coal combustion, and industrial emissions.[58,108,109] In addition, the pyrogenic sources, namely, the motor vehicle emissions from both gasoline and diesel emissions and the leakage of used oil and road dust have been previously identified as the original sources of this group.[124–126] In accordance with previous
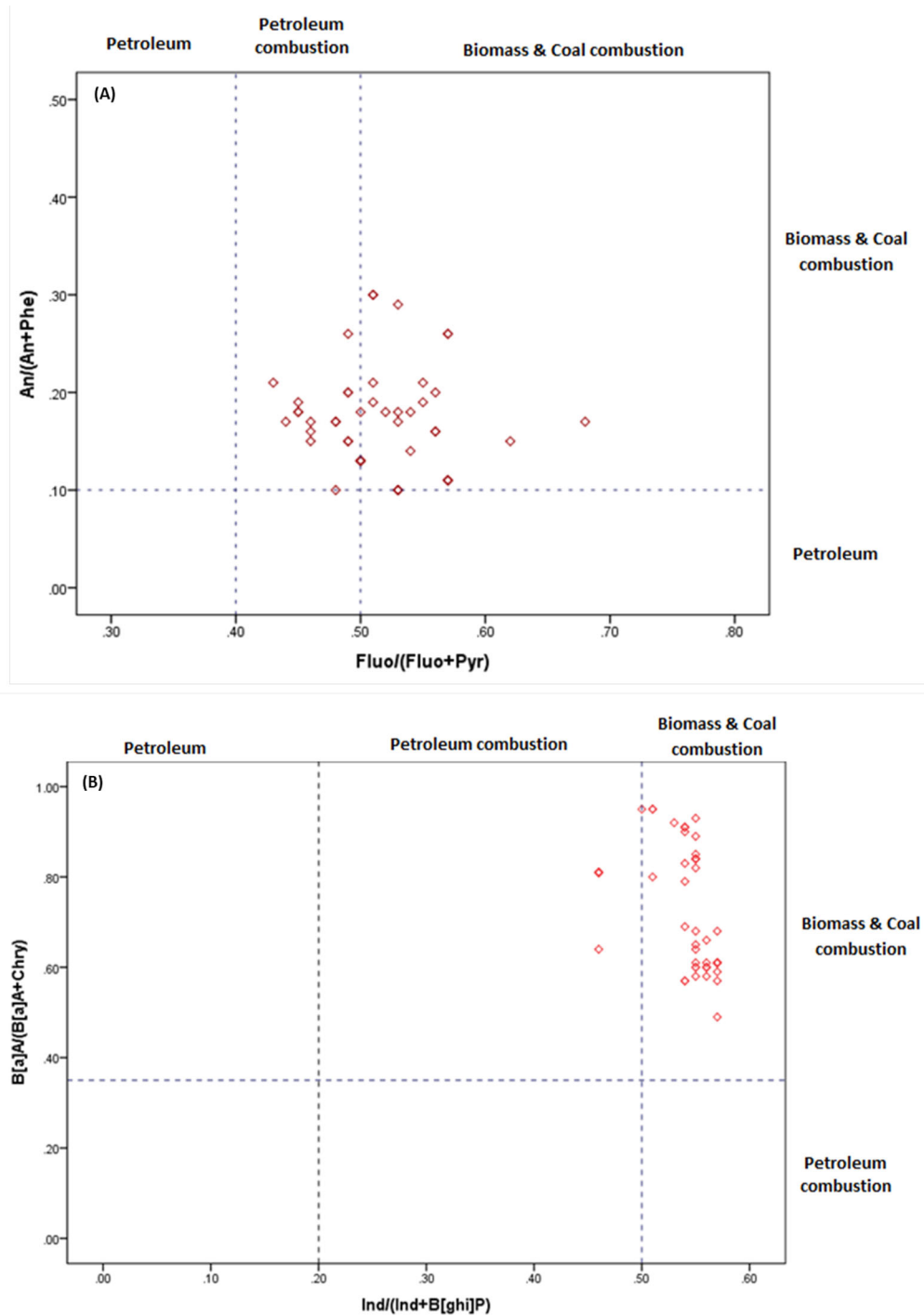
Figure 3. The diagnostic ratios of: (A) Fluo/(Fluo + Pyr) versus An/(An + Phe) and (B) Ind/(Ind + B[ghi]P) versus B[a]A/(B[a]A + Chry) in PM$_{2.5}$ from Doi-Inthanon, Chiang-Mai province.

literature reports, 3–4 ring PAHs have been found in the road dust in urban cities.[127,128] The second group consists of B[b]F, B[a]P, B[k]F, B[e]P, D[a,h]A, Cor, B[a]A and Phe, which are the high-molecular-weight PAHs with 5–6 rings. This group usually originates from pyrogenic
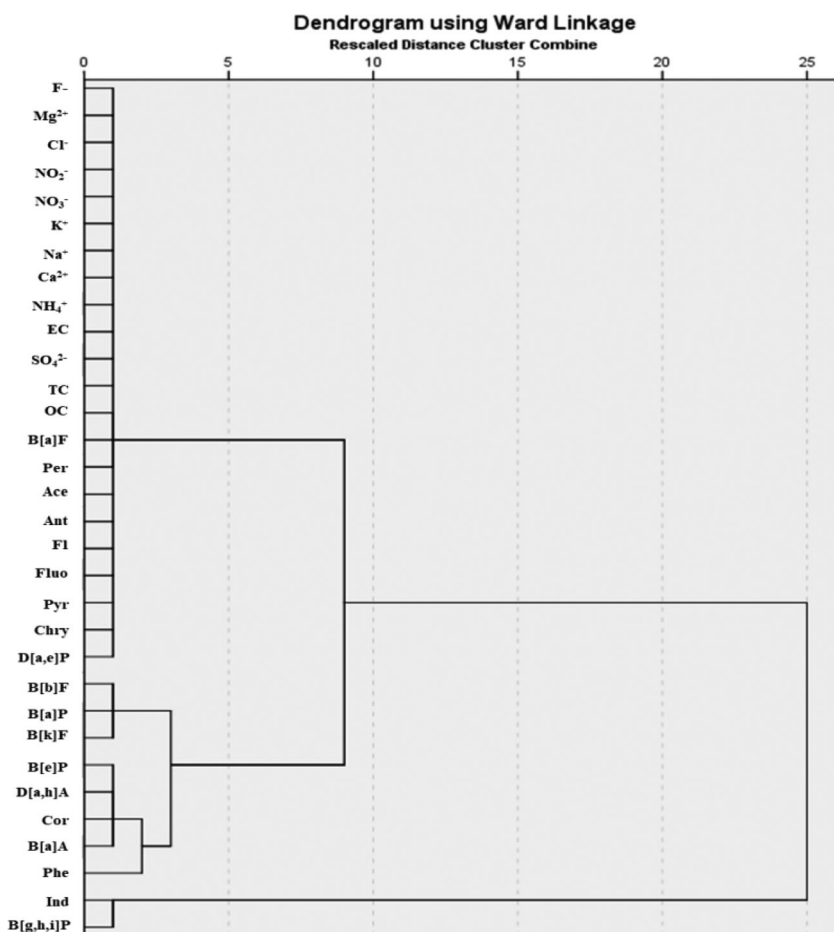
Figure 4. Hierarchical Cluster Analysis (HCA) of carbonaceous compounds, water soluble ionic species and 19 individual PAHs in PM$_{2.5}$ at Doi-Inthanon Chiang-Mai province.

sources that released from motor vehicle, diesel/gasoline engine, road-paving asphalt, crude oil, bitumen[129] and road dust.[130] The last group consists of Ind and B[g,h,i]P, which are the anthropogenic PAHs from the use of petroleum products, motor vehicles, oil combustion and industrial waste incinerators.[120,122] The last group consists of Ind and B[g,h,i]P, which are the anthropogenic PAHs from the use of petroleum products, motor vehicles, oil combustion and industrial waste incinerators.[120,122]

HCA illustrates that mixtures of biomass burning, fossil fuel combustion (vehicles and energy production), coal combustion, and industrial emissions are major sources of PM$_{2.5}$.

3.3.2. Source estimations from PCA

As is widely known, PCA is a multivariate technique. PCA achieves multivariate data reduction by transforming the data into orthogonal components that are linear combinations of the original variables. Hence, PCA reduces multidimensional data into fewer dimensions..[131] In this study, the concentrations of OC-EC, WSIS and 19 individual PAHs from 50 samples were selected as the active variables. The majority of the variance (87.3%) of the scaled data was explained by five eigenvectors/principal components. The first principal component (PC1) explains 51.6% of the total variance, while the second principal component (PC2) accounts for 16.2% of the total

Table 5. Rotated Component Matrix[a] of carbonaceous compounds, water soluble ionic species and 19 individual PAHs in PM$_{2.5}$ at Doi-Inthanon Chiang-Mai province.

	Principal components (PC)				
Compositions	PC1	PC2	PC3	PC4	PC5
TC	.303	**.836**	.284	−.219	.104
OC	.310	**.834**	.296	−.205	.115
EC	.265	**.812**	.227	−.266	.054
F$^-$	.102	**.742**	.156	−.057	−.395
Cl$^-$	−.186	.002	−.105	**.740**	−.329
NO$_2^-$	.070	−.164	−.195	.656	.078
NO$_3^-$	.039	**.818**	.060	.131	.111
SO$_4^{2-}$	.239	.602	.127	−.100	.643
Na$^+$	.061	**.830**	.116	.220	.078
NH$_4^+$	.161	.606	−.028	−.018	**.711**
K$^+$	.263	**.827**	.251	−.145	.340
Mg^{2+}	−.059	.570	−.058	.684	.210
Ca^{2+}	.303	.697	.200	.218	.116
Ace	.118	.204	**.944**	−.123	−.003
Fl	.092	.060	**.817**	.013	.283
Phe	.116	.286	**.918**	−.117	−.103
Ant	.182	.219	**.915**	−.126	−.090
Fluo	.566	.538	.501	−.143	−.013
Pyr	.673	.543	.350	−.139	−.009
B[a]A	.508	.409	.658	−.204	−.023
Chry	**.770**	.520	−.004	−.215	.048
B[b]F	**.914**	.352	.008	−.087	.058
B[k]F	**.925**	.304	.008	−.063	.070
B[a]F	**.946**	.288	.007	−.075	.037
B[e]P	**.929**	.286	.179	−.074	.043
B[a]P	**.924**	.332	−.075	−.096	.052
Per	**.947**	.277	.056	−.071	.063
Ind	**.960**	.016	.149	.017	.063
B[g,h,i]P	**.919**	.055	.327	−.001	.018
D[a,h]A	**.972**	.047	.149	.007	.045
Cor	**.882**	−.076	.296	.056	.050
D[a,e]P	**.861**	−.094	.203	.115	.069
Variance [%]	**51.6**	**16.2**	**10.6**	**5.20**	**3.70**
Estimated source	**Vehicular Exhaust**	**Biomass Burning**	**Diesel Emissions**	**Sea-Salt Aerosols**	**Agricultural Emissions**

[a]Rotation Method: Varimax with Kaiser Normalization.
[b]Bold loading > 0.70.

variance. Interestingly, PC3, PC4, and PC5 describe 10.6%, 5.20%, and 3.70% of the total variance, respectively (see Table 5).

PC1 represents high loading factors of 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, which can be described as the high molecular weight 4-6 ring PAHs. Since these congeners are deeply connected with vehicle exhaust, it is reasonable to conclude that traffic emissions explain 51.6% of the total variance. It is also worth mentioning that B[k]F and B[g,h,i]P have been used as tracers of vehicle emissions.[132,133] While B[g,h,i]P and Cor are related to gasoline vehicles,[134,135] B[a]P, B[k]F and B[g,h,i]P are identified as tracers of diesel/gasoline emissions.[136] Chry and B[k]F can be considered as geochemical markers for identifying coal combustion.[120,137] While B[b]F and Ind can be used as biomarkers for oil combustion, D[a,h]A, Per, B[a]P and B[e]P are widely considered as tracers for discriminating the air pollutants released from industrial stacks.[120]

PC2 is related to biomass burning, accounting for 16.2% of the total variance, with a high loading factor for OC, EC and WSIS. This PC is believed to be the biomass burning source of carbonaceous compositions. OC, EC and K$^+$ are generated from biomass burning. BB emissions contain a significant amount of WSIS, such as NH$_4^+$, K$^+$, and NO$_3^{-}$.[8,17,18,62] Moreover, BB also

emits significant amounts of carbonaceous aerosols that are composed of both OC and EC into the atmosphere.[18,62,65]

PC3 accounts for 10.6% of the total variance and has high loading factors for Ace, Fl, Phe and Ant. Several studies have reported that the most significant PAH compound from diesel engine exhaust was Phe.[138,139] As a consequence, PC3 can be attributed to vehicle exhaust, particularly diesel emissions. This interpretation is consistent with the fact that Pyr and Fluo can also be considered as chemical tracers of vehicle emissions.[139]

PC4, which explains 5.20% of the total variance, is associated with sea-salt aerosols because of relatively high loading factors of Cl^-. The results suggest a strong influence from maritime aerosols in $PM_{2.5}$. This explanation is in good agreement with previous studies conducted in Brisbane, Australia and in Auckland, New Zealand, highlighting Cl^- as a chemical tracer of maritime aerosols.

The last group, PC5, shows a relatively high loading factor from NH_4^+. It is the only WSIS that possesses a high correlation coefficient ($r = 0.71$) with PC5. It is well known that volatilization from fertilizer is the main source of NH_4^+ in particulate matter.[140,141] reported the photochemical formation of SO_2 from both traffic/industrial activities and biomass burning because of the high affinity of SO_4^{2-} for NH_4^+. SO_4^{2-}, NO_3^- and NH_4^+ mainly form in the atmosphere by gas-to-particle conversion from their precursor trace gaseous species (e.g., SO_2, NO_x and NH_3).[111]

4. Conclusions

The results of the one-year monitoring campaign highlight the importance of biomass burning as one of the main contributors of carbonaceous particles in the ambient air of Don-Inthanon, Chiang-Mai province.

The comparatively high OC/EC ratios (e.g., 6.8 ± 3.0), coupled with the high SOC contents (e.g., $8.12 \pm 0.26 \, \mu g \, m^{-3}$) detected at Don-Inthanon, emphasize the dominant effects of biomass/agricultural waste burning in northern Thailand. HCA illustrates that mixtures of biomass burning, fossil fuel combustion (vehicles and energy production), coal combustion, and industrial emissions are major sources of $PM_{2.5}$. It is also to interesting to note that PCA successfully identified five major sources of $PM_{2.5}$, namely, vehicular exhaust, biomass burning, diesel emissions, sea-salt aerosols and agricultural emissions, which accounted for 51.6%, 16.2%, 10.6%, 5.20% and 3.70%, respectively. Overall, this study provides compelling evidence for manage the air pollution such as policy as a priority for national clean air act policies, strategies and plans.

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