

**CHEMICAL CHARACTERIZATION AND SOURCE APPORTIONMENT OF PM_{2.5}
BOUND WITH PRIMARY AND SECONDARY ORGANIC COMPOUNDS DURING
HAZE AND POST-HAZE OVER THE NORTHERN THAILAND**

by

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ABSTRACT

Northern Thailand has been facing recurrent haze events every year during the local summer season over the past few decades. Biomass burning, a well-known major source of this problem resulting from unsuitable agricultural practices, has caused several adverse impacts on human health, the environment, and socioeconomic aspects. Despite efforts, zero burning policy over northern Thailand has not been successfully implemented, as the haze persists for extended periods. In this research, daily PM_{2.5} was collected at a rural site in Chiang Mai, northern Thailand during haze and post-haze periods in 2019. Bulk chemical components and organic tracers were measured, including organic carbon (OC), elemental carbon (EC), water soluble inorganic ions (WSIIs), crustal and trace elements, nonpolar and polar organic compounds. Average mass of PM_{2.5} were 87 ± 36 and 21 ± 11 $\mu\text{g m}^{-3}$ during haze and post-haze, respectively. Mass closure shows that during haze and post-haze, organic matter, water-soluble inorganic ions, and mineral dust were the top three contributors in PM_{2.5} mass. Source apportionment by positive matrix factorization (PMF) with organic tracers identified three different biomass burning sources including general biomass burning, nearby biomass burning (fresh aerosol), and softwood burning (aged aerosol) and three other sources including secondary sulfate, mixed dusts and traffic related emissions, and secondary nitrate, with contributions of 31%, 19%, 13%, 17%, 15%, and 5% during haze and 5%, 32%, 1%, 34%, 26%, and 2% during post-haze, respectively. Estimated lifetime cancer risk from exposure to PM_{2.5} bound PAHs ranged in moderate risk for the whole period. Biomass burning, softwood burning, and diesel combustion were the most contributor to lifetime cancer risk during haze, while diesel and gasoline combustions were the most contributors during post-haze. Several polar organic compounds display secondary organic aerosols characteristic especially dicarboxylic acids and nitrated aromatic phenols. The ratio of malic acid to succinic acid indicates stronger photooxidation during post-haze than haze period. Future study requires a longer sampling period in non-haze and different site environments for comparison and should be applied with remote sensing information. PMF model is recommended for source apportionment with comprehensive chemical species and organic tracers. Both gas and particulate phases Polycyclic Aromatic Hydrocarbons (PAHs) are recommended for lifetime cancer risk assessment.

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CHAPTER ONE:

INTRODUCTION

1.1 Background and research gaps

Haze event has been become a recurrent problem which emerged as a significant public health and environmental concerns of Thailand in the last decade. Haze event is characterized by elevated concentrations of airborne particulate matter (PM) especially fine size PM with diameter less than 2.5 micrometers ($PM_{2.5}$) and other associate pollutants such as sulfur dioxide (SO_2), nitrogen oxides (NO_x), carbon monoxide (CO), metals and polycyclic aromatic hydrocarbons (PAHs) (Chantara et al., 2019; Kayee et al., 2020; Suriyawong et al., 2023; Wiriya et al., 2016). The occurrences of haze are influenced by a combination of factors such as meteorological conditions (P. D. Hien et al., 2002; Sresawasd et al., 2021; Toh et al., 2013), season of the year and monsoon winds (Fujii et al., 2016; Hansen et al., 2019; Khan, Sulong, et al., 2016; Popovicheva et al., 2016), topography features (mountain valley and peninsular) (Kim Oanh & Leelasakultum, 2011; Promsiri et al., 2023), sources and receptor's site environments (Saksakulkrai et al., 2023), these factors complexly control the intensity and duration of haze episodes.

The emitted pollutants from haze events cause impacts on several aspects. Effects on human health, more than 80% of biomass burning derived PM are the PM fine mode (diameter less than 2.5 micron, $PM_{2.5}$) which is inhalable size that can easily penetrate into deep respiratory system and reach to lung (Finch, 2018). Diverse type of toxic pollutants bound with PM can induce acute effects such as inflammation reactions, oxidative stress and genetic toxicity (cancer) (Z. Li et al., 2017) and cause significant premature death (Shi, Zhao, et al., 2018; S. Yin, 2023). Of environmental aspect, PM emitted during haze cause low visibility (H. H. Lee et al., 2017) black carbon in PM alters radiative forcing in the atmosphere consequently lead to anomalous precipitation pattern (W. S. Lee & Kim, 2010). Of the socioeconomic aspect, smoke haze discourages tourism and outdoor activities, leading to a decline in tourist arrivals, lower revenue for tourism-dependent businesses, and a negative impact on the local economy. Moreover, and

indirect impact from exposure to PM on public health result in absence from work and reduce productivity of individual worker, decrease work efficiency and increase of sick leave, eventually cause significant economic losses (Bui & Nguyen, 2022; X. Zhang et al., 2022).

Haze events emit substantial amount of PM_{2.5}, in northern Thailand haze are generally from anthropogenic activities, particularly agricultural practices since Thailand is known as agrarian country, which contribute significantly to the smoke and haze problems. The practice of open burning agricultural fields, crop residues, and forested areas is a common method employed by farmers to clear land for cultivation or manage agricultural waste (Kawichai, Prapamontol, et al., 2022; Khamkaew, Chantara, Janta, et al., 2016b; N. D. Luong et al., 2022; Sirithian et al., 2018; W. Song et al., 2022; Yabueng et al., 2020). Thai government has therefore established a policy to control any type of open burning called “Zero burning policy” since 2016 to prohibit and eliminate open burning over 9 provinces of northern Thailand for approximately 60 days during local summer season (dry season) between February to April every year. Study by Yabueng et al. (2020) has demonstrated that the policy seemed to be accomplished since the average mass concentration of PM_{2.5} had decreased after the zero policy launched, however, they found that the haze duration become being prolonged from 2 months to 3 months. Their finding reflects that the policy solely may not be sufficient to mitigate haze problem, making this as interesting point for the gap of research.

Biomass burning derived PM_{2.5} in northern Thailand has been identified by using source-specific tracers such as K⁺, OC, and an organic marker, levoglucosan (Chantara et al., 2019; Chuang et al., 2016; Khamkaew, Chantara, Janta, et al., 2016b; Thepnuan et al., 2019b). However, since biomass burning in northern Thailand can be mixed with several types of biomasses such as crop residues (rice straw, maize and sugarcane), forest leaf litter, hard wood and softwood, which conventional markers, as mentioned above, cannot make comprehensive separation. Besides from biomass burning, other sources during haze and non-haze period in northern Thailand have been

identified by several studies, such as traffic (Chansuebsri et al., 2022; Kayee et al., 2020; Khamkaew, Chantara, Janta, et al., 2016b; Kraisitnitikul et al., 2024; Pongpiachan et al., 2017a), secondary inorganic aerosols (Chansuebsri et al., 2022; Chantara et al., 2012; Pengchai et al., 2009; Thepnuan et al., 2019a), soil dust and non-exhausts dusts (Chansuebsri et al., 2022; ChooChuay et al., 2022; Kayee et al., 2020; Kraisitnitikul et al., 2024). From these publications, some of them have performed source appointment by receptor models, principal component analysis (PCA) (Chansuebsri et al., 2022; ChooChuay et al., 2022; Khamkaew, Chantara, & Wiriya, 2016) and one used positive matrix factorization (PMF) (Kraisitnitikul et al., 2024). Nevertheless, none of them have employed a comprehensive set of chemical component species of $PM_{2.5}$ to achieve the insight into source of $PM_{2.5}$ during haze and non-haze period especially the major source of haze event, biomass burning. Moreover, according to Hopke (2015), PCA model, which frequently used in Thailand, is inappropriate approach to be used as a receptor model for source apportionment. Therefore, source apportionment of $PM_{2.5}$ in northern Thailand requires more comprehensive in terms of chemical source-specific markers and proper receptor models.

In the aspect of health assessment, PAHs, the carcinogenic pollutants bound with $PM_{2.5}$ from incomplete combustion, have mostly been paid attention by number of previous studies in northern Thailand especially during haze period (Chuesaard et al., 2014; Kongpran et al., 2021; Pongpiachan et al., 2017a; Wiriya et al., 2013a; Yabueng et al., 2020). They performed cancer risk assessment using Benzo(a)pyrene toxicity equivalent concentration (TEQ_{BAP}) of individual PAHs. While some study employed only heavy metals as carcinogen (Kawichai, Bootdee, et al., 2022; Sakunkoo et al., 2022). There has been no study that has done cancer risk assessment by PAHs coupled with heavy metals. Furthermore, making use of source apportionment by PMF model, the obtained contributions of identified source to mass concentration of carcinogenic species (both PAHs and heavy metals) allows to perform source-specific cancer risk assessment. This approach

is a very useful for local policymakers and regulators of northern Thailand to develop targeted strategies to prioritize and mitigate emissions from specific sources.

In addition to biomass burning derived $PM_{2.5}$, organic matter (OM) is the greatest contributor of $PM_{2.5}$ mass especially during haze event. The contribution of OM range more than 25% of $PM_{2.5}$ mass depending on type of PM source and area (Saksakulkrai et al., 2023). OM is composed of a several number of compounds, which can be separated by their polarity characteristics into two main groups: nonpolar and polar organic compounds. Study on polar and nonpolar organic compounds provides an insight into the complexity of atmospheric chemical reactions of the molecular organic compounds in $PM_{2.5}$ such as oxidation, photochemical reactions and gas-to-particle conversion which link to the formation of secondary organic aerosol (SOA) (Angove et al., 2006; Glasius et al., 2000; Kanellopoulos et al., 2020b; Kavouras et al., 1999). Over the past few years, atmospheric scientists have paid attention on these compounds, a number of studies have mostly conducted in East Asia such as China (Cao et al., 2017, 2021; H. Guo et al., 2015; Kang et al., 2018; X. Li et al., 2016; J. Liu et al., 2021; T. Liu et al., 2020; Mu et al., 2021; Q. Wang et al., 2016; S. Zhang et al., 2021), Japan (Ichikawa et al., 2018; Ikemori et al., 2021) and Korea (S.-W. Park et al., 2022), but so far, there has been no study in Thailand. Given the variations in meteorological conditions compared to those previous studies in East Asia, the characteristics of polar and nonpolar organic compounds in northern Thailand may exhibit differently. Consequently, this presents an opportunity to undertake a first characterization into the polar and nonpolar compounds in northern Thailand.

In summary, given the overall research gaps mentioned earlier, this research aims to i) characterize chemical compositions of $PM_{2.5}$, ii) apportion the potential sources of $PM_{2.5}$ by using comprehensive set of chemical components as well as a better source-specific markers for biomass burning, iii) perform source-specific life time cancer risk assessment from PAHs coupled with heavy metals and iv) investigate characteristics of polar and nonpolar organic compounds

involving primary and secondary organic aerosols. All these aims will be examined comparatively between the haze and post-haze period.

1.2 Nonpolar organic compounds

Nonpolar molecules have equal electron distribution in a covalent bond, so the net electrical charge of the molecule is zero. Nonpolar organic compounds in this study include alkanes and polycyclic aromatic hydrocarbons (PAHs), which will be discussed in the following subtopics.

1.2.1 Alkanes

1.2.1.1 Background and previous field study of alkanes

Alkanes are a group of saturated hydrocarbon compounds that consist solely of carbon and hydrogen atoms arranged in a straight or branched chain structure. Aliphatic hydrocarbon like alkanes do not directly cause adverse impacts on human health but alkanes have an important role on atmospheric chemistry as precursor of SOA (J. Li et al., 2022). In the atmosphere, alkanes can be emitted from many sources, from natural source such as plant wax, biogenic volatile organic compounds (BVOCs) and vegetation, and from anthropogenic activities such as the combustion of diesel, gasoline, natural gas, industry processes and coal combustion (Wang et al., 2016; Ichikawa et al., 2018; Caumo et al., 2020; Cao et al., 2021; Fadel et al., 2022; Park et al., 2022). Alkanes consist of different number of carbons from short chain ($C \leq 16$) to long chain ($C > 16$) (J. Yang et al., 2023) which is very useful as an indicator for wide range of combustion sources such as fuels (Caumo et al., 2020) and source of $PM_{2.5}$ (J. Yang et al., 2023). Alkanes emitted in the atmosphere as POA can undergo reactions with atmospheric oxidant such as ozone (O_3), nitrate radical (NO_3),

and hydroxy radicals ($\cdot\text{OH}$), these reactions create highly reactive intermediate molecules such as peroxy radicals ($\text{ROO}\cdot$) (Madhu et al., 2023).

The study on homologue alkanes in field experiment has been done in different durations and areas. A several number of recent field studies were done in China; Beijing, Yang et al. (2023) demonstrated that alkanes varied by season with higher concentrations observed in winter and lower concentrations in summer and fall; Guangzhou, Wang et al. (2016) found similar seasonal and bimodal distribution of alkane homologues with abundant contribution (69%) of high carbon number alkanes ($\text{C}_{24} - \text{C}_{33}$) to total identified alkanes; Xu et al. (2013) investigated alkanes related to the 16th Asian Games and found that The average concentrations of alkanes were much higher in the nighttime (51.6 ng m^{-3}) than during the day (41.3 ng m^{-3}) due to a restriction on motor vehicle operation during the day and the accumulation of pollutants due to temperature inversions at night; in three megacities of Yangtze river delta, Cao et al. (2021) reported total alkane concentrations of 194 ng m^{-3} and 184 ng m^{-3} in Hangzhou and Nanjing, respectively, which are about twice that of Shanghai (84 ng m^{-3}); Sanya, Wang et al. (2015) demonstrated that the concentration of alkanes was generally higher in winter than summer and total concentration of alkanes were 23.8 ng m^{-3} and 5.7 ng m^{-3} in winter and summer, respectively. Of other countries of east Asia, in Japan, Ichikawa et al. (2018) reported at an industrial area of Chiba Prefecture that total alkanes was highest in winter 2016 (21 ng m^{-3}) and seasonal variation was not significant, indicating alkanes were not influenced by seasons. In Korea, Park et al. (2022) observed that the highest concentration of alkanes occurred during Asian dust event, the dominant alkanes carbon number were C_{27} , C_{29} and C_{31} which emitted from biogenic sources. Of other regions such as India; Rani et al. (2023) investigated alkanes in size-segregated aerosols during Diwali Festival and found that the concentration of alkanes was minimum in coarse size fractions while a noticeable difference was observed in fine and sub-micron ranges and 20-36 carbon atoms were found as the most abundant.

1.2.1.2 Carbon preference index

The carbon preference index (CPI) is calculated by dividing the sum of odd number carbon of alkanes (ΣC_{odd}) concentration by the sum of odd number carbon of alkanes concentration (ΣC_{even}) proposed by Bray and Evans (1961). CPI values can be used to identify the sources of n-alkanes in the atmosphere. In natural environments, such as plants and microorganisms, the synthesis of alkanes usually results in a larger fraction of alkanes with odd carbon numbers. Conversely, anthropogenic sources of alkanes, such as the combustion of fossil fuels, industrial emissions, and petroleum combustion, tend to produce a higher proportion of alkanes with even carbon numbers. Hence, when the Carbon Preference Index (CPI) exceeds 1, it indicates a greater abundance of odd-numbered alkanes originating from both plant emissions and low-temperature biomass combustion. A CPI value greater than 5 indicates a predominance of biological sources, while a CPI value between 1 and 5 suggests a mixed source of biogenic and anthropogenic emissions (Iinuma et al., 2007), on the other hand, $\text{CPI} < 1$ indicates the higher contribution of anthropogenic emissions (M. Zheng et al., 2006).

Several studies on alkanes made use of carbon preference index, especially in China, there have been number of reports demonstrating that CPI observed in to 1 indicating the influence of anthropogenic, for example, 1.2, 1.1, and 1.2 found in Shanghai, Hangzhou, and Nanjing, respectively (Cao et al., 2021), CPI ranged from 0.6 to 2.4 (average = 1.1) was observed in Sanya (J. Wang et al., 2015), CPI 1.4 was found during the 16th Asian Games in Guangzhou (H. M. Xu et al., 2013). Of other countries, CPI has also been reported the value close to 1, for example, in Lebanon overall CPI was approximately 1 (Fakhri et al., 2023), in Ichihara, Japan CPI values during the study period ranged from 0.73 to 3.2, with an average of 1.5 (Ichikawa et al., 2018), in Chuncheon, South Korea. The average CPI value was 1.73 with relatively large variation, ranging from 0.57 to 3.99 (S.-W. Park et al., 2022), in India the CPI values were calculated for different size ranges during the Diwali festival and varied in the range of 1.0-2.1, with an average value of 1.6

(Rani et al., 2023). From these studies, it should be noted that alkanes observed are generally influenced by anthropogenic sources.

1.2.2 PAHs

1.2.2.1 Background

Polycyclic aromatic hydrocarbons (PAHs) are a group of organic compounds composed of more than 2 fused aromatic rings which are generally generated from incomplete combustion of organic materials such as oil, coal, petrol, and wood (Abdel-Shafy & Mansour, 2015). PAHs in environments can be produced from various sources and can be found in all parts of environment such as the atmosphere, soil and sediment, the aquatic system, as well as lipid tissue of organisms. Source of PAHs are from both natural and anthropogenic sources (WHO, 1998). The anthropogenic sources of PAHs including incomplete combustion of hydrocarbon fuels, garbage, or other organic substances such as tobacco and plant materials (pyrogenic sources), by oil spills and other petrochemical products (petrogenic sources), or by the post depositional transformation of biogenic precursors (diagenetic sources) (Pongpiachan et al., 2015a). While volcanic eruptions, forest fires, open burning, natural losses or leakage of petroleum or coal deposits are attributed to the natural sources (Abdel-Shafy & Mansour, 2015; Y. Zhang & Tao, 2009). The combustion sources of PAHs are considered as a major primary source which accounted for over 90% of PAHs concentrations in the environment (Howsam & Jones, 1998).

According to U.S Environmental Protection Agency (US-EPA), there are 16 prioritized PAHs including Naphthalene (Nap), Acenaphthene (Ace), Acenaphthylene (Acy), Anthracene (Ant), Fluorene (Flu), Phenanthrene (Phe), Fluoranthene (Flua), Pyrene (Pyr), Benzo(a)anthracene (BaA), Chrysene (Chry), Benzo(b)fluoranthene (BbF),

Benzo(k)fluoranthene (Bkf), Benzo(a)pyrene (BaP), Dibenzo(a,h)anthracene (DahA), Indeno(1,2,3-cd)pyrene (Ind) and Benzo(ghi)perylene (BghiP), they are listed in as Table 1-1 with number of rings. They can be classified into two groups based on their molecular weight: low molecular weight (2 – 3 LMW) PAHs with 2 – 3 aromatic rings and high molecular weight (HMW) PAHs with ≥ 4 aromatic rings.

PAHs have received substantial attention from many previous studies owing to their toxicity as carcinogenic pollutants. Understanding the composition and levels of PAHs in PM_{2.5} can provide valuable insights into the sources and emission patterns of PAHs compounds for assessing the potential risks to human health. Previous studies regarding the level of PAHs have been conducted in many countries and the reported PAHs level varied upon area and duration, for example, China; Beijing, 116 ng m⁻³ in 2003 (J. Zhou et al., 2005), 24.8, 21.0, 0.43 and 22.2 ng m⁻³ in winter 2013, winter 2014, summer 2014 and winter 2015, respectively (H. Yin & Xu, 2018), Guangzhou, annual average 33.9 ng m⁻³ (J. Liu et al., 2015), Xi'an, ranged from 49.6 – 140.0 ng m⁻³ (J. Wang et al., 2017), Nanjing, ranged from 4.03 – 102 ng m⁻³ (Y. Li et al., 2019); in India, Delhi were 96.0 and 45.8 ng m⁻³ in winter and summer (D. P. Singh et al., 2011); while in Thailand PAHs were reported with lower values, Chiang Mai, 25.9 and 4.6 ng m⁻³ in dry season 2010 and 2011, respectively (Wiriya et al., 2013a), in 2010 ranged from 0.6 – 4.14 ng m⁻³ (Chuesaard et al., 2014), haze 2016 was 10.2 ng m⁻³ (Thepnuan & Chantara, 2020), haze 2017 and haze 2018 were 2.6 and 2.0 ng m⁻³, respectively (Yabueng et al., 2020), and haze 2019 was 6.6 ng m⁻³ (Insian et al., 2022).

Table 1-1 U.S.EPA 16 priority PAHs and their ring number, molecular weight and toxicity equivalent concentration (TEQ) from Nisbet and LaGoy, (1992) and U.S.EPA. (1993)

Name	Abbreviation	No. of ring	Molecular weight (g mol ⁻¹)	Nisbet and Lagoy, 1992	U.S.EPA, 1993
Naphthalene	Nap	2	128.7	0.001	-
Acenaphthene	Ace	3	154.21	0.001	-
Acenaphthylene	Acy	3	152.19	0.001	-
Fluorene	Flu	3	166.22	0.001	-
Phenanthrene	Phe	3	178.23	0.001	-
Anthracene	Ant	3	178.24	0.01	-
Fluoranthene	Flua	4	202.26	0.001	-
Pyrene	Pyr	4	202.25	0.001	-
Benz(a)anthracene	BaA	4	228.29	0.01	0.06
Chrysene	Chry	4	228.29	0.01	-
Benz(b)fluoranthene	BbF	5	252.31	0.1	0.07
Benz(k)fluoranthene	BkF	5	252.31	0.1	0.07
Benz(a)pyrene	BaP	5	252.31	1	1
Dibenz(a,h)anthracene	DahA	5	278.35	1	0.6
Benz(ghi)perylene	BghiP	6	276.33	0.01	-
Indeno(1,2,3-cd)pyrene	Ind	6	276.33	0.1	0.08

1.2.2.2 Toxicity equivalent quotient concentration (TEQ) and cancer risk assessment by PAHs

Toxicity equivalent quotient concentration (TEQ) is a measure to assess the combined toxic effects of multiple PAHs based on their individual toxicities as some PAHs are more potent and have more adverse health effects compared to others. TEQ for individual PAHs are determined by multiplying the concentration of each PAH in a mixture by its respective toxic equivalency factors (TEF) based on their relative toxicity compared to a reference compound, benzo(a)pyrene, which is considered as a highly potent and carcinogenic PAH. Equivalency factors are first proposed by Nisbet and LaGoy, (1992) and adapted by U.S.EPA. (1993) (listed in Table 5-1). TEQ_{BaP} is then defined by the following equation

$$TEQ_{BaP} = \sum (PAH \times TEF)$$

Exposure to PAHs has been associated with an increased risk of cancer. Assessing the potential cancer risk associated with PAH exposure is essential for understanding the potency of the problem and guiding regulatory measures to protect public health. Lifetime cancer risk assessment considers several factors, including the toxicity of PAHs, the duration and intensity of exposure, and individual susceptibility (Famiyeh et al., 2021). Incremental lifetime cancer risk (ILCR) is defined as “*the incremental probability of an individual to develop cancer over a lifetime as a result of exposure to a potential carcinogen*” (USEPA., 2019). Obtained TEQ_{BaP} from measured individual PAHs can then subsequently be employed to estimate ILCR as follows:

$$ILCR = CSF \times \frac{TEQ_{BaP} \times IR \times ED \times Ef}{BW \times AT}$$

where CSF, IR, ED, Ef, BW and AT represent slope factor of inhalation, inhalation rate (m³ day⁻¹), exposure duration (year), exposure frequency (day year⁻¹), average body weight (kg) and lifetime average exposure time (year), respectively. Estimated ILCR below the threshold of 10⁻⁶ is considered as negligible risk, 10⁻⁴ > ILCR > 10⁻⁶ is considered as moderate risk and ILCR > 10⁻⁴ is considered as high risk (Famiyeh et al., 2021).

Multiple previous studies have estimated ILCR from measured PM_{2.5} bound PAHs. In Thailand, the ILCR were usually in the medium risk range, for example, studies in northern Thailand especially during haze events, year 2016 was 1 x 10⁻⁴ (Thepnuan & Chantara, 2020), year 2017 and 2018 were 4.71 x 10⁻⁵ and 3.88 x 10⁻⁵, respectively (Yabueng et al., 2020), year 2019 was 1.14 x 10⁻⁴ (Insian et al., 2022), while from combined of gas and particulate bound PAHs ILCR were 9.3 x 10⁻⁴ and 2.5 x 10⁻⁵ for haze and non-haze, respectively, (Kongpran et al., 2021). In China, ILCR were found to be comparatively higher especially in Beijing, were 3.31 x 10⁻³ and 4.39 x 10⁻⁵ in winter and summer (H. Yin & Xu, 2018), while other cities were lower, such as Guangzhou, 5.98 x 10⁻⁴ (J. Liu et al., 2015) and Xi'an, 6.5 and 5.4 x 10⁻⁵ for outdoor and indoor of middle-school (J.

Wang et al., 2017). In Malaysia ILCR were relatively lower than of Thailand and China, in Kuala Lumpur was 2.4×10^{-7} (Sulong et al., 2019a), highest in adult 2.27×10^{-5} (Sulong et al., 2017) and Klang valley, 3×10^{-7} (Jamhari et al., 2014a). Additionally, some recent studies applied receptor model, PMF, by analyzing the concentrations of pollutant species to identify specific emission sources, the PMF model can estimate the contribution of carcinogenic pollutants (PAHs or heavy metals) of each source which can then be used to estimate ILCR. For example, source specific ILCR in China were 9.2×10^{-5} (industrial emissions), 2.6×10^{-5} (diesel and coal combustions), 2.2×10^{-5} (resuspended dust), 1.7×10^{-5} (gasoline combustion) and 6.4×10^{-6} (Tian et al., 2022), in central Tehran were 1.08×10^{-6} (petroleum), 0.66×10^{-6} (natural gas and biomass burning), 1.75×10^{-6} (industrial emission), 2.46×10^{-6} (diesel combustion) and 0.46×10^{-6} (gasoline combustion) (Taghvaei et al., 2018).

1.3 Polar organic compounds

Polar organic compounds comprise of a number of diverse groups of chemical substances, characterized by their polarity due to the presence of functional groups such as hydroxyl (-OH), carbonyl (C=O), or carboxyl (-COOH). In this study, polar organic compounds including saccharides, carboxylic acids, and nitro aromatic phenols will be discussed regarding their background and previous research related to primary and secondary organic aerosols.

1.3.1 Saccharides

Saccharides, a class of organic compounds commonly known as sugars. Saccharides in PM_{2.5} originate from various sources, including both natural and anthropogenic emissions. Natural sources include biogenic emissions from vegetation, such as pollen, plant debris, and fungal spores. Anthropogenic sources related to combustion processes, such as biomass burning, fossil fuel combustion, and industrial activities (Kang et al., 2018; Ren et al., 2020; Xiao et al., 2018).

Anhydrosugars, members of saccharides group, including levoglucosan (1,6 – anhydro- β -D-glucopyranose), mannosan and galactosan, have received very much attention from previous studies in the field of atmospheric chemistry and air pollution research especially levoglucosan, due to its high abundant, stability property in ambient environment, resistant to degradation in the atmosphere and subsequently has a relatively long atmospheric lifetime as a robust organic marker for biomass burning. The formation of levoglucosan occurs specifically during the thermal degradation of cellulose and hemicellulose, major components of biomass (Bhattarai et al., 2019; Simoneit, 2002). This unique chemical compound is released into the atmosphere as a result of biomass burning activities. Its presence in atmospheric samples, particularly in fine particulate matter (PM_{2.5}), serves as a clear indicator of biomass burning contributions to air pollution. Among anhydrosugars isomers, levoglucosan is the major species contribute of more than 90% to total anhydrosugars (Simoneit et al., 1999).

Other isomers, mannosan and galactosan have also been frequently observed along with levoglucosan. As different types of biomasses have different compositions, hardwood contains proportionally more hemicellulose while softwood contains more proportion of cellulose. Levoglucosan is a product of cellulose and hemicellulose pyrolysis while mannosan and galactosan are isomer of hemicellulose, therefore, the ratio of levoglucosan to mannosan can be used to differentiate the type of biomass (W. Li et al., 2021). A higher levoglucosan to mannosan a greater contribution of hardwood or non-softwood biomass burning, such as agricultural residues or crop burning, conversely, a lower ratio suggests a higher proportion of softwood burning contribution. Previous studies demonstrated that the levoglucosan/mannosan ratio higher than 10 indicate hardwood burning (S. Hou et al., 2021), and higher than 40 indicates crop residues burning (P. Fu et al., 2016). Moreover, the ratio of levoglucosan to galactosan can be used to identify the type of woods, straws, grasses, and needles (S. Hou et al., 2021). Fig. 1-1 display an

example use of scatter plot of levoglucosan/mannosan vs. levoglucosan/galactosan to differentiate type of biomass burning.

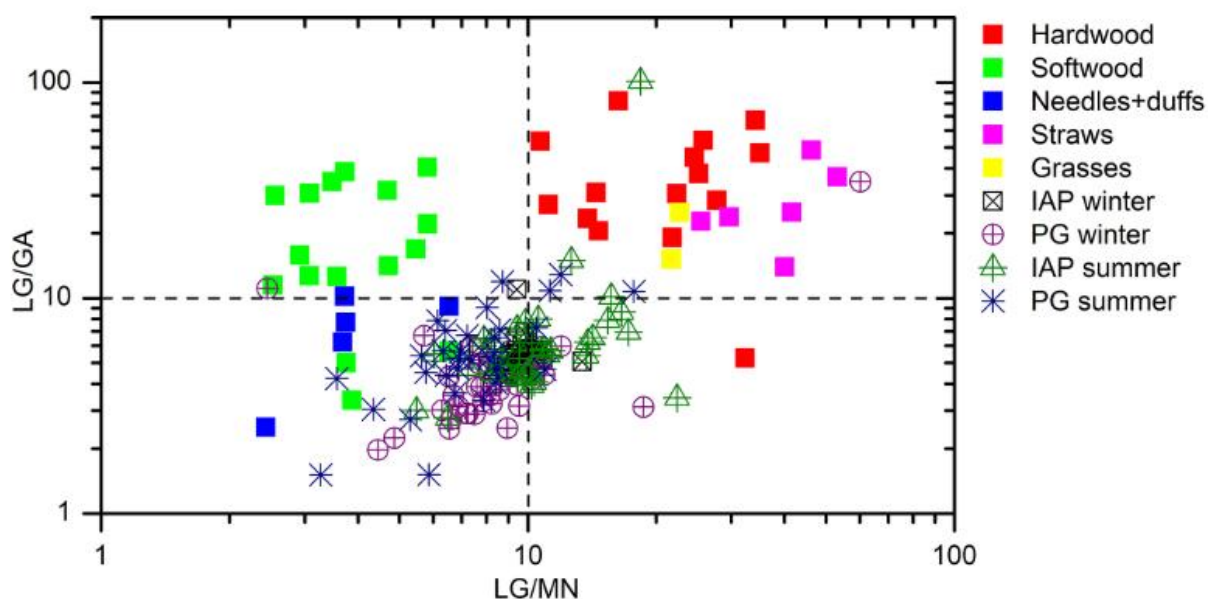


Fig. 1-1 Scatter plot of levoglucosan/mannosan vs. levoglucosan/galactosan. (S. Hou et al., 2021)

Anhydrosugars are considered as primary organic aerosols since they are primarily emitted from thermal decomposition of cellulose. Field experiments during haze events in several recent studies have shown the consistent finding that levoglucosan was the most abundant organic compound in organic matter mass in across region such as Southeast Asian region (Fujii, Kawamoto, et al., 2015; Jaafar et al., 2018; Lan et al., 2021; Nguyen et al., 2016; Thepnuan et al., 2019a; Tsai et al., 2013), in China (Cheng et al., 2013; K. F. Ho et al., 2016; L. Zheng et al., 2018), Atlanta (B. Yan et al., 2008), Australia (Iinuma et al., 2016), California (Y. Liang et al., 2021).

Other members of the saccharides group are sugar alcohols. Previous studies have demonstrated that sugar alcohol can undergo through photooxidation reactions in the atmosphere to form secondary organic aerosols, for example, threitol and erythritol are the products of photodecomposition of 1,3-butadiene in the atmosphere containing NO (Angove et al.,

2006), photooxidation of isoprene (Claeys et al., 2004) and photooxidation of isoprene with acidic conditions (Surratt et al., 2010).

1.3.2 Carboxylic acids

Carboxylic acids are a class of organic compounds that contain a or more carboxyl group (-COOH). Carboxylic acids can be classified based on the number of carboxyl groups present in their structure. Monocarboxylic acids have a single carboxyl group, dicarboxylic acids have two carboxyl groups, and tricarboxylic acids have three carboxyl groups. These different types of carboxylic acids exhibit unique chemical properties and reactivity. Carboxylic acids, especially dicarboxylic acids, have been identified as important components of SOA and serve as tracers for the identification and quantification of SOA sources (Glasius et al., 2000; Hsieh et al., 2008; Kawamura & Bikkina, 2016; Kundu et al., 2010; J. Liu et al., 2021; Q. Yu et al., 2021).

Monocarboxylic acid: Important monocarboxylic acids especially for PM derived from biomass burning are resin acid derivatives, for example, dehydroabietic acid, pinonic acid and pimaric acid. Resin acids are commonly found in the resinous exudates of certain trees, particularly conifers, they are also SOA markers from combustion of softwoods such as pine trees (Fine et al., 2004b, 2004a; Hays et al., 2002; Mancilla et al., 2021). Dehydroabietic acid have been a significant markers for softwood burning and aged aerosols from field studies since its high abundant among resin derivatives (Leithead et al., 2006; Y. Liang et al., 2021; Mancilla et al., 2021; Mikuška et al., 2017; B. Yan et al., 2008) Study by Fine et al. (2004b, 2004a) have clearly demonstrated that dehydroabietic acid is only found in softwood burning with the significant amount compared to other resin derivatives. Besides from biomass burning derived organic markers, 2,3-dihydroxy-4-oxopentanoic acid DHOPA, is also an important SOA tracer for anthropogenic emissions (Srivastava et al., 2018; Q. Wang et al., 2022). Srivastava et al. (2023)

demonstrated that DHOPA is derived from the oxidation of toluene and *m*-xylene under varied NO_x concentrations.

Dicarboxylic acids: There are several important SOA tracers in this group such as oxalic acid (C2), malonic acid (C3), succinic acid (C4), glutaric acid (C5), adipic acid (C6), pimelic acid (C7), suberic acid (C8), azelaic acid (C9), malic acid (hC4), phthalic acid (o-Ph) and isophthalic acid (p-Ph). They have been extensively investigated in both laboratory (Claeys et al., 2004; Glasius et al., 2000; Hatakeyama et al., 1985; Stephanou & Stratigakis, 1993) and field experiments (Cao et al., 2017; Hsieh et al., 2008; Kawamura & Ikushima, 1993; Kawamura & Sakaguchi, 1999; Kundu et al., 2010). It is found that aerosols from different geographical area has different molecular distribution of dicarboxylic acids and found that C2 mass has similar trend with solar irradiation and temperature suggesting significant photochemical oxidation to C2 (Kawamura & Bikkina, 2016). During long range transport, aerosols undergone through aging process, dicarboxylic acids are observed to be greater as the aerosols aged during daytime owing to the an elevated production of dicarboxylic acid from photochemical oxidation (Satsumabayashi et al., 1990). C2 was observed as the most abundant species in aerosol among low molecular weight dicarboxylic acids followed by C3 and C4 (Kawamura & Sakaguchi, 1999). Dicarboxylic acids can also contribute the aerosol nucleation process by binding to sulfate acid and ammonia (W. Xu & Zhang, 2012).

In terms of impact of climate, since their high water solubility and hygroscopic properties, dicarboxylic acids can act as cloud condensation nuclei (Kerminen et al., 1999), in addition, the surface tension of water cloud droplets decrease with the dicarboxylic acids concentration dissolved in the droplets which may alter the formation of cloud (Facchini et al., 2000).

Since succinic acid (C4) can primarily be emitted from biomass burning (Cao et al., 2017), whereas malic acid (hC4) is a secondary organic aerosol (SOA) formed through the

photooxidation of succinic acid (Kawamura & Ikushima, 1993). The ratio between malic acid and succinic acid (hC_4/C_4) has been utilized as an indicator of the strength of photochemical reactions (Kawamura & Sakaguchi, 1999; Q. Yu et al., 2021). Glutaric acid (C_5) and adipic acid (C_6) can be generated through the photochemical oxidation of anthropogenic cyclohexene (Hatakeyama et al., 1985) and aromatic hydrocarbons (Kawamura & Ikushima, 1993). Suberic acid and azelaic acid (C_9) are believed to be produced through the photooxidation of biogenic unsaturated fatty acids (Kaplan & Kawamura, 1987; Stephanou & Stratigakis, 1993). Phthalic (o-Ph) and isophthalic (p-Ph) acids are secondary organic aerosols derived from anthropogenic sources, formed through the aging process of aerosols (J. Li et al., 2020). The ratios of adipic acid to azelaic acid (C_6/C_9) and phthalic acid to azelaic acid (o-Ph/ C_9) can be utilized to distinguish between the contributions of biogenic and anthropogenic sources (Kundu et al., 2010).

1.3.3 Nitro aromatic phenols

Nitro aromatic phenols are a class of organic compounds that contain both a phenolic group (-OH) and one or more nitro ($-NO_2$) groups attached to an aromatic ring, for example, 4-nitrophenol, 4-nitrocatechol, bisphenol A. They have received attention since nitro aromatic phenols contribute to reduced visibility by forming brown carbon particles (Y. Wang et al., 2019; Yuan et al., 2021), influence climate patterns through their radiative effects, and participate in complex chemical reactions in the atmosphere (Y. Chen et al., 2022). Among nitro aromatic phenols, 4-nitrocatechol and 4-nitrophenol have been focused from previous studied since they are the most abundant in this group and as serves as crucial markers for secondary organic aerosols and plays a significant role in atmospheric studies (Li et al., 2020; Yuan et al., 2021; Cai et al., 2022; Zhang et al., 2023). 4-nitrocetachol is a dominant product of SOA from biomass burning derived catechol reaction with OH and NO_3 radicals in the existence of NO_x (Finewax et al., 2018) and due to its long atmospheric lifetime and elevate as the aerosol age (J. Li et al., 2020), 4-nitrocetachol has been proposed as a molecular marker to identify aging process for aged

aerosol (Bertrand et al., 2018; Y. Liang et al., 2021; Q. Wang et al., 2022) which is very useful for study of long range transport of aerosol from biomass burning.

Previous field studies have used 4-nitrocatechol as biomass burning marker in several area such as China (Wang et al., 2019; Li et al., 2020; Yuan et al., 2021), California, USA (Y. Liang et al., 2021), Melbourne, Australia (Iinuma et al., 2016), Japan (Ikemori et al., 2021) and Hongkong (Q. Wang et al., 2022).

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CHAPTER TWO:

AIRBORNE PARTICULATE MATTER IN

SOUTHEAST ASIA: A REVIEW ON

VARIATION, CHEMICAL COMPOSITIONS

AND SOURCE APPORTIONMENT

Airborne particulate matter in Southeast Asia: a review on variation, chemical compositions, and source apportionment

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2.1 Abstract

Southeast Asia region locates in a tropical zone with extensive agricultural activities. Airborne Particulate Matter (PM) in this region is the most important air pollutant, causing millions of premature deaths. This review aims to provide an overview of variation of the levels, chemical compositions, and sources of PM in Southeast Asian countries, and to compare with studies from megacities in other regions. Daily average PM_{2.5} concentrations in Southeast Asia ranged from 11 to 54 $\mu\text{g m}^{-3}$, which are lower than polluted megacities such as Delhi but substantially higher than 24-hr mean air quality guideline by the WHO (15 $\mu\text{g m}^{-3}$). Levels of PM_{2.5} in maritime continental Southeast Asia (Indonesia, Brunei, Malaysia, Philippines, and Singapore) are lower than those of mainland continental Southeast Asia counties (Cambodia, Myanmar, Thailand, Vietnam, and Laos). PM levels are usually two times or higher during haze episodes or dry season than during non-haze days or wet seasons. Organic carbon is a key chemical component in Southeast Asia, contributing to 9 to 52% of PM mass. SO_4^{2-} , NO_3^- , and NH_4^+ are major ions and $\text{NO}_3^-/\text{SO}_4^{2-}$ ratios in Southeast Asia were notably lower than megacities in other regions, implying lower emissions from mobile relative to stationary sources. Source apportionment shows that biomass burning is one of the most important sources particularly during the haze (dry) seasons. Its contribution to PM mass is mostly higher than those in China and India. Road traffic emission was another major contributor in dense-traffic cities such as Bangkok, Kuala Lumpur, and Hanoi. The contribution of secondary aerosols is frequently reported to be lower level than cities from other regions. Sea salt is found in coastal Southeast Asian countries associated with monsoon. Lack of observations of specific trace metals and organic aerosol tracers makes it difficult to quantify the contribution of industrial and fossil fuel combustions to PM. We suggest future PM source apportionment studies in Southeast Asia to make a more comprehensive measurement of both inorganic and organic tracers and apply both chemical mass balance and multivariate receptor models.

2.2 Introduction

Particulate matter (PM) is one of the most important air pollutants. PM is often classified by their aerodynamic diameter: Inhalable (PM_{10}), fine ($PM_{2.5}$), submicrometer (PM_1) and ultra-fine particles ($PM_{0.1}$) referring to particles with aerodynamic diameter less than 10, 2.5, 1 and 0.1 μm , respectively. PM can be produced from both anthropogenic activities such as road traffic, fossil fuel or biomass burning, and natural sources such as dust resuspension, forest fires, pollens, and volcanic eruption; these are called primary particles. PM can also be formed in the atmosphere through chemical reactions (Kelly & Fussell, 2012), which is known as secondary particles.

There is compelling evidence that atmospheric particulate matters cause adverse effects on human health (A. Hassan et al., 2017; A. F. W. Ho et al., 2018; L. T. M. Luong et al., 2020; Noomnual & Shendell, 2017; Shou et al., 2019; Wan Yaacob et al., 2016). Many studies showed exposure to ambient PM from traffic sources (Both et al., 2013; Hieu et al., 2013; Jinsart et al., 2012; Noomnual & Shendell, 2017; Saksena et al., 2008), biomass or open burnings (Pani et al., 2019b; Sulong et al., 2017; Uda et al., 2019), as well as indoor sources (Z. Li et al., 2017) increase health risk and death rate. Premature mortality related to straw burning in China showed a 3% increase with every $10 \mu g m^{-3}$ $PM_{2.5}$ increase (G. He et al., 2020). Long-term exposure of $PM_{2.5}$ from peatland fire and haze events caused 648 mortality cases per year in Kalimantan, Indonesia (Uda *et al.*, 2019). A recent study reveals that eliminating forest and vegetation fires in Southeast Asia mainland could reduce 59,000 premature deaths yearly (Reddington *et al.*, 2021).

Due to the urbanization, industrial and economic growth in the region as well as other parts of the world (D. Yang et al., 2018), $PM_{2.5}$ concentration over Southeast Asia has been consistently increasing from 1999 to 2014 with a rate of $0.02 \mu g m^{-3}$ per year and PM levels exceeded the World Health Organization average annual limit of $PM_{2.5}$ standard ($5 \mu g m^{-3}$) in most

areas of Southeast Asia (Shi, Matsunaga, et al., 2018). Ground-based measurements of PM and other air pollutants have been conducted by many field experiments over Southeast Asian countries or adjacent areas in neighboring downwind countries / regions (e.g., Taiwan) which are affected by PM emitted from Southeast Asia (Yadav *et al.*, 2017), or where the PM from East Asia, such as China affecting PM in Southeast Asia region, due to winter monsoon (Cohen *et al.*, 2010b; Hien *et al.*, 2011).

Although less intensively studied, PM in Southeast Asia is unique across the world due to the tropical and subtropical climate and the large contribution of biomass burning (BB) during the dry seasons. Southeast Asia is well known as a vast agricultural crop region and agricultural residues have annually been disposed on the field and need to be cleared before the new harvest season starts, by a common method, open burning (Kim Oanh *et al.*, 2018). This BB seasonally occurs every year depending on temporal and spatial variations of meteorological factors due to the two distinctive topographies: namely continental and maritime continental Southeast Asia. Continental Southeast Asia encompasses countries with predominantly continental geography, situated away from coastlines. This region includes Cambodia, Myanmar, Thailand, Vietnam, and Laos. In contrast, Maritime Southeast Asia comprises countries primarily located along the coast, distinguished by peninsulas extending into the sea or islands. This group includes Indonesia, Brunei, Malaysia, the Philippines, and Singapore. BB seasons in Southeast Asia generally initiate during local dry season, around February to April, and June to September, for continental and maritime continental Southeast Asia, respectively (Huang *et al.*, 2020; Xian *et al.*, 2013). It is estimated that the emission of PM_{2.5} in topical Asian countries during year 2000 – 2006 accounted approximately for 3.0 – 3.4 Tg yr⁻¹, and the main contributors of BB were Indonesia, India, Myanmar, and Cambodia (Chang & Song, 2010), while during year 2010 – 2015 the emission of PM_{2.5} and PM₁₀ from crop residue burning solely were 2.0 and 1.8 Tg yr⁻¹, respectively, in the

Southeast Asian countries, with the main contributors as Indonesia, followed by Vietnam, Myanmar, Thailand, and Philippines (Kim Oanh *et al.*, 2018).

Several review articles focused on PM over different regions such as Southern Asia (N. Singh *et al.*, 2017), India (Pant & Harrison, 2012), China (Zhu *et al.*, 2018), and Europe (Belis *et al.*, 2013). More recently, Adam *et al.* (2021) reviewed the biomass burning derived PM over Southeast Asia region, including the latest literature from 2010 to 2020. They concluded that vegetation and peatland fires release significant amount of different PM of size and composition, affecting environment, the climate, air quality, and human health in Southeast Asia region. Dahari *et al.* (2021) also compared the inorganic water-soluble ions and trace elements in PM_{2.5} in Malaysia with data from other Southeast Asian countries. However, to date, there has been no comprehensive review on the compositions, sources, and processes of PM in Southeast Asia.

Support information of this chapter is in supplement A

2.3 Spatial and temporal variations of PM concentrations in Southeast Asia

In this section, the levels of PM in Southeast Asian countries will be reviewed considering seasons (haze/non-haze, wet/dry seasons), receptor sites' environment, and long-term trend, which are compared with other megacities.

2.3.1 Annual PM concentrations and primary emission

Long-term PM concentration measurements are rare in Southeast Asian countries, with only a few reports in Kuala Lumpur and Jerantut, Malaysia (Latif *et al.*, 2014; Rahman *et al.*, 2011), Brunei (Dotse *et al.*, 2016), Jakarta, Indonesia (Santoso *et al.*, 2020), Bangkok and Chiang Mai, Thailand (Pengchai *et al.*, 2009; Wimolwattanapun *et al.*, 2011), Hanoi, Vietnam (Cohen *et al.*, 2010a), and mixed 6 Asian cities (Kim Oanh *et al.*, 2006). Data from these studies are summarized

in Table 2-1, which also includes selected results from China and India (Hama et al., 2020; X. Hou et al., 2019). Table 2-1 shows that the daily average PM concentrations in Southeast Asian countries are significantly lower than some of those more polluted megacities in Asia such as China and India, even during the dry season. Due to lack of data, there is no clear trend in the ratio of $PM_{2.5}/PM_{10}$ in Southeast Asia cities but data from Wimolwattanapun *et al.* (2011) and Santoso *et al.* (2020) showed the ratio in Bangkok and Jakarta were much lower than 0.5. This is different to that in Beijing or London where the ratios are much higher.

Data from The World Bank (Fig. 2-1(a)) shows that the annual exposure to $PM_{2.5}$ concentrations in Southeast Asian countries are significantly lower than China and India. But $PM_{2.5}$ levels in all Southeast Asian countries exceeded annual mean standard by US-EPA ($12 \mu g m^{-3}$) and WHO ($5 \mu g m^{-3}$). $PM_{2.5}$ in the maritime continental Southeast Asian countries (Indonesia, Brunei, Malaysia, Philippines, and Singapore) are lower (ranged from 5.9 to $23.6 \mu g m^{-3}$) than those in the mainland continental Southeast Asia (Cambodia, Myanmar, Thailand, Vietnam, and Laos) (ranged from 25.2 to $44.1 \mu g m^{-3}$) (Fig. 2-1(a)). Exposure to $PM_{2.5}$ concentrations showed a clearly decreasing trend in all Southeast Asian countries except in Singapore and Indonesia. However, obtained data from Emission Database for Global Atmospheric Research (EDGAR) (Fig. 2-1(b)) shows that the emission inventory of $PM_{2.5}$ over Southeast Asian countries has been increasing during year 2000 - 2015 in most countries. This may imply that the decreasing exposure does not reflect the same trend for the PM emitted in a country

Table 2-1 Long-term PM concentrations in Southeast Asian countries, China, and India

	Duration	PM size	Daily average PM concentration ± SD (µg m ⁻³) (range: min – max)		PM _{2.5} /PM ₁₀ ratio	Site	References
Malaysia							
Kuala Lumpur	2004 - 2008	PM _{2.5} /PM _{2.5-10}	26.9 ± 9.9 / 22.5 ± 8.2		-	Urban	(Rahman <i>et al.</i> , 2011)
Batu Embun, Jerantut	1997 – 2011	PM ₁₀	37.9 ± 16.8 (9.4 – 276.5)		-	Background	(Latif <i>et al.</i> , 2014)
Brunei , 4 sites							
Belait	2009-2014	PM ₁₀	22.1 ± 11.9		-	-	(Dotse <i>et al.</i> , 2016)
Tutong			18.9 ± 10.8		-		
Temburong			17.0 ± 10.1		-		
Brunei-Muara			11.2 ± 7.6		-		
Indonesia							
Jakarta	2012	PM _{2.5} /PM ₁₀	19.4 / 50.9		0.38	Urban	(Santoso <i>et al.</i> , 2020)
	2013		17.1 / 38.9		0.44		
	2014		17.5 / 46.3		0.38		
	2015		15.5 / 51.2		0.30		
	2016		19.2 / 48.9		0.39		
	2017		19.3 / 50.2		0.38		
	2019		20.7 / 47.7		0.43		
Thailand							
Bangkok	2003	PM _{2.5} /PM ₁₀	19.1 / 52.6		0.46	Urban	(Wimolwattanapun <i>et al.</i> , 2011)
	2004		26.6 / 78.8		0.34		
	2005		23.3 / 54.2		0.43		
	2006		24.3 / 56.1		0.43		
	2007	PM ₁₀	23.2 / 54.5		0.43	Urban	(Pengchai <i>et al.</i> , 2009)
Chiang Mai, 3 sites	Jun 2005 – Jun 2006		52.9 ± 27.3		-		
			45.3 ± 23.1		-		
		54.5 ± 29.2		-			
Vietnam							
Hanoi	Apr 2001 – Dec 2008 (Every Sunday & Wednesday)	PM _{2.5}	54 ± 33		-	Urban	(Cohen et al., 2010a)
6 Asian cities							
Bandung, Indonesia	2001 – 2004	PM _{2.5} /PM ₁₀	Dry	Wet	Dry/Wet	-	(Kim Oanh <i>et al.</i> , 2006)
Bangkok, Thailand			53 / 83	38 / 62	0.64/0.61		
Beijing, China			50 / 76	18 / 33	0.66/0.54		
Chennai, India			168 / 262	104 / 180	0.64/0.58		
Manila, Philippines			46 / 169	42 / 145	0.27/0.29		
Hanoi, Vietnam			44 / 54	43 / 55	0.80/0.78		
			124 / 186	33 / 79	0.67 / 0.42		
India							
Delhi	2014	PM _{2.5} /PM ₁₀	140 ± 87 / 262 ± 139		0.53	Residential	(Hama <i>et al.</i> , 2020)
R K Puram	2015		125 ± 81 / 246 ± 131		0.51		
	2016		136 ± 99 / 271 ± 154		0.50		
	2017		131 ± 91 / 245 ± 238		0.53		
China							
Beijing-Tianjin-Hebei area	2014	PM _{2.5}	94.0		-	-	(X. Hou et al., 2019)
	2015		73.5		-		
	2016		71.2		-		
	2017		70.1		-		
	2018		56.1		-		

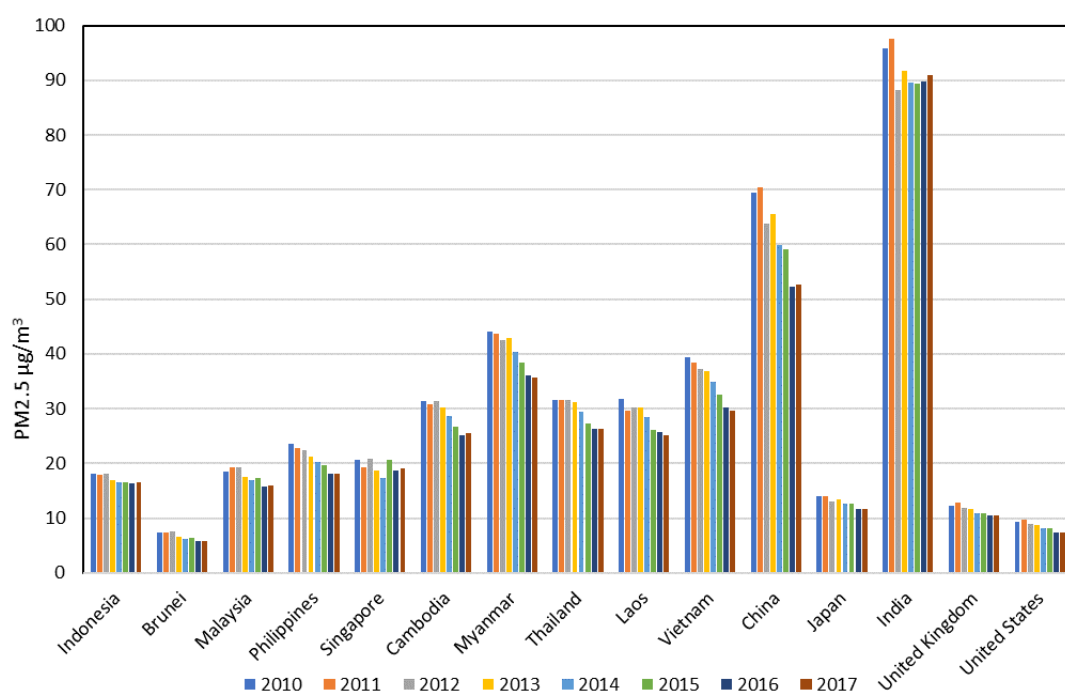


Fig. 2-1 (a) Mean annual exposure of PM_{2.5} (µg m⁻³) in Southeast Asia and selected other counties (The World Bank) during 2010 – 2017

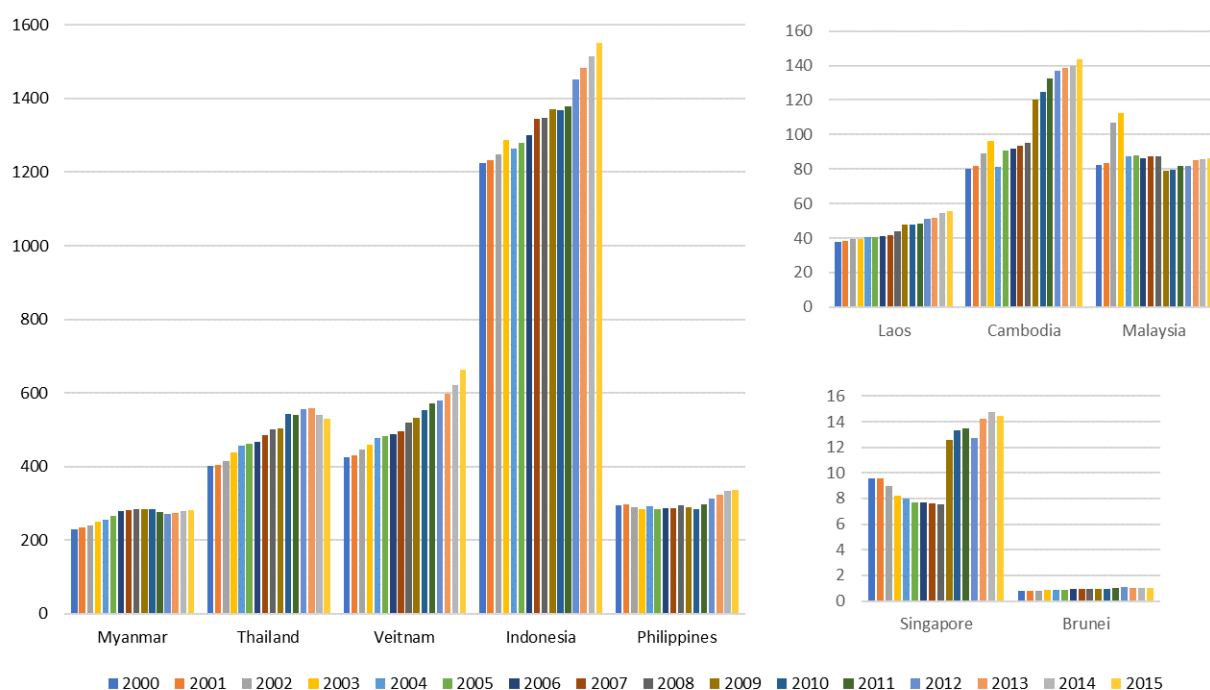


Fig. 2-1 (b) PM_{2.5} primary emission inventory of Southeast Asian Countries (Kt/year) (EDGAR) during 2000 – 2015

2.3.2 PM variations during local wet and dry seasons

PM levels over Southeast Asian countries obtained by a number of previous field studies are compiled in Table 2-2. PM levels relate to smoke-haze episodes in Southeast Asia has been distinctly investigated in 2 different durations:

- i. Dry season over mainland continental Southeast Asia during December to April and
- ii. Dry season over maritime continental Southeast Asia during mid-year between June to September

PM pollution levels are significantly higher during dry seasons across the Southeast Asian countries. In Kuala Lumpur, Malaysia, Jaafar *et al.* (2018) reported $PM_{2.5}$ level at $79 \mu g m^{-3}$ in June 2013 (dry season), while Khan *et al.* (2017) reported lower level at $23 \mu g m^{-3}$ during November 2013 – January 2014 (wet season). Similarly, Tham *et al.* (2019) reported in a 3 years study (2012 – 2015) that $PM_{2.5}$ concentrations in Singapore during dry season ($60.0 \mu g m^{-3}$) were approximately 2 times higher than wet season ($25.2 \mu g m^{-3}$). Very high $PM_{2.5}$ levels, up to $199 \mu g m^{-3}$ were also reported in Singapore. Similar results were also reported for PM_1 and PM_{10} concentrations in 2015 – 2016 (Hapsari Budisulistiorini *et al.*, 2018; Urbančok *et al.*, 2017). PM concentrations over the mainland continental Southeast Asia have been mainly investigated in the Northern part of Southeast Asia in Thailand (Chiang Mai) and Vietnam (Hanoi). Smoke-haze episodes in this region generally occur between December and April with much higher PM levels compared to wet or non-haze season. Tsai *et al.* (2013) categorized PM_{10} concentration above $120 \mu g m^{-3}$ as a threshold for episodic period of smoke-haze in Chiang Mai, Thailand, and reported PM_{10} concentrations during the haze episodic ($139.6 \mu g m^{-3}$) during Feb - Apr in year 2010 which were approximately 2 times higher than non-haze period ($58.1 \mu g m^{-3}$). Similar trend was also found at a rural area in Chiang Mai, where $PM_{2.5}$ level of 69.7 and $25.0 \mu g m^{-3}$ were reported in 2013 during haze and non-haze episodes, respectively (Chuang *et al.*, 2016).

PM_{2.5} levels in north Vietnam were also reported to be more than 2 times higher in dry than wet season. In Hanoi, PM_{2.5} was 51.5 and 18.9 $\mu\text{g m}^{-3}$ in 1999, and 136.9 and 58.1 $\mu\text{g m}^{-3}$ in 2015, during the dry and wet seasons, respectively (Hien *et al.*, 2002; Thuy *et al.*, 2018). Other studies at the rural sites of north Vietnam displayed a similar feature - PM_{2.5} concentrations in dry season were approximately 2 times higher than wet season (Co *et al.*, 2014; Hang & Kim Oanh, 2014).

Comparing to the Northern Southeast Asia, PM concentrations in dry and wet seasons in megacities over the mainland continental Southeast Asia at lower latitude such as Bangkok (Thailand) and Ho Chi Minh city (Vietnam) were reported with lower ranges (wet – dry) (Bangkok: 25 - 57 $\mu\text{g m}^{-3}$, Ho Chi Minh: 27.7 – 38.2 $\mu\text{g m}^{-3}$) compared to Chiang Mai (25 – 142.5 $\mu\text{g m}^{-3}$) and Hanoi (18.9 – 136.9 $\mu\text{g m}^{-3}$). This may be attributed to a stronger influence of biomass burning to PM concentrations during dry season across the Northern Southeast Asia region. Table 2-2 also clearly showed that the PM concentration could reach dangerously high levels during extreme pollution events. For example, the PM_{2.5} reached 191 $\mu\text{g m}^{-3}$ at a rural site in Kampar of Malaysia, 140 $\mu\text{g m}^{-3}$ at an urban site in Chiang Mai, Thailand (Tsai *et al.*, 2013), and hundreds to thousands $\mu\text{g m}^{-3}$ near sources (Betha *et al.*, 2013; Fujii *et al.*, 2014, 2019).

In summary, PM concentrations in Southeast Asia in the dry seasons are usually more than 2 times higher than those of wet seasons.

2.3.3 PM variations in different receptor environments

PM observations at different receptor sites, such as agricultural, rural, urban, suburban, traffic areas, or near sources offer useful information about the factors affecting PM levels in the air. There were few studies measured PM levels near BB sources during burning periods such as in Kalimantan and Sumatra (Indonesia). The sampling sites were located around 10 to 20 meters

from the peatland fire area, and very high PM concentrations were found ranging from hundreds to thousands $\mu\text{g m}^{-3}$ (Table 2-2) (Betha et al., 2013; Fujii et al., 2014, 2019). Another recent study conducted in Chiang Mai, Thailand, has compared PM level between rural site where locates near open burning source and urban site during different seasons. It was found that $\text{PM}_{2.5}$ during dry season was significantly higher at near source site than urban site (Chansuebsri *et al.*, 2022).

PM levels at urban sites are usually reported to be higher than suburban and rural sites. This is attributed to additional PM sources from vehicular and residential emissions at the urban sites. $\text{PM}_{2.5}$ at an urban site in Kuala Lumpur during haze episodes ($72.3 \mu\text{g m}^{-3}$) (Sulong *et al.*, 2017) was higher than at a rural site in Kampar ($55.9 \mu\text{g m}^{-3}$) (Ahmed *et al.*, 2016), while PM_{10} at the urban site was only slightly higher than at the semi-urban site (115 versus $109 \mu\text{g m}^{-3}$ (haze) and 47 versus $44 \mu\text{g m}^{-3}$ (non-haze)) (Samsuddin *et al.*, 2018). Industrial and traffic activities can also contribute to PM increment. For instance, as shown in Table 2-2, Tsai *et al.* (2013) reported that PM_{10} level in Chiang Mai at an industrial site was higher than an urban site; and Santoso *et al.* (2011) reported that both $\text{PM}_{2.5}$ and PM_{10} concentrations in Serpong, Indonesia, at an industrial site were higher than a residential site. Similarly, PM concentrations from 2002 to 2003 in Bangkok at busy traffic sites (Table 2-2) (Chuersuwan *et al.*, 2008) are higher than those at urban sites (Table 2-1) (Wimolwattanapun *et al.*, 2011). However, it should be noted that, this feature is not always true, especially in the areas where biomass burning dominates local sources during smoke-haze periods. For example, in Chiang Mai, year 2015, there was no significant difference between $\text{PM}_{2.5}$ level measured at rural ($118 \mu\text{g m}^{-3}$) and urban site ($113 \mu\text{g m}^{-3}$) (Pani et al., 2019a), and year 2019, in which $\text{PM}_{2.5}$ at rural site (near source) ($115 \mu\text{g m}^{-3}$) was higher than urban site ($76 \mu\text{g m}^{-3}$) (Chansuebsri *et al.*, 2022).

Overall, existing data revealed that the daily average concentrations of PM over Southeast Asian countries are usually lower than those in polluted megacities such as Beijing and Delhi. In contrast to the decreasing $\text{PM}_{2.5}$ exposure over Southeast Asia region, primary emission inventory

of PM_{2.5} has been increasing in most Southeast Asian countries. Furthermore, the variation of PM concentrations is subject to meteorological factors during different seasons (i.e., wind speed, relative humidity and rainfall) and receptor's environments including local sources and long-range transports.

Table 2-2 Reported PM daily average concentrations during haze and non-haze periods in Southeast Asian countries

	Duration	PM size	Daily average PM concentration $\pm$ SD ($\mu\text{g m}^{-3}$) (range: min – max)	Site	References
Malaysia					
Kuala Lumpur	21 – 27 Jun 2013	PM _{2.5}	79.4 (14.5 – 160.9)	Suburban	(Jaafar <i>et al.</i> , 2018)
	12 Nov 2013 – 15 Jan 2014	PM _{2.5}	23 $\pm$ 9 (16 – 55)	Urban	(Khan <i>et al.</i> , 2017)
	2015	PM ₁₀	Haze: 109 $\pm$ 49.5 (14 – 245) Non-haze: 44.3 $\pm$ 18.8 (12.4 – 115)	Semi urban	(Samsuddin <i>et al.</i> , 2018)
	2015	PM ₁₀	Haze: 115 $\pm$ 57.7 (32.4 – 289) Non-haze: 46.6 $\pm$ 13.7 (21.8 – 109)	Urban	(Samsuddin <i>et al.</i> , 2018)
Kampar Singapore	Jun 2015 – Jan 2016	PM _{2.5}	Pre-haze: 24.5 $\pm$ 12.0 (Jun – Aug 2015) Haze: 72.3 $\pm$ 38.0 (Sep – Oct 2015) Post-haze: 14.3 $\pm$ 3.6 (Nov 2015 – Jan 2016)	Urban background	(Sulong <i>et al.</i> , 2017)
	Aug – Oct 2015	PM _{2.5}	55.9 (9.9 – 191.1)	Rural	(Ahmed <i>et al.</i> , 2016)
	May 2012 – Jun 2013 and Jun 2015 – Dec 2015	PM _{2.5}	Non-smoke: 25.2 $\pm$ 9.3 (11.5 – 60.4) Smoke-dominant: 60.6 $\pm$ 40.8 (22.4 – 194.6) Episodic smoke: 199.0 $\pm$ 88.3 (87.8 – 317.0)	Urban	(Tham <i>et al.</i> , 2019)
	10 – 21 Oct 2015	PM ₁	Haze: 53.6 $\pm$ 26.9 (10–28 Oct) Non-haze: 26.0 $\pm$ 0.4 (29–31 Oct) All: 49.8 $\pm$ 27.7	Urban	(Hapsari Budisulistiorini <i>et al.</i> , 2018)
	May 2015 – May 2016	PM ₁₀	Haze: 116 (71 – 323) Non-haze: 46.5 (32 – 70)	Urban	(Urbančok <i>et al.</i> , 2017)
Indonesia					
Kalimantan	19 Sep – 12 Oct 2009	PM _{2.5}	235 – 7817 (6 – 8 h)	Near source	(Betha <i>et al.</i> , 2013)
Riau, Sumatra	11 – 16 Sep 2015	PM _{2.5}	810 $\pm$ 530 (3 – 4 h) 54 – 87 (12h)	Near source Background	(Fujii <i>et al.</i> , 2019)
	16 – 17 May 13 – 17 June 2012	PM _{2.5}	Near source: 7120 $\pm$ 3620 Background: 23.9 $\pm$ 2.53	Near source Background	(Fujii <i>et al.</i> , 2014)
Serpong Setu	Aug – Nov 2008	PM _{2.5} / PM ₁₀	23 (15 – 34) / 72.3 (29 – 77)	Industrial	(Santoso <i>et al.</i> , 2011)
	Aug – Nov 2008	PM _{2.5} / PM ₁₀	21 (13 – 27) / 47.7 (18 – 41)	Residential	(Santoso <i>et al.</i> , 2011)
Thailand					
Chiang Mai	2 Feb – 2 Apr 2010	PM ₁₀	Non-episodic ¹ : 58.1 $\pm$ 30.9 Episodic ² : 139.6 $\pm$ 14.0	Urban	(Tsai <i>et al.</i> , 2013)
	2 Feb – 2 Apr 2010	PM ₁₀	Non-episodic ¹ : 77.4 $\pm$ 25.1 Episodic ² : 142.5 $\pm$ 17.7	Industrial	(Tsai <i>et al.</i> , 2013)
	Feb – Apr 2013	PM _{2.5}	BB ³ : 69.7 $\pm$ 25.1 Non-BB: 25.0 $\pm$ 10.9	Rural	(Chuang <i>et al.</i> , 2016)
	1 – 31 Mar 2015	PM _{2.5}	Haze: 118 $\pm$ 36	Rural	(Pani <i>et al.</i> , 2019a)
	1 – 17 Mar 2015	PM _{2.5}	Haze: 113 $\pm$ 45	Urban	(Pani <i>et al.</i> , 2019a)
	2019	PM _{2.5}	Hot dry season: 76.8 $\pm$ 40.9 Wet season: 20.0 $\pm$ 7.6 Cold dry season: 35.2 $\pm$ 7.5	Urban	(Chansuebsri <i>et al.</i> , 2022)
	2019	PM _{2.5}	Hot dry season: 115.4 $\pm$ 49.5 Wet season: 25.0 $\pm$ 7.1 Cold dry season: 39.1 $\pm$ 8.5	Rural (near source)	(Chansuebsri <i>et al.</i> , 2022)

¹ PM₁₀ < 120 $\mu\text{g m}^{-3}$, ² PM₁₀ $\geq$ 120 $\mu\text{g m}^{-3}$, ³ identified by high K⁺, levoglucosan and OC

Table 2-2 (continue)

	Duration	PM size	Daily average PM concentration $\pm$ SD ($\mu\text{g m}^{-3}$) (range: min – max)	Site	References
Thailand					
Bangkok, 4 sites					
Din Daeng	Feb 2002 – Jan 2003	PM _{2.5} / PM ₁₀	69.0 $\pm$ 28.8 / 108.1 $\pm$ 35.5	Traffic	(Chuersuwan <i>et al.</i> , 2008)
Jan Krasem			40.9 $\pm$ 21.4 / 61.1 $\pm$ 25.2		
Bann Somdej			41.5 $\pm$ 24.6 / 62.1 $\pm$ 30.7		
Bang Na			37.9 $\pm$ 18.9 / 57.6 $\pm$ 23.9		
Bangkok	Jun-Jul 2010 (Wet)	PM _{2.5}	Wet: 25 $\pm$ 6 (16 – 36)	Traffic	(Kim Oanh <i>et al.</i> , 2013)
	Dec 2010 (Dry)		Dry: 57 $\pm$ 13 (34 – 76)		
Vietnam					
Hanoi	Oct 1998 – Mar 1999	PM _{2.5}	Dry: 51.5 $\pm$ 28.5	Urban	(Hien <i>et al.</i> , 2002)
	May 1999 – Jul 1999		Wet: 18.9 $\pm$ 8.0		
			All: 36.1 $\pm$ 1.3		
		PM ₁₀	Dry: 69.8 $\pm$ 52.3		
			Wet: 27.6 $\pm$ 15.0		
			All: 87.1 $\pm$ 3.1		
	Aug 2015 (Wet)	PM _{2.5} / PM ₁₀ /	58.1 / 86.6 / 6.1	Traffic	(Thuy <i>et al.</i> , 2018)
	Oct – Dec 2015 (Dry)	PM ₁	136.9 / 104.7 / 11.9		
	Aug 2015 (Wet)	PM _{2.5} /P	46.8 / 73.9 / 5.97	Mixed	(Thuy <i>et al.</i> , 2018)
	Oct – Dec 2015 (Dry)	PM ₁₀ /PM ₁	70.4 / 104.7 / 5.4		
Ho Chi Minh city	Dec 2007 – Jan 2008	PM _{2.5}	Weekdays: 134 $\pm$ 31	Traffic	(Huong Giang & Kim Oanh, 2014)
	(Dry)		Weekends: 114 $\pm$ 30		
	Mar 2017 – Mar 2018	PM _{2.5}	Dry: 38.2 $\pm$ 14.3 (15.1–89.2)	Road sites	(Hien <i>et al.</i> , 2019)
	Dec - Apr (Dry)		Wet: 40.0 $\pm$ 11.7 (16.4–70.35)		
	May – Nov (Wet)		All: 39.1 $\pm$ 14.9 (15.1–89.2)		
			Dry: 35.8 $\pm$ 10.1 (15.8–58.5)	Residential sites	
			Wet: 27.7 $\pm$ 12.3 (10.4–65.8)		
			All: 31.4 $\pm$ 12.0 (10.4–65.8)		
			Dry: 37.7 $\pm$ 12.4 (16.8–72.0)	Urban background	
			Wet: 33.2 $\pm$ 13.1 (13.3–64.2)		
			All: 35.4 $\pm$ 12.9 (13.3–72.0)		
Sonla	2013	PM _{2.5}	47 $\pm$ 30 (BB from South China)	Rural	(Nguyen <i>et al.</i> , 2016)
			98 $\pm$ 18 (BB from within Southeast Asia)		
			20 $\pm$ 14 (BB from South China Sea)		
Mong Duong	Jul-Aug 2009 (Wet)	PM _{2.5}	Wet: 35 $\pm$ 16	Urban	(Hang & Kim Oanh, 2014)
	Dec 2009-Jan 2010 (Dry)		Dry: 60 $\pm$ 23		
Cam Hai	Jul-Aug 2009 (Wet)	PM _{2.5}	Wet: 27 $\pm$ 12	Rural	(Hang & Kim Oanh, 2014)
	Dec 2009-Jan 2010 (Dry)		Dry: 53 $\pm$ 17		
Tamdao	20 Sep – 8 Oct 2005 (Wet)	PM _{2.5} / PM ₁₀	Wet: 25 $\pm$ 12 / 33 $\pm$ 11	Rural	(Co <i>et al.</i> , 2014)
	17 Dec 2005 – 18 Jan 2006 (Dry)		Dry: 51 $\pm$ 29 / 67 $\pm$ 32		

Dry – Local dry season reported in the literatures

Wet – Local wet season reported in the literatures

2.4 Chemical compositions in PM in Southeast Asian countries

Atmospheric PM is composed of a large number of chemical species depending on their sources and atmospheric chemical processes. A comprehensive understanding in PM chemical composition is important to evaluate PM toxicity (e.g., carcinogenic compounds including PAHs) (Jamhari et al., 2014c), source characteristics (Chantara *et al.*, 2019; Hao *et al.*, 2019), as well as to carry out source apportionment by receptor models.

Chemical characteristics of PM have been investigated in many countries, especially in megacities such as Delhi, Beijing, Los Angeles and London. For Southeast Asia region, chemical contributions of PM have mostly been examined during smoke – haze episodes or dry season. These studies were commonly conducted focusing on major component groups including carbonaceous compounds (organic carbon (OC) and elemental carbon (EC)), water soluble ions (WSIs), and trace metals. Few studies in Southeast Asia have measured specific organic compounds including commonly used biomass burning aerosol tracer such as levoglucosan.

In this section, chemical composition in PM obtained from a number of studies in Southeast Asian countries will be summarized as the three abovementioned main groups, which will be used to illustrate the general pattern across the region and compared with other megacities.

2.4.1 Carbonaceous species in PM

Carbonaceous species are a major component of atmospheric PM, especially during haze seasons where total carbon (TC) in PM_{2.5} increases by factors of 2 – 15 as reported during peat fires event in Indonesia (Tham *et al.*, 2019). The contribution of TC to PM mass is generally higher in smaller size of PM (Thuy *et al.*, 2018) and depends on the influence of different PM sources

(Chuersuwan *et al.*, 2008). OC contributes as high as 70% of the total mass of particles directly emitted from peat fires but with extremely low EC (1% to PM mass) (Jayarathne *et al.*, 2018). Carbonaceous species are also the dominant component in PM from gasoline-fueled vehicles with EC contributing more than 50% of all carbonaceous species (Lin *et al.*, 2020). While in diesel vehicles, carbonaceous species contribute 55.9 – 59.1% to PM_{2.5} (Hao *et al.*, 2019). The contributions of carbonaceous species were found to be relatively low in resuspended road dust, with OC and EC accounting for 12.5% and 2.1% of PM_{2.5} mass, respectively (Chen *et al.*, 2012), and during dust storm (Tao *et al.*, 2013).

OC and EC concentrations over Southeast Asia and other countries were collected from available publications and then calculated as percentage contribution to PM mass and were displayed as box plots (Fig. 2-2); with the full tables available in Table SA-1 and SA-2). In general, percentage contribution of OC to PM mass over Southeast Asian countries, based on annual average or seasons, are higher than those in Beijing and Delhi, as well as in a number of other cities (Fig. 2-2 (a)). Particularly, studies in the Northern Southeast Asia, such as in Chiang Mai and Hanoi, showed notably higher OC contributions (range 19.2 – 41.2 %) and higher OC/EC ratios (range 3.1 – 8.3) than in other regions (Fig. 2-2 (c)). Thepnuan *et al.* (2019) reported that OC accounts for 36.5 % of PM_{2.5} in Chiang Mai, Thailand, and Lee *et al.* (2016) showed OC contributed to 59 % and 58 % of PM_{2.5} mass during haze season 2012 and 2013, respectively, in Sonla, Vietnam (Table SA-1)

In maritime continental Southeast Asia, similar features of OC contribution in PM have also been observed with peatland fire as a specific biomass burning source. Fujii *et al.* (2014) determined OC and EC at background and near peatland fire sites during haze and non-haze seasons in Sumatra Island, Indonesia; their report stated that OC near peat fires contributes up to 71% of PM mass, which was much higher than that in PM at a background site (39.1%). Previous studies in PM chemical characteristics have used levoglucosan as an organic tracer to estimate the

contribution of biomass burning activities. For example Tsai *et al.* (2013) and Thepnuan *et al.* (2019a) indicated that levoglucosan is strongly correlated with OC and K⁺ during haze season in Chiang Mai, Thailand. EC in Southeast Asian countries is usually lower than 10% of PM mass, which is not significantly different to that in other regions. Additional data of mass reconstructions is shown in Fig. 2-3 from selected cities in Southeast Asia during haze season. Organic matter (OM) mass is estimated by multiply OC with factor 1.6 (Chow *et al.*, 2015). The pie charts show OM contributed more than 25% of total contribution and varied upon cities while EC contributed to narrower range between 6 and 8%. This implies a distinction in chemical characteristics of biomass sources over different cities.

OC/EC ratio has also been used by several studies in Southeast Asia to identify carbonaceous sources associated with atmospheric PM. It should be noted that the analysis instruments/methods of OC/EC were different upon studies (Bautista *et al.*, 2015). Uncertainty during OC and EC split point from different methods may result in inconsistent OC/EC ratios. However, we can still see general patterns from previous studies. OC/EC ratios in Southeast Asian countries especially in continental Southeast Asia were obviously higher than in other countries (Fig. 2-2 (c)). This may reflect the major contribution of biomass burning and/or secondary organic aerosols (SOA) to PM mass in this region (Engling *et al.*, 2014). Pani *et al.* (2019) reported that OC/EC ratios at a rural site was higher than at urban site during haze season in Chiang Mai, suggesting the additional contribution of EC from vehicular emissions and coal combustions to PM mass in urban areas. Popovicheva *et al.* (2017) explained that an extremely high OC/EC ratio (145) measured near peat fire source during smoldering phase was attributed to the condensation of semi-volatile organic compounds at low temperature in smoldering plume. Furthermore, ratio of char-EC (EC1 – pyrolytic organic carbon (POC)) to soot – EC (EC2 + EC3) in PM is also useful to identify the contribution of carbonaceous compound due to the char-EC is generated by biomass burning under low temperature combustion, while the soot-EC is formed under higher

temperature such as coal and vehicles combustions (Chuang *et al.*, 2013). Some studies conducted in Southeast Asia employed char-EC/soot-EC ratio to preliminary classify PM source such as (Phairuang *et al.*, 2020) and (Tham *et al.*, 2019).

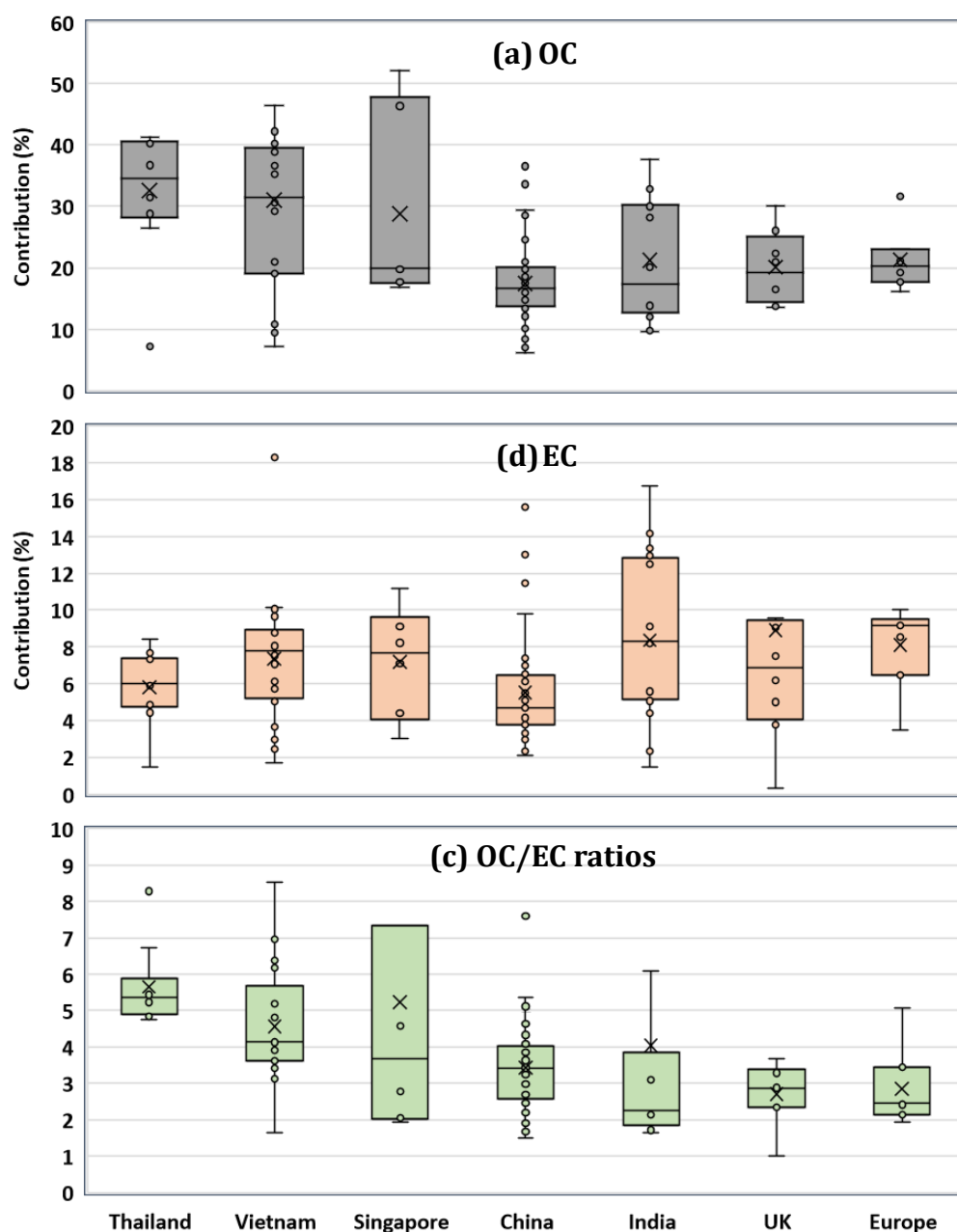


Fig. 2-2 Box plots of contribution (%) of (a) OC, (b) EC to $PM_{2.5}$ mass, and (c) OC/EC ratios in Southeast Asia and other countries ($N < 2$ data is not inclusive). (Upper bar, upper box, middle line, lower box, lower bar, circle dot and cross represent maximum, Q3, median, Q1, minimum, individual data and mean, respectively.)

References; **Thailand** (Chuang *et al.*, 2013; Li *et al.*, 2013; Pani *et al.*, 2018, 2019; Thepnuan *et al.*, 2019; Choochuay *et al.*, 2020), **Vietnam** (Cohen *et al.*, 2010a; Hai and Kim Oanh, 2013; Co *et al.*, 2014; Hang and Kim Oanh, 2014; Son *et al.*, 2016; Lee *et al.*, 2016; Thuy *et al.*, 2018), **Singapore** (Hapsari Budisulistiorini *et al.*, 2018; Tham *et al.*, 2019), **China** (Tao *et al.*, 2013, 2015; Ho *et al.*, 2016; Ji *et al.*, 2016; Li *et al.*, 2016; Niu *et al.*, 2016; Zhang *et al.*, 2016; Shen *et al.*, 2017; Tan *et al.*, 2017; Jiang *et al.*, 2018; Shao *et al.*, 2018; Xu *et al.*, 2018; Liang *et al.*, 2019; Zhao *et al.*, 2019; Lu *et al.*, 2020), **India** (Jain *et al.*, 2020; Kumar & Sunder Raman, 2020; Mandal *et al.*, 2014; Pant *et al.*, 2015; Pipal & Satsangi, 2015; Police, Sahu, *et al.*, 2018; Rengarajan *et al.*, 2011; Sharma & Mandal, 2017), **UK** (Crilley *et al.*, 2015; Pant *et al.*, 2017; Taiwo, 2016; J. Yin *et al.*, 2015), **Europe** (Bressi *et al.*, 2013; Salameh *et al.*, 2015)

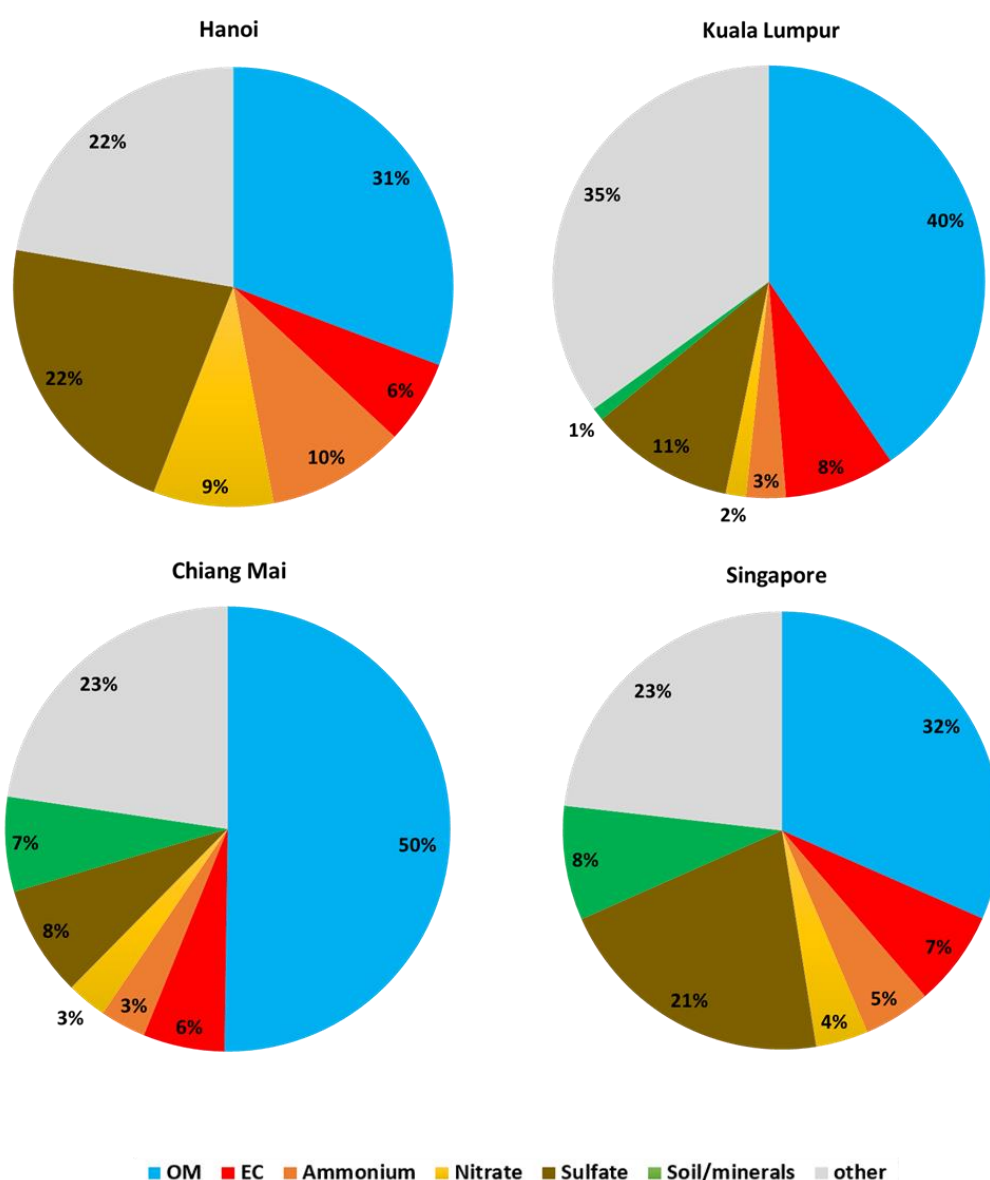


Fig. 2-3 Percentage contributions of chemical components to PM_{2.5} during haze season in selected cities: Hanoi (Hai & Kim Oanh, 2013), Kuala Lumpur (Khan, Sulong, et al., 2016), Chiang Mai (Pani et al., 2018), and Singapore (See et al., 2006). OM is estimated by 1.6OC, Soil is calculated by sum of 2.2Al, 2.49Si, 1.63Ca, 1.94Ti, and 2.42Fe (Chow et al., 2015).

2.4.2 Water-soluble inorganic ions (WSIs) in PM

Another major contributor in PM mass is Water-soluble inorganic ions (WSIs). They consist of several species including cations (Ca^{2+} , Na^+ , K^+ , Mg^{2+} , and NH_4^+)s, and anions (F^- , Cl^- , NO_3^- , PO_4^{3-} , and SO_4^{2-}). SO_4^{2-} , NO_3^- , and NH_4^+ (SNA – Sulfate, Nitrate and Ammonia, respectively) were given much attention due to their high contribution to PM mass (Lu *et al.*, 2020; Sulong *et al.*, 2017).

Fig. 2-4 shows box plots of the percentage contribution of WSIs to PM mass in selected countries (with full table in supplementary Table SA-3 and SA-4). In general, results from different Southeast Asian countries were similar. SNA ions are the most abundant species of WSIs in PM, mostly in a descending order: $\text{SO}_4^{2-} > \text{NH}_4^+ > \text{NO}_3^-$, while K^+ and Na^+ have a similar but smaller contribution to PM mass. Some studies in Southeast Asian countries show relatively low SNA contribution. For example, a study conducted in Singapore by Tham *et al.* (2019) found that the contribution of SO_4^{2-} in $\text{PM}_{2.5}$ during high haze episodes ($\text{PM}_{2.5}=199 \pm 88.3 \text{ ug/m}^3$) (2.9%) was significantly lower than those of non-haze (19.5%) and haze seasons (13.2%) when the $\text{PM}_{2.5}$ concentrations were lower ($25.2 \pm 9.3 \text{ ug/m}^3$; $60.6 \pm 40.8 \text{ ug/m}^3$ for non-haze and haze seasons, respectively). This may be attributed to the very high contribution of total carbon (combination of OC and EC) at 49% to $\text{PM}_{2.5}$ mass during haze episodes associated with peat fire, resulting in lower contribution from SO_4^{2-} . However, as shown in mass reconstruction in Fig. 2-3, SO_4^{2-} is the second highest contributor to $\text{PM}_{2.5}$ and the highest contributor of WSIs group during haze season. Significant contributions of K^+ to WSIs were found during haze event in Chiang Mai, Thailand (Table SA-3). Chantara *et al.* (2019) found that K^+ was the most abundant ions in $\text{PM}_{2.5}$ collected during combustion of various biomass type, including agricultural wastes and forest leaf litters, in a custom-made chamber, followed by Cl^- and SO_4^{2-} . Sillapapiromsuk *et al.* (2013) suggested that

K^+ and Cl^- can be used as potential tracers of agricultural waste burning where fertilizers and herbicides are used.

Fig. 2-4 shows that contributions of SNA in PM are significantly lower in Southeast Asian countries than those of China, India, and Europe regardless of whether in haze or non-haze seasons, especially for NO_3^- and NH_4^+ . For Southeast Asian countries, the contributions of NO_3^- and NH_4^+ in $PM_{2.5}$ are mostly below 6%. While in other countries, they are mostly much more than 6% (Fig. 2-4 (b) and 2-4(c), see also Table SA-3 and SA-4). In addition, NO_3^- to SO_4^{2-} ratios in Southeast Asia are also mostly lower than 0.5 while in other countries they are mostly above 0.5, particularly in more developed cities (Fig. 2-4 (d)), see also Table SA-3 and SA-4). These indicate the disparities of sources and chemical characteristics in PM over Southeast Asia and other regions. NO_3^-/SO_4^{2-} ratio has been used by number of previous studies as a potential indicator for mobile and stationary sources. High NO_3^-/SO_4^{2-} ratio indicates mobile sources dominate over stationary sources (Jiang *et al.*, 2018), while low NO_3^-/SO_4^{2-} , together with other indicator ratios such as high OC/EC, and $nss-K^+/EC$ indicate biomass burning influences (Pani *et al.*, 2018). Another potential reason is that the difference in meteorological condition, for example, temperature and relative humidity. High relative humidity favors sulfate formation (Liu *et al.*, 2019; Wang *et al.*, 2019; Han *et al.*, 2021). Southeast Asia region locates in tropical area surrounding by maritime territories where relative humidity is usually high throughout the year. And higher temperature in Southeast Asian countries does not favor the partition of nitric acid to form nitrate aerosol.

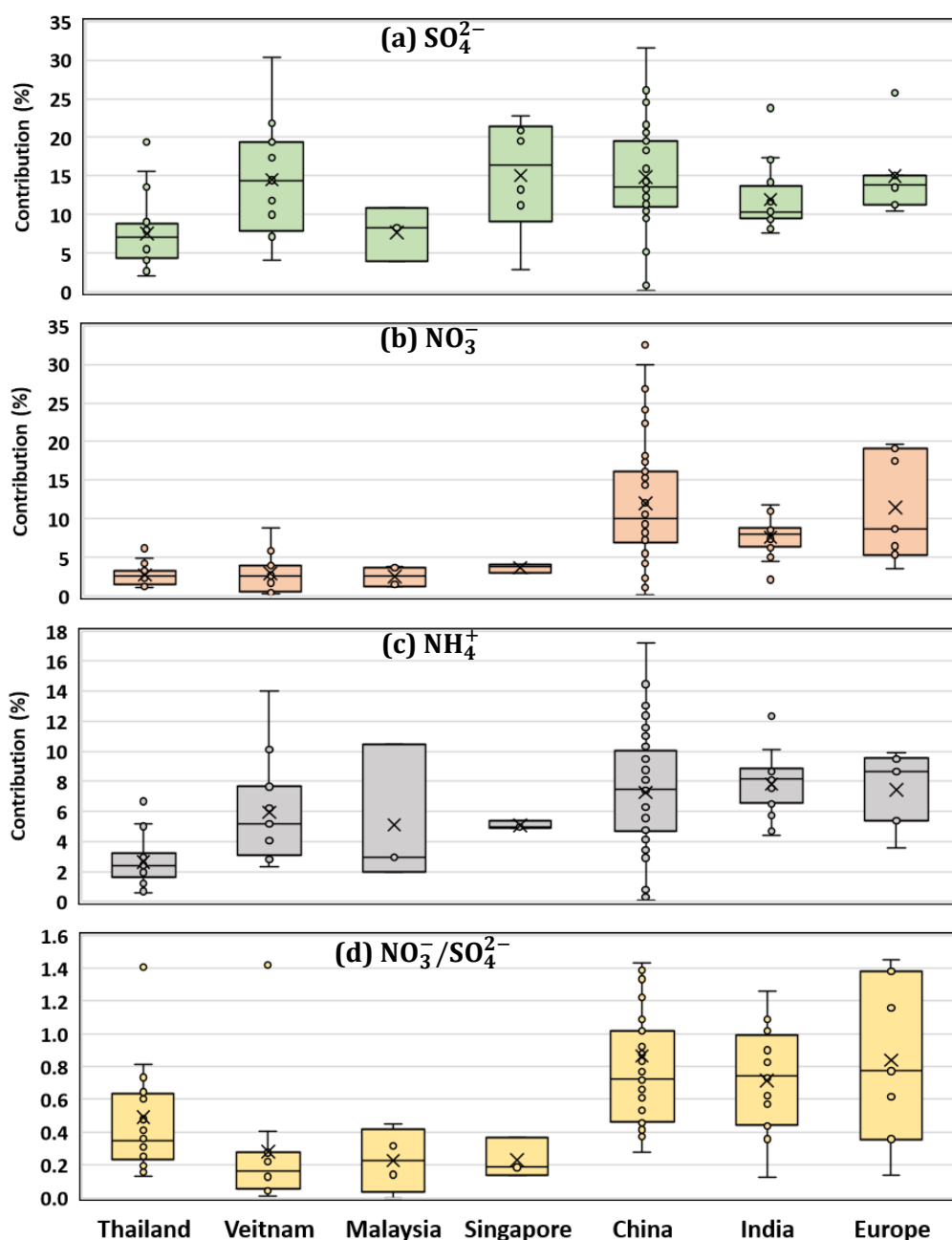


Fig. 2-4 Box plots of contribution (%) of (a) SO_4^{2-} , (b) NO_3^- , (c) NH_4^+ to PM_{2.5} mass, and (d) NO_3^-/SO_4^{2-} ratios in Southeast Asia and other countries (N < 2 data is not inclusive). (Upper bar, upper box, middle line, lower box, lower bar, circle dot and cross represent maximum, Q3, median, Q1, minimum, individual data and mean, respectively.)

References; **Thailand** (Chansuebsri et al., 2022; Choochuay et al., 2020; Chuang et al., 2013; Chuersuan et al., 2008; Khamkaew, Chantara, Janta, et al., 2016b; C. Li et al., 2013; Pani et al., 2018, 2019b; Thepnuan et al., 2019a), **Vietnam** (Hai and Kim Oanh, 2013; Co et al., 2014; Hang and Kim Oanh, 2014; Lee et al., 2016), **Malaysia** (Khan et al., 2016, 2017; Jaafar et al., 2018; Farren et al., 2019), **Singapore** (Hapsari Budisulistiorini et al., 2018; See et al., 2006; Tham et al., 2019), **China** (Tao et al., 2013, 2015; Ho et al., 2016; Li et al., 2016; Niu et al., 2016; He et al., 2017; Shen et al., 2017; Tan et al., 2017; Jiang et al., 2018; Shao et al., 2018; Xu et al., 2018; Rui Zhang et al., 2018; Liang et al., 2019; Zhao et al., 2019; Dong et al., 2020; Lu et al., 2020), **India** (Jain et al., 2020; Kumar & Sunder Raman, 2020; Pant et al., 2015; Rengarajan et al., 2011; Saxena et al., 2017; Sharma & Mandal, 2017), **Europe** (Bressi et al., 2013; Salameh et al., 2015)

2.4.3 Crustal and trace elements in PM

Trace elements contribute little to PM mass, but they are good tracers for apportioning anthropogenic PM sources (Amil *et al.*, 2016). Crustal elements such as Si, Al and Ti are often used as tracers for mineral dust. Species and mass concentrations of trace elements differ among different environments (Liang *et al.*, 2016). Several studies have measured crustal and trace elements over different receptor sites such as industrial (Amil *et al.*, 2016; X. Liang *et al.*, 2019; S. Zhou *et al.*, 2014), traffic (Chuersuwan *et al.*, 2008; Pant *et al.*, 2017), urban (Pani *et al.*, 2018; Rahman *et al.*, 2011), rural (Co *et al.*, 2014), as well as over different seasons and smoke-haze episode (Fujii *et al.*, 2016; Khamkaew, Chantara, Janta, *et al.*, 2016b; See *et al.*, 2006). Oxides of crustal elements (Si, Al, Ti, and Fe) and sea salts (Na, K, and Ca) mostly contribute to mass fraction of coarse PM (PM_{10-2.5}), while heavy metals (e.g., Pb, V, Cd, Cu, Ni, and Zn) are mainly present in fine PM (PM_{2.5}).

In Southeast Asia, few studies measured both trace metals and other major chemical components (such as OC, EC and ions) in PM. Most studies on trace elements in PM focused on smoke-haze episodes. Since there are several trace metal species which cannot be all included, Table 2-3 only presents the major crustal elements including Al, Si, Ca, K, Ti, and Fe, as well as other key elements such as Na, Mg, and Zn (see Table SA-5 and SA-6 in supplement for more data).

Generally speaking, the concentration of crustal elements in Southeast Asia region is much higher than that of the trace elements. This is similar to what was observed in other countries such as China and India (Table 2-3). Higher contribution of crustal elements was found in coarse size PM (PM₁₀) than fine size (PM_{2.5}) regardless of where the samples were collected (e.g., rural, urban or roadside) (Rahman *et al.*, 2011) (see also Table SA-5). In Southeast Asia, Si is the highest contributor among crustal element, similarly, other countries like China and India. PM mass reconstruction in Fig. 2-3 shows the contribution of mineral/soil to PM mass in Southeast Asia is

usually very low but varied across different counties. However, K has distinctly higher contributions in Southeast Asia than other countries, especially in Chiang Mai, Thailand (Khamkaew, Chantara, Janta, et al., 2016b; Pani et al., 2018) (Table 2-3). This is due to the high contribution of biomass burning (K is usually calculated as a fraction of crustal mass by surrogate Fe: $K = 0.6Fe$) (Huang *et al.*, 2014; Chow *et al.*, 2015). Khamkaew *et al.* (2016) found a good correlation between K and $PM_{2.5}$ concentrations and levoglucosan during haze season in Chiang Mai, Thailand, which further supports the fact that the high contributions of K are due to biomass burning.

2.4.4 Summary: chemical compositions and mass closure

General picture of chemical compositions of PM in Southeast Asia can be summarized as follows.

i) contribution of OC to PM mass in this region is obviously higher than those in other megacities such as Beijing and Delhi, especially over continental Southeast Asia region such as Chiang Mai, Thailand, and Hanoi, Vietnam. This implies the influence of biomass burning over this region.

ii) SO_4^{2-} , NO_3^- , and NH_4^+ are the major contributor of ions to PM mass across countries in Southeast Asia. SO_4^{2-} is the second highest contributor especially during haze season. K^+ is also an important tracer ion related to biomass burning which is usually found with high contribution. Lower NO_3^- to SO_4^{2-} ratios in Southeast Asia comparing to those of China and India indicate that stationary sources (such as biomass burning) dominates mobile sources (such as vehicle emissions) and due to high temperature and humidity in this region favor sulfate formation.

iii) Crustal and trace elements contribute little to PM mass in Southeast Asia. Their contributions differ upon different environments such as industry, traffic, or urban. Crustal

elements (Al, Si, Ca, K, Ti, and Fe) are the major contributors in this group and are commonly found to be similar contributions in other countries such as China and India.

In addition to contribution of chemical compositions, mass closure of each component over representative Southeast Asian countries has also shown agreement to the above-mentioned summaries. Comparing to a former study conducted in London, Beijing, and Delhi (Harrison, 2020), correspondence of contribution of OM > ions > soil/minerals is found in Chiang Mai, Kuala Lumpur, Singapore, Beijing and Delhi, while ions > OM > soil/minerals is found in Hanoi and London. This may be attributed to lower influence of biomass burning and higher vehicular emission sources in Hanoi and London. However, it should be noted that data availability in mass closure in the previous reviewed studies over Southeast Asia is limited, only few studies have done completed set of bulk chemicals analysis.

Table 2-3 Percentage contribution of crustal and trace elements (%) in PM_{2.5} mass concentration in Southeast Asia and other countries

Country	Season	Site	Al	Si	Ca	K	Ti	Fe	Na	Mg	Cu	Zn	References
Thailand													
Bangkok	Dry	Traffic	2.77	-	4.32	1.42	-	2.07	2.12	0.68	0.12	1.13	(Chuersuwan <i>et al.</i> , 2008)
Chiang Mai	Haze	Urban	1.83	-	-	3.55	-	-	-	1.08	-	0.43	(Pani <i>et al.</i> , 2018)
	Haze	Urban	1.22	-	-	3.27	-	-	-	1.54	-	0.06	
	Haze	Urban	1.70	-	-	3.14	-	-	-	1.21	-	0.45	
Chiang Mai	Haze	Urban	1.87	-	-	3.54	-	-	-	1.13	0.02	0.49	(Khamkaew, Chantara, Janta, et al., 2016b)
	Haze	Rural	2.00	-	-	3.22	-	-	-	1.02	0.01	0.24	
Vietnam													
Hanoi	Annual	Suburban	0.41	2.00	0.85	1.80	0.06	0.73	0.23	-	0.02	0.90	(Cohen et al., 2010a)
Mong Duong	Wet	Industrial	0.14	0.57	1.14	-	0.03	0.86	-	0.17	0.09	0.29	(Hang & Kim Oanh, 2014)
	Dry	Industrial	0.33	1.33	0.83	-	0.03	0.67	-	0.17	0.03	0.33	
Cam Hai	Wet	Rural	0.15	0.74	0.37	-	0.07	0.74	-	0.01	0.01	0.37	
	Dry	Rural	0.19	0.57	0.75	-	0.04	0.38	-	0.00	0.04	0.38	
Tamdao, Veit	Wet	Rural	1.60	4.80	1.20	-	0.40	1.20	-	-	0.01	0.40	(Co <i>et al.</i> , 2014)
	Dry	Rural	1.96	4.90	1.18	-	0.01	0.39	-	0.20	0.02	0.10	
Malaysia													
Kuala Lumpur	Haze	Suburban	0.07	-	-	0.66	-	0.03	-	0.05	0.01	0.09	(Khan <i>et al.</i> , 2016)
Kuala Lumpur	Wet	Urban	0.61	-	0.35	0.43	-	-	1.43	0.39	0.04	0.05	(Khan <i>et al.</i> , 2017)
Kuala Lumpur	Annual	Urban	0.56	1.19	0.48	1.45	0.03	0.41	1.04	0.45	0.06	0.15	(Rahman <i>et al.</i> , 2011)
Selangor	Haze	Urban	0.40	0.95	0.25	1.97	0.03	0.23	-	-	-	0.13	(Fujii <i>et al.</i> , 2016)
Singapore													
Singapore	Haze	Urban	2.68	0.30	1.53	-	0.03	1.24	-	0.24	0.90	0.75	(See <i>et al.</i> , 2006)
	Non	Urban	2.08	0.44	1.03	-	0.06	1.47	-	0.44	1.46	1.19	
Indonesia													
Jakarta	Annual	Urban	0.48	0.88	0.74	0.98	0.05	0.61	1.45	0.24	0.04	0.65	(Santoso <i>et al.</i> , 2020)
China													
Beijing	Haze	Urban	0.58	-	1.04	1.04	-	0.67	0.55	0.30	0.03	0.39	(R. Shen et al., 2017)
	Non-haze	Urban	2.94	-	3.92	1.57	-	2.55	0.98	1.33	0.04	0.23	
Ningdong (Yinglite)	Winter	Industrial	2.07	-	4.83	0.33	0.06	0.99	0.31	0.57	0.00	0.05	(Liang <i>et al.</i> , 2019)
	Summer	Industrial	4.61	-	12.43	0.52	0.12	2.03	0.34	1.27	0.01	0.06	
Zhengzhou	Haze	Urban	1.36	1.48	1.66	1.09	-	1.18	-	-	0.03	0.22	(Dong <i>et al.</i> , 2020)
	Sandy haze	Urban	3.57	8.23	2.54	1.68	-	2.52	-	-	0.01	0.14	
	Non-haze	Urban	3.28	5.36	6.62	1.70	-	3.12	-	-	0.03	0.32	
Cixi	Haze	Mixed	0.29	0.97	1.04	0.35	0.01	0.31	0.48	0.15	0.02	0.32	(Lu <i>et al.</i> , 2020)
	Non-haze	Mixed	0.52	1.12	0.68	0.40	0.01	0.54	0.67	0.15	0.04	0.37	
India													
Delhi, India	Annual	Urban	2.44	-	3.36	0.87	-	3.01	0.70	1.16	0.68	0.86	(Shridhar <i>et al.</i> , 2010)
	Annual	Rural	2.68	-	3.08	0.92	-	2.19	0.67	1.03	0.21	0.39	
Delhi, India	Summer	Traffic	1.39	3.23	1.34	-	0.12	1.22	-	-	0.03	0.34	(Pant <i>et al.</i> , 2015)
	Winter	Traffic	0.62	0.65	0.40	-	0.03	0.42	-	-	0.03	0.23	
Europe and America													
Barcelona, Spain	Annual	Urban bg	-	-	0.78	0.69	-	0.83	-	0.25	0.04	0.23	(Salameh <i>et al.</i> , 2015)
Marseille, France	Annual	Urban bg	-	-	5.57	1.15	-	1.35	-	0.73	0.09	0.12	
Genoa, Italy	Annual	Urban bg	-	-	0.79	0.71	-	1.01	-	0.26	0.04	0.14	
Thessaloniki, Greece	Annual	Urban bg	-	-	5.49	1.16	-	2.44	-	1.19	0.23	0.21	
Toronto, Canada	Annual	Urban	0.20	-	0.55	0.42	0.02	0.54	-	-	0.03	0.09	(Jeong <i>et al.</i> , 2020)
Los Angeles, US	Annual	Urban	2.55	-	2.85	-	0.25	4.24	-	1.33	0.20	0.08	(Cheung <i>et al.</i> , 2011)

Note: haze – haze event during local dry season; non-haze – outside haze event during dry or wet season

2.5 Source apportionment of PM in Southeast Asian countries

2.5.1 Popular receptor models for PM source apportionment in Southeast Asia studies

Source apportionment of PM provides key information for developing measures to control PM pollution. Receptor models are widely used for source apportionment. Chemical Mass Balance model (CMB) and Positive Matrix Factorization model (PMF) are the most frequently used ones across the world. Fundamentally, the principle of receptor model is that mass conservation can be estimated and source of contaminants in PM can be apportioned by a mass balance analysis which obtained from chemical constituents. A mass balance can then be simply written from chemical species in n number of samples and p independent sources as following equation (Hopke, 2016):

$$x_{ij} = \sum_{k=1}^p g_{ik} f_{kj}$$

where x_{ij} is j^{th} chemical species concentration in i^{th} sample, f_{kj} is concentration of j^{th} chemical species from k^{th} source, g_{ik} is contribution of k^{th} source in i^{th} sample. The approach of analysis in source apportionment of airborne particulate samples is to identify the contribution of sources in samples (g_{ik}) by known species concentrations from chemical analysis (x_{ij}) and chemical source profile (f_{kj}). PMF model has been more frequently used as a source apportionment tool in previous studies than CMB model due to the requirement of chemical source profiles in CMB. CMB is suitable for any source apportionment study which sources and their profiles are known (Hopke, 2016).

In Southeast Asian countries, few studies performed source apportionment of PM. From hundreds of studies regarding atmospheric particles in Southeast Asia region we have reviewed,

only 22 studies investigated the potential sources of PM by receptor models. Among those studies which PM source apportionment were done, the PMF has mostly been applied as the main tools for source apportionment followed by CMB. Overall, Malaysia is the country with the largest number of the PM source apportionment studies, particularly in Kuala Lumpur (Rahman *et al.*, 2011; Khan *et al.*, 2016, 2017; Sulong *et al.*, 2017, 2019). A few studies were also carried out in other Southeast Asian countries including Vietnam (Cohen *et al.*, 2010a; Hai & Kim Oanh, 2013; Hang & Kim Oanh, 2014), Singapore (Engling *et al.*, 2014; Santoso *et al.*, 2011), Thailand (Chuersuwan *et al.*, 2008; Narita *et al.*, 2019; Wimolwattanapun *et al.*, 2011), Indonesia (Santoso *et al.*, 2011), Myanmar (Sricharoenvech *et al.*, 2020), and Philippines (Bagtasa *et al.*, 2018). Here, we compiled available PM source apportionment data in Southeast Asian countries, which are compared with data from other megacities in India and China

2.5.2 PM source apportionment results by receptor models

Based on previous studies in PM source apportionment, we categorize the major sources of PM into 7 main types including Biomass burning (BB), Secondary aerosols (SA), Industrial (IND), Road traffic (excluding road dust) (RT), Soil dust (SD) or Road dust (RD), Sea salt (SS), and Fossil fuel (FF). However, due to the complexity of PM sources which sometimes cannot be clearly separated, we attributed mixed sources as “other” sources. A total of 22 source apportionment studies have been carried out over Southeast Asian countries which are summarized in Table 2-4. Studies conducted in the megacities of India and China are also summarized in a separated Table 2-5. Each of the major source will be discussed in terms of their main chemical profile and contribution to PM mass.

2.5.2.1 Biomass burning (BB)

Biomass burning (BB) haze in Southeast Asian countries occur seasonally, i.e., during December – April in continental countries and during June – September in maritime countries. Hazes are mainly caused by burning of biomasses, such as forest fires over Southeast Asian continent (Boreddy et al., 2018; Le et al., 2014; S. Yin et al., 2019), peatland fires in Sumatra Island, Indonesia (Atwood *et al.*, 2013; Fujii *et al.*, 2015; Lee *et al.*, 2017; Latif *et al.*, 2018; Tham *et al.*, 2019), or various types of agricultural wastes such as rice straw, maize residues, sugarcane by open burnings after harvest season (Kim Oanh et al., 2018; Lasko & Vadrevu, 2018; Phairuang et al., 2019; Pongpiachan et al., 2017a; Popovicheva et al., 2017; Sirithian et al., 2018). On a global scale, agricultural and forest fires contributed 23% and 18%, respectively, to the total BB aerosol emission. While in Southeast Asia, 252-235 million ha (6%) of total forest area was reduced by BB (Yadav *et al.*, 2017). Chang and Song (2010) reported the burned area over tropical Asian countries at 85,2277 km²yr⁻¹ during year 2000 – 2006 and were mostly dense in cropland, forest, grass land, and shrubland. Sillapapiromsuk *et al.* (2013) reported 71 – 88% and 12 – 29% of the burned area during 2010 - 2011 in Chiang Mai, Thailand, was due to forest fires and agricultural land burning, respectively. Similarly, the most recent study in 2019 in the Northern Thailand during burning season indicated that approximately 80% of the total burned area was due to forest fire (8,344 km²) and only 16% was in agricultural area (1,647km²) (Akbari et al., 2021). This is also consistent with a study by Shi and Yamaguchi (2014) who reported that 70% of total burning area over Southeast Asia was in forest.

BB aerosols are primarily composed of organic matter and EC with minor contributions from inorganic ions. OC, K⁺ and levoglucosan are often used as tracer species for source apportionment of BB aerosol (Yin and Harrison, 2008; Khan *et al.*, 2016; Sricharoenvech *et al.*, 2020; Hai and Kim Oanh, 2013). K⁺ appeared to be a more popular tracer, likely due to its easiness to determine. However, levoglucosan can be a more effective tracer of BB because it is a product

from cellulose burning at a temperature above 300 °C. Chantara *et al.* (2019) found in a chamber study that the ratios of levoglucosan/K⁺ from agricultural waste is significantly lower than forest leaf litters which can be potentially used to identify the sources of BB. Thepnuan *et al.* (2019) reported that the ratio of levoglucosan/K⁺ (0.92) and LG/mannosan (20.4) in PM_{2.5} and the high OC content (36.5%) indicated forest and agricultural waste burning were main contributors to PM_{2.5} mass during haze season 2016 in Chiang Mai, Thailand.

K⁺ alone might be insufficient for apportionment of BB source when PM is influenced by other key sources. PMF studies in Kuala Lumpur showed that BB aerosol is often identified as mixed with other sources such as secondary aerosols (Amil *et al.*, 2016; Khan *et al.*, 2017; Sulong *et al.*, 2017) or sea salt (Khan *et al.*, 2016) during haze episodes. This may be attributed to the formation of secondary particles on BB aerosols during their long-range transport (Khan *et al.*, 2016). The receptor sites of these studies are close to the ocean and are hundred miles away from the BB sources in Indonesia. By using levoglucosan as a BB aerosol tracer coupled with Polycyclic Aromatic Hydrocarbons (PAHs), Sulong *et al.*, (2019) observed that 89% of total levoglucosan is related to BB aerosol, which contributes to 22% of PM_{2.5} mass during June 2015 – May 2016 in Kuala Lumpur. PAHs have also been used to determine the contribution of BB in PM. Khan *et al.* (2017) employed PMF by two different sets of chemical compositions, i.e. inorganic compounds and PAHs, and they found that BB aerosol is correlated with high molecular weight PAHs during burning season in Kuala Lumpur. However, other common PAHs sources such as vehicular exhausts or fossil fuel emissions may also contribute to airborne PAHs (ChooChuay, Pongpiachan, Tipmanee, Suttinun, et al., 2020; Pongpiachan et al., 2015b; Wiriya et al., 2013a). PAHs can be used as markers to differentiate between BB and other sources, for example, low molecular weight PAHs (two or three aromatic rings) is usually from natural gas and coal burning (Khan *et al.*, 2017), moderate molecular weight PAHs (three or four aromatic rings) from diesel vehicles, while high molecular weight PAHs (four or more aromatic rings) can be frequently found during haze

season by BB source (Phoothiwut & Junyapoon, 2013) and gasoline vehicle emissions (Tian *et al.*, 2021).

BB aerosol has also been observed during wet season. A study at an industrial site (Mong Dong, Vietnam) showed BB from local sources such as cooking and garden waste burning contributed significantly during wet season, which also contained other trace elements (Fe, Zn, Ca, Cd, Cu) (Hang & Kim Oanh, 2014). Narita *et al.* (2019) applied a CMB model and found that the contribution of BB in Bangkok, Thailand, differed only 5% between dry and wet season at an urban site but differed by 15% at a suburban site (curbside Bangkok). Even at a rural site near coast in Burgos city, Ilocos Norte, Philippines, BB from local source was also found to be as high as 43% in summer and 35% in winter during 2015 - 2016, which is higher than BB from long range transport (3% and 5% in summer and winter) (Bagtasa *et al.*, 2018).

In Southeast Asia, BB is often the most significant source in PM with contributions mostly more than 20%. Lower contributions were recorded in some cities in Vietnam due to less intensive BB activities. The highest contribution was reported up to 75.6% in Singapore during haze event (Table 2-4). Comparing with other studies observed in Asian megacities including Delhi (India), and Beijing (China) (Table 2-5), the contribution of BB in airborne particles over Southeast Asian countries (Table 2-4) is much higher. BB contributes 8% - 18% to PM_{2.5} in Beijing and 11% - 23% in Delhi. This is similar to the data by Zhu *et al.* (2018) who found that BB in China ranged from 8.5% - 18%. Other sources including SIA, soil dust and industrial emission are the top three contributors in PM_{2.5} across China, except in Northeast and Southwest of China where the fossil fuel and biomass burning are more important (Zhu *et al.*, 2018). A review by Pant and Harrison (2012) showed that BB contributed 7% to 20% of PM_{2.5} in India depending on season and location. This agreed well with a global review of recent source apportionment in year 2020 that biomass burning accounted for 16.3%, 10.3% and 14.9% in PM_{2.5} for Southeast Asia, China and South Asia, respectively (Hopke *et al.*, 2020).

2.5.2.2 Secondary aerosols (SA)

Secondary aerosols are formed in the atmosphere from precursor gases such as SO_2 , NO_x , NH_3 and volatile organic compounds (VOCs). SO_2 is generally emitted from the combustion of sulfur contained fossil fuel such as coal and liquid fuels, NO_2 is emitted from high temperature combustion processes, whereas NH_3 is from agricultural sources, animal waste, road traffic, and power plant slips (Behera et al., 2013; Hopke et al., 2020; Pant & Harrison, 2012). VOCs can be from natural biogenic or anthropogenic sources. There are two main types of secondary aerosol including secondary inorganic aerosol (SIA) and secondary organic aerosol (SOA). SIA are often separated into sulfate, nitrate, or mixed in source apportionment studies (Hopke *et al.*, 2020).

Sulfate (SO_4^{2-}), nitrate (NO_3^-) and ammonium (NH_4^+) (known as SNA) are the key species of secondary particulate aerosols due to their significantly high contributions to PM mass. SA formation varies depending on meteorological conditions (sunlight intensity, temperature, humidity) (R. Zhang et al., 2018), precursors concentrations and preexisting particles (X. Fu et al., 2020). Recent studies showed that during haze episodes, secondary sulfate formation is influenced by high relative humidity, and mainly produced via the aqueous – phase oxidation of SO_2 in the liquid water on the surface of particles rather than gas – phase conversion of SO_2 , while secondary nitrate is less impacted by relative humidity (Zhang *et al.*, 2018) and gas – to – particles partitioning is the main pathway control secondary nitrate (Dong *et al.*, 2020). Through neutralization reactions with alkaline species such as NH_4^+ , sulfate and nitrate are converted to secondary particles such as NH_4HSO_4 , $(\text{NH}_4)_2\text{SO}_4$, and NH_4NO_3 (Dong *et al.*, 2020). Secondary nitrate is likely to dominate during winter due to low temperature enhances the formation of NH_4NO_3 while secondary sulfate is higher in summer when photochemical activity is high (Hopke *et al.*, 2020).

A significant number of studies did not clearly identify secondary inorganic sources as they are often mixed with sources. Only 9 out of the 23 studies reported the actual secondary aerosol contribution to PM. As shown in Table 2-4, contributions of secondary aerosols to PM in Southeast Asia varied significantly, ranging from as low as 0.1% in Bangkok, Thailand (Chuersuwan *et al.*, 2008)) to as high as 41.9% in Cam Hai, Vietnam (Hang & Kim Oanh, 2014). The extremely low contribution of secondary aerosol to PM appears unreasonably and may be due to the mixing of sulfate and nitrate in other factors.

In contrast, SA contributions in Beijing and Delhi (Table 2-5) were found to be relatively consistent, which are usually above 20%. This observation is consistent with Hopke *et al.* (2020), who reported that the contribution of mixed SIA in Southeast Asia was 23.9% while in Southern Asia and China were 34.3% and 31%, respectively. Singh *et al.* (2017) reported that SA contributed to 22% of PM at 5 urban stations in South Asia (Delhi, Dhaka, Mumbai, Agra, and Lahore). A review by Zhu *et al.* (2018) showed that SIA is one of the top three sources identified in China. Furthermore, since sulfate formation is favored in high temperature during summer and nitrate formation in lower temperature during winter (Zhang *et al.*, 2013), SIA in India and China has often been separated to sulfate and nitrate sources by the receptor models. However, differences of temperature between winter and summer in tropical Southeast Asian countries are low, which may explain why SA is mostly lumped as total secondary aerosols or mixed secondary inorganic aerosols (SIA).

There are extremely limited data for source apportionment in Southeast Asian countries using secondary organic aerosol tracers. In the case that OC/EC ratio is higher than 2, secondary organic carbon (SOC) can be estimated (ChooChuay, Pongpiachan, Tipmanee, Suttinun, et al., 2020) by using EC-tracer method (Turpin & Huntzicker, 1991). Thuy *et al.* (2018) reported that SOC contributed 43%, 42%, and 28% to PM_{0.1}, PM_{2.5}, and PM₁₀, respectively, in Hanoi, Vietnam. Pani *et al.* (2019) found that SOC in urban site was higher than rural site in Chiang Mai, Thailand,

indicating higher occurrence in the reaction between gaseous organic precursors and the oxidation of volatile organic compounds over urban areas. However, SOC in Southeast Asia PM has not been estimated by receptor models. A recent study conducted in Yangon, Myanmar, identified SOA and SIA as two of the four main sources in PM₁₀ by CMB model (Sricharoenvech *et al.*, 2020). They also found that SIA was seasonally dependent which increased during winter and summer seasons and decreased during rainy season, while SOA seem to be related to temperature which was highest in summer followed by rainy season and winter.

2.5.2.3 Industries (IND)

Industrial sources were identified in several receptor modelling studies. Chemical composition of PM from industrial activities vary with respect to the influence of local or regional types of industry surrounded the receptor sites, production processes, raw materials used, different operating conditions (X. Bi *et al.*, 2018; Pant & Harrison, 2012). A series of heavy metals have been used as tracers to identify industrial sources, for example, V, Cr, Cu, Mn, Cd, Pb, As, and Zn. Unlike biomass burning and secondary aerosols, such tracers for identifying industrial emissions differ according to type of industries. A review study regarding source apportionment of PM emitted from industries have stated that some studies were not able to distinguish industrial emission from other sources and the tracers of general and specific industries vary between studies and source of PM (Taiwo *et al.*, 2014). Many studies have showed various set of tracer apportioned from different industries, for example, Ca and Al are potential tracers for cement emissions, and Fe, Si and K for steel plant emissions (Bi *et al.*, 2018); Hsu *et al.* (2017) indicated that Co, Fe, Mn and Pb can be used as tracers for iron ore and steel industrial emissions; Matawle *et al.* (2014) reported that Co, Cd, Mo, Sb, and V are good tracers for brick kiln processes, Hg and Pb for steel rolling mills, and As and Ni for steel processing industries; Police *et al.* (2018) indicated that Cr and Mn are tracers of metal industrial emissions. Sylvestre *et al.* (2017)

measured trace metals from different steps in steel industry and found Fe, Al, Mn and Zn as likely tracers.

In Southeast Asian countries (Table 2-4), the contributions of industrial sources to PM is highly variable in different receptor modelling studies, ranging from 0.1% at a rural site (Bagtasa *et al.*, 2018) to 25.4% at an urban site during haze season (Sulong *et al.*, 2017). The tracer species for the industrial source are different in different studies. In Kuala Lumpur, Malaysia, Rahman *et al.* (2011) used Al, Si, Mg, K, Cl, V, Cr, Mn, Zn, As, and black carbon (BC) as IND PM tracers while Sulong *et al.* (2017) used Ba, K, Sr, Cd, and V during haze season and Cr, Mn, and As for both haze and wet seasons. In Hanoi, Vietnam, Cohen *et al.* (2010) apportioned 3 different industries including zinc (Zn, Si, Pb), calcium (Ca, H, K, Na, Fe, and heavy metals), iron (BC, Fe, and significant amount of V and Ni), and Hai and Kim Oanh (2013) used Mg^{2+} , Fe, Pb, Zn, Al, Cr and Mn as industrial PM tracers. In Bangkok, Thailand, rather than using metal tracers, ChooChuay *et al.* (2020) used F^- and NO_2^- which could be made from electrolytic aluminum industries or waste incinerations. In Chiang Rai, Thailand, Kayee *et al.* (2020) used Cr, Ni and Mn as industrial tracers. A study in Singapore used low molecular weight PAHs, anthracene (Ant) and fluoranthene (Flt) as indicators for stationary source such as steel and refinery industries (Urbančok *et al.*, 2017). It should be noted that the some studies (ChooChuay, Pongpiachan, Tipmanee, Suttinun, et al., 2020; Kayee et al., 2020; Urbančok et al., 2017) used Principal Component Analysis model (PCA) as a source apportionment tool.

The assignment of industrial source is challenging because different tracers are used in different studies. This makes it difficult to directly compare the contributions of industrial source to those reported in other places such as India and China. The global review by Hopke *et al.* (2020) indicated that this source contributed 15.4%, 6.4%, and 16.8% for Southeast Asia, Southern Asia, and China, respectively. However, Singh *et al.* (2017) reported a 23% contribution of industries to

PM_{2.5} over 5 cities in South Asia, and Zhu *et al.* (2018) argued that even industrial emission is one of the top three sources of PM in China, its contribution varied significantly in different regions.

2.5.2.4 Road traffic (RT) (excluding road dust)

Road traffic emissions are considered one of the most important sources of PM especially in the urban environments. Road traffic was reported in a review study as the 2nd highest contribution of PM in Southeast Asia region at 23%, slightly lower than mixed SIA at 23.9% (Hopke *et al.*, 2020). PM_{2.5} emitted from different vehicle engines have shown distinct chemical profiles depending on fuel types. Elemental carbon (EC) and organic carbon (OC) are the most significant species in engine exhaust PM (Hao *et al.*, 2019; Lin *et al.*, 2020). They are used in combination with other elements for example from source profile studies, Mn, Zn and Fe as tracers for gasoline exhaust particles, as well as organic compounds such as PAHs (Jin *et al.*, 2017). Mn is used as additive in gasoline to increase gasoline octane (Shridhar *et al.*, 2010). Hosseinzadeh-Bandbafha *et al.* (2018) stated that Pt, Ce, Fe, Ba, Mn, and Cu are common metal-based additives in diesel/biodiesel for combustion improvement. Cu and Zn are major additives to lubricant oils and used in wear of brake and tyres (Y. Liu *et al.*, 2019; L. Yu *et al.*, 2013). Pb may also be emitted from wear of brake and tyres or resuspended from road dust (L. Yu *et al.*, 2013). But all of these tracers have many other potential sources including industries.

The contribution of road traffic to PM (Table 2-4) ranged from 10% in Hanoi, Vietnam (Hai and Kim Oanh, 2013) to 65% in Bangi, Malaysia (only PAHs applied in PMF) (Khan *et al.*, 2015). In Kuala Lumpur, Malaysia, Rahman *et al.* (2011) apportioned up to 57.6% (two stroke engine 25.7% and motor vehicle 31.9%) with high load of BC and metallic tracers Fe, Zn, Mn, and Br, and Sulong *et al.* (2017) found an annual contribution of 22.4%, which was higher than during haze season (17.8%). In Hanoi (Vietnam), Cohen *et al.* (2010) apportioned (8 years) this source into 3 different types with a total of 40.2%. BC, Cu, Zn, Br, and Pb were key species, while a contribution

of only 10% to PM was reported from Dec 2006 to Feb 2007 in Hanoi in a study by Hai and Kim Oanh (2013). At a residential site in Serpong, Indonesia, traffic source was found to contribute 30% to PM with high level of Zn, and S due to high sulfur in diesel fuel (Santoso *et al*, 2011). Long term study (5 years) in Bangkok, Thailand, indicated that road traffic was the largest contributor of PM_{2.5} (50 – 70% of PM_{2.5} mass), which is associated with high BC, S, Ca, Si, Fe as major species (Wimolwattanapun *et al*, 2011). Corresponding to another long-term study (2001-2007) in Bandung, Indonesia, where the road traffic emissions were main sources of PM_{2.5} (22% in dry season, 35% in wet season) (Lestari & Mauliadi, 2009).

Hai and Kim Oanh (2013) observed higher contribution of road traffic sources in wet season than dry season at an industrial site (Mong Doun) and a rural site (Cam Hai), Vietnam, which is consistent with the results observed in Bangkok, Thailand (Narita *et al*, 2019). This is attributed to lower contributions of other major sources in PM mass such as secondary aerosol or biomass burning during wet season (Table 2-4). Some studies observed at coastal sites such as in Mong Doun and Cam Hai, Vietnam, showed a high contribution of ship emissions which identified by presence of NaCl and BC (Hang & Kim Oanh, 2014). In Singapore, ship emission was found to contribute 27% of PM_{2.5} during non-haze days (Engling *et al*, 2014), based on high OC, sulfate and PAHs with molecular mass at 178 – 300 amu as tracers.

Road traffic is an important source of PM_{2.5} in China with its contribution ranging from 10.2% (North China) and 21.5% (South China) (Zhu *et al*, 2018). In India, road traffic contributes to 10 – 80% to PM (Pant & Harrison, 2012) and 37% on average in South Asia (N. Singh *et al*, 2017). From Table 2-4, we can see that the contribution of this source to PM in Southeast Asia is usually higher than that in China but close to that in India.

2.5.2.5 Soil dust (SD) and Road dust (RD)

Soil dust contains crustal elements including Al, Ca, Na, Mg, K, Si, Fe and Ti and is emitted into the atmosphere by winds and anthropogenic activities such as constructions while road dust is generally identified by crustal elements coupled with additional metallic elements such as Cu and Sb (brake wear), Zn (tire wear), and Zn, Ba, Ca (lubricant oil) (Hopke *et al.*, 2020). Soil and road dust sources sometimes can be difficult to discriminate due to their complexity of chemical compounds mixed from various types of original sources which accumulate on the ground through dry deposition (Pant & Harrison, 2012).

Soil or road dust are not detected in all receptor modelling studies and sometimes they are identified as being mixed with other sources (Table 2-4). Khan *et al.* (2016) reported a mixed source of soil and road dust characterized by major tracers. Mg (natural dust) coupled with Zn, Cu and Ba (emitted from non-exhaust sources), and moderate level of Ni and V (from lubricant oil), at University Kebangsaan (study site), Kuala Lumpur, in dry season 2013 and wet season 2014. After that, at the same study site, in dry season 2014 Khan *et al.* (2016) separated dust into 2 groups, mixed soil and road dust, and mixed road dust with fuel oil combustion identified by high concentration of road dust tracers with Ni and V emitted from shipping, refining of crude oil and industrial power plants, while soil dust was found with enriched Mg. Santoso *et al.* (2011) reported a source of lead industry mixed with small part of road dust due to the presence of sulfur in the same factor from two residential sites in Serpong, Indonesia, and also identified a separated road dust source. ChooChuay *et al.* (2020) identified a mixed source of sea salt and soil dust by high load of Cl^- and Na^+ (from sea salt), and Mg^{2+} and Ca^{2+} (from soil dust) in Bangkok. In addition, two mixed sources of soil dust with BB, and refuse incineration with road dust were reported in Chiang Rai, Thailand (Kayee *et al.*, 2020).

Studies in Southeast Asia have shown that variations in contributions of soil and road dust depend on seasonal conditions. Low humidity during dry season enhances the disturbances by winds or mechanical activities like constructions and traffics. For example, at a coastal rural site in Philippines, the contributions of fine dust to PM are in this order: winter (15%) > fall (11%) > spring (7%) > summer (3%) (Bagtasa *et al.*, 2018). A study in Kuala Lumpur, Malaysia, found that the contribution of dust in pre-haze, haze and post haze periods were 25.8%, 22.3% and 31.1%, respectively (Sulong *et al.* (2017)). Amil *et al.*, (2016) showed a similar pattern as the highest contribution of mineral dust was observed during haze season at 19%, in comparison to 7% during inter-monsoon. However, Rahman *et al.* (2011) reported that soil dust and road dust did not show seasonal pattern except few high peaks during low rainfall in February and in dry season (April – October). The contribution of soil and road dusts to PM is also highly dependent on the sizes of PM. Lestari and Mauliadi (2009) reported very high contributions of various types of dust contributed to PM₁₀ (29% in dry season and 59% in wet season) in Bandung, Indonesia. Wimolwattanapun *et al.* (2011) showed evidently that construction dust and soil dust were the major sources of coarse PM at both Bangkok (urban) and Pathumthani (suburban) sites but less important in PM_{2.5}. Furthermore, the contribution of the two sources varied depending on the sampling sites. In the same study, Wimolwattanapun *et al.* (2011) reported a higher contribution of road dust in coarse PM at the urban site than at suburban site (25% vs 6%). On the other hand, Narita *et al.* (2019) observed a slightly higher contribution of soil and construction dust at a suburban site (5.1% – 9.2%) than an urban (4.4% – 5.6%) site in Bangkok. This difference in the pattern of dust contribution to PM at the two studies may be due to the chosen locations of the urban and suburban sites which have different local environments.

Overall, the contributions of soil and road dust to PM in Southeast Asian countries vary significantly between sites and season. Long-term studies showed that these sources contributed less than 10% in PM_{2.5} at some locations (Table 2-4) (Cohen et al., 2010a; Rahman et al., 2011;

Wimolwattanapun et al., 2011). This appears to be lower than that estimated by Hopke *et al.* (2020) in their global review where they estimated the dust, contribution to PM in Southeast Asia to be as high as 19.1%. This is probably due to the difference in percentage contribution of this source reported in literatures especially short – term studies where mostly conducted during haze event when contribution of soil/road dust is usually high.

Similar to that in South China, soil and road dust are not the top three sources of PM_{2.5} in published studies Southeast Asian countries (Zhu *et al.*, 2018) (Table 2-5). This is not the case in Delhi India where dust is a key contributor to PM_{2.5}, particularly to the coarse particles (N. Singh et al., 2017) (Table 2-5).

2.5.2.6 Sea salt (SS)

Sea salt is generated by natural processes. During long-range transport, fresh sea salt ages, which could lead to the replacement of chloride by nitrate or sulfate ions (Khan *et al.*, 2017; Sulong *et al.*, 2019). Therefore, sea salt, in some case, represents the sum of fresh and aged sea salts in the aged air masses (Hopke *et al.*, 2020). Previous studies agreed well that this source can be identified by significant amount of Na⁺, Mg²⁺, and Cl⁻ (Pant & Harrison, 2012; N. Singh et al., 2017). The use of Cl⁻ as a sole sea salt tracer may not work due to Cl⁻ emission from biomass burning and coal combustion.

Southeast Asian countries have a variety of topographic features from continental mainland in the north through maritime continental and islands in the south where close to the Indian ocean and the Pacific Ocean. Studies across different geographical areas show that this source is dependent on the location. The closer to the sea/ocean, the higher contribution of sea salt. Significantly higher contribution of sea salt to PM was observed in some Southeast Asian countries such as Malaysia, Indonesia, and Philippines. This source has also been observed with

high seasonal variations. In Kuala Lumpur, Malaysia, Sulong *et al.* (2017) found that sea salt during haze season (1.8% to $PM_{2.5}$) was significantly lower than the whole study period (18.6%). Amil *et al.* (2016) studied between different monsoon and inter-monsoon periods and revealed that sea salt contributed highest to $PM_{2.5}$ in the first inter-monsoon (INT1) during April to early May at 45% and lowest during haze and the second inter-monsoon (INT2) (6%). At a rural site near coastline in Philippines, the contributions of sea salt to $PM_{2.5}$ was in this order: summer (49%) > fall (40%) > spring (15%) > winter (14%) (Bagtasa *et al.*, 2018). While in Bandung, Indonesia, Lestari and Mauliadi (2009) found significant contributions of aged sea salt in PM_{10} during dry season associated with wind directions from Indian Ocean and Java sea.

Similar to other sources, sea salt has frequently been identified as mixtures with other sources especially secondary sulfate and nitrate. This mixtures are sometimes characterized by high contributions of non-sea salt sulfate ($nss-SO_4^{2-}$) and non-sea salt nitrate ($nss-NO_3^-$) (Khan *et al.*, 2016; Khan *et al.*, 2017; Fujii *et al.*, 2017). Hai and Kim Oanh (2013) showed that long marine air mass trajectories were found to be associated with high contributions of the mixed sources of sea salt, sulfate and nitrate. Other studies have also found mixed aged sea salt with industrial sources (Khan *et al.*, 2016), or with dust (ChooChuay, Pongpiachan, Tipmanee, Suttinun, et al., 2020).

Because this source is usually identified as a mixture with other sources and there is a lack of long-term study, only 8 from 23 reported specific sea salt source. Therefore, it is quite challenging to estimate the general contribution of sea salt. In Southeast Asian countries where sea salt was identified, its contribution (Table 2-4) to $PM_{2.5}$ ranged from as low as 1.8% during haze in Kuala Lumpur, Malaysia (Sulong *et al.*, 2017), to as high as 49% during summer at a rural area in Philippines (Bagtasa *et al.*, 2018). Hopke *et al.* (2020) estimated that sea salt contributed at 7.8% to $PM_{2.5}$ mass for Southeast Asia. Comparing to China and India, sea salt has been more often identified in Southeast Asia than those of two countries. Table 2-5 shows that sea salt is not

an important source of PM in China since only the eastern China locates closely to the ocean. This is consistent with the review by Zhu *et al.* (2018). Sea salt is a negligible source to PM in inland India (Pant and Harrison, 2012) but its contribution is not known in coastal cities due to lack of data.

2.5.2.7 Fossil fuel combustion (FF)

Fossil Fuel here refers mainly to coal and heavy oil combustions. Because coal's chemical profile differs depending on the nature of the coal, understanding of its nature is crucial for the accurate source apportionment of coal by receptor models (Hopke *et al.*, 2020). For example, in China, high abundance of Cl^- found in coal combustion which is a distinctive feature mostly observed in the northern China (Tan *et al.*, 2017; Xu *et al.*, 2018), and high abundance of As and Se (Liu *et al.*, 2019; Chen *et al.*, 2020) are reported in coal combustion particles while high abundance of Ni and V are associated with particles from oil combustion, while high Al, Cr, Cl, Fe, Zn, and SO_4^{2-} were found from fossil fuel combustion in India (Sharma & Mandal, 2017). A recent study on source profiles of $\text{PM}_{2.5}$ from three types of coal combustion (coal-fire industrial boiler, power plant and residential stove) found that OC, crustal elements (Al, Ca, Mg, and Si), ions (NH_4^+ , Cl^- , and SO_4^{2-}) and moderate molecular weight PAHs (three and four rings PAHs) were the major contributors in industrial boiler and power plant while residential stove was found with lower crustal elements (Tian *et al.*, 2021).

Studies in Southeast Asia used similar tracers for fossil fuel source. Khan *et al.* (2016) used the As, Ba and Sr for dry season 2013 and wet season 2014 in Kuala Lumpur, Malaysia as coal combustion tracers. They also used As in dry season 2014 with additional element Bi, OC and pyrolysis organic carbon to identify coal combustion. Cohen *et al.* (2010) used Na, S and trace heavy metals such as Co, Cu, Br, and Pb to identify coal combustion in Hanoi, Vietnam. Santoso *et*

al. (2011) also used high value of sulfur to identify coal and residual oil burning from multiple fueled power plants in Serpong, Indonesia.

PAHs have also been used as tracers of fossil fuel derived by receptor models. For instance, in Kuala Lumpur (Malaysia), fluorene, anthracene, fluoranthene, pyrene and benzo[k]fluoranthene (Sulong *et al.*, 2019b), low molecular weight PAHs like naphthalene, acenaphthylene, acenaphthene, fluorene and anthracene (Khan *et al.*, 2015) were used to identify natural gas and coal burnings. In Bangkok (Thailand), Choochuay *et al.* (2020) identified fossil fuel combustion from power plants nearby Bangkok by PCA by using coronene, benzo[g,h,i]perylene, indeno[1,2,3-cd]pyrene, dibenzo[a,h]anthracene and dibenzo[a,e]pyrene as tracers.

Only half of reviewed studies in Southeast Asia have identified this source. This suggests that either fossil fuel combustion is not a major source of PM or it is not specifically identified in the previous studies which mostly did not measure organic tracers. In studies with fossil fuel source, its contribution (Table 2-4) ranged between 5.2% (Singapore, G. Engling *et al.* (2014)) and 28.3% (Kuala Lumpur, Sulong *et al.* (2019)). Hopke *et al.* (2020) estimated that fossil fuel combustion contributed 13.8% in Southeast Asia region, while South Asia and China contributed 7.1% and 10.9%, respectively. A review in China during 2007 – 2016 reported the contribution of fossil fuel ranged from 7% (South China) and 18.1% (Northeast China) with a decreasing trend from 27.2% during 2005 – 2009 to 10.7% during 2010 – 2016 (Zhu *et al.*, 2018).

Table 2-4 Summarized percentage contributions (%) in total PM mass of 7 main sources over Southeast Asian countries

City, Country	Duration	PM	Site	No. of factors	Percentage contribution (%) of factor in PM mass								References
					Biomass burning (BB)	Secondary aerosol (SA)	Industrial (IND)	Road Traffic (RT)	Soil (SD) /road dust (RD)	Sea salt (SS)	Fossil fuel (FF)	Mixed sources and others	
Malaysia													
Kuala Lumpur	2004 - 2008	PM2.5	Urban	6	17.5	-	16.7	57.6	SD: 8.3	-	-	-	(Rahman <i>et al.</i> , 2011)
Kuala Lumpur	Jun 2015 – Jan 2016	PM2.5	Urban	5	-	-	Haze: 25.4 Whole periods: 15.9	Haze: 17.8 Whole periods: 22.4	Haze: 22.3 Whole periods: 4.7	Haze: 1.8 Whole periods: 18.6	-	Haze: 32.8 (SIA + BB) Whole periods: 38.5 (SIA + BB)	(Sulong <i>et al.</i> , 2017)
Kuala Lumpur	Sep – Oct 2015 (haze)	PM2.5	Urban	4	22.3	-	-	49.4	-	-	28.3	-	(Sulong et al., 2019b)
Kuala Lumpur	Jun 2015 – May 2016	PM2.5 (PAHs only)	Urban	4	22.3	-	-	49.4	-	-	28.3	-	(Sulong et al., 2019b)
Kuala Lumpur	24 Jun – 14 Sep 2014	PM2.5	Semi-urban	7	-	-	-	-	SD: 8	-	Coal: 25	RD and FF: 7 BB + SS: 7 SIA With F: 15 RT + SS II: 24 Aged SS + IN: 5	(Khan <i>et al.</i> , 2016)
Kuala Lumpur	Jun – Sep 2013, Jan – Feb 2014	PM2.5	Semi-urban	5	-	SA nitrate: 17	-	-	SD + RD: 13	-	Coal: 19	RT + BB: 31 SS + SA sulfate: 20	(Khan <i>et al.</i> , 2016)
Kuala Lumpur	12 Nov 2013 – 15 Jan 2014	PM2.5 (Inorganic)	Urban	4	-	-	-	-	SD: 24	-	-	SA + BB: 34 FF + RD: 25 SS + SA (nitrate): 17	(Khan <i>et al.</i> , 2016)
Kuala Lumpur		PM2.5 (PAHs)	Urban	3	42	-	-	-	-	-	fossil fuel: 36 Natural gas + coal: 22	-	(Khan <i>et al.</i> , 2017)
Kuala Lumpur	Aug 2011 – Jul 2012	PM2.5	Urban-industrial	5	-	-	-	17	SD: 14	17	-	RT + IND: 10 SA + BB: 42	(Amil <i>et al.</i> , 2016)
Kuala Lumpur	Jun – Sep 2013, Jan – Feb 2014	PM2.5 (PAHs)	Semi-urban	3	-	-	-	Gasoline: 65 Diesel: 19	-	-	Natural gas + coal: 15	-	(Khan <i>et al.</i> , 2015)
Vietnam													
Mong Doung (MD),	July – Aug 2009 (Wet), Dec 2009 – Jan 2010 (Dry)	PM2.5	Industrial	5	Wet: 41 Dry: 20	Wet: 16.4 Dry: 41.6	-	Diesel ship Wet: 24, Dry: 22 Traffic Wet: 17, Dry: 13	-	-	-	Miscellaneous Wet: 1.4 Dry: 3.8	(Hang & Kim Oanh, 2014)
Cam Hai (CM)			Rural	5	Wet: 1.7 Dry: 22	Wet: 41.9 Dry: 38.8	-	Diesel ship Wet: 29, Dry: 15 Traffic Wet: 26, Dry: 14	-	-	-	Miscellaneous Wet: 1.4 Dry: 11	(Cohen et al., 2010a)
Hanoi	Apr 2001 – Dec 2008	PM10	Suburban	10	12	7.8	18.7	40.2	SD: 3.4	-	Coal: 17	-	(Cohen et al., 2010a)
Hanoi	Dec 2006 – Feb 2007	PM2.5	Urban	7	-	SA mixed: 40 SA sulfate rich: 16	6	10	SD: 1	Aged SS: 11	-	Residential: 16	(Hai & Kim Oanh, 2013)
Philippines													
Ilocos Norte	Aug – Sep and Nov 2015, Jan – Feb and Mar 2016	PM2.5	Rural	6	Summer: 43, 3 (LRT) Winter: 35, 5 (LRT)	LRT sulfate Summer: 1 Winter: 8	LRT Summer: 0.1 Winter 22	-	Summer: 3 Winter: 15	Summer: 49 Winter: 14	-	-	(Bagtasa <i>et al.</i> , 2018)

Table 2-4 (continue) Summarized percentage contributions (%) in total PM mass of 7 main sources over Southeast Asian countries

City, Country	Duration	PM	Site	No. of factors	Biomass burning (BB)	Percentage contribution (%) of factor in PM mass							References
						Secondary aerosol (SA)	Industrial (IN)	Road Traffic (RT)	Soil (SD) /road dust (RD)	Sea salt (SS)	Fossil fuel (FF)	Mixed sources and others	
Mainland Southeast Asia	2001 - 2016	PM2.5		3	48	-	-	-	-	-	-	LRT: 55 Anthropogenic: 27	(Yin <i>et al.</i> , 2019)
Thailand													
Bangkok	Jan 2003 – Dec 2007	PM2.5	Urban	6	No percentage reported, sources were ordered: Traffic > Aged sea salt > Biomass burning > Secondary sulfate > Soil > Zn rich								(Wimolwattanapun <i>et al.</i> , 2011)
Pathumthani			Suburban	5	No percentage reported, sources were ordered: Traffic > Biomass burning > Secondary sulfate > Soil > Aged sea salt								
Myanmar													
Yangon	May 2017 – Apr 2018	PM10	Urban	4	4	sources: dust, secondary inorganic aerosol (SIA), secondary organic aerosol (SOA), biomass burning							(Srichaorenvech <i>et al.</i> , 2020)*
Thailand													
Bangkok	Sep 2015 – Mar 2017	PM2.5	Urban	7	Wet: 28.4 Dry: 33.9	Wet: 25.1 Dry: 16.6	Wet: 6.2 Dry: 7.4	Wet: 27.4 Dry: 23.0	Wet: 5.6 Dry: 4.4	Wet: 5.2 Dry: 2.0	-	Wet: 14.7 Dry: 29.0	(Narita <i>et al.</i> , 2019)*
			Suburban	7	Wet: 26.1 Dry: 41.2	Wet: 18.5 Dry: 16.4	Wet: 6.6 Dry: 4.0	Wet: 27.9 Dry: 27.0	Wet: 9.2 Dry: 5.1	Wet: 4.7 Dry: 2.6	-	Wet: 15.2 Dry: 32.6	
Bangkok	Feb 2002 – Jan 2003	PM2.5	Urban 6 sites	6	26 – 41	0.1	-	29 – 32	1 – 2	-	-	Unexplained: 28 – 47 Meat cooking: 4	(Chuersuwan <i>et al.</i> , 2008)*
		PM10	Urban 6 sites	6	28 – 36	0.1	-	22 – 39	RD: 1 – 15	-	-	Unexplained: 23 – 33 Meat cooking: 10	
Bangkok (PCA)	Aug 2017 – Mar 2018	PM2.5	Urban	5	24		4.46	43.7	-	-	Power plants: 6.48	SS + SD: 10.5	(ChooChuay, Pongpiachan, Tipmanee, Suttinun, et al., 2020)**
Bangkok (PCA)	Jan – Apr 2019	PM2.5	Urban	3	-	-	-	14.78	-	12.98		mixture of natural and anthropogenic sources: 54.97	(Kayee <i>et al.</i> , 2020)**
Chiang Rai			Urban	3	-	-	20.96	-	-	-	-	SD and BB: 46.59 Refuse incineration + RD: 18.59	
Indonesia													
Bandung	2001-2007	PM2.5	Urban	9	Wet: 13 Dry: 16	Wet: 26 Dry: 25	Wet: 6 Dry: 24	Wet: 35 Dry: 22	Wet: 20 Dry: -	Wet: - Dry: 13			(Lestari & Mauliadi, 2009)
		PM10	Urban	6			Wet: 41 Dry: 48		Wet: 59 Dry: 29	Wet: - Dry: 23			
Serpong	Aug – Nov 2008	PM2.5	Residential	5	15	-	-	30	RD: 17	-	Coal: 26	IND + RD: 12	(Santoso <i>et al.</i> , 2011)
Riau and Sunatra (CMB)	16 – 22 Mar 2005	PM2.5 (CMB)	Semiurban	5	51	2.9	-	25	-	18.4	-	Petroleum Refinery: 2.7	(Siao <i>et al.</i> , 2007)*
			Urban	5	18.1	7.2		27.5		40.6		Petroleum Refinery: 6.6	(Engling <i>et al.</i> , 2014)*
Singapore	Sep 2006 - Jan 2007	TSP (CMB)	Urban	4	75.6	-	-	Ship emission: 5.3 Diesel Exhaust: 18.6	-	-	-	Petroleum Refinery: 0.5	
				haze									
				4	-	-	-	Ship emission: 27 Diesel exhaust: 62	-	-	Power plant: 5.2	Petroleum Refinery: 5.8	
				Clear days									

*studies employed CMB model

**studies employed PCA model

Table 2-5 Summarized percentage contributions (%) in total PM mass of 7 main sources in India and China

City, Country	Duration	PM	Site	No. of factors	Percentage contribution (%) of factor in PM mass							References	
					Biomass burning (BB)	Secondary aerosol (SA)	Industrial (IN)	Traffic and Vehicle exhaust (TV)	Soil (SD) /road dust (RD)	Sea salt (SS)	Fossil fuel (FF)		Mixed sources and others
India													
Delhi	Jan 2013 – Dec 2016	PM2.5, PM10	Urban	8	23, 19	21.3, 23.2	10.3, 9.9	16, 21	13, 17	6, 7	10, 7		(Jain <i>et al.</i> , 2020)
Delhi	Jan 2010 – Dec 2011	PM10	Urban	7	13.4	21.7	5.5	16.8	20.7	4.6	17.4		(Sharma <i>et al.</i> , 2014)
Delhi	Jan 2013 – May 2014	PM2.5	Urban	7	12.3	23.2	6.3	18.5	22.5	4.1	13.1		(Sharma & Mandal, 2017)
Van Vihar National Park	Jan 2012 – Dec 2013	PM2.5	Park within urban	7	11	28.9			SD: 17.3	0.8	23.6	Pyrolysis carbon-rich aerosol: 15.7	Kumar and Sunder Raman, 2020)
China													
Beijing	15 Dec 2016 – 4 Mar 2017	PM2.5	Urban	6	8	29			RD: 16 SD: 9		Coal: 21	IND + TV: 17	(Xu <i>et al.</i> , 2018)
Beijing	Jan, Apr, Jul, Oct 2000	PM2.5	Urban, 5 sites	7	11	31	6	6	RD: 9		Coal: 19		(Y. Song et al., 2006)
Beijing	Apr, Jul, Oct 2009, Jan 2010	PM2.5	Urban	6	18	SIA: 26	25		SD: 15		Coal: 12	TV + waste incineration: 4	(Zhang <i>et al.</i> , 2013)
Beijing	Nov – Dec 2016	PM2.5	Urban	6	9	SIA: 44	8	18	5		Coal: 16		(Liu <i>et al.</i> , 2019)
Beijing	Jan – Dec 2010	PM2.5	Urban	7	11.3	26.5	6	17.1	SD: 10.4 RD: 12.7 SD: 42.2		15.6		(L. Yu et al., 2013)
Yinglite and Baofeng	3 – 30 Jul 2016, 22 Dec 2016 – 16 Jan 2017	PM2.5	Industrial	4				30.2				BB + waste incineration: 5.1 SIA + coal: 22.5	(Liang <i>et al.</i> , 2019)
Lanzhou	Winter 2012,	PM2.5	Urban	7		33	13	8	MD: 12 - 13		Coal: 29 Power plants: 3		(J. Tan et al., 2017)
	summer 2013					15	10	25	MD: 12 - 13		Coal: 3 Power plants: 3		

2.6 Conclusions and future research questions

PM is a major air pollution problem in Southeast Asia region, which is exacerbated particularly during haze season. This article reviewed PM levels, their chemical components, and PM source apportionment results in Southeast Asia. The key features are summarized as follow:

- Southeast Asia region covers a vast area, with distinctions in climate and topography. Consequently, haze event in this region occurs during local dry season in different months and areas: during Feb – Apr in mainland continent (mostly in Myanmar, Thailand, and Vietnam) and during Jun – Sep in maritime continent (mostly in Malaysia, Singapore, and Indonesia). Long term studies show that $PM_{2.5}$ in most Southeast Asian counties exceeded the 24-hour mean guideline value by WHO ($15 \mu g m^{-3}$) but lower than of megacities in China and India. PM levels during haze episode were more than 2 times higher than non-haze or wet season.
- The most abundant component in $PM_{2.5}$ is organic matter. Contributions of OC in Southeast Asia region were higher than of megacities in China, India, and other European countries, supporting an important role of BB over this region. EC contributed less than 10%, which is not significantly different to other megacities. Sulfate, nitrate, and ammonium ions are the dominant components in water soluble ions with descending order $SO_4^{2-} > NO_3^- > NH_4^+$. Their contributions to $PM_{2.5}$ are lower in Southeast Asia than those of the other megacities, which may be attributed to the very high organic matter contributions derived from BB during haze seasons. NO_3^-/SO_4^{2-} ratios in Southeast Asia were notably lower than 0.5, which is mostly lower than in megacities in China and India and European countries.
- Hundreds of studies conducted PM measurements and their chemical compositions across this region but some of them have performed PM source apportionment. From

these studies, 7 main sources are classified by receptor models including biomass burning (BB), secondary aerosols (SA), industrial emissions (IND), road traffic emission (RT), soil dust (SD) and road dust (RD), sea salt (SS), and fossil fuels (FF). Among these seven sources, BB is the most significant source in Southeast Asia identified by OC together with other BB tracers (K^+ and levoglucosan) especially during haze seasons. The 2nd largest source in Southeast Asia is road traffic especially in urban environment like megacities (such as Hanoi, Bangkok, Kuala Lumpur) or coastal area where is influenced by ship emission (such as Singapore). Secondary aerosol is also an important source identified by SNA ions especially SO_4^{2-} which significantly contributes to PM during haze event over Southeast Asia region. Natural sea salt is identified in Southeast Asian countries that are located near the sea/ocean and strongly related to monsoon. Some sources are difficult to identify due to the lack of common tracers, for example, industry. Such sources are often identified as mixed with other sources such as soil or road dust. Due to a lack of organic tracers, fossil fuel combustion sources are also difficult to be identified/quantified in Southeast Asia.

- Our comprehensive findings have provided benefits for authorities and policy makers in mitigation PM pollution from local to regional scales. The major sources of PM can be prioritized to make a significant emission reduction, for example, road traffic emissions and road/soil dusts can be mitigated to improve air quality of local areas, biomass burnings as the main source for regional area have to be harmonizely mitigated across the region due to its transboundary movements.

The studies that were reviewed in this paper show some degree of confidence to conclude the key features of PM over Southeast Asia region. However, there are several major constrains which should be taken into account for improvement for future studies:

- Most studies in Southeast Asia region were done only in short-term experiments especially during smoke-haze seasons. Additional information to represent long-term trends is needed to fill this gap.
- Few studies provided a more complete set of chemical components, i.e., including OC/EC, WSIs, and crustal/trace metal elements. This sometimes leads to a poor mass closure estimation and provides insufficient data for receptor modelling especially PMF. Moreover, very few studies have done measurement for organic tracers (e.g., PAHs, n-alkanes, levoglucosan, etc.) which are very important constituents for classifying fossil fuel emissions, vehicle emissions, biomass burning as well as secondary organic aerosols.
- PMF is the most popular receptor model for source apportionment, but it requires user's expertise for source identification. Users may find unusual combination of chemical components in classified factors and forced to identify a source. This could result in ambiguous sources as some studies reported mixed or unexplained sources.
- There is a lack of studies in some countries of Southeast Asia region such as Myanmar, Laos, and Cambodia. Studies over the Northern Southeast Asia pointed out that BB usually occurs in the vast area covering these three countries during smoke-haze events. Therefore, it is recommended that this needs to be filled by additional information such as remote sensing data.
- According to Hopke, (2015), PCA is not a really receptor model since it apportions of differences from mean value instead of actual value, therefore, it does not provide source apportionment from pollutants. Since some of studies that are reviewed here have used PCA, it is recommended for future research to adopt PMF, CMB and/or hybrid receptor models to generate robust results.

- Future studies should aim to use multiple receptor models, with both online and offline aerosol data to provide a more reliable source apportionment and a better understanding of uncertainties (Beddows et al., 2015; J. Xu et al., 2021).

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CHAPTER THREE:

METHODOLOGY

3.1 Sampling site

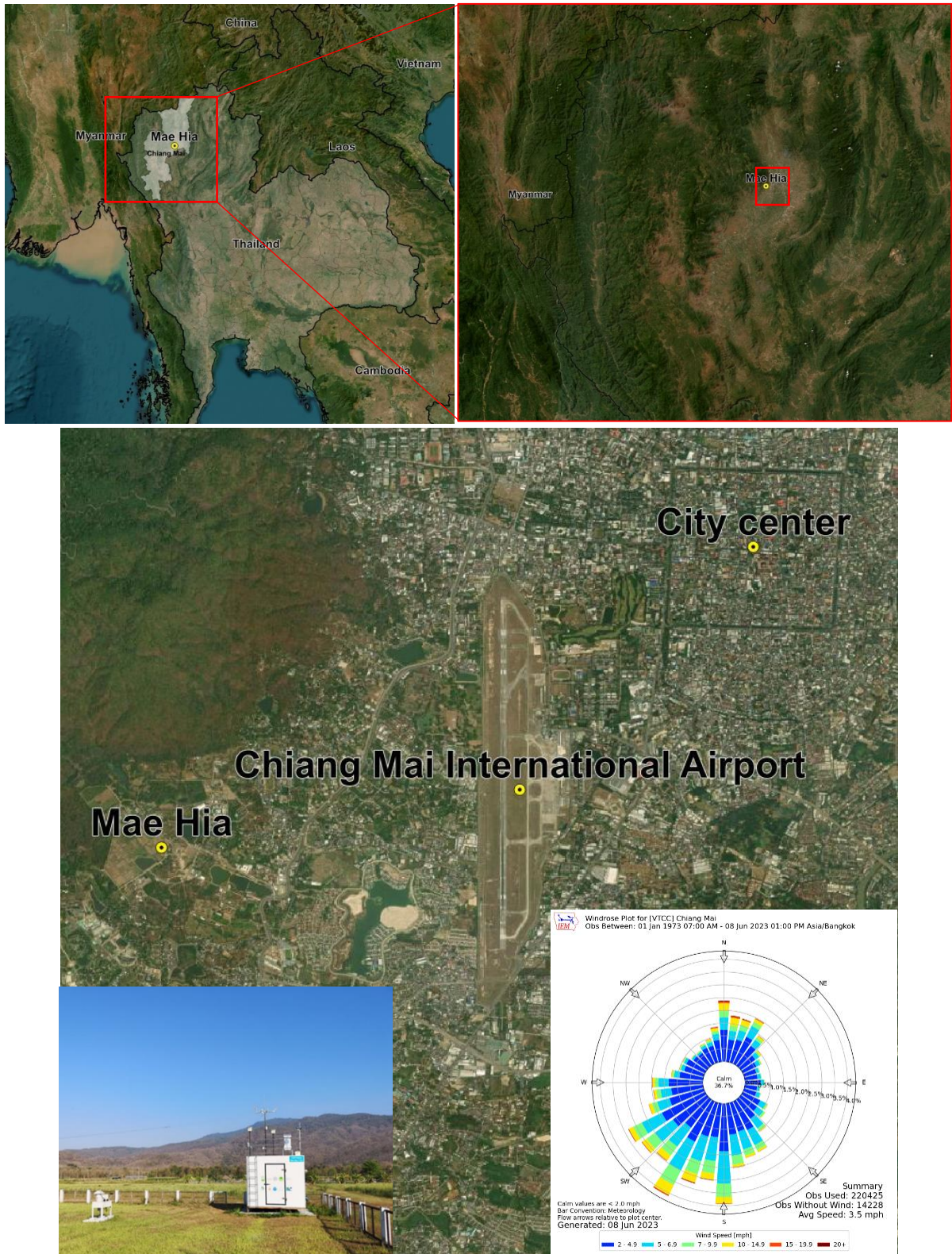


Fig. 3-1 Sampling site location, picture of station on the field and prevailing wind rose diagram averaged from 1973 to 2023 (<https://mesonet.agron.iastate.edu/>)

The sampling site was at Mae Hia Agricultural Meteorological field (18°45'40.93"N, 98°55'54.66"E, 341 meters above sea level) (Fig. 3-1), suburb of the city of Chiang Mai, northern Thailand, situated on agricultural areas adjacent to mountain ranges to the west, approximately 6.5 km away from Chiang Mai city center to the northeast direction and 3 km away from Chiang Mai International airport to the east. On a regional scale, the study site is close to Myanmar from the southwest to the north approximately 110 – 160 km from the sampling site to the borderline between Thailand and Myanmar. Prevailing wind arrives Chiang Mai in two main directions: south to west during February and September and north to northeast during October and January. During the sampling period (17 Feb – 10 Jun) (Fig. 3-1). Trust, there was no major influence from the airport due to prevailing winds. The location and site's environment are representative of rural areas where is rarely affected by vehicles and local residents, providing a clear characteristic of aged air pollutants through long-range transport during haze episode. The city of Chiang Mai is located in a valley topographic area, aligning from north-south direction surrounded by mountain ranges. This characteristic of the topography is of interest to us since air pollutants could be trapped and accumulated within the study area during haze episode. Moreover, Chiang Mai is the largest city of north Thailand and has a population of 1.73 million in 20,107 km² area. 80% of the total area is mountain where mostly covered by deciduous forest. The sampler was installed around 4 meters above the ground level on the rooftop of the station in order to avoid effects from soil/dust resuspension.

3.2 Quartz fiber filter preparation

All quartz fiber filters were pre-baked in oven at 500 °C for at least 8 hours to get rid of organic compound contaminated on the filter, after bake filters were kept in pre-baked aluminum foil prior for sampling.

3.3 PM_{2.5} Sampling collection

PM_{2.5} was collected from ambient air on quartz fiber filters (8" x 10", Whatman) using high volume sampler (SIBATA high volume air sampler) at flow rate 1,000 liters/min for 24 hours. The sampling was carried out every day at 9AM between 17th February - 19th April, and 11th May – 10th June 2019 for haze and post-haze episodes, respectively. Prior to sampling, the filters were baked at 500°C for 8 hours to remove contaminated organic substances, then transferred to desiccator to remove moisture for 24 hours before being pre-weighed. After sampling, the collected PM_{2.5} on quartz fiber filters were then kept in aluminum foils and transferred into desiccator for 24 hours and post-weighed under controlled temperature and humidity conditions to determine PM_{2.5} mass concentration. Finally, the sampled filters were stored in a freezer at -20°C until chemical analysis. In order to validate the sampling method, we performed a comparison of PM_{2.5} sampling alongside with the high-volume sampler, by collecting PM_{2.5} on PTFE filters using mini volume sampler (flow rate at 5 liters/min) which was installed next to the high-volume sampler. The total number of samples collected were 93; 62 and 31 for haze and post-haze episodes, respectively. Daily PM_{2.5} concentration higher and lower than 100 µg m⁻³ will be identified as episodic haze event (EH) and low haze (LH), respectively.

3.4 Bulk chemical analysis

3.4.1 PM_{2.5} mass gravimetric measurement

In addition to measuring PM_{2.5} mass, we performed a comparison of PM_{2.5} mass between collecting using high volume (HV) sampler (the main sampling method used in this study) and mini volume sampler (Minivol). The two samplers were installed approximately three meters away from each other at the same altitude on the site's roof. Fig. 3-2 (left and right) displays scatter plots of PM_{2.5} mass collected by HV and Minivol at Mae Hia site, and between collected by HV at

Mae Hia site and Chiang Mai City center by PCD (<http://air4thai.pcd.go.th>), respectively. A strong correlation coefficient ($R^2 = 0.99$) and a slope of 0.71 were found for the measured $PM_{2.5}$ mass from HV vs. Minivol at Mae Hia site (Fig. 3-1 (left)), displaying that $PM_{2.5}$ mass collected from Minivol was much lower than that collected by HV ($t_s > t_c$, 95% confidence interval). This is due to the difference in adsorption capability in Teflon filters used in Minivol and quartz fiber filter used in HV. Quartz fiber filter acts as an adsorbent for volatilized particle organics, gaseous organics, and ambient particle organics, and retain them better than the Teflon filters (Parshintsev et al., 2011; Turpin et al., 1994). Volatile organics and semi-volatile organic compounds are likely to loss from the Teflon filter due to high temperature during pre-monsoon season, in other word, or during dry season when which haze episodes occur (Bhardwaj & Sunder Raman, 2022). The scatter plot between HV at Mae Hia and Chiang Mai city center also shows a strong correlation ($R^2=0.98$) (Fig. 3-1 (right)) with a slightly higher $PM_{2.5}$ mass in the city center (slope = 1.04), which may be attributed to the additional $PM_{2.5}$ mass from vehicular and residential emissions.

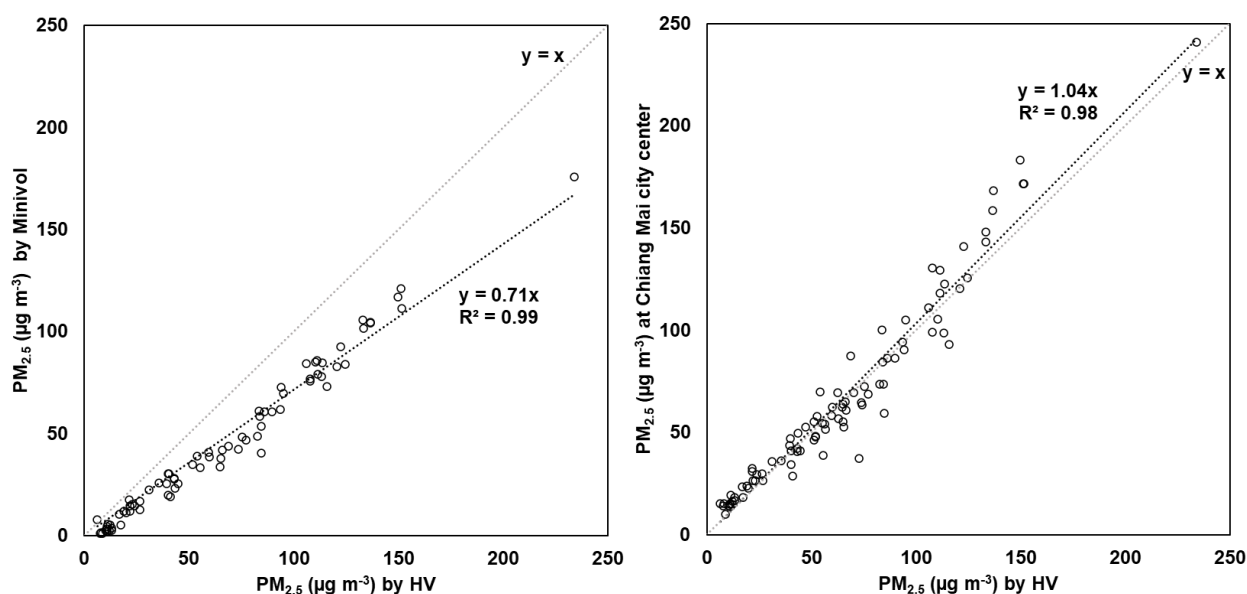


Fig. 3-2 Scatter plots of measured $PM_{2.5}$ mass; collected by HV and Minivol at MH site (left), and between collected by HV at MH site and at Chiang Mai city center (right) (PCD site)

3.4.2 Organic carbon (OC) and Elemental carbon (EC)

Carbonaceous contents of PM_{2.5} were analyzed by thermal-optical analysis with DRI model 2015 OC/EC Carbon Analyzer employing IMPROVE_A protocol thermal-optical transmittance. This analysis uses of the thermal refractivity of EC which is not volatilized in an inert atmosphere (pure He) at temperature below 700 °C. EC can only be oxidized to gas phase by oxidation in an oxygenated atmosphere at temperature higher than 340 °C (Petzold et al., 2013) while OC can be volatilized in an inert atmosphere as temperature increases. The analysis therefore consists of two progressive heating steps as follow; i) inert atmosphere phase or Helium mode – this step a fraction of OC will be heated and volatilized in non-oxygenated condition made by pure He gas, and ii) oxidizing atmosphere or oxygenated mode – this step oxygen gas (O₂) (mixed of 98% He/ 2% O₂) is introduced to analytical system where EC is initially oxidized to carbon dioxide gas (CO₂). OC and EC evolve on the 0.5 cm² filter samples punch in inert atmosphere and oxidizing atmosphere, respectively. For thermally derived carbon fractions, IMPROVE_A defines temperature plateaus as follows: 140°C (OC1), 280°C (OC2), 480°C (OC3), and 580°C (OC4) in helium (He) mode; 580°C (EC1), 740°C (EC2), and 840°C (EC3) in oxygenated mode (mixture of 98% He and 2% O₂). Carbon fractions: OC, EC, and total carbon (TC) can be calculated as follows:

$$OC = OC1 + OC2 + OC3 + OC4 + POC$$

$$EC = EC1 + EC2 + EC3 - POC$$

$$TC = OC + EC$$

where POC is pyrolyzed organic carbon and is defined as the OC content measured at the beginning of oxygenated mode until laser transmittance returns to its original value at the start point of analysis.

The unit of OC and EC analyzed by DRI was $\mu\text{g Carbon cm}^{-2}$ which was then converted into $\mu\text{g m}^{-3}$ by following equation;

$$OC \text{ or } EC(\mu\text{g m}^{-3}) = \frac{C (\mu\text{g cm}^{-2}) \times \text{deposit area (cm}^2\text{)}}{\text{total sampling volume (m}^3\text{)}}$$

3.4.3 Water soluble inorganic ions (WSIIs)

Water soluble inorganic ions (WSIIs) including cations (ammonium (NH_4^+), potassium (K^+), magnesium (Mg^{2+}), and calcium (Ca^{2+})) and anions (chloride (Cl^-), sulphate (SO_4^{2-}), and nitrate (NO_3^-)) were analyzed using ion chromatography (IC) by Dionex Integrion HPIC and Dionex ICS 1100 for cation and anion, respectively. The extraction procedure is described as follows: the filter samples were punched into a polypropylene vial and extracted by 10 ml deionized water (DI) in sonication bath at 25°C for 60 minutes and shaken overnight. In order to remove any residues, the extracted samples were then filtered through 0.45µm pore size syringe filter into new vials. Then the clean extracted samples were kept in cold room at 4°C prior to analysis.

The unit of WSIIs analyzed was $\mu\text{g ml}^{-1}$ which was then converted into $\mu\text{g m}^{-3}$ by following equation;

$$\text{Ions } (\mu\text{g m}^{-3}) = \frac{\text{Ions } (\mu\text{g ml}^{-1}) \times 10 \text{ ml} \times \text{deposit area } (\text{cm}^2)}{\text{punch area } (\text{cm}^2) \times \text{total sampling volume } (\text{m}^3)}$$

3.4.4 Crustal and trace elements

Crustal elements and trace elements (total 22 elements including Mg, Al, Ca, Ti, Fe, V, Cr, Mn, Co, Ni, Cu, Zn, As, Rb, Sr, Cd, Sb, Ba, Ce, W, and Pb) were analyzed by Inductively Coupled Mass Spectrometry (ICP-MS). Filter samples extraction procedure is briefly described as follows: 9 cm² filter samples punches were emerged in 10 ml of 67% of ultra-pure nitric acid (HNO_3) in microwave digestion vessels. The vessels were put in Mars microwave digestion system (CEM) using filter-membrane program where temperature increases at ramp time of 15 minutes to reach the max temperature at 200°C and hold at this temperature for 15 minutes before cooling to 30°C. Once the acid digestion was completed, the digested concentrated acid samples were then diluted

16.67 times by deionized water in order to meet the required 2% HNO₃ concentration for analysis with ICP-MS. The 2% HNO₃ acid digested solution samples were then filtered using 0.45 µm pore size syringe filters into metals-free centrifuge tubes and kept in the fridge at 4°C prior to analysis.

The units of elements analyzed was ng ml⁻¹ which was then converted into ng m⁻³ by following equation;

$$\text{Element (ng m}^{-3}\text{)} = \frac{\text{Element (ng ml}^{-1}\text{)} \times 16.67 \text{ (dilution factor)} \times 10 \text{ ml} \times \text{deposit area (cm}^2\text{)}}{\text{punch area 9 (cm}^2\text{)} \times \text{total sampling volume (m}^3\text{)}}$$

3.4.5 Organic compounds

33 polar and 33 nonpolar organic compounds were extracted from collected PM_{2.5} on quartz fiber filter. Extraction procedure performed as follow: 10.5 cm² punched filter was put in glass vial then was spiked with a known amount of a mixture of surrogate standards (full list in Table 3-5 and 3-6) 20 µL of 1 ng µL⁻¹. Filter sample was extracted with 7 mL of methanol and acetonitrile mixture (ACN:MeOH, 1:1, v:v) in a sonication bath at 25 °C for 20 min, followed by filtration using 0.45 µm pore size filter to remove filter fiber residuals. 5.5 ml of filtered extract solution was then concentrated to dryness by a gentle nitrogen blowing and then dissolved with 100 µL of acetonitrile. The extract solution was then split into two vials for nonpolar and polar organic compounds. First half of final extract sample (50 µL) was transferred to a new vial and kept in the freezer prior to analysis of nonpolar organic compounds. Another half of final extract sample was also transferred to a new vial to perform derivatization by adding with 50 µL of N, O-bis(trimethylsilyl) trifluoroacetamide (BSTFA) with 1% trimethyl-chlorosilane (TMS) then heated at 60 °C for 30 min, the derivatized compound was then kept in the freezer. Finally, 5 µL of 10 ng L⁻¹ Tetradecane – d₃₂ was added into both polar and nonpolar extracts as internal standard prior to the analysis by GC/MS.

Analysis was performed by using GC/MS (Agilent 7890A GC coupled to 5975C MS) in electron ionisation mode (EI, 70 eV). 1 µl of extracts were inject in the splitless mode. Compounds were separated in a Restek Rxi-5MS capillary column (30 m × 0.25 mm × 0.25 µm). The temperature program in the analysis as follows: held at 70 °C for 1 min, then ramped to 260 °C by 5 °C/min, followed by ramped to 320 °C by 10 °C/min and then held for 10 minutes. The carrier gas was Helium (He) with flow rate 1.5 ml/min throughout the analysis and the transfer line was heated at 300 °C. The temperature of Mass Spectro quad, Mass Spectro source, and inlet were 150, 230, and 300 °C, respectively. The analysis of both nonpolar and polar organic compounds was performed in Selected Ion Monitoring (SIM) mode. The details of SIM mode configurations including masses of ions monitored, retention time, dwelling time, and acquisition windows are listed in Table 3-1 and 3-2 for nonpolar and polar organic compounds, respectively.

By employing internal calibration, each organic compounds were calculated by the ratio of analyte's response (A_a) to surrogate standard's response (A_s) and the ratio of analyte concentration (C_a) to surrogate standard concentration (C_s) as equation;

$$C_a \text{ (pg ul}^{-1}\text{)} = \left(\left(\frac{A_a}{A_s} \right) - \text{intercept} \right) \times \left(\frac{C_s \text{ (pg ul}^{-1}\text{)}}{\text{slop}} \right)$$

Then the concentration is converted to unit of ng m⁻³ by equation;

$$\text{Compound (ng m}^{-3}\text{)} = \frac{C_a \text{ (pg ul}^{-1}\text{)} \times \text{final volume 105 (ul)} \times \text{dilution factor (2.54)} \times \text{deposit area (cm}^2\text{)}}{\text{punch area 10.5 (cm}^2\text{)} \times \text{total sampling volume (m}^3\text{)} \times 1000 \text{ (pg/ng)}}$$

Note that the final volume of nonpolar compounds was 55 ul.

It should be noted that this study performed analysis of organic compounds which were obtained from particulate phase only. However, organic compounds are present in the atmosphere in both particulate and gas phases to different degrees. They largely depend on meteorological

condition, typically temperature. Gas-phase compounds are found to be increasing in summertime or tropical region, while compounds in particulate phase are dominant during winter (Abdel-Shafy & Mansour, 2015; M. Li et al., 2020). Some studies perform measurement in both gas and particulate phases according to specific objectives, for example, to investigate gas/particle partitioning (M. Li et al., 2020) or to evaluate toxicity of PAHs (Kongpran et al., 2021). Due to constraints on field measurement in this study, it was not feasible to conduct the collection of gas-phase organic compounds, which may subsequently influence source attribution in some degree. Nevertheless, the combination information from organic compound in particle phase with an information from other essential components also provides a robust outcome.

Table 3-1 GC-MS analysis program configuration in SIM mode for nonpolar organic compounds

	Start time	Compound	Retention time	Sim ion(s)	Dwell time
Group 1	9.30	Naphthalene	10.67	128.0	50
Group 2	11.00	Tridecane-d28	12.45	50, 66, 82	50
Group 3	16.30	Acenaphthylene-d8	17.78	160.2	50
		Acenaphthylene	17.85	152.2	50
Group 4	20.00	C16	21.27	57, 226.4	50
		C17	23.98	57, 240.5	50
		C18	26.61	57, 254.5	50
Group 5	25.00	Phenanthrene	27.00	178.2	50
		Anthracene	27.29	178.2	50
		C19	29.13	57, 268.5	50
Group 6	28.70	Eicosane-d42	30.75	50, 66, 82	50
		C20	31.55	57, 282.6	50
		C21	33.87	57, 296.6	50
Group 7	32.50	Fluoranthene	33.91	202.2	50
Group 8	34.00	Pyrene-d10	35.09	212.3	100
		Pyrene	35.20	202.2	100
		C22	36.10	57, 310.6	50
		C23	38.23	57, 324.7	50
		C24	40.28	57, 338.7	50
Group 9	40.50	Pentacosane-d52	41.39	50, 66, 82	50
		C25	42.24	57, 352.7	50
		Benz(a)anthracene	42.32	228.3	50
		Chrysene	42.53	228.3	50
		C26	44.14	57, 366.8	50
		C27	45.97	57, 380.8	50
		C28	47.73	57, 71, 85	50
Group 10	47.20	Benz(b)fluoranthene	48.23	252.3	50
		Benz(k)fluoranthene	48.37	252.3	50
		C29	49.43	57, 71, 85	50
Group 11	49.05	Benz(a)pyrene	50.30	252.3	50
		C30	51.07	57, 71, 85	50
		C31	52.65	57, 71, 85	50
		C32	54.20	57, 71, 85	50
Group 12	53.00	Indeno(1,2,3-cd)pyrene	55.00	276.3	50
		Dibenz(a,h)anthracene	55.18	278.3	50
		C33	55.79	57, 71, 85	50
Group 13	56.00	Benz(ghi)perylene	56.22	276.3	50
		C34	57.66	57, 479	50

Table 3-2 GC-MS analysis program configuration in SIM mode for polar organic compounds

	Start time	Compound	Retention time	Sim ion(s)	Dwell time
Group 1	8.00	Succinic acid-d4	14.37	251	50
		Succinic acid	14.44	247	50
		Nitrophenol-d4	16.81	155, 200	50
Group 2	18.00	Malic acid	18.72	233, 245	50
		D-threitol	18.78	103, 217, 307	50
		Meso-erythritol	18.96	189, 205, 217	50
		DHOPA	19.27	189, 218, 349	50
		Adipic acid	19.35	172, 185, 204, 217	50
Group 3	19.50	(1S)-(+)-Ketopinic acid	19.62	137, 226, 239	50
		4-Nitrophenol	19.69	196, 211	50
		cis-Pinonic acid	20.05	117, 125, 171	50
Group 4	21.30	Pimelic acid	21.69	125, 155, 289	50
		2-Methyl-4-nitrophenol	21.69	210, 225	50
Group 5	22.50	Galactosan	22.8	204, 217	50
		Mannosan	23.29	204, 217, 333	50
Group 6	23.00	Levogluconan	23.74	204, 217, 333	50
		Phthalic acid-d4	23.67	299	50
		Phthalic acid	23.71	221, 295, 310	50
		Suberic acid	23.81	129, 187, 303	50
Group 7	24.40	Syringaldehyde	24.15	224, 239, 254	50
		4-Nitrocatechol	24.89	284, 299	50
		Isophthalic acid	25.24	295, 310, 221	50
Group 8	25.50	Azelic acid	25.94	129, 201, 317	50
		1-Pentadecanol	27.04	117, 129, 285	50
Group 9	28.50	Glutaric acid	29.01	117, 145, 299, 314	50
		Palmitic acid	30.9	117, 129, 145, 313	50
Group 10	31.20	Hemimellictic acid	31.97	147, 249, 411	50
		Trimelitic acid	32.72	147, 249, 411	50
		Trimesic acid	33.68	147, 249, 411	50
Group 11	33.80	Linoleic acid	33.91	262, 337	50
		Oleic acid	33.99	117, 264, 339	50
		Bisphenol A	34.09	341, 357, 372	50
		Stearic acid	34.44	117, 132, 341, 356	50
Group 12	34.70	Pimaric acid	35.72	121, 359, 374	50
		Isopimaric acid	36.33	241, 256, 374	50
		Dehydroabietic acid	37.12	239, 357, 372	50
Group 13	40.00	Cholesterol	44.81	129, 329, 458	50

3.4.6 Quality assurance and quality control (QA/QC) for bulk chemical analysis

For the part of QA/QC, all chemical compositions from bulk analysis were corrected by field blanks and lab/reagent blanks that were treated through the same procedures to the filter samples. Contaminants in field blanks are also listed in table 3-3, 3-4, 3-5, and 3-6 for WSIs, elements, nonpolar and polar organic compound, respectively. The contaminants were calculated by an average of 10 field blanks ($n = 10$). Details on the QA/QC for bulk chemical analysis of each component are described as follows:

- The analysis of OC/EC was reassured by using known concentration sucrose every 10 analyses to monitor the instrument's stability and was daily performed system baking and auto calibration by methane gas to clean and optimize the instrument's system. The minimum detection limits (MDLs) of OC/EC are $0.43 \mu\text{g cm}^{-2}$, $0.12 \mu\text{g cm}^{-2}$, and $0.49 \mu\text{g cm}^{-2}$, for OC, EC, and TC, respectively. Contaminants of OC/EC in field blanks were 0.30 ± 0.07 and $0.03 \pm 0.05 \mu\text{g m}^{-1}$ for OC and EC, respectively.
- To determine the recovery (%) of water extraction of WSIs analysis, the known concentration of cation and anion standards mixture were spiked on pre-baked filters and extracted in a similar procedure described in 3.4.3. Overall, more than 80% WSIs recoveries obtained. The limit of detection (LOD) for WSIs was calculated by 3 times the SD of replicated deionized water blank. The recoveries, contamination in field blanks and LOD of WSIs are shown in Table 3-3.
- The recovery (%) of metallic elements from acid extraction was determined by a standard reference material (NIST1649a) obtained from the National Institute of Standard and Technology (NIST), USA. SRM was treated through acid extraction by the same procedure as the filter samples as described in 3.4.4. The LOD of acid extraction and analysis by ICP-MS was calculated by 3 times the SD of replications of 2% HNO_3

blank. The recoveries, contamination in field blanks and LOD of crustal and trace elements are shown in Table 3-4.

- Organic tracers were reassured by using known concentration of the standards spiked 20 µl on pre-baked blank filter and performed through the same extraction procedure as described in 3.4.5 for both nonpolar and polar compounds to quantify percentage recovery (known concentration of spiked standards were listed in Table 3-5 and 3-6 for nonpolar and polar organic compounds, respectively). In addition, standard reference material, urban dust diameter size below 65 µm (NIST 1649b) was also used to quantify and compare to those certified concentration reported by NIST and previous studies. NIST 1649b was weighed at 50 mg and extracted through the same extraction procedure as described in 3.4.5 (3 and 6 replications for nonpolar and polar compounds, respectively). The recoveries, certified and reported concentrations in literatures and concentration obtained from this study are listed in Table 3-5 and 3-6 for nonpolar and polar organic compounds, respectively. Note that there has been no certified concentration for target polar organic compounds in NIST 1649b.

Table 3-3 Recoveries, and contaminants in field blank of water-soluble inorganic ions

	Recoveries (%)	±SD	Field blank contamination ($\mu\text{g m}^{-3}$)	±SD	LOD ($\mu\text{g m}^{-3}$)
Anions					
Cl^-	99.5	6.0	0.17	0.01	0.10
NO_3^-	90.0	1.2	0.17	0.02	0.34
SO_4^{2-}	83.9	2.1	0.27	0.07	0.27
Cations					
NH_4^+	96.7	4.6	0.12	0.05	0.03
K^+	89.4	5.4	0.17	0.01	0.07
Mg^{2+}	98.6	2.4	0.21	0.01	0.08
Ca^{2+}	92.6	3.9	0.39	0.17	0.01

Table 3-4 Recoveries, and contaminants in field blank of crustal and trace elements

Elements	Recoveries (%)	±SD	Field blank contamination (ng m^{-3})	±SD	LOD (ng m^{-3})
Mg	77.3	0.02	64.9	14.6	12.4
Al	43.1	0.03	73.3	48.3	35.0
Ti	41.1	0.03	1.9	0.9	1.1
V	78.7	0.03	0.1	0.1	0.1
Cr	27.6	0.02	2.6	1.4	0.2
Mn	92.6	0.04	0.8	0.4	0.7
Fe	81.4	0.02	29.5	12.2	4.3
Co	89.2	0.16	0.1	0.0	0.3
Ni	85.8	0.08	0.7	0.4	0.1
Cu	82.8	0.06	10.7	27.1	0.2
Zn	123.8	0.51	35.3	24.0	3.5
As	94.1	0.05	0.1	0.2	0.3
Rb	38.7	0.02	0.1	0.1	0.0
Sr	60.9	0.06	0.8	0.7	1.1
Sb	66.4	0.04	0.1	0.1	0.1
Ce	52.5	0.08	0.3	0.4	0.1
Pb	95.2	0.02	0.9	0.7	0.1

Table 3-5 Surrogate standards used, relative response factors, standard spiked concentration, spiked recoveries, limit of detection, field blank contaminations and the concentration of NIST 1649b (mg kg⁻¹) obtained from this study, certified values, and literatures for nonpolar organic compounds

Surrogate standards used	Compounds	Relative response factor	Spiked concentration (ng µl ⁻¹)	Recovery (%)	±SD	LOD (ng m ⁻³)	Field blank contamination (n = 10) (ng m ⁻³)	±SD	NIST SRM 1648b (mg kg ⁻¹)					
									This study (n = 3)	±SD	Certified value	(Viteri et al., 2021)	(Li et al., 2010)	(Clark et al., 2015)
Tridecane-d28	C16	5.11	1.73	127.3	14.5	0.19	ND	-	ND	-	-	-	-	-
	C17	3.90	1.73	115.3	19.2	0.19	ND	-	ND	-	-	-	-	-
Eicosane-d42	C18	4.88	1.73	82.9	4.9	0.07	ND	-	ND	-	-	-	-	-
	C19	5.01	1.73	82.7	4.8	0.09	ND	-	ND	-	-	-	-	-
	C20	5.28	1.73	81.4	4.6	0.23	0.60	0.01	1.1	0.1	1.9	-	-	-
	C21	4.68	1.73	90.8	5.3	0.21	0.71	0.01	ND	-	-	-	-	-
	C22	4.72	1.73	92.6	6.0	0.23	1.18	0.01	2.7	0.3	5.2	-	-	-
	C23	4.59	1.73	96.2	9.1	0.21	1.77	0.02	8.1	0.3	16.0	-	-	-
Pentacosane-d52	C24	4.80	1.73	114.0	7.8	0.21	1.37	0.02	10.1	0.4	-	-	-	9.5
	C25	5.00	1.73	104.3	10.0	0.20	1.70	0.03	29.7	0.4	-	-	-	68.0
	C26	3.98	1.73	87.2	10.9	0.24	2.52	0.05	37.5	0.4	-	-	-	67.0
	C27	3.03	1.73	68.7	11.3	0.25	2.33	0.03	32.4	1.3	-	-	-	65.0
	C28	2.91	1.73	58.6	11.0	0.34	3.22	0.08	18.3	0.5	-	-	-	39.0
	C29	1.86	1.73	60.3	11.1	0.45	1.54	0.01	21.7	2.6	-	-	-	76.0
	C30	1.57	1.73	57.7	6.3	0.84	2.27	0.01	6.8	0.8	-	-	-	24.0
	C31	0.97	1.73	51.7	6.3	0.90	3.07	0.04	7.0	1.1	-	-	-	46.0
	C32	0.31	1.73	65.7	4.1	2.58	ND	-	4.4	0.4	-	-	-	32.0
	C33	0.22	1.73	77.7	2.7	3.19	ND	-	ND	-	-	-	-	-
	C34	0.12	1.73	81.8	2.2	3.32	ND	-	ND	-	-	-	-	-
Acenaphthylene-d8	Naphthalene	1.82	1.64	61.6	9.8	0.06	ND	-	0.2	0.0	-	1.2	-	-
	Acenaphthylene	2.13	1.64	81.2	1.6	0.06	ND	-	0.2	0.0	-	0.3	0.4	-
	Phenanthrene	2.83	1.64	118.4	8.6	0.02	ND	-	1.3	0.1	-	3.7	5.5	-
	Anthracene	1.94	1.64	115.7	8.3	0.02	ND	-	0.4	0.0	-	0.5	0.8	-
Pyrene-d10	Fluoranthene	3.41	1.64	74.4	0.5	0.45	ND	-	3.4	0.0	6.2	3.9	6.5	-
	Pyrene	3.55	1.64	75.2	0.8	0.39	ND	-	2.7	0.0	5.0	6.1	5.4	-
	Benz(a)anthracene	2.67	1.64	80.0	3.9	0.56	ND	-	1.4	0.1	2.1	1.5	2.5	-
	Chrysene	1.95	1.64	84.1	3.5	0.64	ND	-	4.6	0.0	3.0	2.6	4.9	-
	Benz(b)fluoranthene	2.59	1.64	91.9	7.4	0.09	ND	-	4.3	0.2	6.2	4.4	6.6	-
	Benz(k)fluoranthene	2.61	1.64	85.5	4.9	0.06	ND	-	2.1	0.1	1.7	1.1	2.3	-
	Benz(a)pyrene	1.46	1.64	89.5	7.5	0.07	ND	-	1.2	0.1	2.5	2.5	2.7	-
	Indeno(1,2,3-cd)pyrene	2.21	1.64	101.5	10.7	0.11	ND	-	2.0	0.3	2.9	2.5	3.6	-
	Dibenz(a,h)anthracene	1.78	1.64	103.4	10.3	0.13	ND	-	0.3	0.2	0.3	0.4	0.4	-
	Benz(ghi)perylene	1.05	1.64	98.4	10.6	0.76	ND	-	5.2	0.8	0.9	5.7	5.4	-

ND – not detected

Table 3-6 Surrogate standards used, relative response factors, standard spiked concentration, spiked recoveries, limit of detection, field blank contaminations and the concentration of NIST 1649b (mg kg⁻¹) obtained from this study and literatures for polar organic compounds

Surrogate standards used	Compounds	Relative response factor	Spiked concentration (ng µl ⁻¹)	Recovery (%)	±SD	LOD (ng m ⁻³)	Field blank contamination (n = 10) (ng m ⁻³)	±SD	NIST SRM 1648b (mg kg ⁻¹)					
									This study (n = 6)	±SD	(Albinet et al., 2019)	(Lanzafame et al., 2021)	(Louchouart et al., 2009)	(Gao et al., 2018)
Succinic acid-d4	Succinic acid	1.51	0.96	69.0	3.1	0.30	ND	ND	103.3	5.8	111.0	115.1	-	-
	Malic acid	2.36	0.90	143.7	9.3	0.24	ND	ND	0.9	0.4	-	-	-	-
	D-threitol	2.96	0.90	51.2	5.5	0.23	ND	ND	0.9	0.1	-	-	-	-
	Meso-erythritol	0.45	0.90	60.8	5.6	0.24	ND	ND	12.7	1.0	-	-	-	-
	DHOPA	1.35	0.91	102.9	2.6	0.24	ND	ND	0.2	0.0	1.2	1.8	-	-
	Adipic acid	0.77	0.98	106.7	13.6	0.25	ND	ND	15.8	0.9	-	-	-	-
Nitrophenol-d4	4-Nitrocatechol	0.64	0.91	76.2	16.3	0.28	ND	ND	2.8	0.4	-	-	-	-
	Bisphenol A	6.43	0.89	101.4	21.6	0.23	0.17	0.01	1.3	0.3	-	-	-	-
	4-Nitrophenol	1.30	0.90	91.9	2.7	0.22	ND	ND	9.8	0.8	-	85.4	-	-
	2-Methyl-4-nitrophenol	0.87	0.90	98.8	5.2	0.24	ND	ND	4.0	0.4	-	-	-	-
(1S)-(+)-Ketopinic acid	cis-Pinonic acid	1.05	0.98	78.3	4.2	0.24	ND	ND	0.8	0.0	5.9	5.9	-	-
	Pimelic acid	1.12	0.98	82.6	10.5	0.22	ND	ND	14.1	0.3	-	-	-	-
	Galactosan	5.77	0.90	51.9	15.2	0.21	ND	ND	2.3	0.1	-	-	4.8	-
	Mannosan	2.40	0.90	53.4	5.1	0.24	ND	ND	5.8	0.4	-	-	16.7	-
	Levoglucozan	8.05	0.97	110.3	48.9	0.77	0.58	0.16	59.7	4.0	-	-	160.5	165.0
	Suberic acid	0.80	0.97	70.7	12.8	0.24	ND	ND	39.2	1.2	-	-	-	-
	Glutaric acid	2.72	0.98	87.1	30.0	0.18	0.27	0.11	14.1	1.8	-	-	-	-
	Hemimellictic acid	2.53	0.98	66.2	4.7	0.23	ND	ND	9.0	1.2	-	-	-	-
	Trimelitic acid	3.90	0.96	86.6	5.8	0.24	ND	ND	13.8	1.6	-	-	-	-
	Trimesic acid	11.38	0.98	87.1	8.4	0.24	ND	ND	0.3	0.1	-	-	-	-
	Linoleic acid	1.11	0.94	100.6	30.2	0.19	0.88	0.42	ND	-	-	-	-	-
	Dehydroabietic acid	10.38	0.91	133.9	21.3	0.33	1.24	0.36	8.4	1.6	-	-	-	1.7
	Pimaric acid	1.40	0.93	168.8	21.6	0.32	ND	ND	20.7	4.7	-	-	-	-
	Isopimaric acid	2.04	0.98	112.1	25.6	0.13	ND	ND	0.9	1.4	-	-	-	-
Phthalic acid-d4	Phthalic acid	-	0.97	80.7	2.3	0.23	ND	ND	64.0	2.0	89.7	129.9	-	-
	Isophthalic acid	-	0.98	105.2	7.1	0.23	ND	ND	10.0	0.3	-	-	-	-
	Azeliac acid	-	0.97	94.1	13.4	0.21	ND	ND	107.6	4.0	-	-	-	-
	Syringaldehyde	-	0.91	100.4	50.2	0.30	ND	ND	ND	-	-	-	-	-
External calibration	Cholesterol	-	0.98	89.8	18.8	0.47	0.48	0.05	0.2	0.2	-	-	-	-
	Palmitic acid	-	0.98	115.2	36.9	0.35	0.86	1.71	437.8	244.3	-	-	-	-
	Oleic acid	-	0.94	61.2	30.5	0.32	0.44	0.14	45.8	25.1	-	-	-	-
	Stearic acid	-	0.89	70.3	50.4	0.34	1.83	3.19	235.9	129.7	-	-	-	-
	1-Pentadecanol	-	0.98	65.9	23.9	0.27	0.62	0.00	ND	-	-	-	-	-

ND – not detected

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CHAPTER FOUR:

**CHEMICAL CHARACTERIZATION AND
SOURCE APPORTIONMENT OF PM_{2.5} DURING
HAZE AND POST-HAZE PERIODS IN
NORTHERN THAILAND**

Chemical characterization and source apportionment of PM_{2.5} during haze and post-haze periods in Northern Thailand

4.1 Abstract

Northern Thailand has been facing recurring haze events yearly, causing adverse impacts on health and environments. The sources of the haze particles remain poorly quantified, particularly due to the lack of data on organic tracers. This study first determined the chemical composition of PM_{2.5}, including both inorganic and organic compounds during both haze (17 February and 19 April) and post-haze (11 May and 10 June 2019) periods at a rural site of Chiang Mai, Northern Thailand, and then applied positive matrix factorization (PMF) to apportion their sources. Average PM_{2.5} concentrations during haze ($87 \pm 36 \mu\text{g m}^{-3}$) was more than three times higher than post-haze ($21 \pm 11 \mu\text{g m}^{-3}$). OM was the largest contributor in PM_{2.5} mass followed by water soluble inorganic ions (WSIIs) and mineral dust during both haze and non-haze periods. The availability of data on Levoglucosan, K⁺, Dehydroabietic acid (DHAA), Cl⁻ and 4-Nitrocathechol (4-NC) enabled us to identify three different biomass burning sources, including biomass burning, nearby biomass burning (fresh aerosol) and softwood burning (aged aerosol). Other sources include secondary sulfate, mixed dusts and traffic and nitrate. Source apportionment results show that combined total biomass burning contributed at 63% of PM_{2.5} mass during haze period, while nearby biomass burning (32%) contributed comparably to secondary sulfate (34%) and mixed dust (26%) during post-haze period.

4.2 Introduction

Among several pollutants emitted in the smoke plume, airborne particulate matter with aerodynamic diameter less than 2.5 micron ($PM_{2.5}$) is one of the most important pollutant owing to its high abundant content in size distribution especially from biomass burning (Dattamudi et al., 2020; Nicolaou et al., 2021), its ability to contain higher mutagenic components compared to larger particles ($>PM_{2.5}$) (Mudipalli, 2015) and directly penetrate into the lower respiratory system which consequently cause inevitable adverse impacts on human health such chronic respiratory, lung cancer, cardiovascular disease (A. Hassan et al., 2017; Nicolaou et al., 2021; Uda et al., 2019) and premature mortality (G. He et al., 2020; Shi, Zhao, et al., 2018; Uda et al., 2019).

Haze events has regularly taken place in Southeast Asia region, which is mostly from biomass burning, it is found that Indonesia and Myanmar had the highest number of annual fires, and recurrent fires every year with the highest in Laos (95.8%), Cambodia (78.7%), Thailand (75.0%) and Myanmar (75.1%). The highest percentage of fires in cropland from rice, maize and sugarcane agricultural waste burning was found in Thailand (51.7%), followed by Timor Leste (48.7%) and Philippines (45.0%) (Vadrevu et al., 2019). Haze in Thailand is one of the most environmental issues that has been extensively paid attention over the last decades which largely derived from biomass burning and traffic emission depending on the region of the country (Suriyawong et al., 2023). Forest fire dominated over the Northern region (73%) while agricultural waste burning predominated in the Central region (52%) of Thailand (Punsompong et al., 2021). Northern Thailand is one of the region facing reoccurrence of haze yearly during the local dry season between January to April (Saksakulkrai et al., 2023; Suriyawong et al., 2023; Van et al., 2022). In Chiang Mai, a province in Northern Thailand, forest burning and agricultural waste burning accounted for 71-88% and 12-29% of total burned area, respectively, (Sillapapiromsuk et al., 2013). Chiang Mai, is generally known as a capital city of Northern Thailand, second largest

city in Thailand with population of 1.8 million that attracts 7 million tourists per year (Pardthaisong et al., 2018), located on a flat plain surrounded by mountain ranges called “Chiang Mai basin”. Such bowl-shaped valley topographical feature is known as episode-prone coincides with a certain specific meteorological conditions, temperature inversion and stagnant winds, favoring air pollutants can easily be trapped and accumulated inside the area due to restricted dispersion (Kim Oanh & Leelasakultum, 2011). Moreover, bigger scale climate condition, El Nino-Southern Oscillation (ENSO), also plays a significant role on the weather of Asia Pacific countries, as El Nino phenomenon diminishes humidity input from Pacific Ocean to the area, promoting dry and droughts conditions which indirectly results in more occurrence and intensified wild fire while La Nina phenomenon causes opposite consequences (Dattamudi et al., 2020; Marlier et al., 2013). It is clearly seen that open biomass burning is the most contributor to haze issue in Thailand especially the Northern region, to control such problem, Thai government has then implemented an initiative regulation over 9 provinces over Northern region of Thailand called “zero-burning” policy in year 2013 to reduce every type of open burning during a period of 60 days in dry season (February to April). The zero-burning policy seemed to be successfully accomplished, since a recent study showed that open burning decreased after the policy implemented, however, it contributed in prolonged haze period from 2 months (mid-February to mid-April) to 3 months (mid-February to mid-May) (Yabueng et al., 2020).

Chemical compositions of $PM_{2.5}$ during haze episode in Chiang Mai has been investigated in several previous studies (ChooChuay et al., 2022; Chuang et al., 2013; Khamkaew, Chantara, & Wiriya, 2016; Khamkaew, Chantara, Janta, et al., 2016b; Pani et al., 2018, 2019b; Pongpiachan et al., 2017a; Tao et al., 2020; Thepnuan et al., 2019a, 2019b) some studies have performed PM mass closure (Chuang et al., 2016; Pani et al., 2019b) and source apportionment of PM by using a sole set of water soluble inorganic ions (WSIIs) (Chansuebsri et al., 2022), crustal and metallic elements (Kayee et al., 2020; Kraisitnitikul et al., 2024), organic compounds such as polycyclic

aromatic hydrocarbons (PAHs) (Wiriya et al., 2013b), or WSIs coupled with carbonaceous species organic carbon (OC) and elemental carbon (EC) (Pongpiachan, Wang, Apiratikul, et al., 2022) and other methods (Kawichai, Prapamontol, et al., 2022; Punsompong & Chantara, 2018), none of these have performed source apportionment by employing complete set of bulk chemical analysis (combination of OC/EC, WSIs, crustal and metallic elements) except a study conducted in a neighboring province, Chiang Rai (N. D. Luong et al., 2022). To the best of our knowledge, apart from a popular primary organic aerosol (POA) molecular marker for biomass burning, levoglucosan, there has been no study in Chiang Mai performed source appointment with secondary organic aerosol (SOA) molecular markers to evaluate SOA contribution which may simultaneously be formed via heterogeneous chemical reactions during long-range transport of smoke plume before arriving the receptor site. An additionally inclusive SOA molecular markers to the bulk chemical components could potentially provide an insight into $PM_{2.5}$ sources (Srivastava et al., 2018).

This study aims to (i) characterize chemical components of $PM_{2.5}$ including carbonaceous species, water soluble inorganic ions, crustal and trace elements and organic molecular markers, (ii) identify potential source of $PM_{2.5}$ and apportion them by positive matrix factorization model (PMF), and (iii) demonstrate relationship between $PM_{2.5}$ sources and their original pathway through long-range transport of air mass by back trajectory analysis. These results will be discussed based on haze and post-haze periods with a focus on specific features during heavy haze events ($PM_{2.5}$ exceeded $100 \mu g m^{-3}$) versus light haze ($PM_{2.5}$ below $100 \mu g m^{-3}$).

4.3 Methodology

4.3.1 Sampling site

As described in 3.1

4.3.2 PM_{2.5} sampling collection

As described in 3.3

4.3.3 Bulk chemical analysis

As described in 3.4 and three organic tracers, namely levoglucosan, 4-nitrocatechol and dehydroabietic acid were analyzed as described in 3.4.5

4.3.4 Quality assurance and quality control (QA/QC) for bulk chemical analysis

As described in 3.4.6

4.3.5 PM_{2.5} mass closure

In order to compare between gravimetric mass of PM_{2.5} and sum of measured mass of chemical components, the basis of mass closure equation was adopted to achieve the objective as follows (Chow et al., 2015)

$$[\text{PM}_{2.5}] = [\text{OM}] + [\text{EC}] + [\text{Inorganic ions}] + [\text{Mineral dust}] + [\text{Sea salt}] + [\text{Trace elements}] + [\text{other}]$$

Where; $[\text{OM}] = f^* [\text{OC}]$

[EC], [OC] and f are concentration of EC, OC and OC to OM conversion factor, respectively. Here, $f = 1.6$ (Chow et al., 2015)

$$[\text{Inorganic ions}] = [\text{SO}_4^{2-}] + [\text{NO}_3^-] + [\text{NH}_4^+]$$

$$[\text{Mineral dust}] = 1.89[\text{Al}] + 2.14[\text{Si}] + 1.4[\text{Ca}] + 1.67[\text{Ti}] + 1.43[\text{Fe}] \text{ (Andrews et al., 2000)}$$

Since Si was not measured in our study, Si can be estimated by $3.41[\text{Al}]$ as average ratio of Si to Al in the earth's crust, therefore $[\text{Mineral dust}] = 10.69[\text{Al}] + 1.63[\text{Ca}] + 1.94[\text{Ti}] + 2.42[\text{Fe}]$

$$[\text{Sea salt}] = 1.8[\text{Cl}^-] \text{ (White, 2008)}$$

[Trace elements] = sum of concentration of trace elements and

[other] = unidentified components

4.3.6 PM_{2.5} Source apportionment

Source apportionment of PM_{2.5} was performed by employing PMF model version 5.0 developed by United States Environmental Protection Agency (US EPA). Mass balance can be simply written from chemical species in n number of samples and p independent sources as following equation (Hopke, 2016):

$$x_{ij} = \sum_{k=1}^p g_{ik} f_{kj} + e_{ij}$$

where x_{ij} is j^{th} chemical species concentration in i^{th} sample, f_{kj} is concentration of j^{th} chemical species from k^{th} source, g_{ik} is contribution of k^{th} source in i^{th} sample, e_{ij} is residual which not fit by the model in i^{th} sample and j^{th} species. The goal of the analysis is to identify the contribution of

sources in samples (g_{ik}) by known species concentrations from chemical analysis (x_{ij}) and chemical source profile (f_{kj}). The best value of g_{ik} and f_{kj} are obtained with a minimum value of Q ;

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left(\frac{e_{ij}}{u_{ij}} \right)$$

where u_{ij} is the uncertainty of j^{th} species in i^{th} sample.

4.3.7 Data preparation

Two data files including (1) concentration (in $\mu\text{g m}^{-3}$ unit) of chemical species and (2) uncertainties were used as input data. The concentration data below detection limit and missing value were treated as explained elsewhere by Polissar et al. (1998):

$$x_{ij} = d_j/2 \quad \text{if } x_{ij} < d_j$$

$$x_{ij} = \tilde{x}_j \quad \text{if } x_{ij} \text{ is missing values}$$

where d_j is detection limit of j^{th} species and $\tilde{x}_j$ is arithmetic mean of j^{th} species. Uncertainties were calculated as:

$$u_{ij} = \sqrt{(f_j + x_{ij})^2 + (0.5 \times d_j)^2} \quad \text{if } x_{ij} > d_j$$

$$u_{ij} = d_j \times 5/6 \quad \text{if } x_{ij} \leq d_j$$

$$u_{ij} = 4 \times \tilde{x}_j \quad \text{if } x_{ij} \text{ is missing values}$$

where f_j is error fraction of j^{th} species.

In addition, chemical species used as input data for PMF were carefully selected. Major species contributed to $\text{PM}_{2.5}$ mass, species as specific source markers were selected. The species with more than 50% of the data below detection limit and missing data were removed. Finally, a total of 25 species were applied to the model namely, $\text{PM}_{2.5}$, OC, EC, SO_4^{2-} , NO_3^- , NH_4^+ , Cl^- , K^+ , Ca^{2+} , Mg^{2+} , Al, Ti, V, Mn, Fe, Ni, Cu, As, Rb, Sb, Ba, Ce, Pb, levoglucosan, dehydroabietic acid and 4-nitrocatechol.

4.3.8 Model setting and optimum results selection

4 – 7 number of factors were performed to find the most optimum set of factor profiles (Fig. SB-6 to SB-8 show the trial run of 4, 5 and 7 factors). S/N ratios of each species were used to categorize input species as strong ($S/N > 2$), weak ($0.2 \leq S/N < 2$) and bad ($S/N < 0.2$). Correlation coefficient (r) between measured and modelled concentrations was obtained from the trial run, the species with very poor correlation coefficient was also set as weak. A seed run with the lowest $Q(\text{robust})$ and $Q(\text{true})$ difference was selected, then a rotational tool, Fpeak, was used to reduce rotational ambiguity on base run. Fpeak with change in $Q(\text{robust}) < 5\%$ was selected as suggested by PMF user guide (Norris et al., 2014). Bootstrap (number of 100) was also used to explore and anticipate disproportionate effects and effects of rotational ambiguity (Norris et al., 2014). Finally, final profiles were carefully selected with the following criteria: factors must have low correlation to one another (no similarity across profiles), more than 80% of Fpeak bootstrap match in the same factor profile ensuring the robustness of the solution, the final solution should be realistic and physically explainable based on previous literatures of source profile chemical characteristic.

4.3.9 Back trajectory analysis

To examine smoke haze plume movement arriving at sampling site, long-range transport of air masses was modelled by back trajectory analysis using Hybrid Single-Particle Lagrangian Integrated Trajectory version 5.2 (HYSPLIT) developed by Notional Oceanic and Atmospheric Administration (NOAA). NCEP/NCAR reanalysis between February and June 2019 was used as meteorological data. Site location was set as mentioned in section 2, at 750 meters above mean sea level as the site is approximately 340 meters above sea level and to avoid error by ground friction from model bias. Trajectories were set to run every 6-hour intervals for backward route of -72 hours. Summarized back trajectories are illustrated in Fig. 4-12 to 4-14.

4.4 Results and discussion

Support information of this chapter is in supplement B

4.4.1 PM_{2.5} mass concentration

Temporal variations of daily average (24hr) of PM_{2.5} concentration of the study site (Mae Hia (MH)) in Chiang Mai and other neighboring provinces across Northern region of Thailand are displayed in Fig. 4-1. The patterns of PM_{2.5} concentrations at all sites show a similar variation with our study site in both during haze and post-haze episodes with moderate to high correlation coefficients (R^2) (0.52 – 0.97) (Table SB-1) between at MH and other measuring sites over neighboring provinces obtained from The Department of Pollution Control (PCD) (<http://air4thai.pcd.go.th>), indicating that the haze and post-haze events spatially took place in a regional scale. It is well known that haze events over upper Southeast Asian countries (Thailand, Myanmar, Laos) generally occurs from February to May especially in March when coinciding with open burning season (Van et al., 2022).

Daily average $PM_{2.5}$ concentrations were exceeded 24hr average $PM_{2.5}$ for Thailand National Ambient Air Quality standard ($37.5 \mu g m^{-3}$) prescribed by PCD (<https://www.pcd.go.th>) and the World Health Organization (WHO) 24hr average standard ($15 \mu g m^{-3}$) throughout haze episode with the highest, lowest, and averaged daily $PM_{2.5}$ concentrations of 233.9, 40.1, 86.6 ($SD \pm 36.2$) $\mu g m^{-3}$, respectively. While during post-haze episode, the daily $PM_{2.5}$ were generally lower than Thailand's standard for the most dates except 5 days during 11 and 17 – 20 May; however, they were still greater than WHO's standard between 11 and 28 May, with the highest, lowest, and averaged of 43.0, 6.3, 20.8 ($SD = \pm 11.5$) $\mu g m^{-3}$, respectively. In this study, we categorized episodic haze events (EH) when daily $PM_{2.5}$ concentration greater than $100 \mu g m^{-3}$, 4 episodic haze events were indicated during 12 – 16 Mar (EH1), 22 – 25 Mar (EH2), 29 Mar – 1 Apr (EH3-1) and 2 – 5 Apr (EH-2), and 10 – 12 Apr (EH4).

$PM_{2.5}$ mass in Chiang Mai was measured during haze seasons by several previous studies, for instance, $69.7 \mu g m^{-3}$ in 2013 (Chuang et al., 2016) at a rural site (near source), $118 \mu g m^{-3}$ and $108 \mu g m^{-3}$ in 2015 for a rural site (near source) and an urban site (Pani et al., 2019a), and $64 \mu g m^{-3}$ in 2016 at an urban site (Thepnuan et al., 2019a). Daily average of $PM_{2.5}$ mass from current study differs to most of those reported in above mentioned studies, implying that there were difference factors controlling $PM_{2.5}$ mass level such as meteorological effects, rainfall, emissions, El Nino phenomenon, land use, and transboundary pollution (Dahari et al., 2021). A study conducted to investigate relationships between meteorological factors and $PM_{2.5}$ and PM_{10} during haze periods specially in the Northern Thailand, revealed that $PM_{2.5}$ and PM_{10} display positive correlations with temperature and negative correlation with humidity (Sirithian & Thanatrakolsri, 2022). Furthermore, severe haze events over Southeast Asian countries were recorded during the El – nino years including 1997-1998, 2006, 2013, 2015, and 2019 (current study's year) (Van et al., 2022) which may attribute to notably high $PM_{2.5}$ concentrations in year 2015 and 2019 (current study).

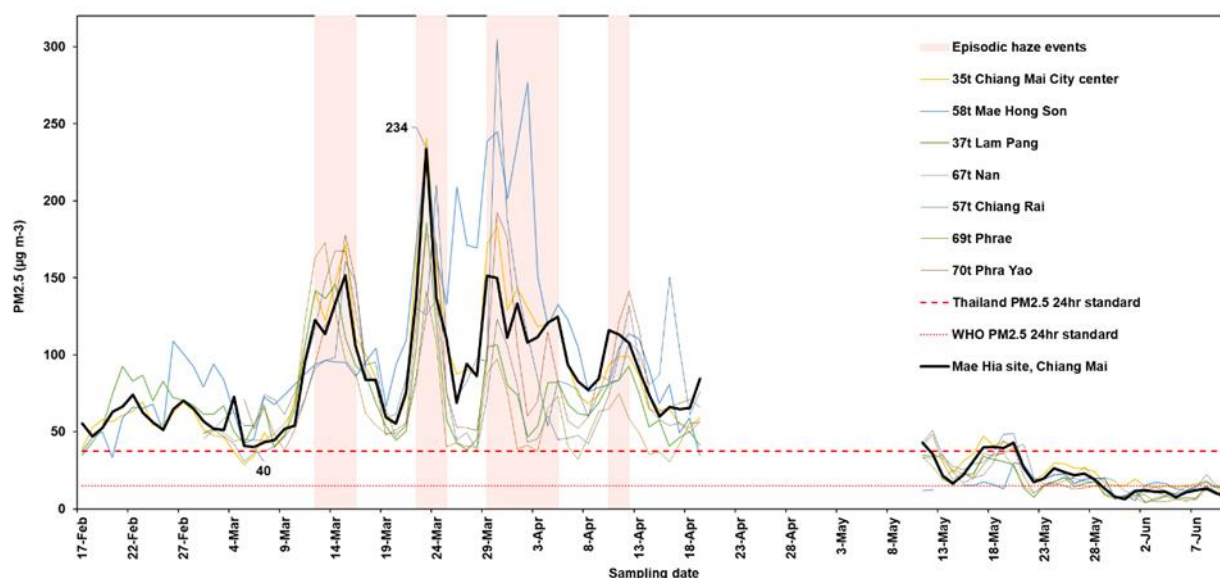


Fig. 4-1 Temporal variations of daily average concentration of PM_{2.5} at Mae Hia site in Chiang Mai and other sites in neighboring provinces in Northern Thailand compared to WHO and Thailand PM_{2.5} 24hr standards.

4.4.2 PM_{2.5} chemical components

Bulk chemical analysis was performed comprising of carbonaceous species (OC and EC), water soluble inorganic ions (WSIIs), elements (crustal and trace elements), and organic tracers (levoglucosan, 4-nitrocatechol (4-NC), and dehydroabietic acid (DHAA)). Their statistical average, SD and percentage contribution to PM_{2.5} mass are shown in Table 4-1, separated into five durations including episodic haze (PM_{2.5} > 100 µg m⁻³), low haze (PM_{2.5} > 100 µg m⁻³), entire haze (sum of both episodic and low haze), post-haze and entire study period, as well as the ratios of OC to EC, char-EC to soot-EC, K⁺ to levoglucosan, SO₄²⁻ to NO₃⁻, Fe to Al and Zn to Al. Mass ratios of PM_{2.5} and its chemical components between low haze to episodic haze, and post-haze to entire haze are shown in supplementary Fig. SB-1.

Table 4-1 Statistical information of chemical components during episodic haze, low haze, entire haze, post-haze and entire study

	Episodic (PM _{2.5} > 100 µg m ⁻³)			Low haze (PM _{2.5} < 100 µg m ⁻³)			Entire haze			Post - haze			Entire period		
	Average	±SD	% in PM _{2.5}	Average	±SD	% in PM _{2.5}	Average	±SD	% in PM _{2.5}	Average	±SD	% in PM _{2.5}	Average	±SD	% in PM _{2.5}
PM _{2.5} (µg m ⁻³)	128.6	28.5		66.2	15.4		86.6	36.2		20.9	11.5		64.7	43.4	
Carbonaceous species (µg m⁻³)															
Total OC	55.0	16.2	42.8	23.0	8.3	34.7	33.5	19.1	38.7	5.9	2.8	28.5	24.3	20.4	37.6
OC1	3.9	1.6	3.0	0.7	1.2	1.0	1.7	2.0	2.0	0.0	0.0	0.0	1.1	1.8	1.8
OC2	9.4	3.1	7.3	4.2	1.5	6.3	5.9	3.3	6.8	0.9	0.5	4.5	4.3	3.6	6.6
OC3	16.5	4.5	12.9	7.9	2.4	11.9	10.7	5.3	12.4	2.3	0.9	10.9	7.9	5.9	12.2
OC4	8.7	1.8	6.8	5.5	1.4	8.2	6.5	2.2	7.5	1.5	0.8	7.1	4.8	3.0	7.5
POC	16.6	6.7	12.9	4.8	2.7	7.2	8.6	7.1	10.0	1.3	0.7	6.1	6.2	6.8	9.5
Total EC	5.8	1.5	4.5	3.3	0.9	5.0	4.1	1.6	4.8	0.9	0.6	4.2	3.0	2.1	4.7
EC1	5.4	1.5	4.2	3.1	0.8	4.7	3.8	1.5	4.4	0.6	0.5	3.0	2.8	2.0	4.3
EC2	0.4	0.1	0.3	0.2	0.1	0.3	0.3	0.1	0.3	0.2	0.1	1.1	0.3	0.1	0.4
EC3	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.2	0.6	0.1	0.2	0.1
Char-EC	5.4	1.5	4.2	3.1	0.8	4.7	3.8	1.5	4.4	0.6	0.5	3.0	2.8	2.0	4.3
Soot-EC	0.4	0.2	0.3	0.2	0.1	0.4	0.3	0.1	0.3	0.2	0.2	1.2	0.3	0.2	0.4
Water soluble ions (µg m⁻³)															
SO ₄ ²⁻	9.0	2.2	7.0	6.6	1.9	9.9	7.4	2.3	8.5	3.4	2.6	16.4	6.0	3.0	9.3
NO ₃ ⁻	6.6	2.5	5.1	3.3	1.3	5.0	4.4	2.4	5.1	0.8	0.3	4.0	3.2	2.6	4.9
NH ₄ ⁺	3.2	0.8	2.5	2.2	0.7	3.4	2.6	0.9	3.0	1.0	0.9	4.6	2.0	1.2	3.2
K ⁺	1.9	0.4	1.5	1.0	0.3	1.4	1.3	0.6	1.5	0.2	0.1	0.9	0.9	0.7	1.4
Ca ²⁺	1.4	0.6	1.1	0.8	0.3	1.2	1.0	0.5	1.2	0.1	0.1	0.5	0.7	0.6	1.1
Mg ²⁺	0.4	0.2	0.3	0.2	0.1	0.2	0.2	0.2	0.3	0.0	0.0	0.1	0.2	0.2	0.3
Cl ⁻	0.5	0.2	0.4	0.3	0.1	0.4	0.4	0.2	0.4	0.2	0.0	1.0	0.3	0.2	0.5
Organic tracers (ng m⁻³)															
Levogluconan	2185.1	917.7	1.7	717.3	479.2	1.1	1203.0	959.3	1.4	100.7	73.1	0.5	835.6	940.7	1.3
4-Nitrocatechol	285.0	222.3	0.3	60.8	73.8	0.1	136.3	176.7	0.2	5.6	6.5	0.0	92.7	156.7	0.1
Dehydroabietic acid	41.3	39.5	<0.1	9.9	8.8	<0.1	20.4	27.8	<0.1	0.8	1.9	<0.1	13.9	24.5	<0.1
Elements (ng m⁻³)															
Al	1312.6	416.1	1.0	1025.3	200.1	1.5	1117.6	314.7	1.3	309.0	281.8	1.5	848.1	488.3	1.3
Fe	903.1	238.2	0.7	664.5	147.9	1.0	740.8	211.7	0.9	203.9	181.1	1.0	561.9	324.3	0.9
Ti	50.4	17.8	<0.1	37.1	8.0	0.1	41.3	13.4	<0.1	11.4	10.1	<0.1	31.3	18.8	<0.1
Zn	111.0	146.1	0.09	88.8	141.9	0.13	96.1	142.4	0.11	96.0	163.5	0.46	96.1	147.0	0.15
Mn	43.3	10.3	<0.1	31.3	8.1	<0.1	35.2	10.5	<0.1	7.4	5.4	<0.1	25.9	16.0	<0.1
V	2.4	0.6	<0.1	2.1	0.3	<0.1	2.2	0.5	<0.1	0.9	0.7	<0.1	1.8	0.8	<0.1
Ni	3.0	0.9	<0.1	2.2	0.4	<0.1	2.5	0.7	<0.1	1.2	0.6	<0.1	2.0	0.9	<0.1
Cu	6.8	4.3	<0.1	4.1	3.0	<0.1	4.8	3.6	<0.1	3.6	5.5	<0.1	4.4	4.3	<0.1
As	1.5	0.5	<0.1	1.0	0.3	<0.1	1.2	0.4	<0.1	0.5	0.3	<0.1	1.0	0.5	<0.1
Rb	6.5	1.5	<0.1	3.8	1.1	<0.1	4.6	1.8	<0.1	1.1	0.7	<0.1	3.4	2.3	<0.1
Sb	0.4	0.1	<0.1	0.4	0.2	<0.1	0.4	0.2	<0.1	0.2	0.1	<0.1	0.4	0.2	<0.1
Ba	9.9	2.8	<0.1	7.0	1.4	<0.1	8.0	2.4	<0.1	1.8	1.1	<0.1	6.0	3.5	<0.1
Ce	2.1	1.1	<0.1	1.5	0.4	<0.1	1.7	0.8	<0.1	0.6	0.5	<0.1	1.3	0.9	<0.1
Pb	14.1	3.9	<0.1	12.2	6.1	<0.1	12.8	5.5	<0.1	5.0	3.1	<0.1	10.2	6.1	<0.1
Total crustal elements ^a (µg m ⁻³)	5.9	1.2	4.5	3.9	0.7	5.9	4.5	1.3	5.2	1.0	0.9	4.9	3.4	2.0	5.2
Total trace elements ^b (µg m ⁻³)	0.3	0.3	0.2	0.2	0.1	0.2	0.2	0.1	0.2	0.1	0.1	0.4	0.1	0.2	0.2
OC/EC	7.8	2.2		7.6	2.4		7.7	2.2		9.7	1.9		6.9	1.7	
Char-EC/Soot-EC	14.6	5.2		3.1	2.6		10.8	7.1		15.0	5.8		14.4	5.0	
Levogluconan/K⁺	0.6	0.3		0.5	0.4		0.6	0.3		0.8	0.3		0.5	0.2	
SO₄²⁻/NO₃⁻	1.1	0.4		0.8	0.4		0.9	0.5		0.7	0.3		0.8	0.4	
Fe/Al	0.7	0.1		0.7	0.1		0.7	0.1		0.9	0.7		0.7	0.4	
Zn/Al	0.1	0.1		0.1	0.1		0.1	0.1		0.3	0.8		0.2	0.5	

Total crustal elements^a – sum of Mg, Al, K, Ca, Ti and FeTotal trace elements^b – sum of V, Cr, Mn, Co, Ni, Cu, Zn, As, Rb, Sr, Cd, Sb, Ba, Ce, W and Pb

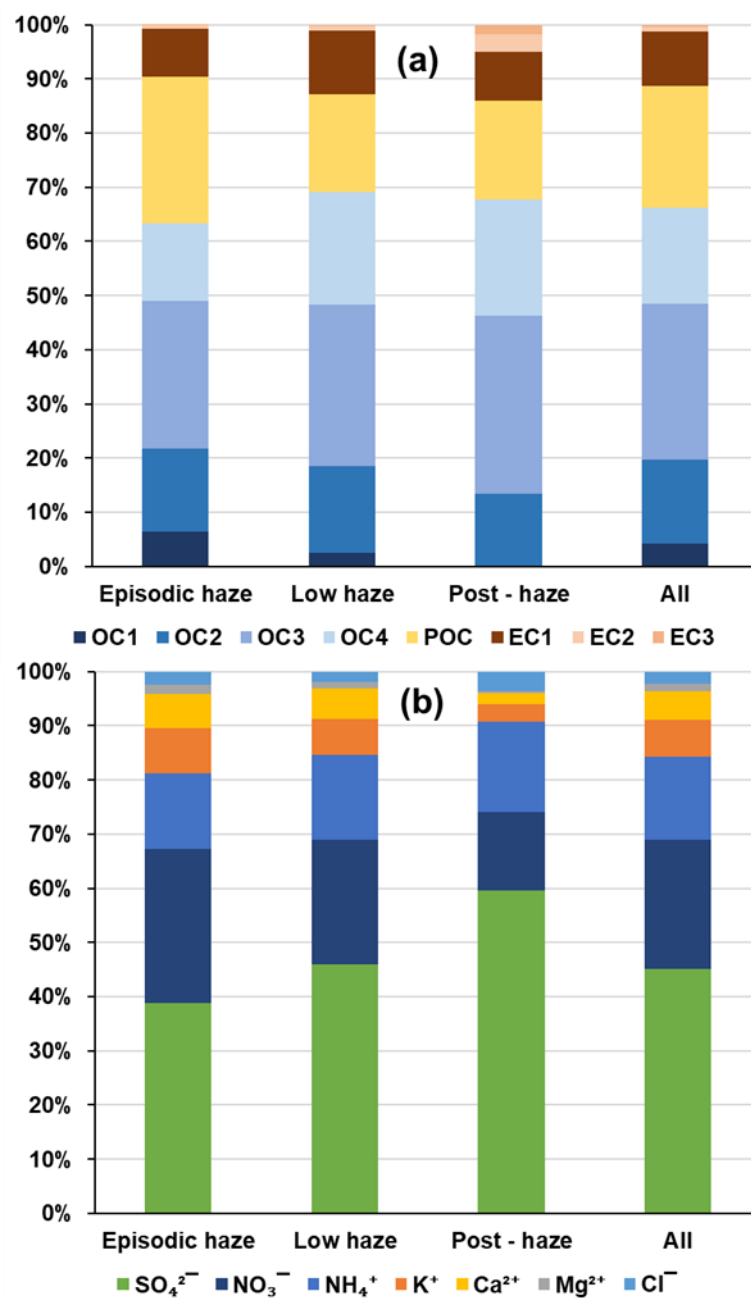


Fig. 4-2 Percentage contributions of carbonaceous species (a) and water-soluble inorganic ions (WSIIs) (b) during episodic haze ($PM_{2.5} > 100 \mu g m^{-3}$), low haze ($PM_{2.5} < 100 \mu g m^{-3}$), post-haze and entire period of study.

4.4.2.1 Carbonaceous species

Carbonaceous species are a major component of $PM_{2.5}$ mass especially during haze events (Chow et al., 2015; C. Te Lee et al., 2016; Saksakulkrai et al., 2023; Tham et al., 2019; Y. Zhang et al., 2015). In this study, OC is the largest contributor of $PM_{2.5}$ mass in every period while total EC contributed much lower than OC; their concentration and percentage contribution to $PM_{2.5}$ mass of OC and EC are listed in Table 4-1. The contributions of OC and EC similar to those reported by previous studies in Chiang Mai (ChooChuay et al., 2022; Chuang et al., 2013; Pani et al., 2018, 2019b; Tao et al., 2020; Thepnuan et al., 2019a). Among carbonaceous species, total OC (sum of OC1-4 and pyrolyzed organic carbon (POC)) contributed more than 80% of total carbon (TC) (Fig. 4-2 (a)) and OC3 was the most dominant fraction, while EC contributed less than 15% of TC. The high contribution of OC indicates biomass burning was a main source of $PM_{2.5}$ corresponding to other studies in Chiang Mai (ChooChuay et al., 2022; Pani et al., 2019b; Thepnuan et al., 2019a) and other cities in such as Indonesia (Fujii, Tohno, et al., 2015), and Singapore (Tham et al., 2019). During low haze and post-haze periods, the sequence of carbonaceous fractions was $OC3 > OC4 > POC > OC3 > OC1 > EC1 > EC2 > EC3$, while during episodic haze was slightly changed in order as $POC = OC3 > OC2 > OC4 > OC1 > EC1 > EC2 > EC3$. Mass ratios between low haze to episodic haze (Fig. S4-1) show that POC and OC1 had substantially increased by factors 5.8 and 3.5, respectively while $PM_{2.5}$ mass increased by factor 1.94. The enhancements of OC1 and POC indicate smoldering condition during episodic events due to lower temperature favors the emission of lower molecular weight organic compounds such as OC1 and OC2 (Cui et al., 2022; Huboyo et al., 2016; Santiago-De La Rosa et al., 2022). The great POC fraction suggests a presence of water-soluble organic compounds (WSOC) or humic-like substances in PM, a notable feature that can be observed during episodic haze events (Tham et al., 2019).

OC/EC ratio has widely been used to identify carbonaceous source, however, OC/EC ratios in this study did not show significant differences in all periods (Table 4-1) Char-EC (EC1 – POC) to

soot-EC (EC2 + EC3) ratio have also been used in several studies due to more efficiency to identify the burning condition between flaming or smoldering (Huboyo et al., 2016; Lim et al., 2012; Santiago-De La Rosa et al., 2022; Tham et al., 2019) and PM source between biomass burning and motor vehicle emission (Chuang et al., 2013; Dejchanchaiwong et al., 2020; Suradi et al., 2021; Thuy et al., 2018). Proportionally higher char-EC than soot-EC is commonly observed at lower temperature combustions in smoldering phase; on the other hand, higher proportion of soot-EC is found at high temperature combustion in flaming phase of biomass burning or engine combustions (C. Te Lee et al., 2016). Char-EC/soot-EC lower than 1 indicates vehicular emission while char-EC/soot-EC higher than 3.9 was an indicator for biomass burning (Dejchanchaiwong et al., 2020). The mean char-EC/soot-EC ratios (Table 4-1) in the present study were episodic haze > low haze > post-haze. Scatter plots between char-EC and soot-EC are displayed in Fig. 4-3. Char-EC/soot-EC plots of both haze groups cluster close to the biomass burning line with distribution at higher value for episodic haze than low haze, suggesting that biomass burning was the major source during haze and was mainly from smoldering phase during episodic haze, whereas char-EC/soot-EC in post-haze cluster around vehicular exhaust line, suggesting vehicular exhaust was one of the main sources during post-haze. Previously, char-EC/soot-EC ratios were reported by several studies, for example, very high ratio of 34.6 at a site very close to biomass burning (Pani et al., 2019b), ratio of 9.4 at a sampling site from smoldering phase (Chuang et al., 2013), and ratio of 14.9 associating with biomass burning over continental Southeast Asia region (C. Te Lee et al., 2016), while lower ratios of 4.9 and 6.9 were reported in big cities of Ho Chi Minh City, (Tran et al., 2023), and Bangkok (ChooChuay, Pongpiachan, Tipmanee, Suttinun, et al., 2020), respectively.

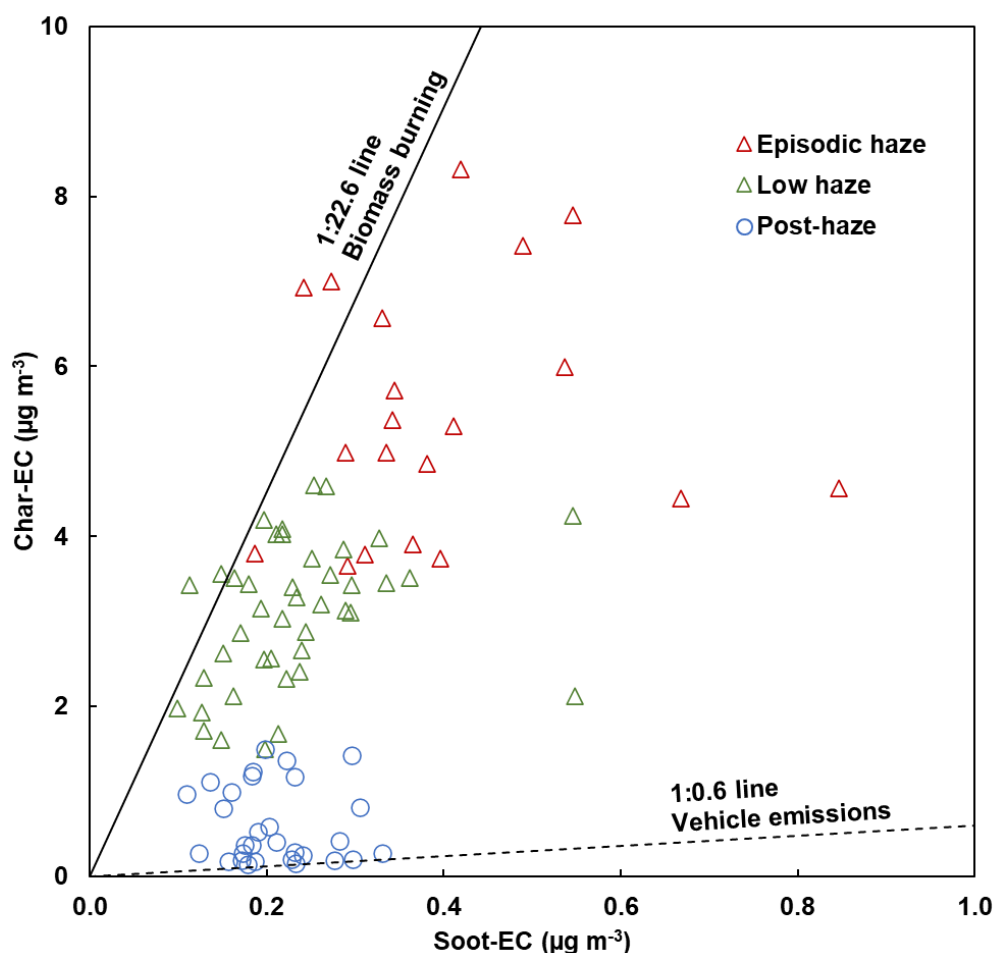


Fig. 4-3 Scatter plots between soot-EC and char-EC during episodic haze ($\text{PM}_{2.5} > 100 \mu\text{g m}^{-3}$), low haze ($\text{PM}_{2.5} < 100 \mu\text{g m}^{-3}$) and post-haze. Reference lines of biomass burning and vehicular exhaust (soot-EC : char-EC, 1:22.6 and 1:0.6, respectively) adopted from a study by Fujii et al. (2016)

4.4.2.2 Water soluble inorganic ions (WSIIs)

Water soluble inorganic ions are also a major group of chemical components of PM mass. WSIIs average concentrations in each episode are shown in Table 4-1. By descending order, the concentration of WSIIs during episodic haze and low haze were similar as $\text{SO}_4^{2-} > \text{NO}_3^- > \text{NH}_4^+ > \text{K}^+ > \text{Ca}^{2+} > \text{Cl}^- > \text{Mg}^{2+}$, while during post-haze were $\text{SO}_4^{2-} > \text{NO}_3^- > \text{NH}_4^+ > \text{Cl}^- > \text{K}^+ > \text{Ca}^{2+} > \text{Mg}^{2+}$. Sulfate (SO_4^{2-}), nitrate (NO_3^-) and ammonium (NH_4^+) or known as SNA were the top three contributors for all sampling periods, similar to those previous studies in Chiang Mai (Chansuebsri et al., 2022; Chantara et al., 2012) but differed to WSIIs obtained from urban areas in Thailand

such as Bangkok (Ahmad et al., 2022; ChooChuay, Pongpiachan, Tipmanee, Suttinun, et al., 2020; Rattanapotanan et al., 2023) where SNA ions were not the main contributors. Percentage contribution of SNA ions to total ions (Fig. 4-2 (b)) during episodic (81%) and low haze (85%) were lower than post-haze (91%), this may be attributed to the higher contributions of other minor ions such as K^+ , Ca^{2+} and Mg^{2+} had substantially increased during haze episodes (Fig. S4-1 (b)).

The correlation coefficients between cations and anions in episodic haze, low haze and post-haze are shown in Table SB-2. NH_4^+ had a very strong correlation with SO_4^{2-} in all episodes, indicating secondary sulfate is one of the major sources of $PM_{2.5}$ for the entire study period, consistent with studies in Chiang Mai where indicated secondary sulfate as a major source of PM (Chantara et al., 2012; Khamkaew, Chantara, Janta, et al., 2016a; Thepnuan et al., 2019b; Tsai et al., 2013). NO_3^- had a strong correlation with Ca^{2+} during episodic haze and low haze, indicating that secondary nitrate derived from biomass burning in the form of calcium nitrate ($Ca(NO_3)_2$) (more details and formation reactions of calcium nitrate are explained in section 3.5.6). Furthermore, strong correlation between Cl^- and other salts (K^+ , Mg^{2+} and Ca^{2+}) in episodic and low haze episodes may be attributed to the formation of chlorine salts (KCl , $CaCl_2$ and $MgCl_2$) form by biomass burning and soil dust (Jiang et al., 2018).

Ratio of NO_3^- to SO_4^{2-} has been consider as a potential indicator to identify stationary and mobile sources. Higher NO_3^-/SO_4^{2-} indicates mobile sources dominate over stationary sources (Jiang et al., 2018), while lower ratio indicates biomass burning (Pani et al., 2018). Average NO_3^-/SO_4^{2-} ratio in post-haze > episodic haze > low haze and higher ratio in post-haze suggests either higher influence of mobile source. Fig. 4-4 shows box plots of the NO_3^-/SO_4^{2-} ratios of this study comparing with Southeast Asian countries, China, India and some European countries. The ratios in this study were comparable to those reported in Thailand but were relatively higher than of Southeast Asian countries. It is a clear noticeable that the ratios of this study and Southeast

Asian countries were lower compared to those from China, India and European countries, this may be attributed to a greater contribution from dense traffic in megacities such as Beijing and Delhi lead to substantial NO_x emission and meteorological condition where low temperature in such countries favors the partitioning of gaseous NO_x into NO_3^- formation on PM (Q. Xu et al., 2019).

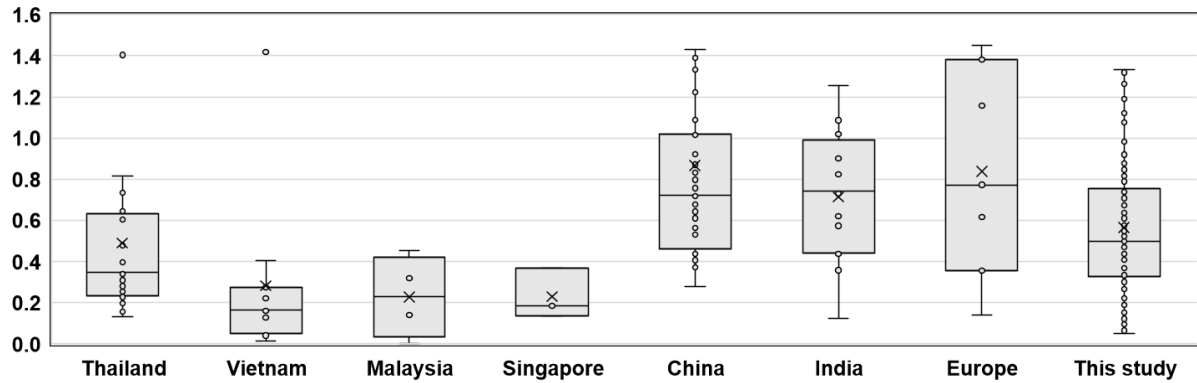


Fig. 4-4 Box plots of $\text{NO}_3^-/\text{SO}_4^{2-}$ ratios in selected Southeast Asian countries, China, India and Europe. Information obtained from Saksakulkrai et al. (2023)

4.4.2.3 Crustal elements and trace metals

The concentrations and percentage contribution of total crustal and total trace elements during episodic haze, low haze and post-haze are shown in statistical Table 4-1.

Since crustal elements are commonly found in Earth's crust, their content in $\text{PM}_{2.5}$ is generally higher compared to trace elements for all episodes. Both crustal and trace elements had a similar descending order of concentrations as episodic haze > low haze > post-haze, which were consistent with previous literatures comparing metals content between haze vs. non-haze condition (Betha et al., 2013; Kayee et al., 2020; Kraisitnitikul et al., 2024). The concentration of crustal elements was in descending order as $\text{K} > \text{Al} > \text{Fe} > \text{Ca} > \text{Mg} > \text{Ti}$ for every episode. In Fig. S4-1(b), elevated mass ratio during haze episode of crustal elements (4.4) was greater than of trace elements (1.3), suggesting a huge contribution of natural soil dust during haze episode. Rubidium (Rb) was the only one trace element had very strong correlation with other biomass

burning tracers (correlation matrix in Table SB-3), OC ($R^2 = 0.83$), K⁺ ($R^2 = 0.90$) and levoglucosan ($R^2 = 0.76$) during haze episode, suggesting that Rb was emitted from biomass burning. Rb has been used as alternative biomass burning tracer in a number of previous studies (Lindgren et al., 2006; Zhang et al., 2013; Amil et al., 2016; Massimi et al., 2020) yet has never been used as a tracer for biomass burning in Ching Mai. Previous studies have shown that Rb is associated with K in soil and plants, and Rb has been used as a tracer of K⁺. The level of Rb⁺ in plants, fungi, and animals is influenced by soil chemical properties and the amount of K⁺ in soil through food webs. pH in soil regulates Rb in plants and animals, high acidity in soil (low pH) causes K⁺ leaching and loss from soil body. This low availability of K⁺ in soil is then compensated by substantially increased uptake of Rb by plants and fungi. Rb elevates and propagates in the food webs and being stored in organisms tissue (Tyler, 1997; Drobner & Tyler, 1998; Nyholm & Tyler, 2000). This would be of interest to propose Rb as a new potential biomass burning tracer that can differentiate biomass from different source's areas where chemical soil's properties are different, for example, forest (low K⁺) vs. agricultural areas (high K⁺).

Fe to Al and Zn to Al ratios have been used to indicate natural crustal dusts or anthropogenic-related dusts (Kayee et al., 2020; Z. Shen et al., 2016). Fe/Al and Zn/Al ratios are shown in statistical Table 4-1, Fe/Al ratio during post-haze (0.88) was slightly higher than those in episodic haze (0.7) and low haze (0.65), while Zn/Al ratio was significantly higher in post-haze (0.31) than episodic (0.07) and low haze (0.09), the elevated ratios of Fe/Al and Zn/Al during post-haze suggest that anthropogenic-related dusts had influence to PM_{2.5} mass in some extent. Kayee et al. (2020) reported Zn/Al ratios in Chiang Rai (neighboring province to Chiang Mai) ranged from 0.07 to 0.15 and 0.08 to 0.31 for haze and non-haze, respectively, which were close to Zn/Al obtained from the current study. Furthermore, several trace elements had very strong correlations ($R^2 > 0.8$) in post-haze period as shown in Table SB-3 compared to haze period, suggesting traffic-related emissions contribution during post-haze.

4.4.2.4 Organic tracers

Levoglucosan, 4-Nitrocatechol (4-NC) and Dehydroabietic acid (DHAA) are organic tracers associated with biomass burning. Levoglucosan has been well known as general wood burning tracer for primary organic aerosol (POA) (Bhattacharai et al., 2019; Janoszka & Czaplicka, 2019), while 4-NC and DHAA have been proposed for secondary organic aerosol (SOA) from aging process (Bertrand et al., 2018; Q. Wang et al., 2022) and softwoods burning (B. Yan et al., 2008), respectively. Their concentrations and percentage contributions to PM_{2.5} mass during each episode are shown in statistical Table 4-1; in general, their concentrations had similar trends as descending order: episodic haze > low haze > post-haze. Level of Levoglucosan observed in this study was similar to those reported in Chiang Mai during haze episode (Khamkaew, Chantara, Janta, et al., 2016b; Thepnuan et al., 2019a; Tsai et al., 2013). Levoglucosan, 4-NC and DHAA had substantially increased during entire haze episode compared to post-haze (Fig. S4-1(b)) by factors of 11.9, 24.1 and 26.5, respectively.

Ratio between Levoglucosan and K⁺ can be served as indicator to differentiate type of biomass burning, levoglucosan/K⁺ below 1 indicates the combustion of crop residues or agricultural waste burning while above 1 indicate the combination of softwoods and hardwoods burnings (Cheng et al., 2013). As shown in Table 4-1, only the levoglucosan/K⁺ ratio during episodic haze event had the ratio higher than 1, while during low haze and post-haze were 0.65 and 0.68, respectively, implying that less crop residue burning and more hardwoods/softwoods burning when extreme haze occurs. However, levoglucosan/K⁺ ratio cannot be used to separate softwood and hardwood burning. Previous studies demonstrated that levoglucosan and DHAA can be found in the aerosol emitted from the burning of softwood with proportionally higher contribution of DHAA than of levoglucosan, while only levoglucosan is found from hardwoods burning (Bhattacharai et al., 2019; Fine et al., 2004b, 2004a; Hays et al., 2002; Leithead et al., 2006), ratio between levoglucosan/DHAA was then used in this study as an indicator to display the

influences of hardwoods versus softwoods burnings. In addition, 4-NC as a SOA tracer is generally formed during aging process of biomass burning aerosol (Finewax et al., 2018), the ratio of Cl⁻ to 4-NC were then proposed to indicate fresh and aged aerosols. The ratios of levoglucosan to K⁺, levoglucosan to DHAA and Cl⁻ to 4-NC are displayed as scattered plots to one another in Fig. 4-5 (a, b and c) during low haze and different episodic haze events. From scatter plots, we can roughly indicate characteristics of low haze and episodic haze periods as follow:

- i) During low haze: the levoglucosan/K⁺ ratios were mostly below 1, indicating that majority of biomass burning derived aerosol were emitted from agricultural waste burning, the ratio of levoglucosan/DHAA distributed in a broad range from 37 to 225 which could be mixed of hardwoods and softwoods burning, and the ratio of Cl⁻/4-NC distribute in a higher range, implying that higher proportion of fresh aerosols associated with this period. In addition, fire hot spots observed during low haze (Fig. 4-11) are predominantly located within northern Thailand, primarily associated with agricultural fields (Fig. 4-15).
- ii) During episodic haze: the levoglucosan/K⁺ ratios greater than or equal to 1 were observed in EH3-1, EH3-2 and most of EH2, suggesting hardwoods and/or softwoods burning, while the ratio below 1 found in EH1 and EH4 linked to agricultural waste burning. Softwood burning associated with the ratio of levoglucosan/DHAA below 50 were found in EH3-1 and EH3-2, while more contribution of hardwoods burning found associated with the higher ratios in EH2, EH4 and EH1. Fire hot spot maps during EH3-1 and EH 3-2 (Fig.4-10) show a highly concentrated intensity of fire to the southwest of the sampling site over borderline between Thailand and Myanmar. These fires are associated with forest areas at high altitudes (Fig. 4-15). All episodic haze events (EH1-4) had lower range (0.6 – 6) of Cl⁻/4-NC ratio compared to low haze (3 – 17), implying that higher proportion of aged aerosols associated with episodic haze.

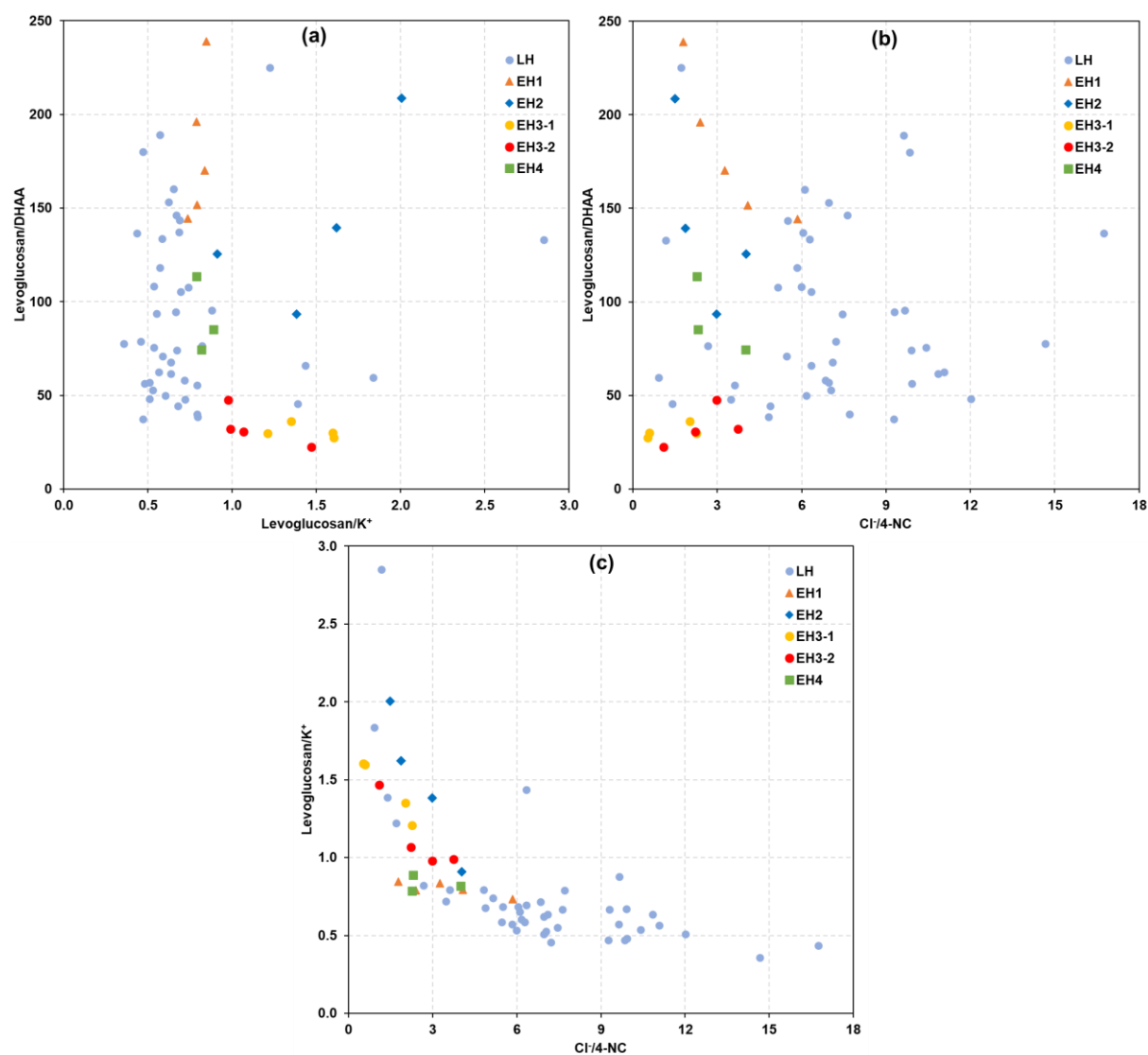


Fig. 4-5 Scatter plots between Levoglucosan/K⁺ and Levoglucosan/DHAA ratios (a), Cl⁻/4-NC and Levoglucosan/DHAA ratios (b) and Cl⁻/4-NC and Levoglucosan/K⁺ ratios (c)

4.4.3 PM_{2.5} Mass closure

In order to compare the measured PM_{2.5} mass concentration with the total mass concentration of measured chemical components, the concept of reconstructed mass (Chow et al., 2015) was adopted by applying equation (A). Organic matter (OM) during haze and post – haze episodes were calculated by multiplying with OC to OM conversion factors ($f = 1.6$). A linear regression of daily gravimetric mass of PM_{2.5} and reconstructed mass by all components is plotted in Fig. SB-2. The reconstructed mass shows very strong correlation to PM_{2.5} mass with a coefficient of determination (R^2) = 0.99 from whole study period by $N = 92$ ($R^2 = 0.98$ and 0.95 for haze and post – haze, respectively), indicating that a very good agreement between the reconstructed mass and the gravimetric mass of PM_{2.5}. Mass closure of PM_{2.5} for haze and post-haze episodes are shown in Fig. 4-6. Percentage contribution of chemical components to PM_{2.5} mass by descending order were: [OM] > [WSIIs] > [Mineral dust] > [EC] > [Other] > [Sea salt] > [Trace elements] for haze episode, and [OM] > [WSIIs] > [Mineral dust] > [Other] > [EC] > [Sea salt] > [Trace elements] for post-haze episode. OM was the most abundant component for both episodes but contributed higher in haze episode (61.9%) than post-haze episode (45.6%) due to biomass burning. Mineral dust (14.3%) and WSIIs (16.5%) during haze episode were relatively comparable, while during post-haze episode, WSIIs were significantly higher (22.5%). This characteristic of SO_4^{2-} was also found during haze episode 2014 in Chiang Mai, Pani et al. (2018) reported that the contribution of Ammonium Sulphate ($(\text{NH}_4)_2\text{SO}_4$) to PM_{2.5} mass was highest (18%) during low biomass burning and was lowest (6%) during intense biomass burning. This may be attributed to favorable meteorological conditions (lower temperature and higher relative humidity) during lesser biomass burning influence like post-haze episode promote an association of SO_4^{2-} and NH_4^+ to form secondary aerosol. EC contributed approximately 4-5% to PM_{2.5} mass in both episodes, suggesting that the sampling site was not influenced by coal combustion or vehicular emissions. Sea salt contributed very little to PM_{2.5} mass with a slight increase during

post-haze episode, implying to negligible influence by marine aerosols since sampling period was conducted between local summer (haze) when which prevailing winds mostly originated from continent. Tracers contributed approximately less than 0.5% to $PM_{2.5}$ mass for both episodes with no significant difference between both episodes.

Comparing to prior studies conducted in Chiang Mai, Thailand, results from our study during haze and post – haze episodes have shared agreements with Pani et al. (2018) and Pani et al. (2019) that organic matter (OM) was the major contributor to $PM_{2.5}$ mass followed by water soluble inorganic ions (WSIIs) and mineral dust. However, Pani et al. (2019) reported mineral dust less than 10% and EC between 4 – 10%, while our study found approximately 14-16% and 4-5% for mineral dust and EC, respectively. Another study performed by Tao et al. (2020) at an urban site in Chiang Mai at which $PM_{2.5}$ sampling period of their study (27 Feb – 22 May) and this study (17 Feb – 19 Apr) were overlapped during dry season 2019. A similar pattern of chemical components in $PM_{2.5}$ mass closure during haze episode was obtained in both studies. OM was the main chemical component to $PM_{2.5}$ mass at 60% contribution, followed by WSIIs and mineral dust at between 14-18% contribution and EC at 3%. OM or some studies refer as organic carbon (OC) have been reported as the most contributor to $PM_{2.5}$ mass in Chiang Mai by several literatures such as; Chuang et al. (2013) OC (41%); Thepnuan et al. (2019) OC (36.5%); Pani et al. (2019) OC (43%); and Pani et al. (2018) OM ranged from 59 – 72% to $PM_{2.5}$ mass.. WSIIs especially Sulphate, Nitrate, and Ammonium (known as SNA) ions have also been reported in several literatures as the second highest contributor to $PM_{2.5}$ (Chuang et al., 2013; Thepnuan et al., 2019; Chansuebsri et al., 2022). Specifically high contributions of OM and WSIIs to $PM_{2.5}$ have not only been found in Chiang Mai, Thailand, but have also been reported in several publications across Southeast Asian region such as Vietnam (Co et al., 2014; C. Te Lee et al., 2016; Popovicheva et al., 2016), Malaysia (Fujii et al., 2016; H. Hassan et al., 2021; Latif et al., 2018), Singapore (See et al., 2006; Hapsari Budisulistiorini et al., 2018), Indonesia (Siregar et al., 2022).

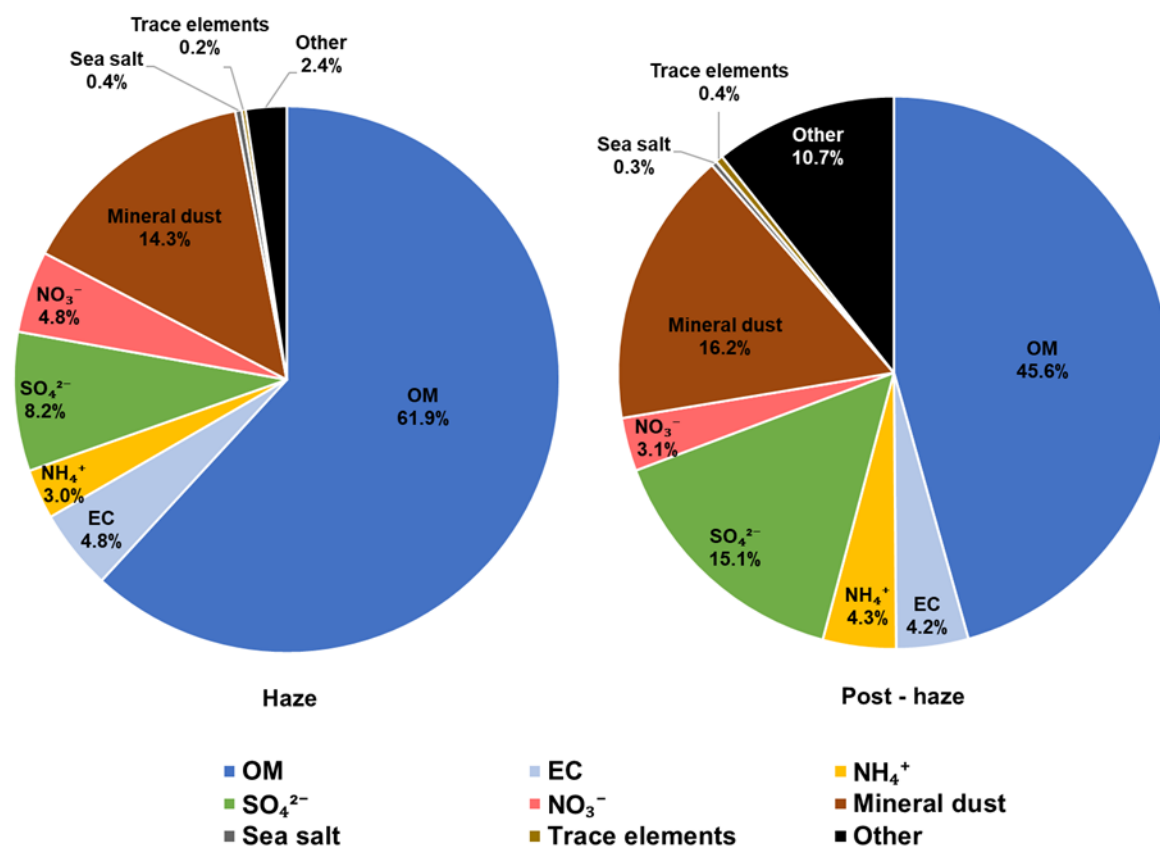


Fig. 4-6 PM_{2.5} mass reconstructions of haze and post-haze episodes

4.4.4 Source apportionment by PMF model

Source apportionment was performed for the whole period of study by employing a comprehensive set of bulk chemical analysis. 6 main factors were identified namely biomass burning, mixed of fresh biomass burning and traffic emission, softwoods burning (aged aerosol), secondary sulfate, mixed dusts and traffic emission, and secondary nitrate. The abovementioned biomass burning derived sources were distinguished by 3 organic molecular makers including Levoglucosan, Dehydroabietic acid (DHAA) and 4-Nitrocatechol (4-NC) as key markers for general biomass burning, softwoods burning and aged aerosol, respectively, which will be discussed later in this section. To the best of our knowledge, DHAA and 4-NC were used for the first time in this study for source identification of PM_{2.5} in Chiang Mai, Thailand. Chemical source profiles, temporal

variation of source concentration contributed in PM_{2.5} mass, and percentage contribution of each source during haze and post-haze are shown in Fig. 4-7, 4-8, and 4-9, respectively. Regional fire hot spot maps during episodic haze (EH) and low haze (LH) are displayed in Fig. 4-10 and 4-11, respectively. Backward trajectories are displayed in Fig. 4-12, 4-13, and 4-14 for 72hr EH, 48hr EH, and 48hr LH, respectively.

4.4.4.1 General biomass burning

This source was identified as a main biomass burning based on broad contributions of the source to several biomass burning tracers including OC (25%), EC (23%), Levoglucosan (19%), DHA (12%), Rb (46%), and especially K⁺ (52%). Levoglucosan (a product of burnt cellulose) and K⁺ (a product of combusted wood lignin) have been frequently used as biomass burning tracers from previous studies in many countries as well as in Chiang Mai, Thailand (Chuang et al., 2013; Khamkaew et al., 2016b; Thepnuan et al., 2019; Chansuebsri et al., 2022). A significantly higher K⁺ content in this source than those of other biomass derived sources may indicate agricultural burning was dominant over forest leaf litter (Chantara et al., 2019). In addition, Rb has never been reported as a biomass tracer in Thailand but was reported in other countries such as China (R. Zhang et al., 2013), Malaysia (Amil et al., 2016) and Italy (Massimi et al., 2020). Measured Rb from this study correlated very well with PM_{2.5} ($R^2 = 0.86$) and OC ($R^2 = 0.80$), therefore Rb was considered as one of biomass tracers.

The overall contribution of general biomass burning (Fig. 4-9) accounted for 28% which was the highest source contributor among other sources to PM_{2.5} mass. Contributions during haze and post-haze accounted for 31% and 5%, corresponding to average concentration of 26.6 $\mu\text{g m}^{-3}$ and 1.2 $\mu\text{g m}^{-3}$, respectively. During haze period, temporal distribution of the main biomass burning source was higher than those of other biomass burning derived sources (nearby biomass burning and softwoods burning) (Fig. 4-8) during both LH and EH, especially during episodic

events (red highlighted durations in Fig. 4-8) when distribution peaks were found in sync with the four events. Recent study in Chiang Mai reported by Chansuebsri et al. (2022) during haze episode year 2019 (the same duration of this study) that biomass burning being mixed with secondary inorganic aerosol (SIA) accounted for 35% by employing principal component analysis (PCA) for source apportionment. While ChooChuay et al. (2020), by using PCA, reported vehicular exhaust was the most contributor source in $PM_{2.5}$ followed by biomass burning. This is due to the difference in duration of sampling campaign since the latter study performed an annual sampling between March 2017 and March 2018, though, haze season over the Northern Thailand usually lasts for approximately 3 months in dry season between February and April (Saksakulkrai et al., 2023). However, according to the study by Kraisitnitikul et al. (2024) in Chiang Mai where $PM_{2.5}$ was collected during haze episode in La nina year (2017) and El nino year (2019), has found that biomass burning significantly contributed higher in year 2019 resulting from extreme weather in El nino year caused more occurrences of open burning.

4.4.4.2 Nearby biomass burning (fresh aerosol)

The significant contribution of this source to chlorine (Cl^-) was notably found to be very high, accounting for 68% to total Cl^- contribution. This source also has moderate proportions of biomass burning markers including OC (23%), EC (17%), Levoglucosan (14%), Rb (15%). NO_3^- (25%), Ni (30%), Cu (30%), together with high Cl^- -suggesting contribution of fresh aerosol from biomass burning. Cl^- has been considered as a key marker for fresh aerosol especially from biomass burning (Chantara et al., 2019) and sometimes coal combustions (J. Tan et al., 2017; X. Xu et al., 2018). Studies performed simulation of burning of several types of biomass in chamber and estimate their composition contributed in the emitted $PM_{2.5}$, K^+ and Cl^- were commonly found as highest among water soluble ions (Chantara et al., 2019; Ni et al., 2017; S. S. Park et al., 2013). This suggests that fresh aerosol from biomass burning not only contains a significant amount of K^+ but also Cl^- . However, during aging process of aerosol in the ambient atmosphere, Cl^- in chlorine salt

such as KCl is chemically replaced by other anions such as SO_4^{2-} and NO_3^- as the aerosol transports before reaching the receptor site, Cl^-/K^+ can then decrease in aged aerosol (C. Li et al., 2015).

Fresh aerosol of biomass burning contributed 21% to $\text{PM}_{2.5}$, the second highest contributor. During haze and post-haze this source contributed at 19% and 32% which corresponding to average concentration of $14.6 \mu\text{g m}^{-3}$ and $7.2 \mu\text{g m}^{-3}$, respectively. Temporal distribution during haze episode shows lower base (mostly $< 20 \mu\text{g m}^{-3}$) compared to those of the general biomass burning source (mostly $> 20 \mu\text{g m}^{-3}$) (Fig. 4-8). It should also be noted from Fig. 4-8 that the contribution of this source during post-haze episode prolonged throughout post-haze episode, implying background fresh emission such as local biomass burning. During episodic events ($\text{PM}_{2.5} > 100 \mu\text{g m}^{-3}$), the distribution reached its peak in the episodic 1 – 3 (Fig. 4-8). 48-hour backwards trajectories during four episodic events (Fig. 4-13)) show circulating patterns during episodic haze (EH) 1, 2, and 3-1, implying lower wind speed during these events. Moreover, Chiang Mai locates in a flat plain basin surrounded by mountain ranges, together with such pattern of air mass movements and low wind speed resulting in inhibiting dispersion of pollutants and enhancing accumulation of fresh aerosol from biomass burning within the basin (Sresawasd et al., 2021).

4.4.4.3 Softwoods burning (aged aerosol)

This source makes a very high contribution to biomass burning derived organic markers; Levoglucosan (49%), dehydroabietic acid (68%), and 4-nitrocatechol (73%), with significant level of OC (29%), and EC (24%). Low contribution to K^+ (7%) may be attributed to the smoldering phase associated with this factor, K^+ tends to be highly emitted during flaming phase compared to smoldering phase, while levoglucosan conversely tends to be found higher during smoldering than flaming condition (Z. Zhang et al., 2017). Unexpectedly high contributions to Cl^- (31%) may be attributed to the model bias on factors separation as Cl^- from fresh aerosol may partly correlated

well to this source especially during episodic events 2 and 3-1. Softwood burning can be identified by an organic molecular marker, dehydroabietic acid (DHAA), a direct product from softwood burning and a secondary product from oxidation of resin acid in atmospheric condition (B. Yan et al., 2008). DHAA has been used in a number of previous studies regarding biomass burning derived PM to identify softwood burning (Fujii, Tohno, et al., 2015; Leithead et al., 2006; Nguyen et al., 2016; Simoneit, 2002; Viana et al., 2008; Y. Zhang et al., 2013) and aged aerosol (B. Yan et al., 2008). Levoglucosan is well known as a good primary organic aerosol (POA) marker made by biomass burning, while 4-nitrocatechol (4-NC) has been proposed as secondary organic aerosol (SOA) markers from aged aerosol from biomass burning (Bertrand et al., 2018; Finewax et al., 2018; Q. Wang et al., 2022) due to the most abundant content among nitro-aromatic compounds from biomass burning derived PM (Claeys et al., 2012; Iinuma et al., 2016; Y. Wang et al., 2019). 4-NC is produced by the reactions of catechol (a direct emitted product from biomass burning) with OH and NO₃ radicals in the presence of NO_x condition during aging process of aerosol (Finewax et al., 2018). In the current study, we employed DHAA and 4-NC as softwood burning and aged aerosol markers for the first time in Chiang Mai, Thailand.

Overall, the softwood burning (aged aerosol) contributed 12% to PM_{2.5}. During haze episode, this source contributed 31% equivalent to 12 µg m⁻³, while during post-haze contributed negligible at 1% or 0.1 µg m⁻³ to PM_{2.5} mass. Temporal distribution (Fig. 4-8) displays a well associated contribution of this source with two episodic events 2, 3. Backward trajectories during episodic events related to the air mass arriving the sampling site from south to southwest directions during episodic events 2 (EH2) and 3 (EH3-1 & EH3-2) (Fig. 4-12 and 4-13), this may infer to the area where softwoods such as coniferous trees densely situate (Fig. 4-15). It can be observed in fire hotspot maps of EH2, EH3-1, and EH3-3 (Fig. 4-10) displaying highly concentrated in forest area (Fig. 4-15) along the border of Thailand and Myanmar at west - southwest to the sampling site. Scatter plots between levoglucosan/DHAA and Cl-/4-NC ratios (Fig. 4-5 (b)) in

section 4.4.2 shows that those of episodic 3 (EH3-1 and EH3-2) were cluster at lower ratio comparing to those of low haze (LH) and other episodic events, implying contribution of softwood burning and aged aerosol. While those of episodic 2 (EH2) show poor cluster with levoglucosan/DHAA ratio but well cluster with Cl-/4-NC ratio, suggesting that aerosol in EH2 associated more in hardwoods burning but still aged before arriving the sampling site. A previous study conducted during wildfire in Northern California, has also found that DHAA and nitroethanol were categorized in the same timeline factor which mean that they had correlation to a highly extent (Y. Liang et al., 2021). Softwoods burning over Chiang Mai was identified in previous studies by using the ratios of Levoglucosan to its monomers (mannosan and galactosan) or K⁺ (Chantara et al., 2019; W. Song et al., 2022; Thepnuan et al., 2019a).

4.4.4.4 Secondary Sulfate

Secondary sulfate was generally identified by high contribution to NH₄⁺ (75%) and SO₄²⁻ (70%) of the total species mass. Secondary sulfate can be produced from both gas- and aqueous-phase process, however, the formation of secondary sulfate is influenced by relative humidity (RH) and mainly formed by aqueous-phase. Sulfate is generally formed by being oxidized and catalyzed with metals on wet surface of preexisting particles under high RH especially during haze episodes, reported studies in China (W. Li et al., 2011; X. Liu et al., 2015; X. Wang et al., 2012; R. Zhang et al., 2018). While some studies in Southeast Asian countries have shown discrepancy where secondary inorganic aerosol (sulfate and nitrate) contributions were lower in haze than non-haze episodes (Van et al., 2022), for example, in Malaysia (Amil et al., 2016) and Singapore (Tham et al., 2019). Lower contributions of metallic elements such as Pb (40%), V (33%), and As (24%) indicate that secondary sulfate may partly come from industrial or traffic-related emissions (Chansuebsri et al., 2022; Chantara et al., 2012; Pengchai et al., 2009).

In this study, total contribution of secondary sulfate to $PM_{2.5}$ mass accounted for 19% to $PM_{2.5}$ mass, while during haze and post-haze were 17% and 34% (Fig. 4-9), equivalent to $15 \mu g m^{-3}$ and $7 \mu g m^{-3}$, respectively. Temporal distribution of secondary sulfate contribution was typically higher during haze than post-haze episodes (Fig. 4-8) but did not show peaks during episodic events. It is notable that, even the absolute concentration was higher in haze ($15 \mu g m^{-3}$) than post-haze ($7 \mu g m^{-3}$), but the percentage contribution of secondary sulfate during post-haze was significantly higher. This may be attributed to very low contributions by other sources particularly biomass burning and more influences from local residential burning or nearby traffic together with high RH during post-haze (early wet season) favors the formation of secondary sulfate. A long term study (5 years) in the same site location as this study revealed that $(NH_4)_2SO_4$ was the major compound in the study area (Chantara et al., 2012). Many studies have agreed that SO_4^{2-} is a major component in water soluble ions for PM collected in Chiang Mai (Chansuebsri et al., 2022; ChooChuay et al., 2022; H. Y. Huang et al., 2020; Khamkaew, Chantara, Janta, et al., 2016a; Pani et al., 2018; Thepnuan et al., 2019b). Study in source apportionment of PM in Chiang Mai have performed by using principle component analysis (PCA) to identify secondary sulfate; Pengchai et al. (2009) identified secondary sulfate at 6.5%, Khamkaew et al., (2016a) identified contribution at 32% from combustion sources; while ChooChuay et al. (2022) did not clearly identify secondary inorganic aerosol sources, this may be ascribed that secondary inorganic aerosols are potentially found being mixed with other sources rather than a sole source (Saksakulkrai et al., 2023).

4.4.4.5 Mixed dusts, traffic related emissions

This factor is associated with high contributions to several elements: Al (52%), Ti (47%), and Fe (41%) (crustal elements), Cu (60%), and Pb (27%), indicating contributions of both soil and road dusts. Furthermore, significant contribution of many trace metals including Ni (56%), Ce (49%), Ba (44%), V (41%), and Mn (40%), together with a higher contribution of EC (21%) than OC (6%), suggesting contribution of traffic emissions. Therefore, this factor was considered

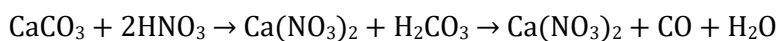
as mixed dusts and traffic-related emissions owing to the complexity in chemical composition of soil/road dust sources, sometimes they were found as being mixed with other sources (Saksakulkrai et al., 2023). Soil dust contains several crustal elements including Al, Si, Na, Ca, K, Mg, Ti and Fe, while road dusts contains those crustal elements and additional heavy metallic elements such as Zn from tyre wear, Sb and Cu from brake wear, and Ba, Zn, and Ca from lubricant oil (Hopke et al., 2020), Mn is used as an additive to improve gasoline rating (Shridhar et al., 2010) and also brake wear (Jain et al., 2020). Pb was an additive in gasoline fuel but no longer being used since its toxicity, thus Pb can be a tracer of non-exhaust traffic source like road dust (Khan, Sulong, et al., 2016; Pant & Harrison, 2012). Ni coupled with V are widely used as markers for oil combustion and road traffic emission (Amil et al., 2016; Cohen et al., 2010a; Fujii et al., 2017; Kayee et al., 2020; Khan et al., 2017; L. Yu et al., 2013). Ba is used to indicate both exhaust and non-exhaust traffic-related sources (Sharma & Mandal, 2017; L. Yu et al., 2013).

Contribution of mixed dust and traffic emissions to PM_{2.5} mass was 16% for a whole period of study. The contributions during haze and post-haze were 15% and 26% accounted for 13 µg m⁻³ and 6 µg m⁻³, respectively. Temporal distribution over both episodes (Fig. 4-8) clearly shows that this source did not vary with the intensity of haze events. Ni and V, as an indication of traffic emission (Kayee et al., 2020), during haze and post-haze show that correlations (R^2) was strong in post-haze ($R^2 = 0.87$) but was weak in haze ($R^2 = 0.43$), as well as several strong correlations ($R^2 > 0.8$) of trace metals (Table SB-3), implying a higher influence of traffic during post-haze period. The following studies in Chiang Mai also reported these sources; Kraisitnitikul et al. (2024) found a small contribution of traffic emission tracers at Mae Hia, Chiang Mai (same as current study's site), Pengchai et al. (2009) identified vehicular emission (13.1%), soil (9%), and road dust (7.8%); Chansuebsri et al. (2022) reported soil dust at 17.3% and 19.8% of two rural sites, and 22.6% at an urban site, ChooChuay et al. (2022) separately identified vehicular exhaust (51.6%) and diesel emissions (10.6%) by making use of 19 PAHs as source-specific markers,

however, dusts were not identified. It worth mentioning here that, in comparison among previous studies, by means of using a comprehensive set of source-specific tracers provide a better separation in source apportionment especially very complex sources such as industries, vehicular exhausts, and mixed dusts.

4.4.4.6 Secondary Nitrate

Likewise secondary sulfate, secondary nitrate is generally identified by high contribution of the factor to NO_3^- ion, it can also be identified with other cations such as NH_4^+ (Jain et al., 2020; Kumar & Sunder Raman, 2020; Y. Song et al., 2006; X. Xu et al., 2018; D. Yang et al., 2018). However, in the condition that NH_4^+ is not sufficient to neutralize SO_4^{2-} and NO_3^- , calcium sulfate or nitrate may be present (Cesari et al., 2019). In this study, NO_3^- and Ca^{2+} of this source contributed very high at 52% and 48% of total NO_3^- and Ca^{2+} , respectively, implying that this secondary nitrate source was in the form of calcium nitrate ($\text{Ca}(\text{NO}_3)_2$). To the best of our knowledge, calcium nitrate was classified as secondary nitrate source for the first time in our present study for Chiang Mai, Thailand. Ca can be found as calcite mineral or calcium carbonate (CaCO_3), a common component in natural dust particles especially mountain areas in the northern Southeast Asia. Ca is also one of the most vulnerable elements that can easily go through several chemical reactions in a polluted environment. Calcium nitrate is not usually found in pure dust particles, it forms in the atmosphere by the chemical transformation of soil dust containing calcium carbonate in acidic condition (nitric acid) (Goel et al., 2020; Naimie et al., 2022). Krueger et al. (2003) observed the conversion of calcium carbonate particle to deliquescent calcium nitrate via reaction with nitric acid (HNO_3) in the first laboratory-based experiment as the following reactions:



Tan et al. (2016) studied the heterogeneous reactions of NO_2 with $\text{CaCO}_3 - (\text{NH}_4)_2\text{SO}_4$ mixtures, they found that NO_2 reacts with CaCO_3 particles can form calcium nitrate under dry and wet conditions and the presence of $(\text{NH}_4)_2\text{SO}_4$ promotes calcium nitrate formation under suitable relative humidity (RH 40% - 60%).

In the present study, the total contribution of secondary nitrate to $\text{PM}_{2.5}$ mass was 5%, the lowest contributor overall. During haze and post-haze this source contributed 5% and 2% (Fig. 4-9), equivalent to concentrations of $4 \mu\text{g m}^{-3}$ and $0.5 \mu\text{g m}^{-3}$, respectively. The correlation between Ca^{2+} and NO_3^- was very strong during haze ($R^2 = 0.91$) but weak during post-haze ($R^2 = 0.51$) (Table SB-2), the average concentration of NO_2 , SO_4^{2-} , and RH during haze and post-haze were 15 and 7 ppb, 7.3 and $3.4 \mu\text{g m}^{-3}$, and 47% and 67%, respectively. These conditions were corresponding to the study by Tan et al. (2016) as mentioned above, and were favorable for the formation of calcium nitrate during haze episode. Temporal distribution of secondary nitrate in Fig. 4-8 shows a higher contribution over haze episode and reached the peaks in episodic event (EH1) and (EH3-1). We suspected that soil dust containing calcite mineral (CaCO_3) was burned from the ground and emitted into the atmosphere during haze events. Moreover, forest fire may elevated during episodic haze and emitted a substantial of Ca from leaf litter burning (Akbari et al., 2021). Secondary nitrate can also be formed by the reaction of sea salt such as NaCl with nitric acid (Naimie et al., 2022), however, Chiang Mai locates in the continent approximately 300 km. away from the sea so the contribution of sea salt in this case was negligible. Previous studies in Chiang Mai usually reported this source as mixed secondary inorganic aerosols (SIA, mixture of sulfate and nitrate) (Chansuebsri et al., 2022; ChooChuay et al., 2022), thus it is not applicable in regards of comparison since no study reported a sole secondary nitrate source. Nevertheless, secondary nitrate was observed in other countries, for example, Bhopal, India (4.1%) (Kumar & Sunder Raman, 2020), Hong Kong (13%) (X. H. H. Huang et al., 2014), Greece (9%) (Mantas et al.,

2014), Beijing, China (14%) (Y. Song et al., 2006), New York, USA (6.3%) (Y. Wang et al., 2012), and Seoul, Korea (20.9%) (Heo et al., 2009).

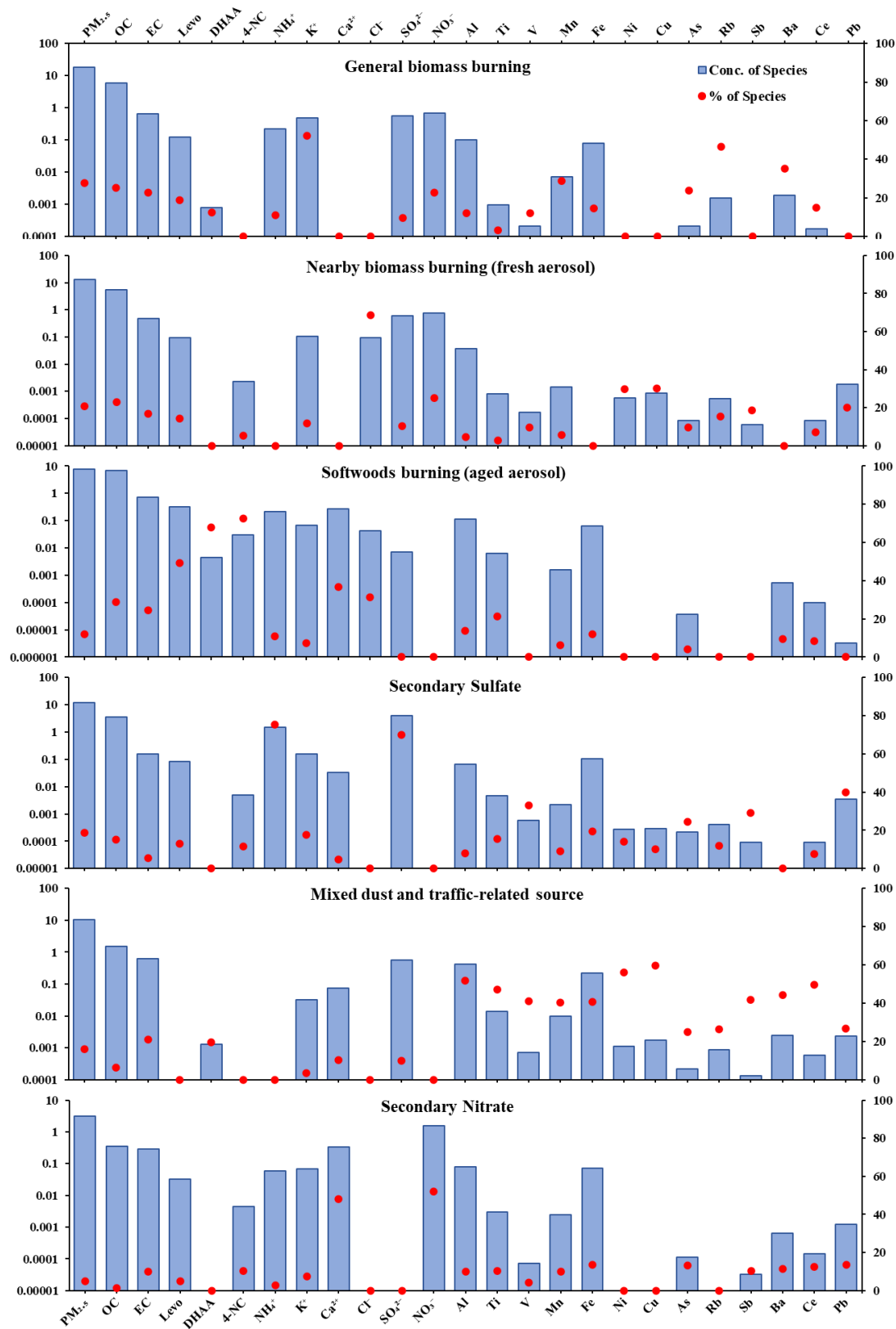


Fig. 4-7 Chemical source profiles. Bar charts represent concentrations of components (left axis) and red dots represent percentage contribution of components (right axis).

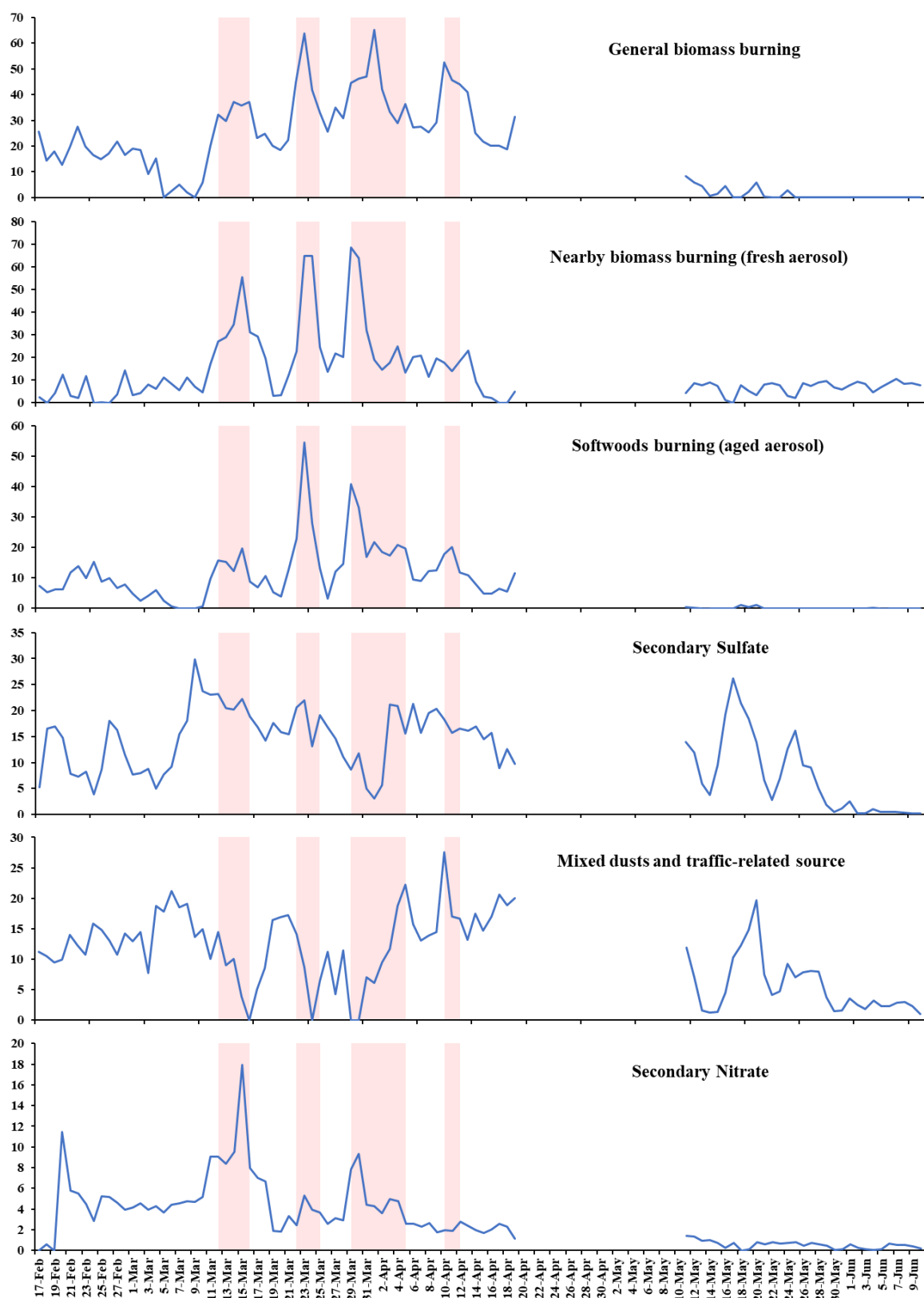


Fig. 4-8 Factor contribution concentration ($\mu\text{g m}^{-3}$) during haze and post-haze. Pink highlighted areas are episodic events ($\text{PM}_{2.5} > 100 \mu\text{g m}^{-3}$).

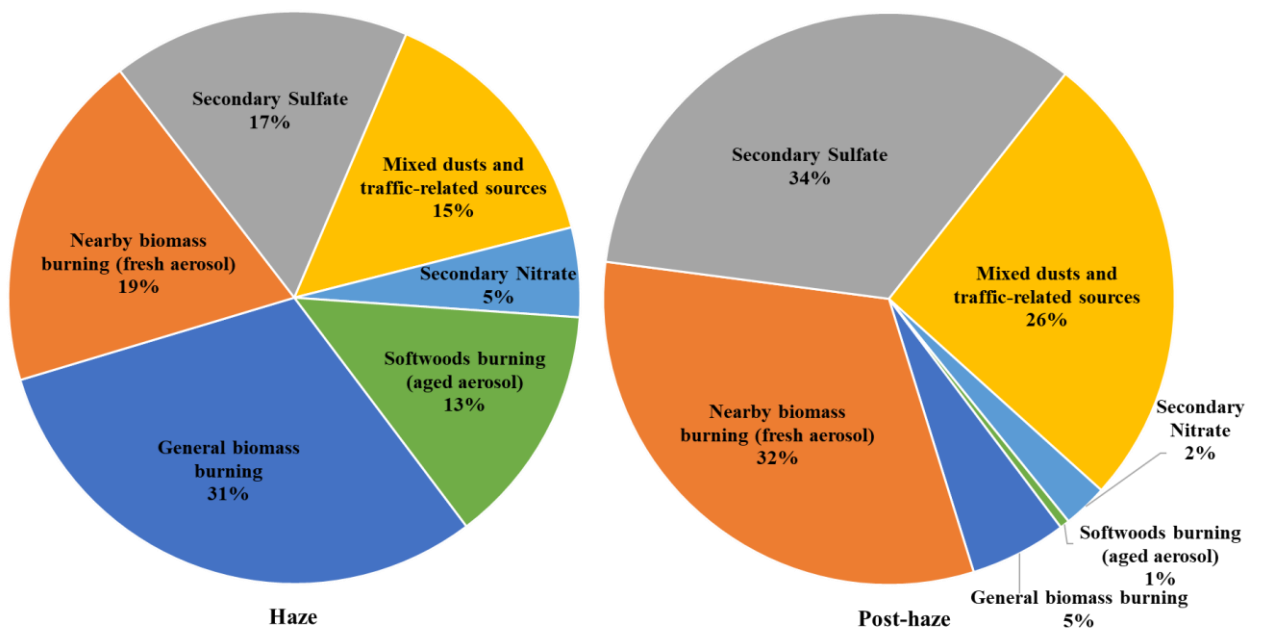


Fig. 4-9 Percentage contributions of 6 identified sources during haze (left) and post-haze (right).

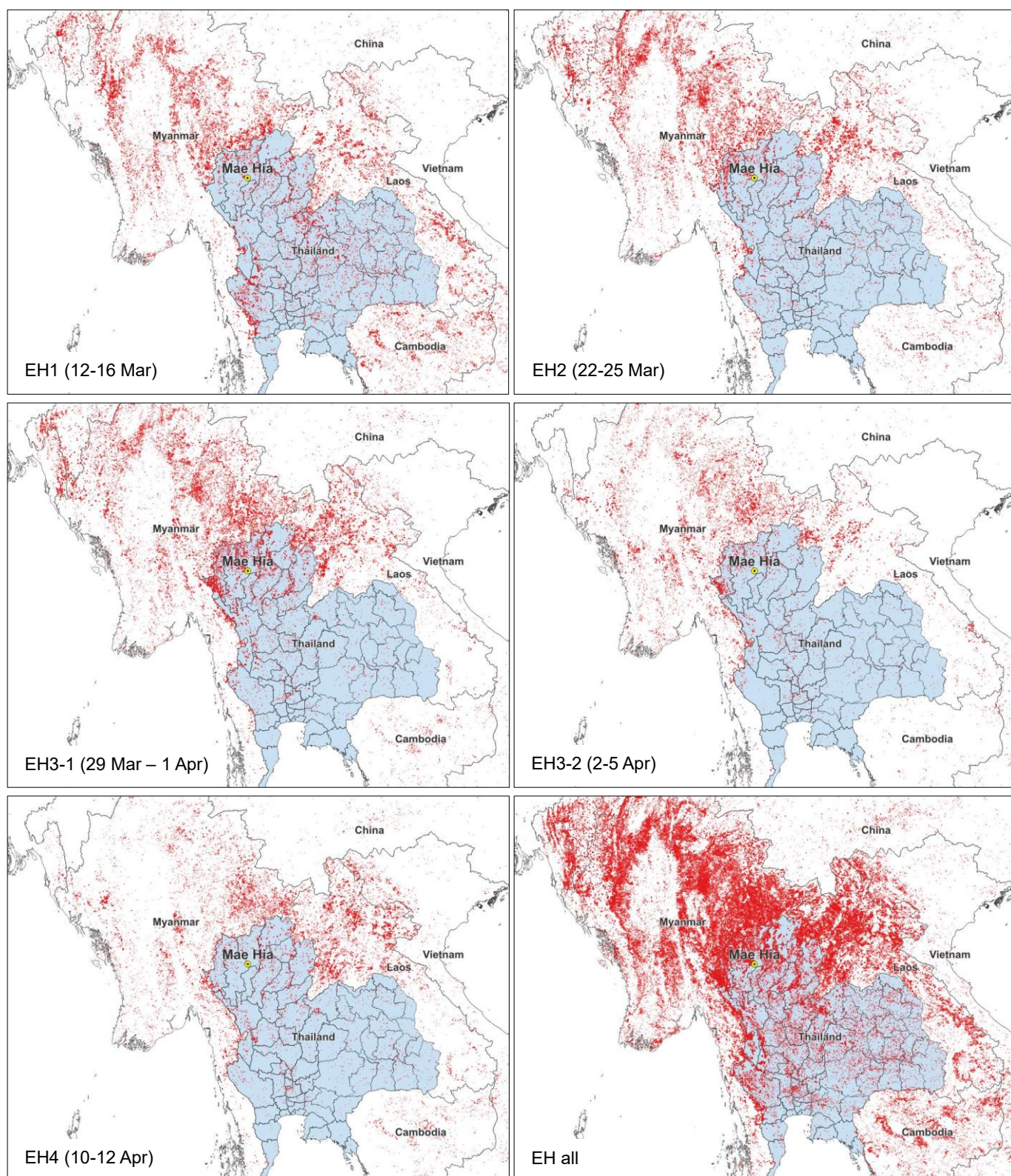


Fig. 4-10 Regional fire hot spot maps during episodic haze events (EH) ($\text{PM}_{2.5} > 100 \mu\text{g m}^{-3}$), and overall EH (EH all). Information from <https://firms.modaps.eosdis.nasa.gov/>. Data source: SUOMI VIIRS C2

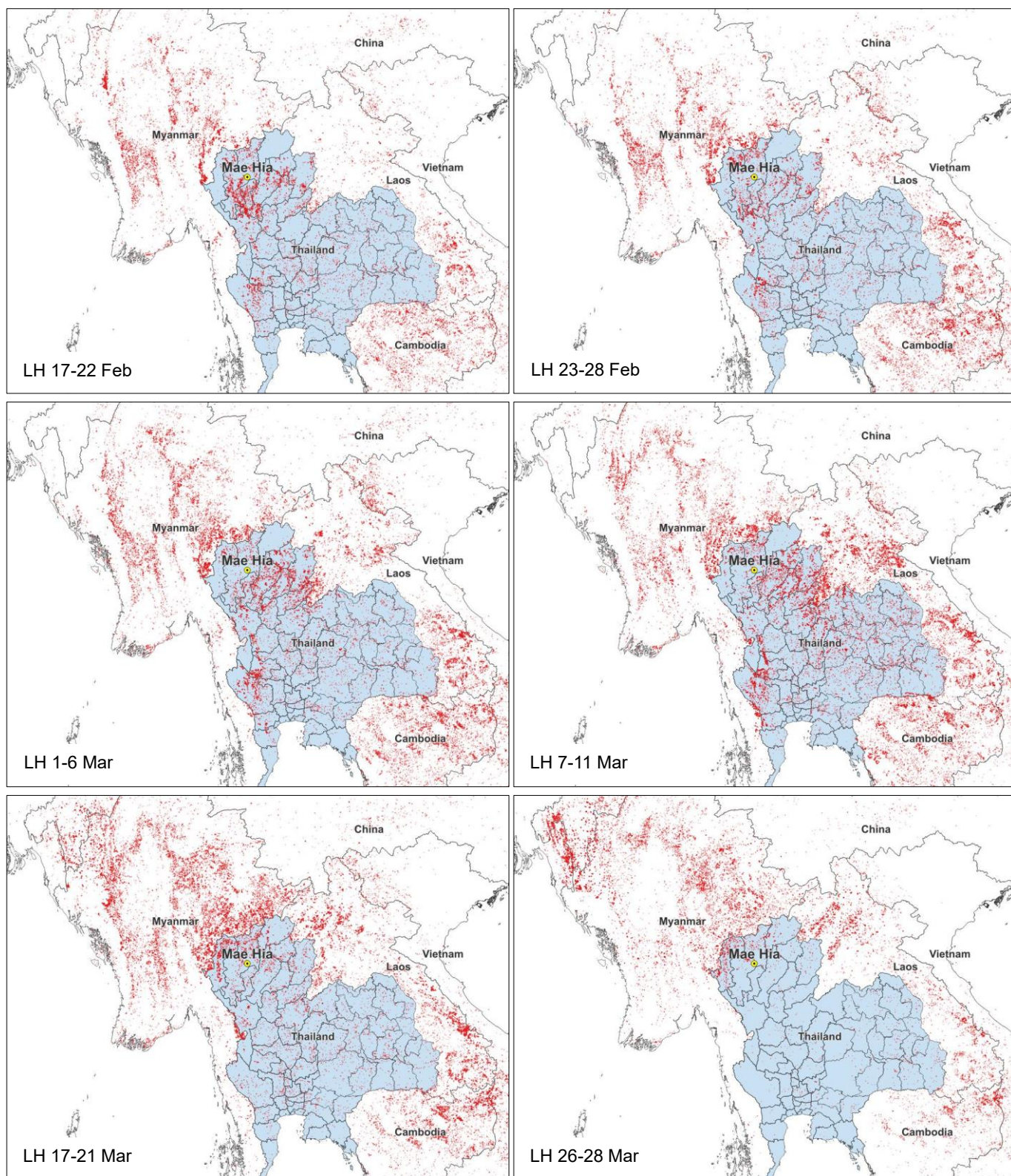


Fig. 4-11 Reginal fire hot spot maps during low haze events (LH) ($PM_{2.5} < 100 \mu g m^{-3}$). Information from <https://firms.modaps.eosdis.nasa.gov/>. Data source: SUOMI VIIRS C2

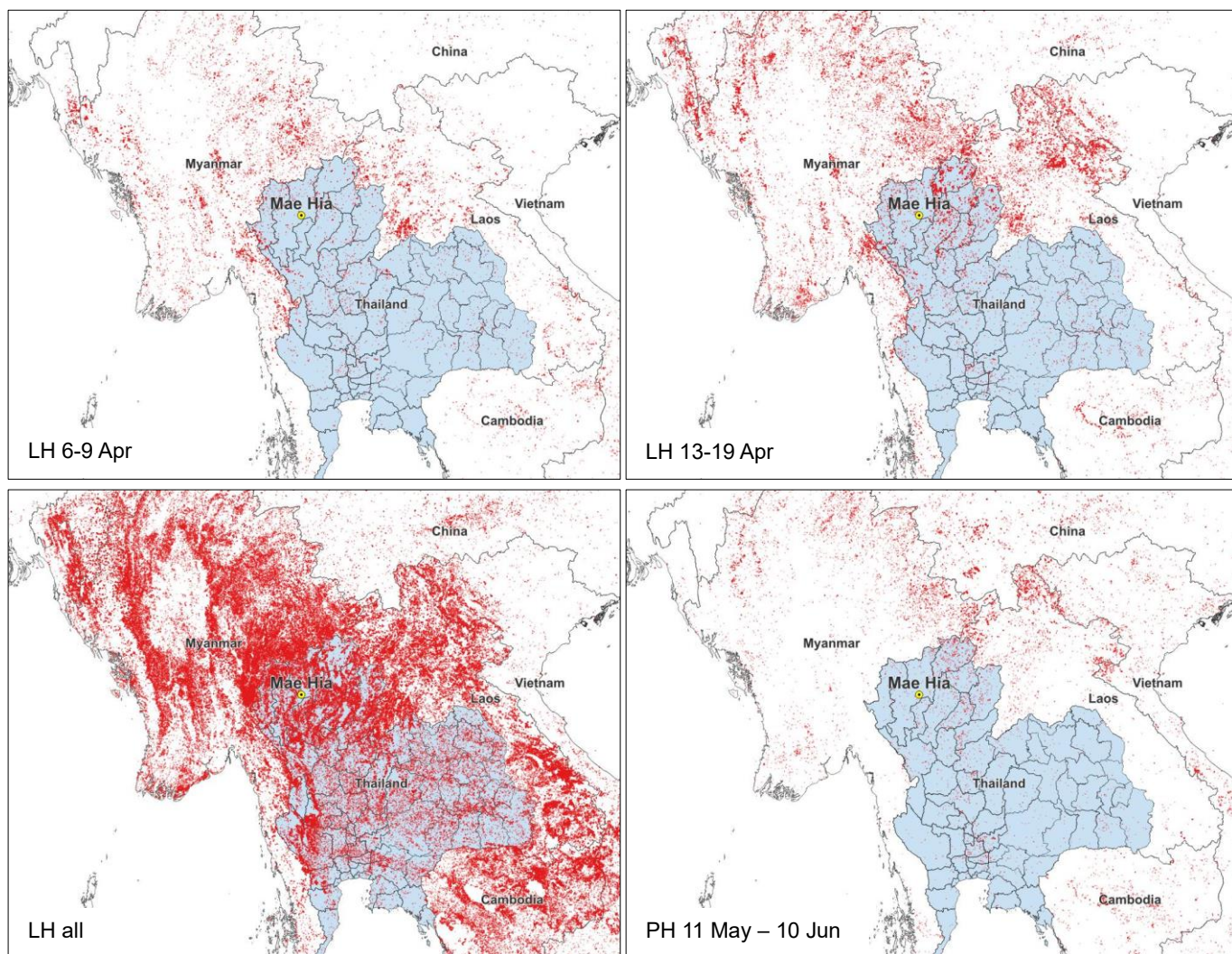


Fig. 4-11 (Continue) Regional fire hot spot maps during low haze events (LH) ($\text{PM}_{2.5} < 100 \mu\text{g m}^{-3}$), overall low haze (LH all), and post-haze (PH). Information from <https://firms.modaps.eosdis.nasa.gov/>. Data source: SUOMI VIIRS C2

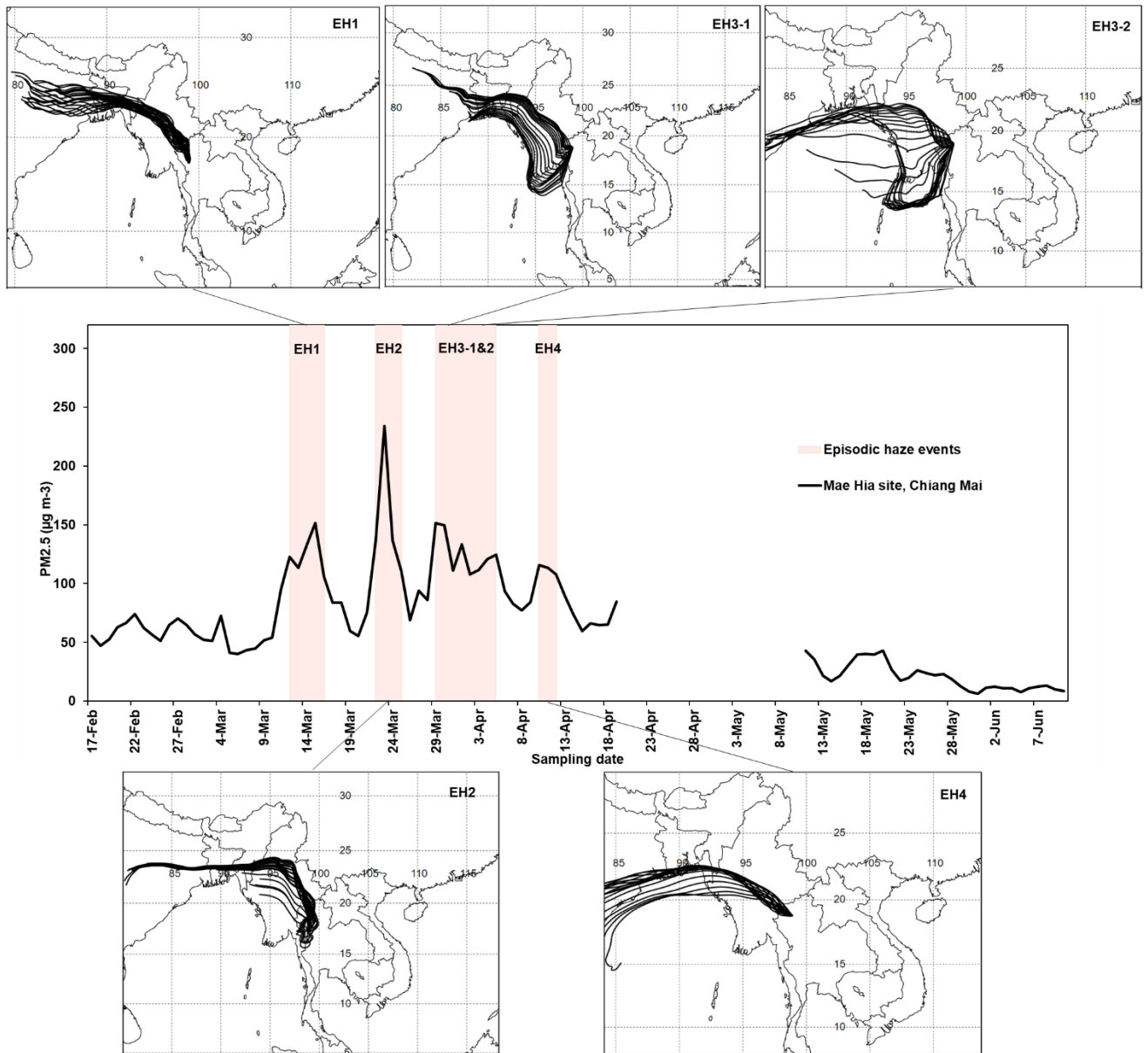


Fig. 4-12 72-hr Backward trajectories every 3-hr interval in different episodic haze events

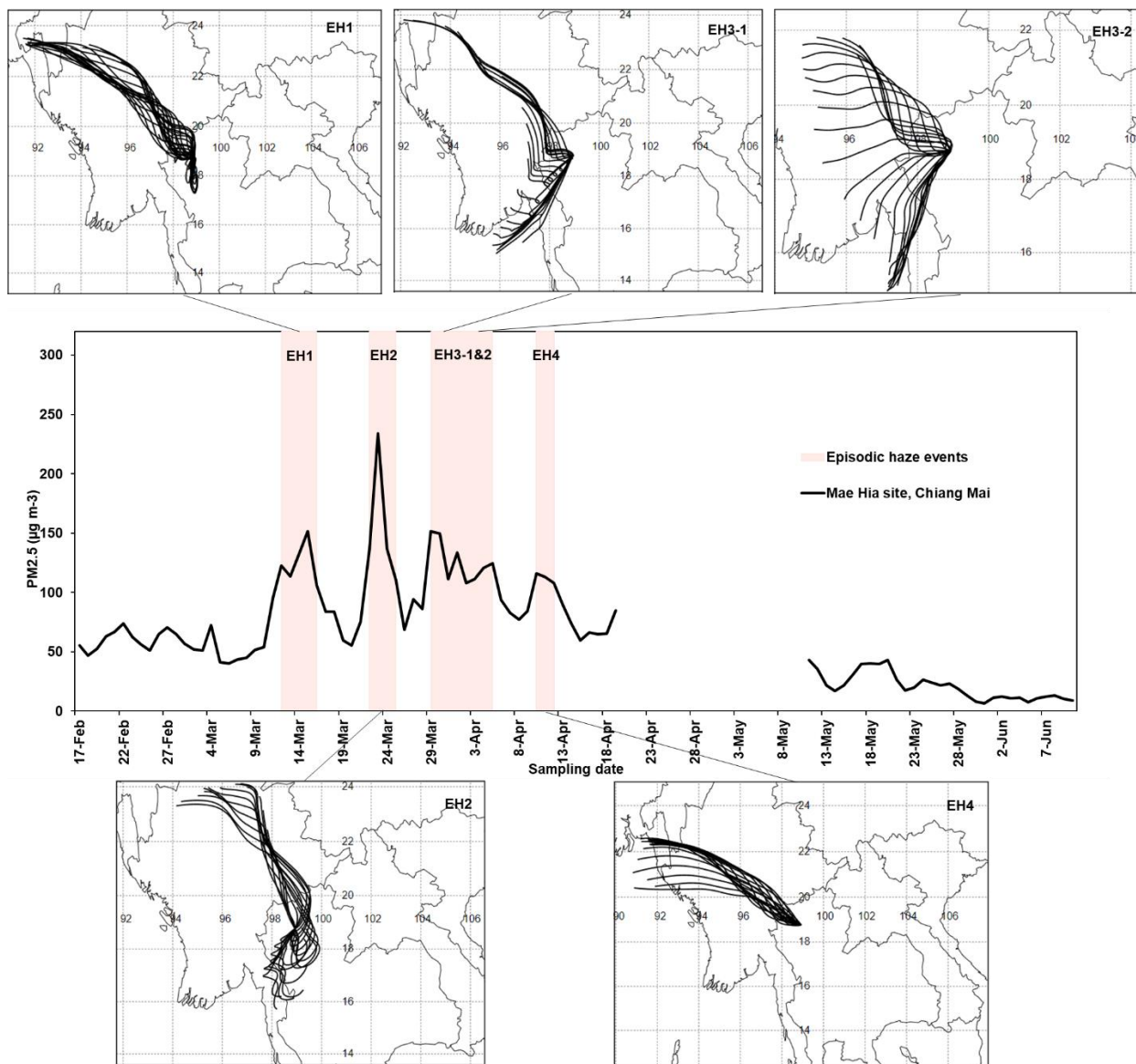


Fig. 4-13 48-hr Backward trajectories every 3-hr interval in different episodic haze events

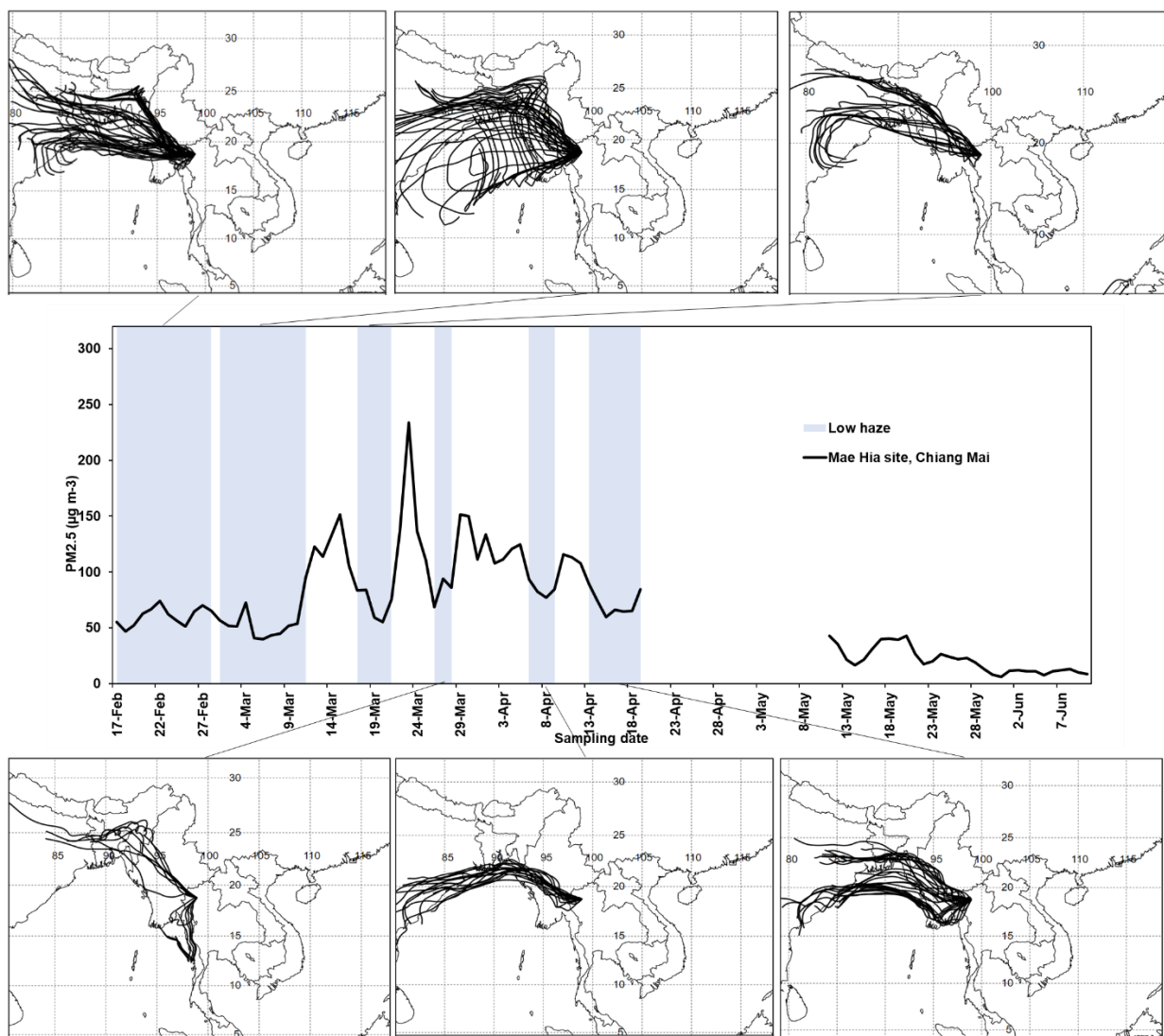


Fig. 4-14 48-hr Backward trajectories every 3-hr interval during low haze

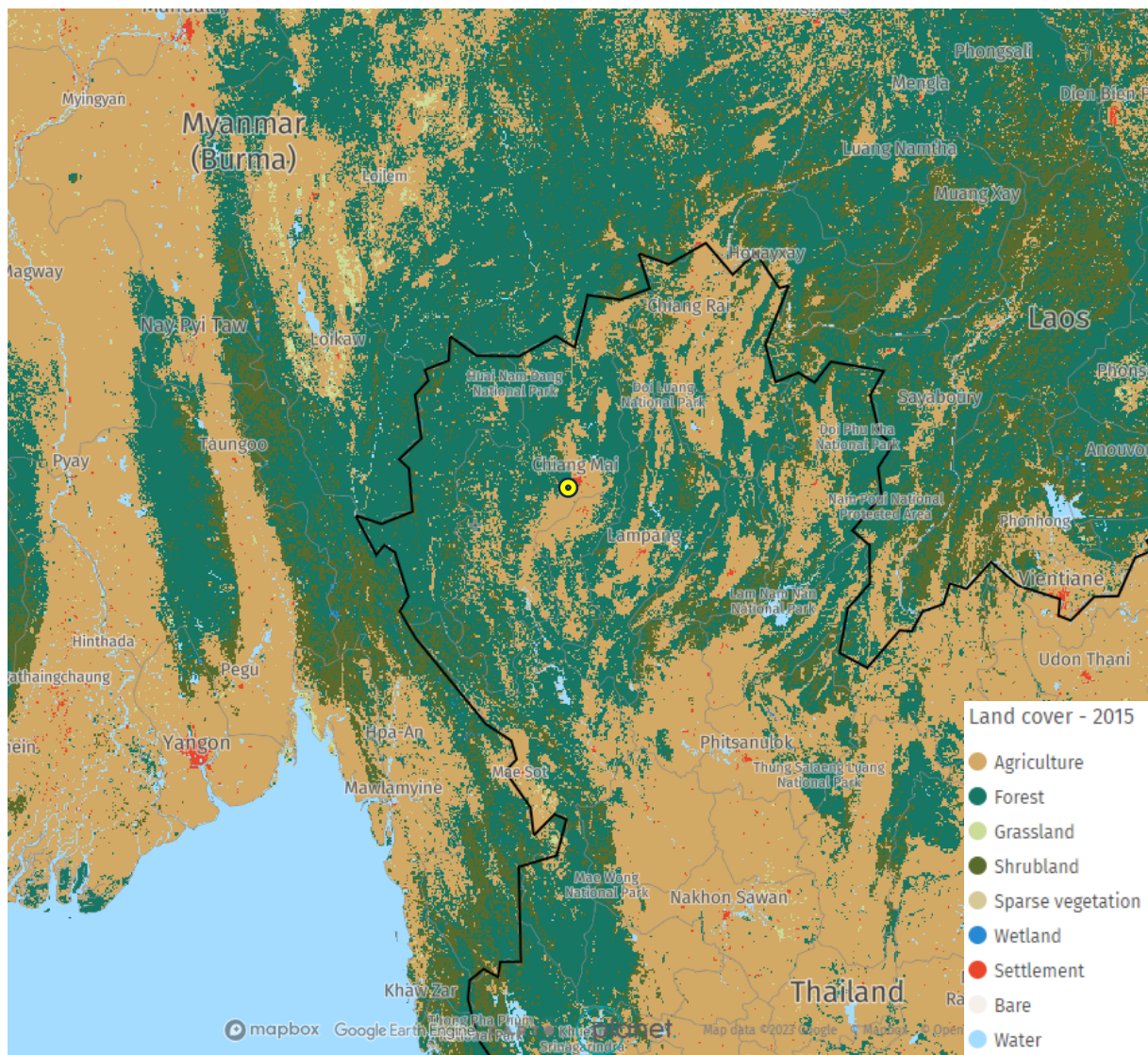


Fig. 4-15 Land use cover map year 2015 over northern Thailand and neighboring countries.
Source: <https://www.globalforestwatch.org/>

4.5 Conclusion

This study performed characterization of chemical components of PM_{2.5}, PM_{2.5} mass reconstruction and identified potential source of PM_{2.5} by positive matrix factorization model (PMF) associated with their original pathway through long-range transport of air mass by back trajectory analysis. The results of chemical characteristics showed that OC was the greatest contributor with OC3 dominated in carbonaceous species due to biomass burning, the ratios of soot-EC/char-EC reveal biomass burning in haze period, sulfate, nitrate and ammonia (SNA) were the three major components in WSIs group. The ratios between organic tracers and ions; Levoglucosan/K⁺, Levoglucosan/Dehydroabietic acid (DHAA), Cl⁻/4-Nitrocatechol (4-NC) were very useful for identification of fresh and aged aerosol and different type of biomass burning. Crop residuals burning associated with low haze episode while softwoods burning associated with smoldering phase during episodic haze events (PM_{2.5} > 100 µg m⁻³). Mass closure illustrated OM was the largest contributor to PM_{2.5} mass followed by SNA ions, soil dust and EC during both haze and post-haze periods. Source apportionment by PMF model identified 6 sources of PM_{2.5} including general biomass burning, nearby biomass burning (fresh aerosol), softwoods burning (aged aerosol), secondary sulfate, mixed dusts and traffic-related sources and secondary nitrate. Three biomass burning sources contributed approximately 60% during haze period, while nearby biomass burning, secondary sulfate and mixed dust and traffic-related sources contributed relatively similar at approximately 30% during post-haze period. The finding also highlights the usefulness of applying organic tracers into chemical characterization and source apportionment which provides an insight aspect regarding biomass burning for the future study in Thailand and other countries in Southeast Asian region.

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CHAPTER FIVE:

FINE PARTICULATE MATTER (PM_{2.5})-

BOUND POLYCYCLIC AROMATIC

HYDROCARBON (PAHs) IN NORTHERN

THAILAND AND THEIR SOURCE-SPECIFIC

CANCER RISK ASSESSMENT

Fine particulate matter (pm_{2.5})-bound polycyclic aromatic hydrocarbon (PAHs) in northern Thailand and their source-specific cancer risk assessment

5.1 Abstract

Polycyclic aromatic hydrocarbons (PAHs) in Fine particulate matter (PM_{2.5}) are a significant risk factor to human health. This study aims to characterize PAHs in fine particles during haze (17 Feb to 19 Apr) and post-haze (11 May to 10 Jun) periods in northern Thailand in 2019 and estimate their lifetime cancer risks. The results show that the concentration of PAHs typically varied along with PM_{2.5} mass, but the total measured PAHs (Σ PAHs) are anti-correlated with PM_{2.5} mass. Σ PAHs mass during episodic haze, low haze and post-haze periods were 13.2 ± 3.6 , 9.7 ± 1.3 and 7.9 ± 2.2 ng m⁻³, respectively. High molecular weight PAHs had strong correlations with levoglucosan, a tracer for biomass burning aerosol, during episodic haze periods (PM_{2.5} > 100 μ g m⁻¹) while lower molecular weight PAHs had moderate correlations with PM_{2.5} mass during low haze. Lifetime cancer risk (LCR) of fine PAHs ranged in moderate risk with values of 4.21×10^{-5} and 0.67×10^{-5} for haze and post-haze periods, respectively. Source specific LCR during haze periods were highest from diesel exhaust (1.35×10^{-5}) followed by softwood burning (1.23×10^{-5}), biomass burning (0.60×10^{-5}), mixed dusts (0.51×10^{-5}), gasoline (0.26×10^{-5}). Diesel emissions, biomass burning and softwoods burning were the main contributors of sources specific LCR during the haze period, while diesel and gasoline exhaust were the main contributors for the post-haze period. Our results suggest that reducing diesel exhaust and softwood burning are the most effective ways to mitigate cancer risks caused by fine particle PAHs.

5.2 Introduction

Haze problem in northern, Thailand, has been considered as a challenging problem due to its annual recurrence during local dry season (Saksakulkrai et al., 2023; Van et al., 2022). Fine particulate matter (PM_{2.5}), a significant air pollutant, is substantially emitted by the haze phenomenon which, simultaneously, bound with several harmful chemical components, typically polycyclic aromatic hydrocarbons (PAHs), a group of aromatic hydrocarbons formed during incomplete combustion of both natural and anthropogenic sources. PAHs bound to PM_{2.5} allows to be inhaled deep into the lower respiratory system and can then be absorbed by lung tissue leading to several adverse health issues such as acute (eye irritation, inflammation and nausea) and chronic effects (damage on DNA, developing of lung cancer) (Franco et al., 2008; Kim et al., 2013).

There are more than a hundred different PAHs exist in the complex of environment, according to U.S Environmental Protection Agency (US-EPA), 16 PAHs are classified as priority pollutants including naphthalene (Nap), Acenaphthene (Ace), Acenaphthylene (Acy), Anthracene (Ant), Fluorene (Flu), Phenanthrene (Phe), Fluoranthene (Flua), Pyrene (Pyr), Benzo(a)anthracene (BaA), Chrysene (Chry), Benzo(b)fluoranthene (BbF), Benzo(k)fluoranthene (Bkf), Benzo(a)pyrene (BaP), Dibenzo(a,h)anthracene (DahA), Indeno(1,2,3-cd)pyrene (Ind) and Benzo(ghi)perylene (BghiP). They can also be classified into two group: low molecular weight compounds (LMW) (less than four benzene rings), and high molecular weight compounds (HMW) (four or more benzene rings) (Kim et al., 2013). PAHs are generally produced by three major source type in the environment namely pyrogenic (incomplete combustion of organic compounds at high temperatures 350°C to more than 1200°C with an absence of oxygen or low oxygen conditions such as engine combustion, oil combustion and wood in fireplace or forest), petrogenic (maturation of crude oil without combustion, for example oil spill or evaporation) and biological (production from some type of plants and microorganisms during the degradation of organic

matters) (Abdel-Shafy & Mansour, 2015). Previous studies have demonstrated that LMW PAHs with higher vapor pressure are generally dominant in gas phase while high HMW PAHs with low vapor pressure are dominant in particle phase (Dat & Chang, 2017; Insian et al., 2022; Kongpran et al., 2021; Lv et al., 2016; Mahasakpan et al., 2023). Although, in regard to toxic equivalent (TEQ_{BaP}) to the highest carcinogenic rank PAH, Benzo(a)pyrene, HWM PAHs have higher TEQ compared to those of LMW. Therefore, HMW bound in particulate phase are usually more concerned due to their contribution to a higher toxicity in airborne particles. Moreover, PAHs are generally associated with different size of airborne particulate matters, studies on particulate matter size distribution of PAHs have demonstrated a similar finding that majority of PAHs were found increase in concentration with decrease in particle size especially fine mode of particles (usually diameter smaller than 2-3 micron) (X. Bi et al., 2005; Gupta et al., 2011; G. Shen et al., 2010; J. Zhou et al., 2005).

Besides PAHs, PM-bound with heavy metals has also been investigated in as an important toxic components together with PAHs (Ahmad et al., 2022; C. Bi et al., 2020; Pandey et al., 2013; Tian et al., 2022; Wu et al., 2022). Hazardous metals such as Cd, Ni, Cr, Co, Pb and As have been received attention as cancer-causing elements from several previous studies regarding inhalation risk assessment of airborne particles (Alias et al., 2020; F. Guo et al., 2022; J. Liu et al., 2019; Qin et al., 2020; X. Wang et al., 2018). Heavy metals can be emitted from biomass burning (Kayee et al., 2020; Pongpiachan, Wang, Chetiyakornkul, et al., 2022; Wu et al., 2022) and considerably from anthropogenic emissions such as traffic-related sources (exhaust and non-exhausts emission) and industry (Alias et al., 2020; Fang et al., 2021; R. H. Yan et al., 2022). Recent studies on health risk assessments have integrated a source apportionment receptor model, Positive Matrix Factorization (PMF), to examine source-specific cancer and non-cancer risks (Sulong et al., 2017; Taghvaei et al., 2018; Tian et al., 2022). The results of source-specific cancer risk assessments can help prioritize mitigation actions for policymakers by identifying the major contributors to $PM_{2.5}$ -

related cancer risks, regulators and policymakers can develop targeted strategies to reduce emissions from specific sources.

The hazardous heavy metals coupled with PAHs applied with source-specific cancer risk assessment approach are then very useful for the area where haze is prominent like northern Thailand. However, previous studies in Thailand have conducted health risk assessment by only applying a sole set of either PAHs (Insian et al., 2022; Kongpran et al., 2021; Pengchai et al., 2009; Pooltawee et al., 2017; Wiriyaa et al., 2013a) or heavy metals (Kawichai, Bootdee, et al., 2022; Sakunkoo et al., 2022) and, to the best of our knowledge, there has been no study applied source-specific risk assessment in this particular area. Therefore, in order to address this research gap, this study aims to perform characterization of 16 main US-EPA PAHs and to carry out the cancer risk assessment of PM_{2.5} bound PAHs and heavy metals in the comparative aspects between haze and post-haze and between source-specific outcomes.

5.3 Methodology

5.3.1 PM_{2.5} site and sampling collection

As described in 3.1 and 3.3.

5.3.2 Chemical analysis

5.3.2.1 PAHs and heavy metals analysis

As described in 3.4.4 and 3.4.5

5.3.2.2 Quality assurance and quality control

As described in 3.4.6

5.3.3 PAHs and metals source apportionment by PMF model

Source apportionment of PM_{2.5} was performed by employing PMF model version 5.0 developed by United States Environmental Protection Agency (US EPA). Mass balance can be simply written from chemical species in n number of samples and p independent sources as following equation (Hopke, 2016):

$$x_{ij} = \sum_{k=1}^p g_{ik} C_{kj} + e_{ij}$$

where x_{ij} is jth chemical species concentration in ith sample, C_{kj} is concentration of jth chemical species from kth source, g_{ik} is contribution of kth source in ith sample, e_{ij} is residual which not fit by the model in ith sample and jth species. Source-specific marker species were also used to achieve better separation, OC, EC, Levoglucosan, dehydroabietic acid (DHAA), SO_4^{2-} , NO_3^- , NH_4^+ , key crustal elements (Al, Ti and Fe), and some trace elements (Cu, Zn, Pb, Mn, Ba and Pb) were taken as input for PMF model.

PAHs and heavy metals concentrations (C_{kj}) obtained from resolved solution of PMF model were used to estimate lifetime cancer risk with which will be described in the next section.

5.3.4 Health risk assessment

5.3.4.1 Benzo(a)pyrene toxicity equivalent concentration (TEQ_{BaP})

To assess the carcinogenic of PAHs, toxicity equivalent concentration to BaP is proposed. The concentrations of PAH congeners are converted to TEQ based on their relative toxicities to BaP as equation

$$TEQ_{BaP} = \sum(PAH_j \times TEF_j) \quad (1)$$

where PAH_i represents the concentration of j PAH individual congener and TEF_j represents the toxic equivalent factor of j PAH. Each PAH has a lower toxicity level and will be multiplied by different TEF values as proposed by Nisbet and LaGoy, (1992). Eq. (1) is then expressed as

$$TEQ_{BaP} = 0.1(BaA + BaF + BkF + Ind) + 0.01(Ant + BghiP + Chry) + 0.001(Ace + Acy + Flua + Flu + Nap + Phe + Pyr) + DahA + BaP$$

5.3.4.2 Chronic daily intake (CDI)

A carcinogenic health risk refers to the likelihood of an individual developing cancer of any kind due to long-term exposure to substances or factors that have the potential to cause cancer. According to US EPA, (2009), chronic daily intake of j species is estimated by

$$CDI_j (\mu kg^{-1} day^{-1}) = \frac{C_{kj} \times IR \times ED \times Ef}{BW \times AT}$$

where C_{kj} is concentration j species (PAHs and heavy metals (Co, Cr, Ni, As, and Cd)) from k source (for source-specific assessment), IR is inhalation rate (average for adult is 16.2 m³ day⁻¹) (Tian et al., 2022), ED is exposure duration (average exposure is considered as lifetime of 70 years) (US

EPA, 2009), Ef is exposure frequency (90 and 30 days year⁻¹ for haze and post-haze periods, respectively), BW is body weight (average weight for adult is 65.7 kg)(Tian et al., 2022) and AT is averaging time (ED x 365 days).

It is recommended that bioavailability or bioaccessibility concentration of toxic substances are used in regard to health risk assessments. Bioavailability and bioaccessibility of toxic substances associated with airborne particulate matters (PM) are different depending on several factors such as concentrations, seasons, study areas, sources, and other components in PM which toxic substance can be reacted with (Y. Li et al., 2019; Luo et al., 2021; Wang et al., 2021; Xie et al., 2019; Zhao et al., 2021). Therefore, in line with prior research, evaluating the bioavailability of hazardous substances in PM is necessary in vitro studies, such as simulated epithelial lung fluid assessments. However, due to certain constraints within this study, such evaluations were not conducted, and as of now, there are no published representative values specific to Thailand. Consequently, this study relied on the total concentrations of polycyclic aromatic hydrocarbons (PAHs) and heavy metals as an upper threshold for health risk assessment. The knowledge gap for Thailand lies in the unexplored realms of bioavailability and bioaccessibility of PAHs and heavy metals.

5.3.4.3 Lifetime cancer risk (LCR)

Lifetime cancer risk can be estimated by multiplying chronic daily intake (CDI) with inhalation cancer slope factor CSF_j of j species.

$$LRC_j = \sum_i^j (CDI_i \times CSF_i)$$

Generally, lifetime cancer risk at 1 chance in 1 million (10^{-6}) is threshold value of the risk, LRC that is below 10^{-6} is considered as negligible risk, while between 10^{-6} and 10^{-4} is moderate risk and more than 10^{-4} is high risk of cancer, respectively. The cancer slope factor for each species was calculated as described in supplementary section SA.

5.4 Results and discussion

Support information of this chapter is in supplement C

5.4.1 Concentrations of PAHs in different sampling periods

Temporal distributions concentration of individual PAHs during haze (17 February to 19 April) and post-haze (11 May to 10 June), 2019, are displayed in Fig. 5-1 together with $PM_{2.5}$ concentration and the ratio between daily total PAHs to $PM_{2.5}$ mass ($\sum PAHs/PM_{2.5}$, $\times 10^5$ and $\times 10^4$ for haze and post-haze, respectively). Overall, the distribution of $\sum PAHs$ concentrations shows a similar trend in variation with the $PM_{2.5}$ mass during haze but does not show this characteristic during post-haze. However, the ratio of $\sum PAHs/PM_{2.5}$ show a decreasing trend as the $PM_{2.5}$ mass increase which was noticeable in both haze and post-haze periods.

Table 5-1 shows statistical data on average concentrations of individual PAHs and $PM_{2.5}$ mass. 14 of 16 PAHs were detected during haze period (fluorene and acenaphthene not detected) while 13 of 16 PAHs were detected in post-haze (except naphthalene, fluorene and acenaphthene not detected). $\sum PAHs$ mass during heavy haze, low haze and post-haze were 13.2 ± 3.6 , 9.7 ± 1.3 and 7.9 ± 2.2 $ng\ m^{-3}$, respectively.

$\sum PAHs$ concentration determined in this study is higher than most of the previous studies (see Table 5-2) except by a study PAHs analyzed on PM_{10} (Wiriya et al., 2013a) and a study of 9 urban sites over northern Thailand (Kongpran et al., 2021). These studies agree that $\sum PAHs$

during haze periods were notably higher than non-haze periods. Comparing to other countries, Σ PAHs in this study was substantially lower than PAHs in megacity such as Beijing, China (Gao et al., 2016) and Delhi, India (D. P. Singh et al., 2011), but higher than the Σ PAHs reported from Kuala Lumpur, Malaysia (Jamhari et al., 2014b; Khan et al., 2015) and Cordoba, Argentina (Amarillo et al., 2017).

The percentage contributions of individual PAHs and their different number of aromatic rings to total PAHs between episodic haze events ($PM_{2.5} > 100 \mu g m^{-3}$), low haze ($PM_{2.5} < 100 \mu g m^{-3}$) and post-haze are illustrated in Fig. 5-2. Overall, 4-rings PAHs contributed the greatest mass followed by 5-rings, 6-rings and 3-rings PAHs. It was clearly observed that 5- and 6-rings PAHs had the highest contribution during episodic hazes (54.7%) followed by low haze (48.8%) and post-haze periods (43.2%). According to a chamber study regarding PM-bound PAHs derived from the combustion of several types of biomass collected in northern Thailand, higher molecular weight PAHs (4-6 rings) are proportionally higher than those of low molecular weight PAHs (Wiriya et al., 2016). This characteristic has been reported by several field experiment studies associated with biomass burning derived PM (Rajput et al., 2011, 2014; H. L. Wang et al., 2008; Wiriya et al., 2013a; Yabueng et al., 2020). PAHs are also associated with vehicular exhaust, including both gasoline and diesel engine emissions. Emission from diesel engines is associated with low to medium molecular weight PAHs, while the emission from gasoline trends to associate with high molecular weight PAHs. Previous studies reported the contribution of PAHs to $PM_{2.5}$ emitted from diesel vehicles were at 59%, 38%, and 2.9%, while the contribution of PAHs to $PM_{2.5}$ emitted from gasoline vehicles were at 66.6%, 18.7%, and 14.42% for 2 – 3, 4, 5 – 7 rings PAHs, respectively. (Lin, Li, Amesho, et al., 2019; Lin, Li, Shangdiar, et al., 2019) The contributions of Ant, Phe, Flua, Pyr, BaA, Chry, BkF, BaP and DahA to total PAHs follow this pattern: post-haze > low haze > episodic haze, while those of the BbF, Ind and BghiP follow a different variation: episodic haze >

low haze > post-haze. Similarly, Insian et al. (2022) reported that BbF and BghiP were the greatest contributors of total PAHs in Chiang Mai, northern Thailand, during haze period (Feb – Apr 2019).

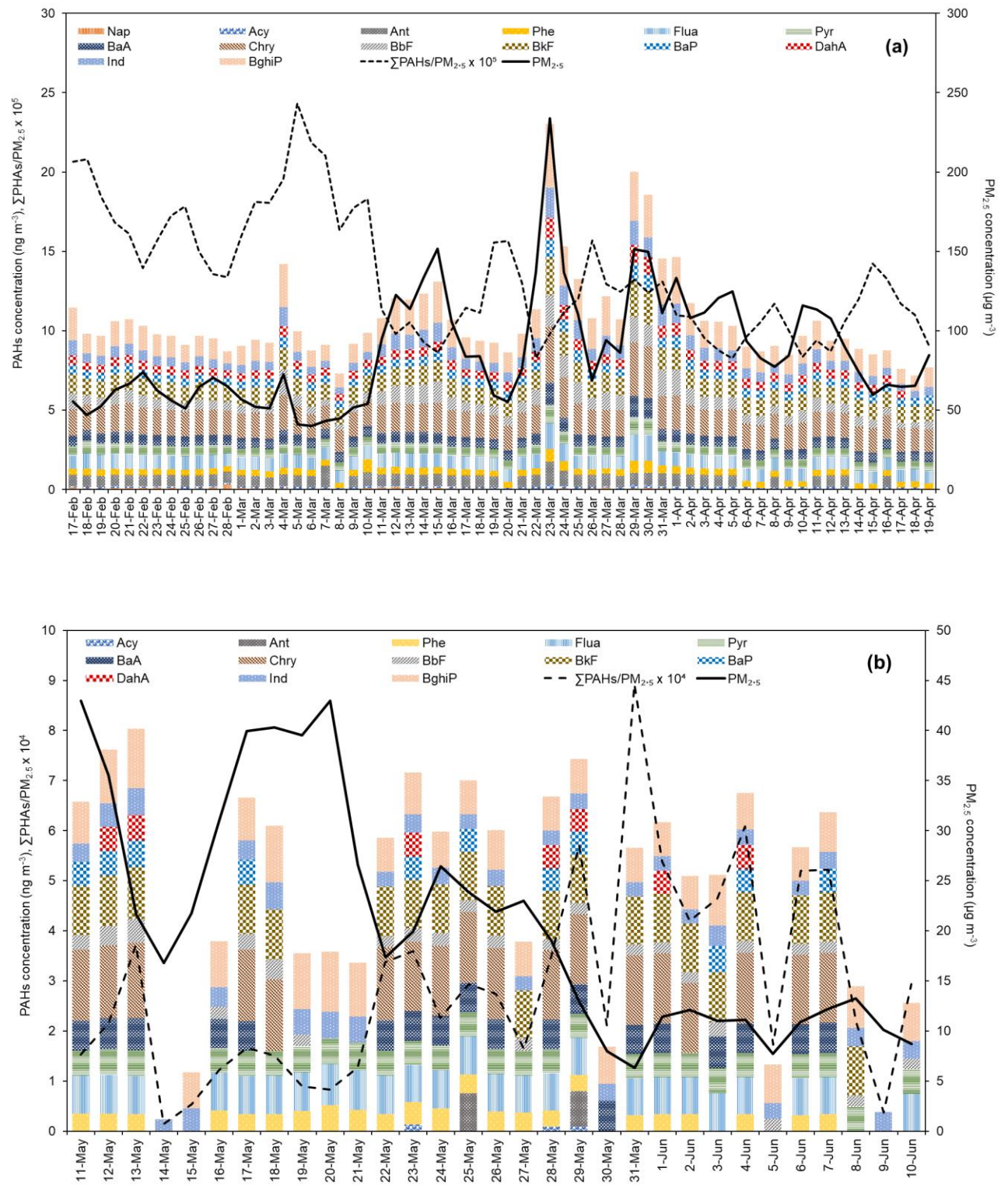


Fig. 5-1 Temporal variations of daily concentrations of individual PAH, PM_{2.5}, and total PAHs to PM_{2.5} ratios during haze (a) and post-haze periods (b)

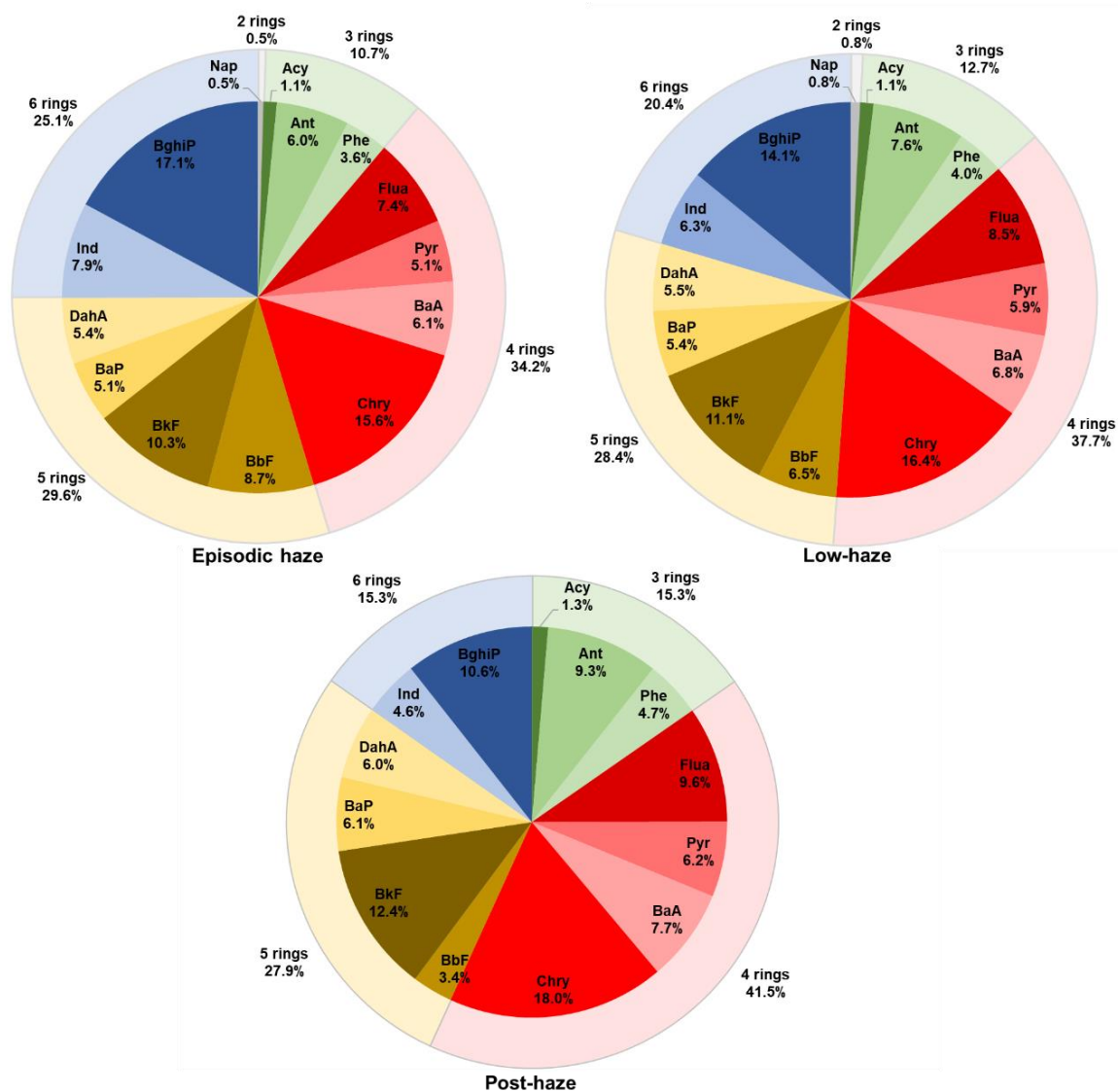


Fig. 5-2 Percentage contribution of individual PAHs and of different number of aromatic rings to total PAHs in episodic haze, low haze and post-haze.

Table 5-1 Average concentration and standard deviation of PM_{2.5} and individual PAHs during episodic haze (PM_{2.5} > 100 µg m⁻³), low haze (PM_{2.5} < 100 µg m⁻³), entire haze, post-haze and entire study periods

	Abbreviation	rings	Episodic haze (PM _{2.5} > 100 µg m ⁻³)	±SD	Low haze (PM _{2.5} < 100 µg m ⁻³)	±SD	Entire haze	±SD	Post-haze	±SD	All	±SD
PM_{2.5} (µg m⁻³)			128.6	36.2	66.2	15.4	86.6	36.2	20.9	11.5	64.7	43.4
PAHs (ng m⁻³)												
Napthalene	Nap	2	0.06	0.03	0.08	0.05	0.07	0.05	ND	-	0.07	0.05
Acenathene	Ace	2	ND	-	ND	-	ND	-	ND	-	ND	-
Acenaphthylene	Acy	3	0.15	0.03	0.11	0.01	0.12	0.03	0.11	0.03	0.12	0.03
Fluorene	Flu	3	ND	-	ND	-	ND	-	ND	-	ND	-
Anthracene	Ant	3	0.80	0.17	0.74	0.11	0.76	0.14	0.73	0.03	0.76	0.14
Phenanthrene	Phe	3	0.47	0.13	0.39	0.07	0.42	0.10	0.37	0.05	0.40	0.09
Fluoranthene	Flua	4	0.99	0.29	0.83	0.04	0.88	0.18	0.76	0.02	0.85	0.16
Pyrene	Pyr	4	0.67	0.19	0.58	0.06	0.61	0.12	0.49	0.02	0.57	0.12
Benz(a)anthrazene	BaA	4	0.81	0.24	0.66	0.05	0.71	0.15	0.60	0.02	0.68	0.14
Chrysene	Chry	4	2.06	0.61	1.60	0.12	1.75	0.42	1.41	0.03	1.67	0.39
Benz(b)fluoranthene	BbF	5	1.16	0.37	0.63	0.22	0.80	0.37	0.26	0.07	0.65	0.40
Benz(k)fluoranthene	BkF	5	1.36	0.41	1.08	0.06	1.17	0.27	0.98	0.02	1.12	0.25
Benz(a)pyrene	BaP	5	0.67	0.20	0.53	0.04	0.57	0.13	0.48	0.03	0.56	0.13
Dibenz(a,h)anthracene	DahA	5	0.72	0.23	0.53	0.04	0.60	0.16	0.47	0.02	0.58	0.16
Indeno(1,2,3-cd)pyrene	Ind	6	1.05	0.32	0.61	0.17	0.75	0.30	0.37	0.09	0.62	0.31
Benz(ghi)perylene	BghiP	6	2.27	0.66	1.37	0.37	1.66	0.64	0.84	0.17	1.40	0.66
2 rings			0.06	0.03	0.08	0.06	0.07	0.05	ND	-	0.05	0.05
Σ3 rings			1.42	0.36	1.24	0.37	1.30	0.40	1.21	0.28	0.88	0.53
Σ4 rings			4.53	1.32	3.67	0.24	3.95	0.86	3.27	1.26	3.37	1.30
Σ5 rings			3.91	1.15	2.77	0.39	3.14	0.90	2.20	0.83	2.47	1.28
Σ6 rings			3.31	0.97	1.99	0.54	2.42	0.94	1.20	0.33	1.99	0.99
ΣPAHs			13.23	3.64	9.67	1.27	9.67	1.27	7.88	2.25	8.75	3.84

Table 5-2 comparison of PAHs concentrations from previous study in northern Thailand and other selected countries.

Study site	Site environment	Sampling period	PM ($\mu\text{g m}^{-3}$)	PAH concentration (ng m ⁻³)																	References
				Nap	Ace	Acy	Flu	Phe	Ant	Flua	Pyr	BaA	Chry	BbF	BkF	BaP	DahA	Ind	BghiP	Total	
Northern Thailand	9 urban sites	Haze, 2013	-	-	-	-	0.08	0.18	0.06	-	0.26	0.34	0.48	-	0.08	<0.01	ND	<0.01	<0.01	2.44	(Pongpiachan et al., 2017b)
Northern Thailand	12 urban sites	Non-haze, 2012	-	-	-	-	0.08	0.19	0.02	-	0.18	0.46	0.33	-	0.01	0.02	ND	<0.01	<0.01	3.11	
Northern Thailand	12 urban sites	Haze, 2018	105.3	ND	ND	ND	ND	1.7	1.0	2.9	0.6	3.2	3.5	5.4	1.8	7.0	1.4	7.2	6.8	42.70	(Kongpran et al., 2021)
Northern Thailand	sub-urban	Non-haze, 2018	14.2	ND	ND	ND	ND	0.04	0.02	0.10	0.01	0.10	0.1	0.20	0.03	0.20	0.01	0.10	0.10	0.90	(Yabueng et al., 2020)
Northern Thailand	sub-urban	Haze, 2017	38.2	0.07	<0.01	0.03	0.02	0.06	0.04	0.12	0.16	0.14	0.15	0.24	0.26	0.27	0.15	0.47	0.37	2.57	
Northern Thailand	sub-urban	Haze, 2018	37.9	<0.01	<0.01	ND	<0.01	<0.01	<0.01	0.02	0.04	<0.01	0.01	0.12	0.16	0.10	0.24	0.76	0.55	2.02	
Western Thailand	urban	Haze, 2016	61.6	0.02	0.02	0.1	0.06	1.04	0.31	0.11	0.02	0.29	0.06	0.84	0.20	0.70	0.65	0.85	1.21	6.32	(Janta et al., 2020)
Western Thailand	urban	Non-haze, 2016	13.8	ND	ND	0.06	0.02	0.99	0.21	0.05	0.16	0.16	ND	0.16	0.02	0.21	0.26	0.22	0.26	2.59	
Northern Thailand	urban	Feb, 2010 (dry season)	93.5	-	-	-	0.24	-	-	-	0.31	0.14	0.35	0.71	0.37	0.61	0.03	0.71	0.65	4.14	(Chuesaard et al., 2014)**
Northern Thailand	urban	Aug, 2010 (wet season)	25.1	-	-	-	0.02	-	-	-	0.04	0.02	0.03	0.09	0.05	0.08	ND	0.16	0.20	0.70	
Northern Thailand	urban	Apr, 2010 (haze)	104.9	1.2	ND	ND	1.91	ND	ND	2.78	0.55	2.41	2.10	0.15	3.34	2.40	0.15	5.05	3.85	25.87	(Wiriya et al., 2013a)*
Northern Thailand	urban	Aug-Nov, 2010 (non-haze)	13.3	0.37	ND	0.01	0.17	0.19	0.17	0.21	0.15	0.22	0.23	0.3	0.25	0.04	ND	0.44	0.36	3.12	
Cordoba, Argentina	urban	Aug 2011-2013	98.4	1.91	0.10	-	0.36	0.28	<0.01	0.21	0.12	0.19	0.16	0.39	0.26	0.16	0.05	-	0.37	4.50	(Amarillo et al., 2017)*
Beijing, China	5 urban sites	Nov, 2013	117.8	-	-	2.72	1.09	6.5	4.84	28.47	20.49	36.94	37.32	28.10	30.66	33.37	5.49	10.78	3.43	250.2	(Gao et al., 2016)
Kuala Lumpur, Malaysia	urban	Dec, 2010 – Mar, 2011 (NE monsoon)	39.7	ND	ND	ND	ND	0.04	0.03	0.07	0.08	0.07	0.12	0.36	0.20	0.26	0.13	0.66	0.94	2.97	(Jamhari et al., 2014b)*
Kuala Lumpur, Malaysia	Industry	Jan – Feb, 2014 (NE monsoon)	26.1	ND	ND	ND	ND	0.05	0.02	0.01	0.05	0.04	0.06	0.37	0.21	0.25	0.09	0.22	0.87	2.24	
Kuala Lumpur, Malaysia	semi-urban	Jun – Sep, 2013 (SW monsoon)	-	0.10	0.18	0.04	0.09	0.06	0.06	0.16	0.12	0.06	0.13	0.75	0.33	0.37	0.22	0.51	0.81	3.85	(Khan et al., 2015)
Kuala Lumpur, Malaysia	semi-urban	Jan – Feb, 2014 (NE monsoon)	-	0.08	0.07	0.03	0.04	0.02	0.02	0.04	0.04	0.03	0.05	0.42	0.18	0.23	0.11	0.22	0.31	1.85	
Delhi, India	urban	Winter 2007-2008	61.8	2.10	0.60	2.20	1.20	1.20	3.20	3.80	2.10	3.60	3.70	11.80	13.60	9.90	2.80	18.70	15.50	96.0	(D. P. Singh et al., 2011)
Delhi, India	urban	Summer 2008	39.4	1.80	0.40	1.30	0.80	1.00	1.60	2.70	1.50	1.80	2.80	5.40	6.90	5.10	2.10	4.20	6.40	45.8	

5.4.2 Correlation coefficients of individual PAHs with PM_{2.5} and levoglucosan during haze and post-haze periods

To investigate the relationship of individual PAHs with PM_{2.5} and an organic biomass burning marker (levoglucosan (Levo)), Pearson's correlation was applied. Table 5-3 shows the correlation coefficient (R^2) of PAHs with PM_{2.5} and with Levoglucosan. Overall, higher molecular weight PAHs had a stronger relationship with PM_{2.5} and levoglucosan than lower molecular weight PAHs during the entire haze period. In the case of episodic haze versus low haze (PM_{2.5} > 100 vs. < 100 $\mu\text{g m}^{-3}$), disparities in the relationship of PAHs with PM_{2.5} and of PAHs with levoglucosan were observed: while PAHs (except Nap) had moderate to very strong correlations with PM_{2.5} and levoglucosan during episodic haze events, PAHs had weak or no correlations with PM_{2.5} and had moderate to very strong with levoglucosan during low haze periods. It is likely that other sources (e.g., vehicular emissions) contribute significantly to PM_{2.5}-bound PAHs during low haze periods, in addition to the commonly observed biomass burning aerosol (Pongpiachan et al., 2017a). This is particularly true for lower molecular weight PAHs. While in the case of post-haze period, 3-4 rings (Phe, Flua and Pyr) and 5 rings PAHs (BbF and BkF) had a moderate correlation with PM_{2.5} and levoglucosan, while 6-rings PAHs (Ind and DahA) had moderate and strong correlation with PM_{2.5} and levoglucosan, respectively, indicating less influence from biomass burning and more from vehicular emissions. These results agree with prior studies regarding biomass burning derived PM-bound PAHs conducted during haze and non-haze periods in the northern Thailand (Chuesaard et al., 2014; Thepnuan et al., 2020; Wiriya et al., 2013a), western Thailand (Janta et al., 2020) and east China (Mao et al., 2018). Tian et al. (2021) reported that 3 – 4 rings PAHs had proportionally higher abundances in diesel vehicles than gasoline vehicles.

Table 5-3 Pearson's correlation coefficients (R^2) between PAHs with $PM_{2.5}$ and levoglucosan (Levo) in different periods. $R^2 > 0.8$ is in bold.

	Episodic haze		Low haze		Entire haze		Post-haze		All	
	$PM_{2.5}$	Levo	$PM_{2.5}$	Levo	$PM_{2.5}$	Levo	$PM_{2.5}$	Levo	$PM_{2.5}$	Levo
Nap	0.11	-0.21	0.07	0.14	-0.08	-0.1	ND	ND	-0.08	-0.1
Acy	.72**	.81**	0.18	0.17	.76**	.79**	ND	ND	.76**	.79**
Ant	.85**	.69**	-0.24	-0.11	.40**	.39**	ND	ND	.40**	.39**
Phe	.75**	.74**	-0.11	-0.08	.54**	.55**	.48*	0.37	.53**	.56**
Flua	.77**	.68**	-0.16	0.06	.62**	.62**	.67**	.61**	.66**	.68**
Pyr	.78**	.67**	-0.18	-0.04	.58**	.57**	.52**	.45*	.69**	.67**
BaA	.78**	.75**	0.06	.45**	.67**	.72**	0.15	0.26	.68**	.75**
Chry	.77**	.79**	0.18	.58**	.72**	.79**	0.38	0.36	.74**	.82**
BbF	.69**	.87**	.31*	.81**	.77**	.92**	.50*	.58**	.86**	.94**
BkF	.79**	.76**	0.29	.81**	.72**	.77**	0.43	.53*	.72**	.80**
BaP	.77**	.85**	0.28	.57**	.73**	.81**	0.16	0.24	.73**	.82**
DahA	.81**	.84**	.41**	.65**	.77**	.83**	ND	ND	.77**	.84**
Ind	.77**	.86**	.44**	.88**	.83**	.93**	.61**	.76**	.88**	.94**
BghiP	.76**	.85**	.38*	.81**	.80**	.91**	.61**	.72**	.87**	.93**
$\Sigma 2$ -rings	0.11	-0.21	0.07	0.14	-0.08	-0.1	ND	ND	-0.08	-0.1
$\Sigma 3$ -rings	.79**	.74**	-0.22	0.09	.44**	.51**	-0.06	-0.1	.67**	.65**
$\Sigma 4$ -rings	.78**	.75**	0.03	.39*	.68**	.72**	0.01	-0.14	.72**	.71**
$\Sigma 5$ -rings	.80**	.87**	0.26	.76**	.79**	.89**	0.06	-0.02	.84**	.86**
$\Sigma 6$ -rings	.77**	.86**	.40**	.84**	.81**	.92**	.57**	.63**	.88**	.93**
Σ LMWPAHs	.79**	.71**	-0.2	0.11	.42**	.49**	-0.06	-0.1	.67**	.64**
Σ HMWPAHs	.81**	.85**	0.3	.77**	.80**	.88**	0.21	0.05	.85**	.85**
Σ PAHs	.82**	.85**	0.19	.70**	.78**	.87**	0.23	0.06	.85**	.84**

** Correlation is significant at the 0.01 level (2-tailed)

* Correlation is significant at the 0.05 level (2-tailed)

ND – No data (n < 50%)

$\Sigma 2$ -rings = Nap

$\Sigma 3$ -rings = Acy + Ant + Phe

$\Sigma 4$ -rings = Flua + Pyr + BaA + Chry

$\Sigma 5$ -rings = BbF + BkF + BaP

$\Sigma 6$ -rings = Ind + BghiP

Σ LMWPAHs = $\Sigma 2$ -rings + $\Sigma 3$ -rings

Σ HMWPAHs = $\Sigma 4$ -rings + $\Sigma 5$ -rings + $\Sigma 6$ -rings

5.4.3 Lifetime cancer risk assessment

Understanding the relationship between hazardous components such as PAHs and heavy metals and lifetime cancer risk associated with inhaling $PM_{2.5}$ is important to support air pollution and public health management. Lifetime cancer risk from this study was estimated based on measured PAHs during haze and post-haze and compared with that from and metals. Source-specific risks were also obtained from the PMF-LCR model.

5.4.3.1 Lifetime cancer risks from measured PAHs and metals during haze and post-haze periods

Measured LCR from PAHs and heavy metals during different periods are listed in Table 5-4. During the haze period, all estimated LCR are above the threshold risk of 1 chance in million (10^{-6}) and ranged from 2.53×10^{-5} to 9.57×10^{-5} , while during post-haze period, approximately 93% of LCR were above the threshold ranged from 3.20×10^{-7} to 6.66×10^{-6} . Overall, LCR estimated from PAHs was higher than those obtained from heavy metals in every period. It is also clearly observed that, although Σ LCR of all periods show a moderate cancer risk (10^{-6} to 10^{-4}), Σ LCR of episodic haze ($\text{PM}_{2.5} > 100 \mu\text{g m}^{-3}$) and entire haze were approximately 8 and 6 times higher than post-haze, which accounted to the risk of cancer raised from less than 7 to 52 chances in millions of populations. This emphasizes the significant health risk of haze phenomena.

Comparing to previous studies in northern Thailand, Chuesaard et al. (2014) reported the cancer risk below the threshold value in 2010, Wiriya et al., (2013) reported the cancer risk of 5.31×10^{-5} in year 2011, Pongpiachan et al. (2015) reported 6×10^{-6} in year 2013, Thepnuan and Chantara, (2020) indicated significant high risk at 1×10^{-4} in year 2016, Yabueng et al., (2020) estimated the risk of 4.71×10^{-5} and 3.88×10^{-5} in year 2017 and 2018, respectively. Coupled with gas phase PAHs, Kongpran et al. (2021) reported very high cancer risk at 9.3×10^{-4} during haze in year 2018. Insian et al. (2022) reported a higher risk of 1.14×10^{-4} and 6.80×10^{-5} in year 2019 at rural and urban sites, respectively; they also found that the cancer risk substantially increased with smaller size PM owing to a higher proportion of carcinogenic PAHs bound with smaller size PM. It should be noted that the severity of cancer risk from haze events over northern Thailand in the past decade has remained significantly moderate to high level, highlighting the failure to effectively address and mitigate the persistent haze problem.

Study in other parts of Thailand, for example, southern Thailand, has reported a significantly lower LCR compared to this study, during haze (6.7×10^{-8}) and non-haze (4.3×10^{-8}),

which may be attributed to fairly low PAHs contents ($< 2.5 \text{ ng m}^{-3}$) bound with $\text{PM}_{2.5}$ (Mahasakpan et al., 2023), while in western Thailand, ICR were reported with high risk of 10.02×10^{-5} during haze (Janta et al., 2020). Comparing to other countries, LCR from this study was mostly lower than those reported in China; Nanjing, (4.44×10^{-4} to 3.79×10^{-4}) (Y. Li et al., 2019) Xi'an (1.4×10^{-4} to 5.6×10^{-3}) (Bandowe et al., 2014) Guangzhou (5.98×10^{-4}) (J. Liu et al., 2015) and Beijing (4.39×10^{-5} to 3.31×10^{-3}) (H. Yin & Xu, 2018), and India; Delhi (1.02×10^{-4} to 7.88×10^{-4}) but higher than Shiraz, Iran (1.6×10^{-5}) (Kermani et al., 2021), central Tehran (2.8×10^{-5}) (Taghvaei et al., 2018), Thessaloniki, Greece (1.4×10^{-7} to 1.4×10^{-6}) (Sarigiannis et al., 2015), and Kuala Lumpur, Malaysia (2.27×10^{-5}) (Sulong et al., 2017) and (3.41×10^{-7}) (Jamhari et al., 2014c).

Table 5-4 Estimated lifetime cancer risk ($\times 10^{-5}$) (LCR) by measured LCR and by PMF-LCR source specific risk between entire haze, post-haze, episodic haze and low haze.

	Measured LCR ($\times 10^{-5}$)			Source-specific LCR ($\times 10^{-5}$)						
	LCR-PAHs	LCR-Metals	Σ LCR	Gasoline	Diesel	Biomass burning	Softwoods burning	Mixed dusts	Secondary sulfate	Σ LCR
Entire haze	3.34	0.87	4.21	0.26	1.35	0.60	1.23	0.51	0.19	4.14
Episodic	4.04	1.16	5.21	0.25	1.07	1.09	2.00	0.55	0.20	5.17
Low haze	3.00	0.72	3.73	0.27	1.48	0.36	0.83	0.48	0.18	3.59
Post-haze	0.56	0.10	0.67	0.28	0.47	0.21	0.16	0.04	0.25	1.42

5.4.3.2 PAHs and heavy metals source apportionment

By applying with PMF model, and adopted the specific source markers (OC, EC, Levoglucosan, dehydroabietic acid (DHAA), SO_4^{2-} , NO_3^- , NH_4^+ , key crustal elements (Al, Ti and Fe), and some trace elements (Cu, Zn, Pb, Mn, Ba and Pb)), 6-factors were the best solution outcome by the model including biomass burning, softwoods burning, gasoline combustion, diesel combustion, mixed dusts and secondary aerosol (sulfate). Fingerprint and chemical source profile are illustrated in Fig. SC-1 and SC-2. Briefly, each source was identified as follow:

Gasoline combustion: identified by significant load of high molecular weight PAHs including Ind (29%) and BghiP (28%) and low molecular weight PAHs Nap (30%) (Khan et al.,

2015; Soleimani et al., 2022; Sulong et al., 2019a; Taghvaei et al., 2018; Tian et al., 2022) and high contribution on several metals Cu (52%), Ni (28%), Zn (26%) and Cr (21%) (Hao et al., 2019).

Diesel combustion: identified by a broad range of significant contribution of PAHs BkF (64%), Phe (56%), BaA (55%), Chry (55%), BaP (52%), Flua (49%) and Pyr (46%) (Khan et al., 2015; Soleimani et al., 2022; Taghvaei et al., 2018; Tian et al., 2022), with low contribution from metals (Hao et al., 2019).

Biomass burning: classified by significant contributions to levoglucosan (37%), OC (36%), EC (26%), high molecular weight PAHs Ind (26%) and BghiP (23%) (Sulong et al., 2019a) and very high Ant (80%) may be attributed to smoldering condition in biomass burning (Chuesaard et al., 2014; Jenkins et al., 1996).

Softwoods burning: this source contributed very high load of a softwood burning marker, DAHH (71%) (Leithead et al., 2006; Nguyen et al., 2016; B. Yan et al., 2008), levoglucosan (51%), OC (40%), EC (37%).

Mixed dusts: identified by high contribution of crustal elements Al (39%), Ti (39%), Fe (38%) and other metals Cr (37%), Mn (36%), Co (39%), Ni (22%), As (52%), Ba (40%).

Secondary sulfate: identified by high contribution of SO_4^{2-} (57%) and NH_4^+ (62%).

The concentrations of PAHs and heavy metals obtained from these 6 identified sources were then weight-calculated to daily concentration by multiplying with daily contribution of each source.

5.4.3.3 Source-specific lifetime cancer risk

Average source-specific lifetime cancer risks during different periods are shown in Table 5-4. Temporal variations and daily percentage contribution of source-specific lifetime cancer risk (LCR) are shown in Fig. 5-3 (a) and (b), respectively. And Percentage contribution of source specific LCR to total LCR during episodic haze, low haze and post-haze periods are shown as pie charts in Fig. 5-4. Scatter plots between measured LCR and PMF-LCR in Fig. SC-3 show a strong correlation ($R^2 = 0.91$), indicating that LCR from both approaches were in well agreement. Overall, source specific LCR ranged in moderate risk between 10^{-6} to 10^{-4} . LCR were in descending order as follow; entire haze: diesel > softwood burning > biomass burning > mixed dusts > gasoline > secondary sulfate; episodic haze: softwoods burning > biomass burning > diesel > mixed dusts > gasoline > secondary sulfate; low haze: diesel > softwoods burning > mixed dusts > biomass burning > gasoline > secondary sulfate; and post-haze: diesel > gasoline > biomass burning > softwoods burning > secondary sulfate > mixed dusts. It can be then noted that the emissions from diesel engine, softwoods burning and biomass burning were major sources of LCR during haze while diesel and gasoline emissions were major sources of LCR during post-haze.

It is observed that diesel engine emissions was a dominant source of LCR in low haze (1.48×10^{-5}) and post-haze (0.47×10^{-5}) which contribute at 41% and 33% to total LCR, respectively. This is due to light/heavy diesel vehicles are commonly used for public transportation or agricultural sector in Thailand (Kim Oanh et al., 2010), consequently contributing to LCR throughout study period as can be seen from Fig. 5-3 (b). Furthermore, the proportion of hazardous chemical components like PAHs to total PM from diesel fuel combustion are higher than biomass burning (Y. Zhang & Tao, 2009) as shown in Fig. 5-4, resulting in higher LCR from diesel vehicular sources. In the case of gasoline, LCR derived from gasoline had varied in narrow ranged ($0.25 - 0.28 \times 10^{-5}$) but made a significant contribution during post-haze where the contributions increased from 6% in haze to 20% in post-haze. Overall, during post-haze periods, LCR derived by vehicular emissions were dominant.

Open burning of biomass emits considerably amount of $PM_{2.5}$ (Suriyawong et al., 2023). Combined LCR from both biomass and softwoods burnings was 1.83×10^{-5} during haze periods and 3.09×10^{-5} during heavy haze periods, accounted for 44% and 60% to total LCR. LCR of biomass burning had increase initially during the 1st to 3rd episodic events, while later, LCR derived from softwoods burning had substantially elevated after 21 Mar especially during the 2nd to 4th episodic haze events (Fig. 5-3 (a) and (b)). Total LCRs from both open burnings after 2nd episodic haze caused considerably high LCR. On the other hand, during post-haze period, biomass and softwoods burning had lesser impact on cancer risk, only occasionally high LCR contribution from biomass burning which could be from local burning.

LCR derived from mixed dusts was observed to be a minor group in haze and was negligible in post-haze. A slightly higher LCR of mixed dusts was found in episodic haze (0.55×10^{-5}) than low haze (0.48×10^{-5}) while post-haze was very low (0.04×10^{-5}). However, the contribution of LCR from mixed dusts to total LCR during haze period (11-12%) were significantly higher than post-haze (3%), implying that haze phenomenon and/or meteorological conditions in local dry season played a role involving on mixed dusts emission.

A significant increase of source specific LCR from haze to post-haze was found in the case of LCR derived from secondary sulfate, LCR of this source was highest during post-haze (0.25×10^{-5}). Its LCR contribution during post-haze (17%) approximately a 4-fold increased from episodic haze (4%) and low haze (5%), the highest change in contribution among source specific LCR. An elevated LCR was found in the beginning to middle stage of post-haze period in May (Fig. 5-3 (a) and (b)). The contribution of LCR from SA-PAHs (16%) was significant (Fig. 5-4), this may be attributed to the PAHs adsorbed on the $PM_{2.5}$ during transport (Tian et al., 2022). Recent studies have demonstrated that organosulfate (secondary organic sulfate) can be formed by sulfate aerosol with organic compounds, i.e., PAHs (Ding et al., 2022; Kanellopoulos et al., 2022; Riva et al., 2015).

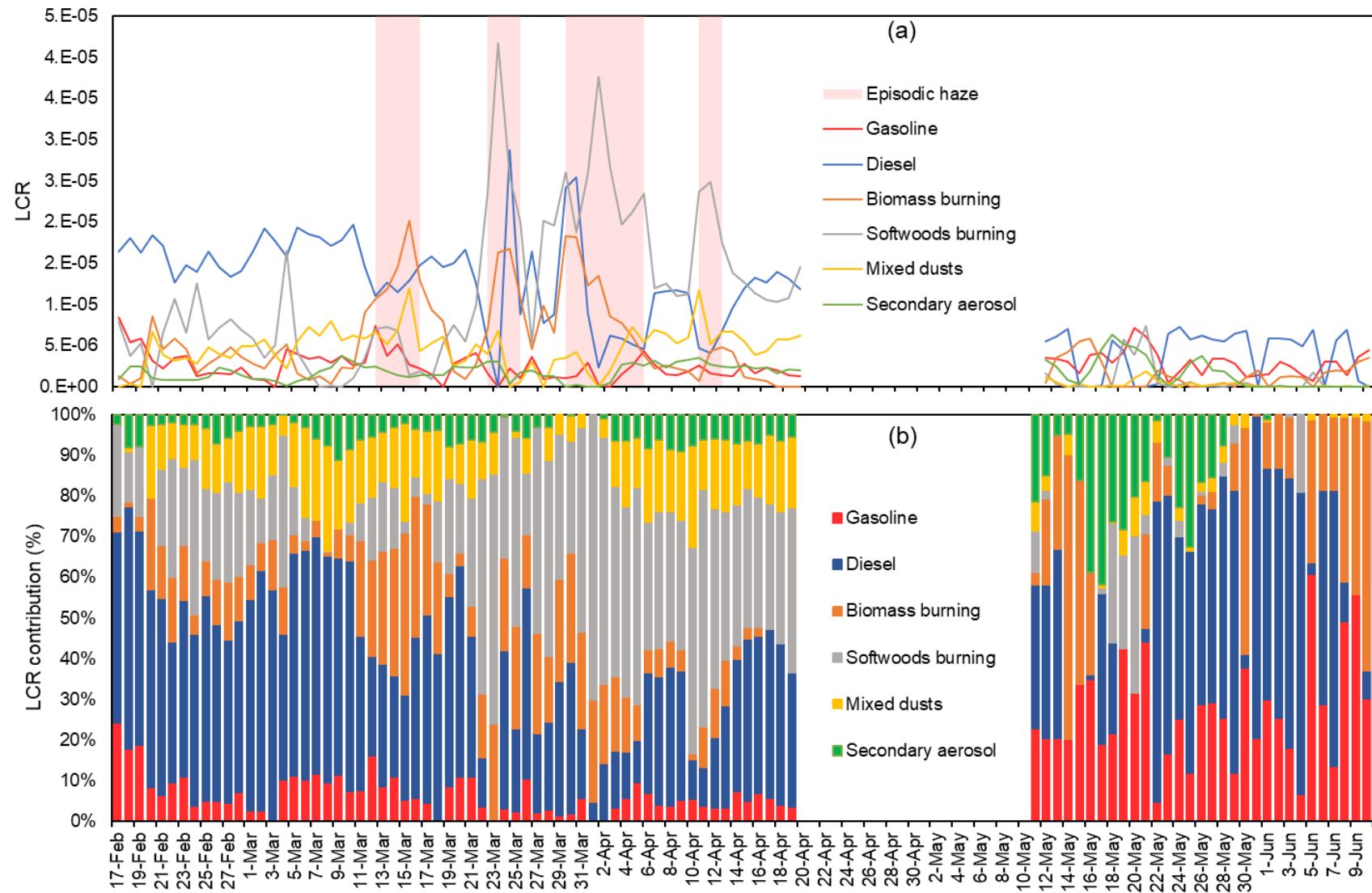


Fig. 5-3 Temporal variations (a) and daily percentage contribution to total LCR (b) of source-specific lifetime cancer risk (LCR)

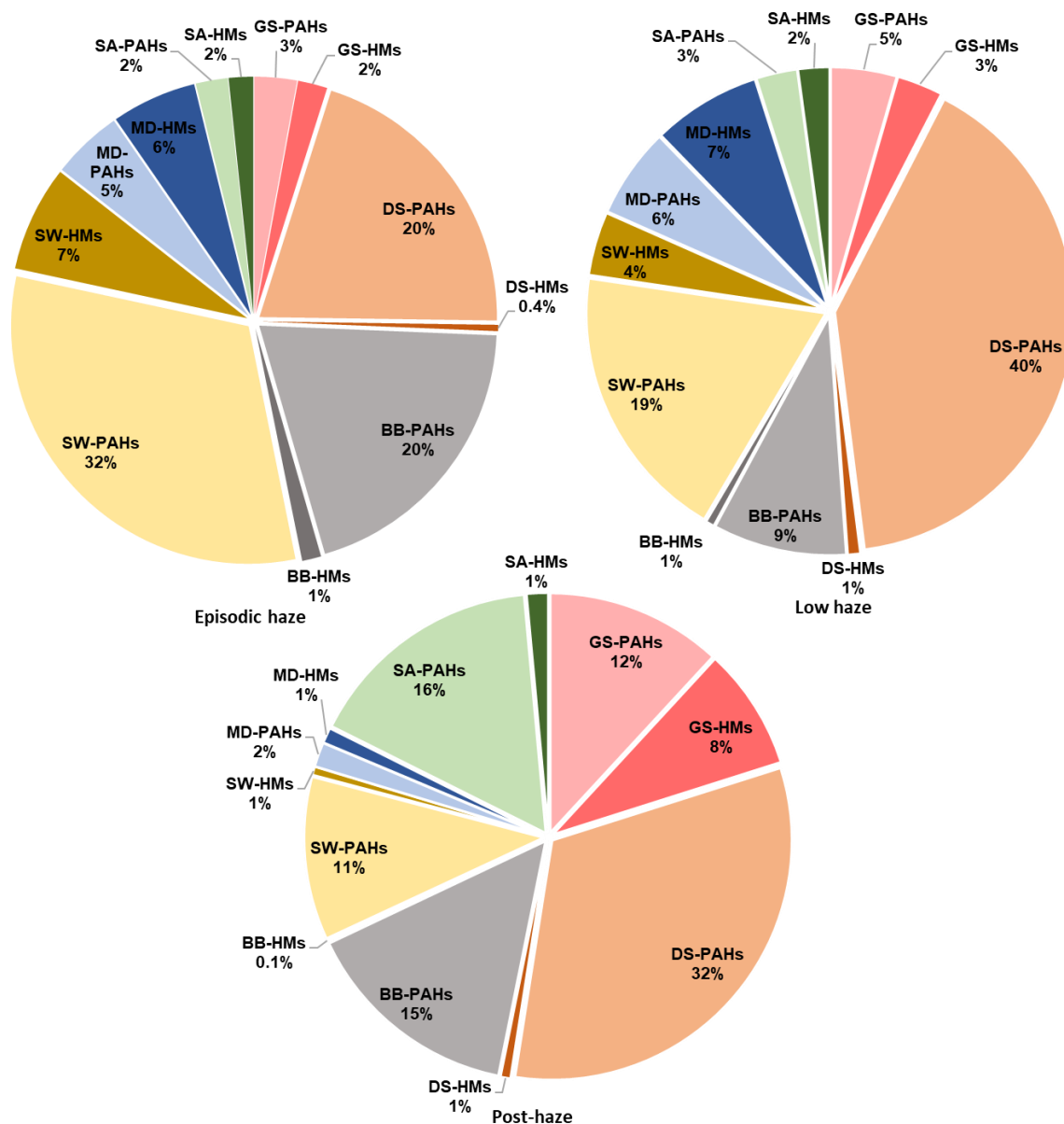


Fig. 5-4 Percentage contribution of source specific LCR to total LCR during episodic haze, low haze and post-haze. Gasoline (GS), diesel (DS), biomass burning (BB), softwood burning (SW), mixed dusts (MD) and secondary sulfate (SA). Suffix -PAHs and -HMs mean source specific LCR calculated from PAHs and heavy metals, respectively

5.4.3.4 Implications in cancer risk mitigation

The outcomes obtained from PMF-LCR provide a useful insight into the relative importance of different sources in contributing to overall lifetime cancer risk, identifying the specific sources of carcinogens, and quantifying their contributions is essential for developing effective strategies for cancer risk mitigation. Further information on LCR of individual heavy metals and PAHs by specific sources during different periods is displayed in Fig. SC-4. Based on the information obtained from section 5.4.3.3 the mitigation could be performed by following approaches:

Haze period: the top three highest contributors to total LCR in episodic and low haze were diesel emission, softwood burning and biomass burning (Fig. 5-4). Fig. SC-4 also show that LCR calculated from diesel-obtained PAHs were mostly highest in several individual PAHs followed by softwood burning-obtained PAHs and biomass burning-obtained PAHs, owing to that PAHs are a minor group of components in woods burning (Fine et al., 2004a, 2004b). By reducing the contribution of these three sources during haze period 50% could reduce total LCR during haze period from 3.87×10^{-5} to 2.28×10^{-5} , accounting for 41% reduction. However, in practical terms, since reducing open biomass burning activities can consequently lead to reducing mixed dusts emission from burned soil and resuspended dust as well, this may then indirectly result in additional reductions. Softwood burning (wildfire in coniferous forest areas) has a significant risk but refers to which may be difficult to control.

Post-haze: biomass burning during this period had less influence compared to diesel and gasoline emissions. The contribution of LCR derived from these two vehicular emissions combined was 53% of total LCR (Fig. 5-4). LCR calculated from heavy metals in gasoline was highest of all sources, and LCR calculated from PAHs in diesel emissions was also highest (Fig. SC-4), suggesting that reducing these emission sources would be more effective and be practically

achievable. Reducing 50% of these emission sources could result in 21% reduction in total LCR of post-haze period from 1.13×10^{-5} to 0.90×10^{-5} .

5.5 Conclusion

This study aims to perform characterization of 16 main US-EPA PAHs and to carry out the cancer risk assessment of $PM_{2.5}$ bound PAHs and heavy metals in the comparative aspects between haze and post-haze and between source-specific outcomes as well as provide an example implication to mitigate cancer risk caused by specific sources. $\Sigma PAHs/PM_{2.5}$ ratio displays an inverse relationship trend implying that the haze contributed to particulate-bound PAHs in some extent. $\Sigma PAHs$ mass during episodic haze, low haze and post-haze were 13.2 ± 3.6 , 9.7 ± 1.3 and $7.9 \pm 2.2 \text{ ng m}^{-3}$, respectively. 5 – 6 rings PAHs associated with intensity of haze especially BbF, Ind and BghiP. Disparities in correlation between PAHs and $PM_{2.5}$ and levoglucosan imply the distinction of PAHs sources, high molecular weight PAHs had strong correlations with levoglucosan episodic haze while lower molecular weight PAHs had moderate correlations with $PM_{2.5}$ during low haze, implying mixed sources of PAHs. Lifetime cancer risk (LCR) ranged in moderate risk with value of 4.21×10^{-5} and 0.67×10^{-5} for haze and post-haze, respectively. Source specific lifetime cancer risk obtained from PMF-LCR revealed 6 sources including gasoline, diesel, biomass burning, softwoods burning, mixed dusts and secondary sulfate. Diesel emissions, biomass burning and softwoods burning were the 3 main contributors of sources specific LCR during haze period, while two vehicular emissions (diesel and gasoline) were main contributors for post-haze period. Our suggestion on mitigation LCR to reduce emission from those main sources by 50% could result in 41% and 21% reduction of LCR during haze and post-haze periods, respectively.

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CHAPTER SIX:

**CHARACTERIZATION OF PM_{2.5} BOUND
ORGANIC COMPOUNDS DURING HAZE AND
POST-HAZE PERIODS IN NORTHERN
THAILAND**

Characterization of PM_{2.5} bound organic compounds during haze and post-haze periods in northern Thailand

6.1 Abstract

Organic matter is the single largest component in fine particles (PM_{2.5}) during haze events in northern Thailand. However, we know little about the chemical composition of those compounds, from both primary organic aerosol (POA) emissions or formed as secondary organic aerosol (SOA) in the atmosphere. Daily PM_{2.5} were collected during haze (17 Feb – 19 Apr) and post-haze (11 May – 10 Jun) 2019 and were analyzed separately for polar and non-polar compounds by GC-MS. Total mass of measured nonpolar and polar compounds were 121.1 ± 34.2 , 83.6 ± 27.1 and 53.1 ± 12.0 ng m⁻³ and $3,563.0 \pm 1,337.8$, $1,278.6 \pm 693.5$ and 313.3 ± 133.1 ng m⁻³ during episodic haze, low haze and post-haze, respectively. High molecular weight PAHs contributed approximately 90% of total PAHs while high molecular weight alkanes (C_≥26) contributed 75-80% of total alkanes. Carbon preference index (CPI) indicates the dominant influence of biogenic and anthropogenic source during haze and post-haze, respectively. In polar compounds, levoglucosan was the most abundant saccharide group and all identified compounds, and 4-nitrocatechol and malic acid were the most abundant compounds of nitrated aromatic phenols and carboxylic acids. Anhydrosugars and nonpolar organic compounds were important POA while dicarboxylic acids and nitrated aromatic phenols were the most abundant group of SOA compounds followed by monocarboxylic acids and tricarboxylic acids. The ratios of malic acid to succinic acid (hC4/C4) and Σ SOA/ Σ POA suggest a higher photooxidation during post-haze than during haze period. This study provides a first comprehensive information on characteristics of polar and nonpolar organic compounds in different period of haze and post-haze in northern Thailand.

6.2 Introduction

Haze events resulting from biomass burning activities have become a reoccurrence environmental issue globally, affecting on air quality, climate and human health (J. Chen et al., 2017; S. Yin, 2023). Biomass burning such as wildfires, agricultural residues burning and open burning for land clearing, these events emit a diverse type of organic compounds, including both polar and nonpolar organic species, which contribute to the formation of primary (POA) and secondary (SOA) organic aerosols (Hapsari Budisulistiorini et al., 2018; Kavouras et al., 1999; Lyu et al., 2019). During biomass burning, POA is directly emitted into the atmosphere as particles bound with organic compounds derived from incomplete combustion processes of biomass such as anhydrosugars and polycyclic aromatic hydrocarbons (PAHs) (Bertrand et al., 2018; J. Li et al., 2020) while SOA is formed through complex chemical reactions by photooxidation of several range of precursors such as aromatic carbons (C. L. Chen et al., 2018; Kleindienst et al., 2012; Riva et al., 2017), toluene and xylene (Qi et al., 2020; Srivastava et al., 2023), terpenes (Ng et al., 2007), isoprene (biogenic volatile organic compound) (Carlton et al., 2009) phenol (Finewax et al., 2018). Some SOA such as oxygenated PAHs and nitro PAHs (derivatives of PAHs) are even more harmful than their parent PAHs (J. Wang et al., 2017). Some SOA also play a significant role on radiative forcing, for example, nitro aromatic compounds, which cause impacts on climate by scatter or absorb solar radiation or act as cloud condensation nuclei (Ichikawa et al., 2018; Shrivastava et al., 2017; Tsigaridis & Kanakidou, 2018).

Field studies on nonpolar organic compounds generally focus on alkanes, PAHs. Many studies have correspondingly concluded that alkanes are the largest abundant in this group followed by PAHs (He et al., 2010; Xu et al., 2013; Wang et al., 2015; Wang et al., 2016; Cao et al., 2021). The distinction in odd and even number of alkanes provides an indicator for separation of alkanes sources, odd number alkanes associate with biogenic source while even number alkanes are emitted from anthropogenic sources such as petrogenic. The ratio between total mass of odd

number alkanes to total mass of even number alkanes, known as carbon preference index (CPI), is then used to differentiate alkanes sources in previous studies (Fadel et al., 2021; Kang et al., 2018; S.-W. Park et al., 2022). Studies of polar organic compounds involving haze events have paid attention on several group of compounds such as nitro aromatic and nitrophenol (Cai et al., 2022; X. Li et al., 2016; Yuan et al., 2021; R. Zhang et al., 2023) carboxylic acids especially dicarboxylic acids and aromatic acids (Cao et al., 2017; Glasius et al., 2000; Hsieh et al., 2008; Kundu et al., 2010; Q. Yu et al., 2019), these compounds are mainly SOA formed through photooxidation in the atmosphere. Certain dicarboxylic acid compounds offer an information about the degree of photooxidation intensity, such as malic acid to succinic acid (Kawamura & Sakaguchi, 1999; Q. Yu et al., 2021). Thus, gaining comprehensive insight into characteristics of both polar and nonpolar organic compounds could then subsequently be advantageous in elucidating formation mechanism, chemical transformation and sources of POAs and SOAs.

Over the past decade, northern Thailand have been suffering from recurrent haze event during the local summer season mainly due to biomass burning (Pardthaisong et al., 2018; Suriyawong et al., 2023). Previous studies regarding PM_{2.5} bound organic compounds in northern Thailand have been mainly paying attention to biomass burning tracers such as levoglucosan (Chantara et al., 2019; Thepnuan et al., 2019b, 2020; Tsai et al., 2013) and carcinogenic compounds such as PAHs (Insian et al., 2022; Mahasakpan et al., 2023; Thepnuan & Chantara, 2020; Yabueng et al., 2020) and their derivatives, nitro-PAHs (Chuesaard et al., 2014) while study of other polar and nonpolar organic compounds as primary and secondary molecular markers are scarce. To fill this gap of knowledge, this study then aims to comprehensively investigate characteristics of polar and nonpolar organic compounds and their characterizations involving POA and SOA during haze and post-haze. To the best of our knowledge, this study was conducted for the first time in northern Thailand.

6.3 Methodology

6.3.1 Sampling site and PM_{2.5} collection

As described in 3.1 and 3.3.

6.3.2 Polar and non-polar organic extraction and analysis by GC-MS

As described in 3.4.5 and 3.4.6

6.3.3 Quality control and Quality assurance

As described in 3.4.6

6.4 Results and discussions

Support information of this chapter is in supplement D

6.4.1 Overview

Statistical data of nonpolar and polar organic compounds is shown in Table SD-1 and SD-2, respectively, in supplement D. Nonpolar organic compounds were divided into two subgroups: polycyclic aromatic hydrocarbons (PAHs) and alkanes, polar organic compounds were divided into four subgroups: saccharides, nitrate aromatic phenols, carboxylic acids and others. The contribution of both nonpolar and polar groups shows higher concentrations with PM_{2.5} mass. The concentration during episodic, haze and post-haze were 121.1 ± 34.2 , 83.6 ± 27.1 and 53.1 ± 12.0 ng m⁻³ for nonpolar compounds and 3563.0 ± 1337.8 , 1278.6 ± 693.5 and 313.3 ± 133.1 ng m⁻³ for polar compounds, respectively. Their contributions are shown in Fig. 6-3 and 6-4. Overall, both nonpolar and polar compounds that were analyzed in this study contributed a range

between 3 – 4% of organic matters (OM) of PM_{2.5} for the whole study periods. The concentration of nonpolar compounds in PM_{2.5} in China were comparable to that determined in this study. Cao et al. (2021) reported concentration ranged from 103 – 243 ng m⁻³, accounted for 1.6 – 2.3% of organic carbon mass (OC), Wang et al. (2016) reported from 2 – 536 ng m⁻³ (0.1 – 3% of OC mass), Xu et al. (2013) reported range from 19 – 223 ng m⁻³, Wang et al. (2015) reported range from 2 – 74 ng m⁻³ (less than 1% of OC mass), while much lower in Japan, Ichikawa et al. (2018) reported range from 5 – 69 ng m⁻³ (<1% of OC mass), and in Korea, Park et al. (2022) reported 25 ng m⁻³ (< 1% of OC mass). For polar organic compounds, this study had relatively higher concentrations especially during haze period: Kanellopoulos et al. (2020) reported range 38 – 323 ng m⁻³ in eastern Mediterranean, Mu et al. (2021) reported range 3 – 86 ng m⁻³ in Jinzhong, He et al. (2020) reported average concentrations at 828 ng m⁻³ during episodic 321 ng m⁻³ during non-episodic and Ren et al. (2020) reported range 307 – 1,313 ng m⁻³, in Shanghai, China.

6.4.2 Nonpolar organic compounds

6.4.2.1 PAHs

Average concentrations of individual PAHs are shown in Fig. 6-1 and the contribution of low molecular weight (LMW) and high molecular weight (HMW) PAHs to total PAHs are shown in Fig. 6-1. Overall, it is clearly shown that the intensity of haze phenomena led to increased concentrations of PHAs especially PAHs with more than 4 rings (HMW). The concentrations of LMW and HMW were 1.4 ± 0.4 , 1.1 ± 0.4 , 0.5 ± 0.3 and 11.7 ± 3.3 , 8.4 ± 1.1 , 4.7 ± 2.0 ng m⁻³ for episodic haze, low haze and post-haze, respectively. Although, in comparison of contribution to total PAHs (Fig. 6-3), PAHs did not show significant differences between those of LMW and HMW (9 – 12% and 91 – 88% for LMW and HMW, respectively) during different periods. As discussed previously in chapter 3, it is noted that only contributions of BkF and BghiP had increased as haze intensity, and as shown in Fig. 6-1(a), average values of both BkF and BghiP had substantially

elevated from post-haze to low haze and episodic haze. More details of PAHs are discussed previously in Chapter 5 section 5.4.1 and 5.4.2.

6.4.2.2 Alkanes

Measured alkanes in this study were C16 – C34, the alkanes with C number below 16 and above 34 were not detected more than 40% of overall data. Average concentrations of individual alkanes are shown in Fig. 6-1 and Table SD-1 for different periods. Average concentrations of total alkanes were 107.9 ± 31.4 , 74.1 ± 26.6 , 48.8 ± 10.7 ng m⁻³ in episodic haze, low haze and post-haze, respectively. According to average value of alkanes shown in Fig.6-1, concentration of low molecular weight alkanes ($\Sigma C_{\leq 25}$) was lower than high molecular weight alkanes ($\Sigma C_{\geq 26}$) for all periods. Total concentration of odd number alkanes (ΣC_{odd}) was higher than even number alkanes (ΣC_{even}) during episodic and low haze while ΣC_{odd} was lower than ΣC_{even} during post-haze. C29 was the highest homologue alkane in episodic haze and low haze while C28 was the highest alkane in post-haze. An enrichment of lower molecular weight alkanes ($C < 26$) have been reported from several studies as being associated with the emissions from vehicular exhausts especially diesel engines (Cao et al., 2021; Fraser et al., 1999; Perrone et al., 2014) while higher molecular weight ($C > 26$) with significant odd carbon number are mainly from leaf wax (Kang et al., 2018) especially C27, C29 and C31 (Bush & McInerney, 2013) which can be emitted to the atmosphere by forest fires and biomass burning (De Oliveira Alves et al., 2014; Simoneit, 2002).

Carbon preference index (CPI) is defined as the ratio between the sum of the odd number carbon alkanes concentration to the sum of the even number carbon alkanes concentration. Average CPI during episodic haze, low haze and post-haze were 1.2 ± 0.2 , 1.2 ± 0.2 and 0.9 ± 0.2 , respectively. Temporal distribution of CPI throughout the study period is demonstrated in Fig. 6-2, CPI during haze period mostly varied above 1 with maximum at value of 1.8 while during post-haze varied below 1 with lowest value of 0.6. The CPI obtain from this study agreed well with previous studies regarding alkanes emitted during haze events from biomass burning, the studies

have reported CPI values higher than 1 (J. He et al., 2010; Kang et al., 2018; J. Li et al., 2020; Viana et al., 2008). In natural environments, such as plants and microorganisms, the biosynthesis of alkanes typically produces a higher proportion of odd number alkanes while anthropogenic sources of alkanes, such as fossil fuel combustion, industrial emissions and petroleum combustions tend to generate a higher proportion of even number alkanes. Therefore, $CPI > 1$ indicates higher proportion of odd alkanes emitted from plants as well as from low-temperature biomass combustion (Iinuma et al., 2007); on the other hand, $CPI < 1$ indicates the higher contribution of anthropogenic emissions (M. Zheng et al., 2006).

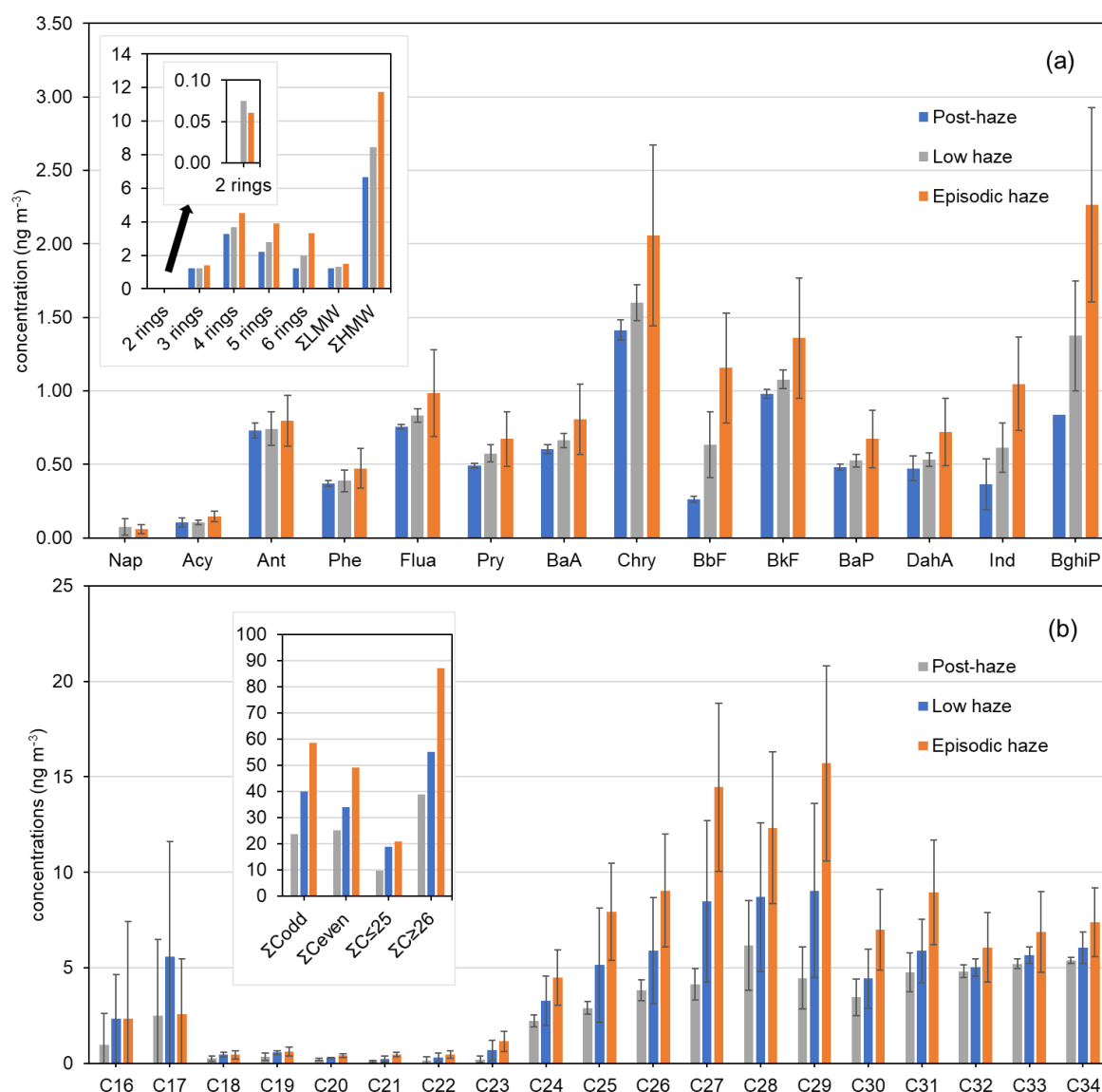


Fig. 6-1 Average concentrations of PAHs (a) and alkanes (b) in post-haze, low haze and episodic haze. Σ LMW is sum of Nap, Acy and Ant, Σ LMW is sum of Ant, Phe, Flua, Pyr, BaA, Chry, BbF, BaF, BaP, DahA, Ind and BghiP. Codd is sum of odd number carbons (C17 – C33) and Ceven is sum of even number carbons (C16 – C34)

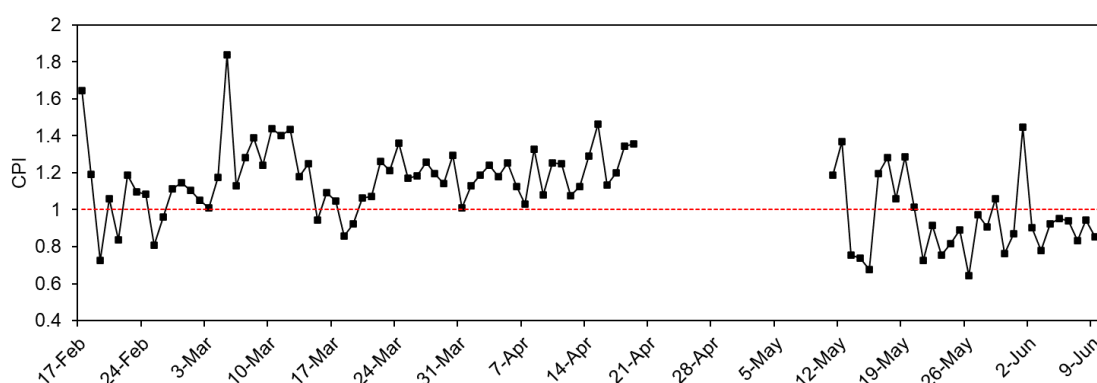


Fig. 6-2 Temporal variation of daily carbon preference index (CPI) for entire study period

6.4.3 Polar organic compounds

6.4.3.1 Saccharides

Measured saccharides compounds in this study were divided into two groups, anhydrosugars (levoglucosan, mannosan and galactosan) and sugar alcohols (D-threitol and meso-erythritol). The concentrations of saccharide compounds are listed in Table SD-2 and their concentrations distribution for each individual compound is displayed in Fig. 6-5. Total saccharide was 2561.7 ± 1026.7 , 873.1 ± 533.4 , and 147.2 ± 86.6 ng m⁻³ for episodic haze, low haze and post-haze, respectively, and the concentration of individual compounds in this group increased with concentration of PM_{2.5}. As a significant biomass burning marker, levoglucosan was the dominant compound in the saccharide group and among polar and nonpolar compounds in every period (Fig. 6-5). The second highest in saccharide group was meso-erythritol. It should be noted that while levoglucosan is a primary anhydro sugar directly emitted from biomass burning, meso-erythritol is a sugar alcohol and secondary organic aerosol (SOA) product from photodecomposition of 1,3-butadienen with nitric oxides in the atmosphere (Angove et al., 2006; Jaoui et al., 2014). Fig. 6-4 shows the contributions of individual compounds to total saccharide, it is found that levoglucosan had an increase trend as 69%, 82% and 87% in pos-haze, low haze and episodic haze, , while meso-erythritol decreased from 25% to 6% from post-haze to episodic haze, other primary anhydro sugars (mannosan and galactosan) and sugar alcohol (D-threitol) did not have significant changes.

Pearson's correlation matrix of saccharide compounds is shown in Table SD-4, all anhydro sugars had very strong correlation to one another in both haze and post-haze, suggesting that they were emitted from the same source (biomass burning). While meso-erythritol had strong correlations with anhydro sugars only in haze period, implying that biomass burning contributes to the formation of meso-erythritol as secondary organic aerosol.

6.4.3.2 Nitrate aromatic phenols

The concentration of total nitrated aromatic phenol was 315.2 ± 227.4 , 69.1 ± 75.8 and 8.4 ± 8.2 ng m⁻³ during episodic haze, low haze and post-haze, respectively (Table SD-2). 4-nitrocatechol was the most abundant compound in this group followed by 4-nitrophenol, 2-methyl-4-nitrophenol and bisphenol A. 4-nitrocatechol and 4-nitrophenol have been commonly reported as the most abundant compounds in nitrophenol group (Cai et al., 2022; Claeys et al., 2012; Iinuma et al., 2016; Y. Wang et al., 2019; R. Zhang et al., 2023). From Fig. 6-5 and Table SD-2, the concentrations of all nitrated aromatic phenols increased with haze events except bisphenol A. In the case of contribution to total nitrate aromatic phenol, an increasing trend appeared with 4-nitrocatechol by which concentrations raised from 67% to 94% from post-haze to episodic haze, respectively (Fig. 6-4) while other compounds including 4-nitrophenol, 2-methyl-4-nitrophenol and bisphenol A had different trend where their contributions decreased as the intensity of haze accrued.

Correlation coefficients shown in Table SD-4 display that, during haze, 4-nitrophenol, 2-methyl-4-nitrophenol and 4-nitrocatechol had very strong correlations with one another and with all species of anhydrosugars, indicating that biomass burning highly influenced SOA formation of this group, while during post-haze, only 4-nitrocatechol showed very strong correlation with anhydrosugars. Bisphenol A had no relationship to any other compounds in both haze and post-haze.

6.4.3.3 Carboxylic acids

Measured carboxylic acids in this study were separated by their number of carboxyl functional groups into three subgroups including mono-, di-, and tri- carboxylic acids and one group of fatty acids. Total average concentrations of carboxylic acids during episodic haze, low haze and post-haze were 661.6 ± 163.6 , 325.6 ± 116.2 and 153.5 ± 63.5 ng m⁻³, second highest

mass among polar organic compounds. Compared to the total contribution of carboxylic acid, as shown in Fig.6-4, dicarboxylic acids were the major contributors at 57%, 74% and 78%, followed by monocarboxylic acid at 4%, 8% and 11% during post-haze, low haze and episodic haze, respectively. Dicarboxylic acids received high attention in previous studies due to their potential as tracers for secondary organic aerosol formation (Cao et al., 2017; Hsieh et al., 2008; Kaplan & Kawamura, 1987; Kawamura & Ikushima, 1993; J. Liu et al., 2021; Q. Yu et al., 2021). Fatty acids had a high significant contribution during post-haze at 35%. Significant contribution of dicarboxylic acids to total polar organic compounds from this study were consistent with reported by previous studies in China (H. Guo et al., 2015; Ren et al., 2020).

Monocarboxylic acid: The average concentrations of total monocarboxylic acid (cis-pinonic acid, dehydroabietic acid, pimaric acid, isopimaric acid and DHOPA) during episodic haze, low haze and post-haze were 73.5 ± 46.3 , 25.9 ± 11.9 and 5.8 ± 3.1 ng m⁻³, respectively. Dehydroabietic acid was the most abundant compound in this group followed by DHOPA, while the rest compounds including cis-pinonic acid, pimaric acid and isopimaric acid had mass concentration less than 5 ng m⁻³. Strong to very strong correlations (Table SD-4, R^2 ranged 0.70 – 0.83) of dehydroabietic acid, pimaric acid and DHOPA were found with anhydrosugars, indicating that these three compounds were highly influenced by biomass burning during haze period as SOA.

Dicarboxylic acids: This group comprises of number of acids namely succinic (C4), glutaric (C5), adipic (C6), pimelic (C7), suberic (C8), azelaic (C9), malic (hC4), phthalic (o-Ph) and isophthalic (p-Ph) acids. Total dicarboxylic acid mass concentrations were 466.2 ± 124.7 , 221.3 ± 90.2 and 76.7 ± 36.5 ng m⁻³ during episodic haze, low haze and post-haze, respectively. Malic acid was the greatest abundant in dicarboxylic acid and among overall carboxylic group, followed by succinic acid, this findings were similar to as reported previously in literatures (Claeys et al., 2010; Hsieh et al., 2008). The concentration of malic acid exceeded succinic acid approximately 2 times for haze and 4 times for post-haze period. The following compounds in descending order of mass

concentration were phthalic acid > azelaic acid > adipic acid > suberic acid > isophthalic acid > pimelic acid > glutaric acid. During haze, dicarboxylic acids had strong to very strong correlations with anhydrosugars (Table SD-4, R^2 ranged from 0.66 – 0.96), suggesting that biomass burning mainly contributed to this group. Moreover, very strong correlations were found between several dicarboxylic acids except glutaric acid and suberic acid, implying significant SOA formation.

Tricarboxylic acids: There are three measured compounds in this group including hemimellitic, trimellitic and trimestic acids. Total mass concentration of tricarboxylic acids was 85.9 ± 22.2 , 42.6 ± 15.9 and 17.5 ± 8.9 for episodic haze, low haze and post-haze, respectively. Hemimellitic and trimellitic were comparable in mass concentrations. Tricarboxylic acid had moderate correlations with anhydrosugars but strong correlations to one another within tricarboxylic acid group.

Fatty acids: Four fatty acids were measured in this group; palmitic, linoleic, oleic and stearic acids. Fatty acid was the only group whose total mass concentration was not influenced by haze phenomena, during episodic haze, low haze and post-haze was 36.0 ± 16.6 , 35.8 ± 32.2 and 53.6 ± 57.2 ng m⁻³, respectively. Palmitic and stearic were abundant fatty acids in this group. No significant correlation was found between fatty acids and anhydrosugars and other groups of carboxylic acid. Although, fatty acids had very strong correlations with cholesterol and 1-pentadecanol during post-haze, implying cooking sources.

6.4.3.4 Others

Syringaldehyde (aldehyde), cholesterol and 1-pentadecanol (alcohol) were also measured in this study. Their mass concentrations are listed in Table SD-2. Syringaldehyde mass concentration varied with haze while cholesterol and 1-pentadecanol did not significantly change. Only syringaldehyde had moderate correlations with anhydrosugars. Syringaldehyde is emitted

from biomass burning (Fujii, Tohno, et al., 2015; W. Li et al., 2021) especially from hardwoods (Fine et al., 2004b, 2004a)

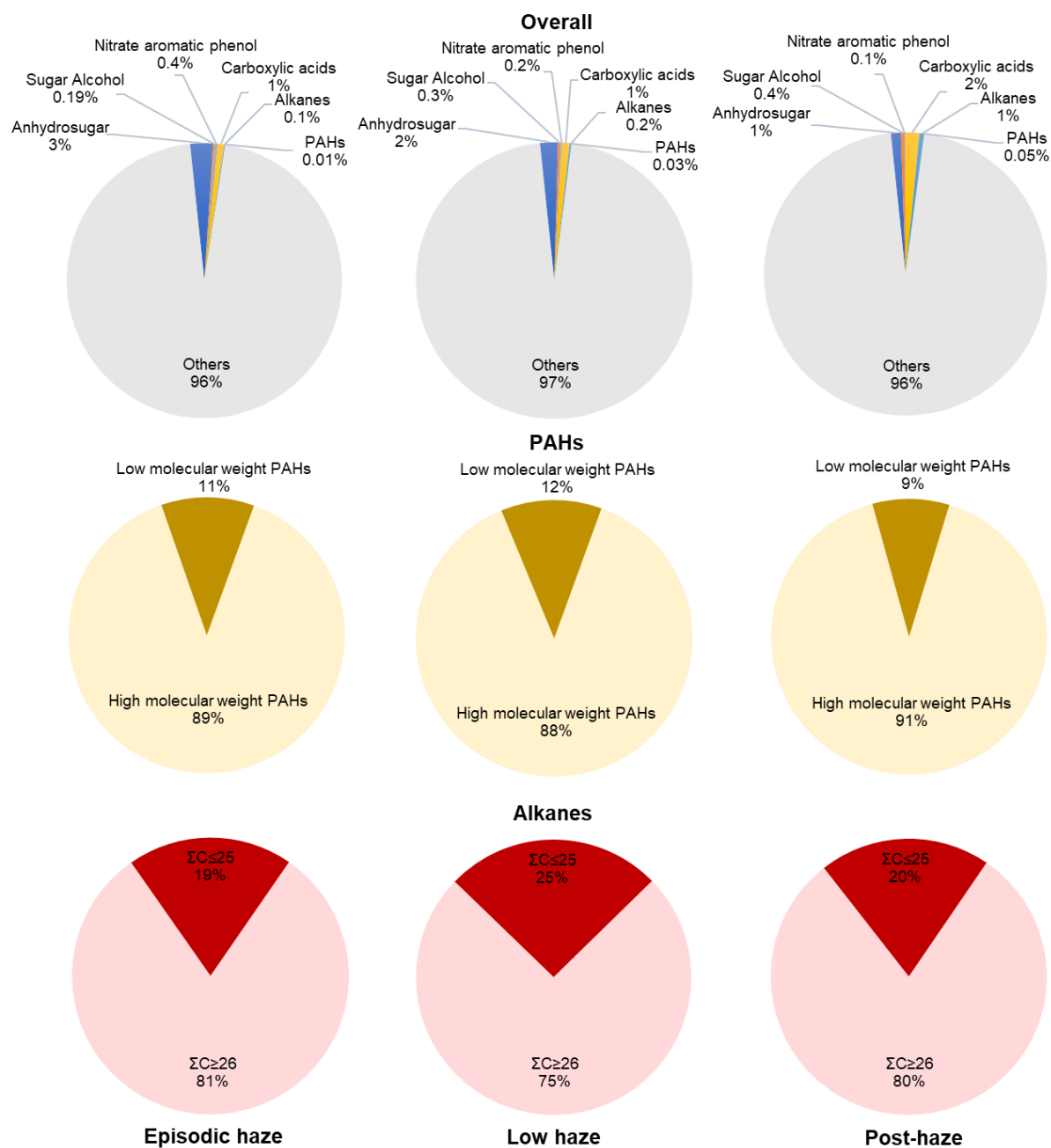


Fig. 6-3 Contributions of overall organic compounds and non-polar organic compound groups of PAHs and Alkanes during episodic haze (left), low haze (middle) and port-haze (right)

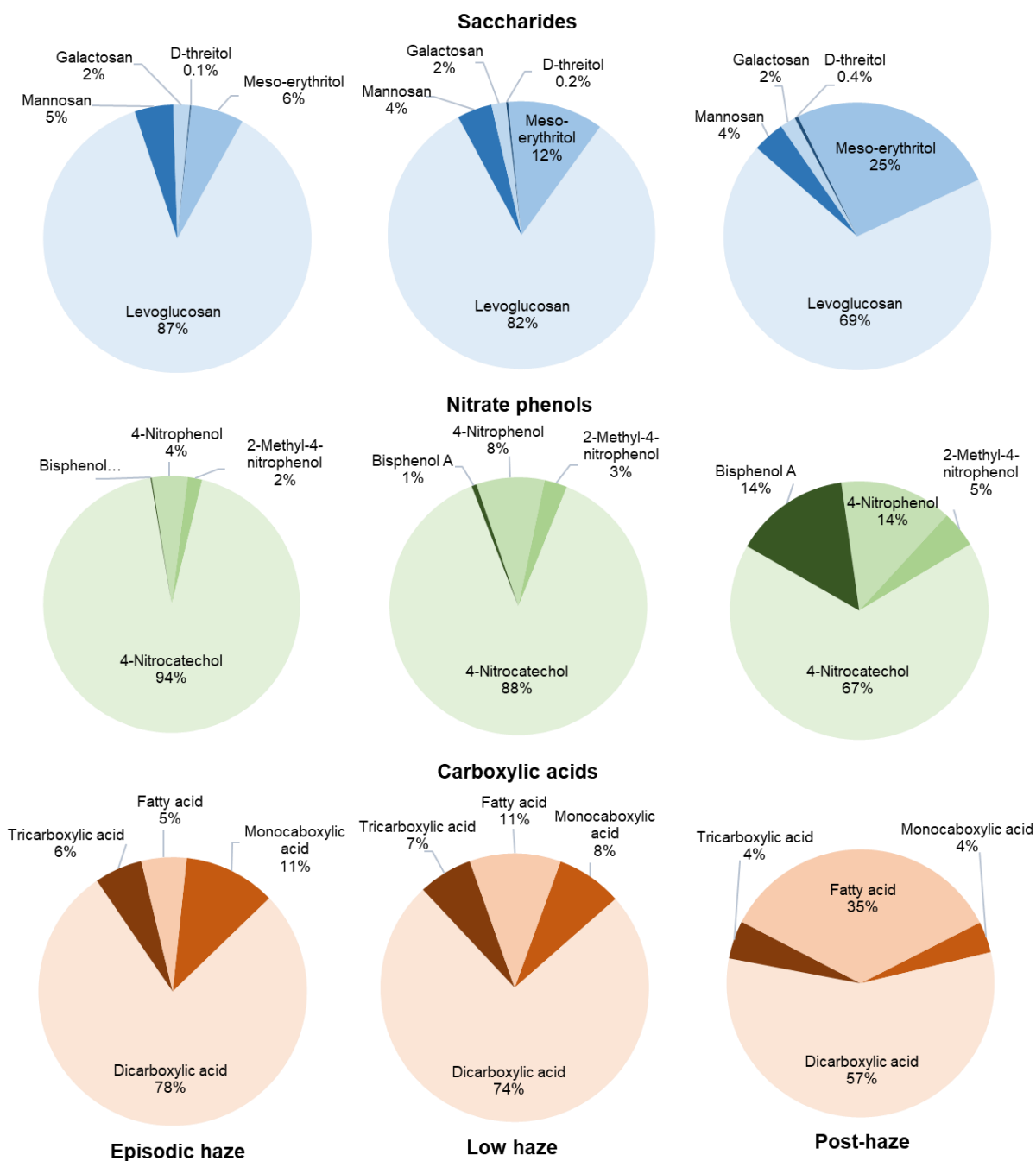


Fig. 6-4 Contributions of polar organic compound groups of saccharides, nitrate phenols and carboxylic acids during episodic haze (left), low haze (middle) and port-haze (right).

6.4.4 Variation of primary and secondary organic compounds, comparing haze and post-haze periods by mass incremental ratio

During haze period, average mass concentration of PM_{2.5} had increased 4 times higher than average mass during post-haze as shown in Table SD-3 (incremental ratio of PM_{2.5} was 4.2). Most of measured polar and nonpolar organic compounds elevated with an increase of PM_{2.5} mass. The increment ratios of nonpolar organic compounds ranged from 1 – 4.1 (Table SD-1) which were lower than the increment ratio of PM_{2.5}. Measured nonpolar compounds (alkanes and PAHs) are usually considered as primary organic compounds which are directly emitted from emission sources. Lyu et al. (2019) demonstrated that PM_{2.5} mass increased approximately 4 times from non-haze to haze days, but the mean concentration of primary organic compounds typically elevated by a factor of around 2.

In the case of polar organic compounds, increment ratios had massively high variation (ranged from 0.4 to 24.1, Table SD-2) since polar compounds comprised of both primary and secondary organic compounds. It is worth noting that biomass burning markers such as anhydrosugars and syringaldehyde (Simoneit, 2002), although they are primary organic compounds, had very high increment ratios (11.9, 11.2, 9.7 and 14.7 for levoglucosan, mannosan, galactosan and syringaldehyde respectively). This may be attributed to the major contribution of biomass burning to PM_{2.5} during haze period in this study and small contribution during post-haze period. Meso-erythritol, the second highest mass in the saccharide group in this study, had an increment ratio of 3.3; meso-erythritol is a secondary organic aerosol from photodecomposition of 1,3-butadiene (Angove et al., 2006; Jaoui et al., 2014).

Dicarboxylic acids have been extensively investigated from several previous studies due to their significant role involving secondary organic aerosols (Cao et al., 2017; Hsieh et al., 2008; Kawamura & Bikkina, 2016; J. Liu et al., 2021; Q. Yu et al., 2021; Y. Zheng et al., 2021). As shown in Table SD-2, most of compounds had an increment ratio above 4, implying a possibility for SOA

formation. Dicarboxylic acids can be originated from a number of sources. Succinic (C4) acid directly relate to biomass burning (Cao et al., 2017) while malic acid (hC4) is an SOA formed by photooxidation of succinic acid (Kawamura & Ikushima, 1993). The ratio between malic acid to succinic acid (hC4/C4) has been used to indicate the intensity of photochemical reaction (Kawamura & Sakaguchi, 1999; Q. Yu et al., 2021). Fig. 6-6 (a) illustrates temporal variation of mass concentrations of malic and succinic acids and their ratio (hC4/C4); the ratio was slightly suppressed during episodic events (Table 6-1, average ratio 1.5) while at the beginning to mid of post-haze the ratio were substantially elevated (average ratio 4.0), indicating that the latter period had stronger photooxidation than during the haze period. During extremely high haze, solar radiation is reduced by smoke plume results in lesser photooxidation; and conversely, clear sky enhances the reaction (Wu et al., 2020; Q. Yu et al., 2019). Glutaric acid (C5) and adipic acid (C6) can be formed by the photochemical oxidation of anthropogenic cyclohexene (Hatakeyama et al., 1985) and aromatic hydrocarbons (Kawamura & Ikushima, 1993). Suberic acid and azelaic acid (C9) are considered to form by the photooxidation of biogenic unsaturated fatty acid (Kaplan & Kawamura, 1987; Stephanou & Stratigakis, 1993). Phthalic (o-Ph) and isophthalic (p-Ph) acids are SOA from anthropogenic sources and formed by aging process of aerosols (J. Li et al., 2020). The ratios between adipic acid to azelaic acid (C6/C9) and phthalic to azelaic acid (o-Ph/C9) can then be used to differentiate the influence of biogenic and anthropogenic sources (Kundu et al., 2010). Temporal distributions of adipic acid, phthalic acid and azelaic acid and their ratios are shown in Fig. 6-6 (b) and (c), C6/C9 and o-Ph/C9 ratios during post-haze (3.4 and 0.9) were higher than haze (1.7 and 0.8) (Table 6-1), indicating higher anthropogenic influence during post-haze.

Monocarboxylic acids also play a significant role especially in biomass burning derived SOA, dehydroabietic acid, pimaric acid and isopimaric acid are secondary organic markers and resin acid derivative products from combustion of softwoods burning (Fine et al., 2004b, 2004a; Hays et al., 2002; Mancilla et al., 2021); their increment ratios were 12.8, 1.9 and 15.6 respectively.

DHOPA, a secondary organic aerosol from anthropogenic emission formed by toluene and m-xylene oxidation (Srivastava et al., 2023) associate with biomass burning (T. Liu et al., 2020), had an increment ratio of 4.5. Cis-pinonic acid, formed by photooxidation of monoterpene with ozone (Glasius et al., 2000; Kavouras et al., 1999), had the ratio of 3.2. While tricarboxylic acids had incremental ratios below 4 for all compounds, 3.8, 3.7 and 3.5 for hemimellitic acid, trimellitic acid and trimesic acid, respectively. Although tricarboxylic acids have been observed associated with secondary formation of aerosols (X. He et al., 2018; Mu et al., 2021).

Significantly high increment ratios were found with nitrated aromatic phenols, which were 24.1, 7.2 and 8.3 for 4-nitrocatechol, 4-nitrophenols and 2-methyl-4-nitrophenol, respectively. These compounds have been considered as secondary organic aerosols derived from biomass burning (Bertrand et al., 2018; Finewax et al., 2018; X. Li et al., 2016; Y. Liang et al., 2021) especially 4-nitrocatechol had substantially elevated during episodic haze. 4-nitrocatechol and 4-nitrophenol and are usually observed as the most abundant compounds in nitroaromatic group (Y. Wang et al., 2019; Yuan et al., 2021). 4-nitrocatechol is also an SOA product from the reaction of catechol with OH in the presence of NO_x (Finewax et al., 2018) and has been used as a molecular tracer for aged aerosols (Bertrand et al., 2018; Q. Wang et al., 2022). While bisphenol A associated with primary emission from the incineration of plastic waste (Munyaneza et al., 2022) had incremental ratio of 0.4.

The lowest increment ratios were observed from fatty acids group with value below 2, suggesting that compounds this group are primarily emitted as primary organic aerosols and are unlikely to have involved biomass burning during the study period. No significant change in mass concentrations implies to background emissions, for example, fatty acids are usually reported to be emitted from cooking (Fadel et al., 2021; Mu et al., 2021).

To display a broad picture of variation of POA and SOA throughout the whole period of study, total primary (Σ POA) and total secondary (Σ SOA) organic compounds and their ratio

($\Sigma\text{SOA}/\Sigma\text{POA}$) are plotted in Fig. 6-6 (d). It can be clearly seen that, even though the absolute mass of SOA was higher in haze than post-haze, the $\Sigma\text{SOA}/\Sigma\text{POA}$ of haze period was lower than of post-haze, implying that the formation of SOA was proportionally higher during post-haze than haze period; in addition, SOA formation was also slightly suppressed during episodic haze ($\text{PM}_{2.5} > 100 \mu\text{g m}^{-3}$). This finding is consistent with a recent study performed in Bangkok, capital city of Thailand, $\text{PM}_{2.5}$ and O_3 (oxidizing agent for SOA formation) had negative correlation suggesting that high $\text{PM}_{2.5}$ attenuate sunlight and inhibit the photooxidation of O_3 in the atmosphere resulting in lessen SOA formation (Uttamang et al., 2023).

Table 6-1 Ratios of phthalic acid to azelaic acid, adipic acid to azelaic acid, malic acid to succinic acid and ΣSOA to ΣPOA during episodic haze, low haze and post-haze, respectively.

	Episodic	Low haze	Post-haze
Phthalic acid/Azelaic acid (Ph/C9)	1.5	1.8	3.4
Adipic acid/Azelaic acid (C6/C9)	0.7	0.8	0.9
Malic acid/Succinic acid (hC4/C4)	1.5	2.5	4.0
$\Sigma\text{SOA}/\Sigma\text{POA}$	0.4	0.5	1.1

ΣSOA – sum of D-threitol, meso-erythritol, 4-nitrophenol, 2-methyl-4-nitrophenol, 4-notrocatechol, dicarboxylic acids (except succinic acid), cis-pinonic acid, dehydroabietic acid, pimaric acid, isopimaric acid, and DHOPA
 ΣPOA – sum of PAHs, alkanes, anhydrosugars and fatty acids

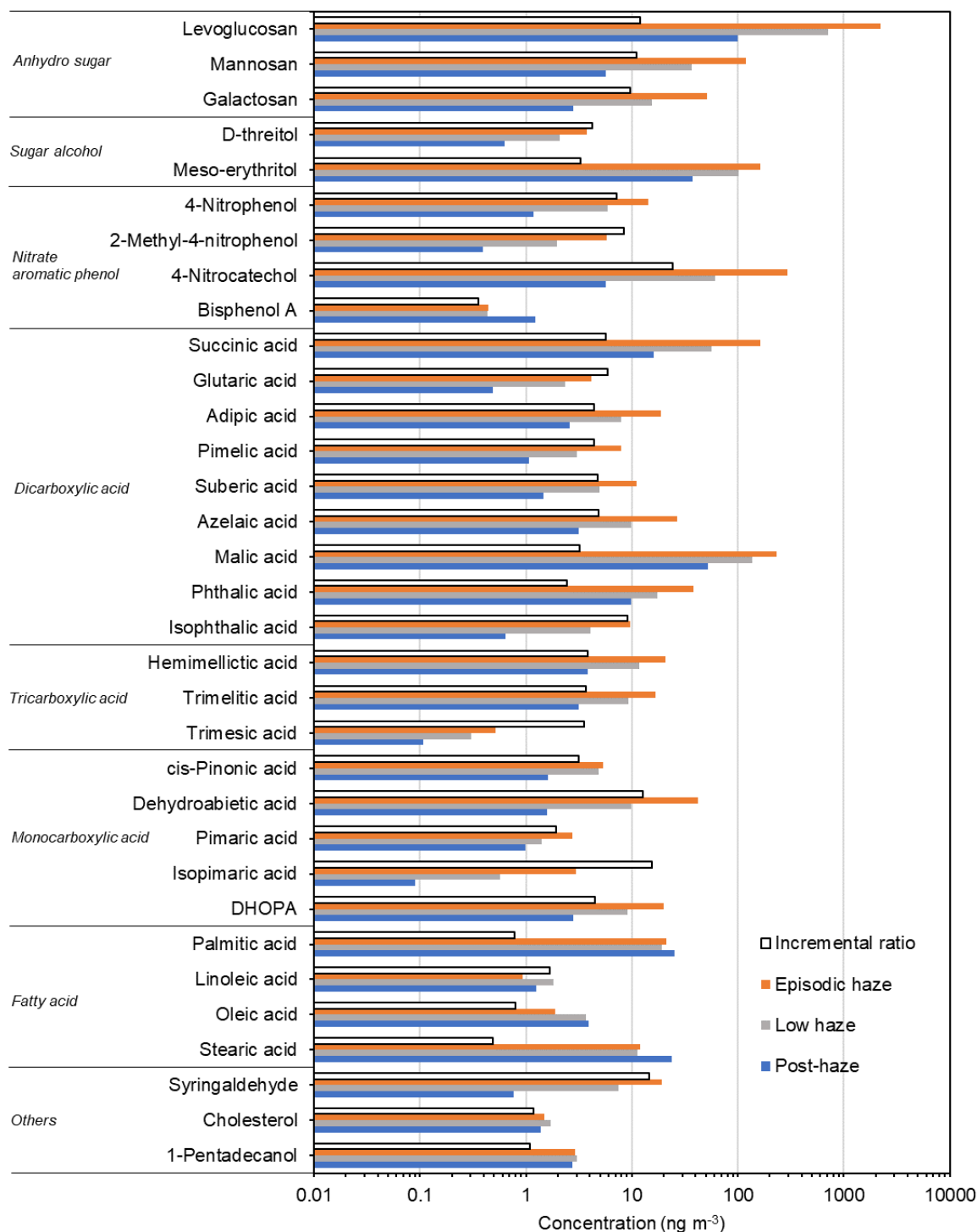


Fig. 6-5 Average concentration of polar organic compounds during episodic haze, low haze and post-haze. Incremental ratio is calculated by concentration of entire haze divided by concentration of post-haze

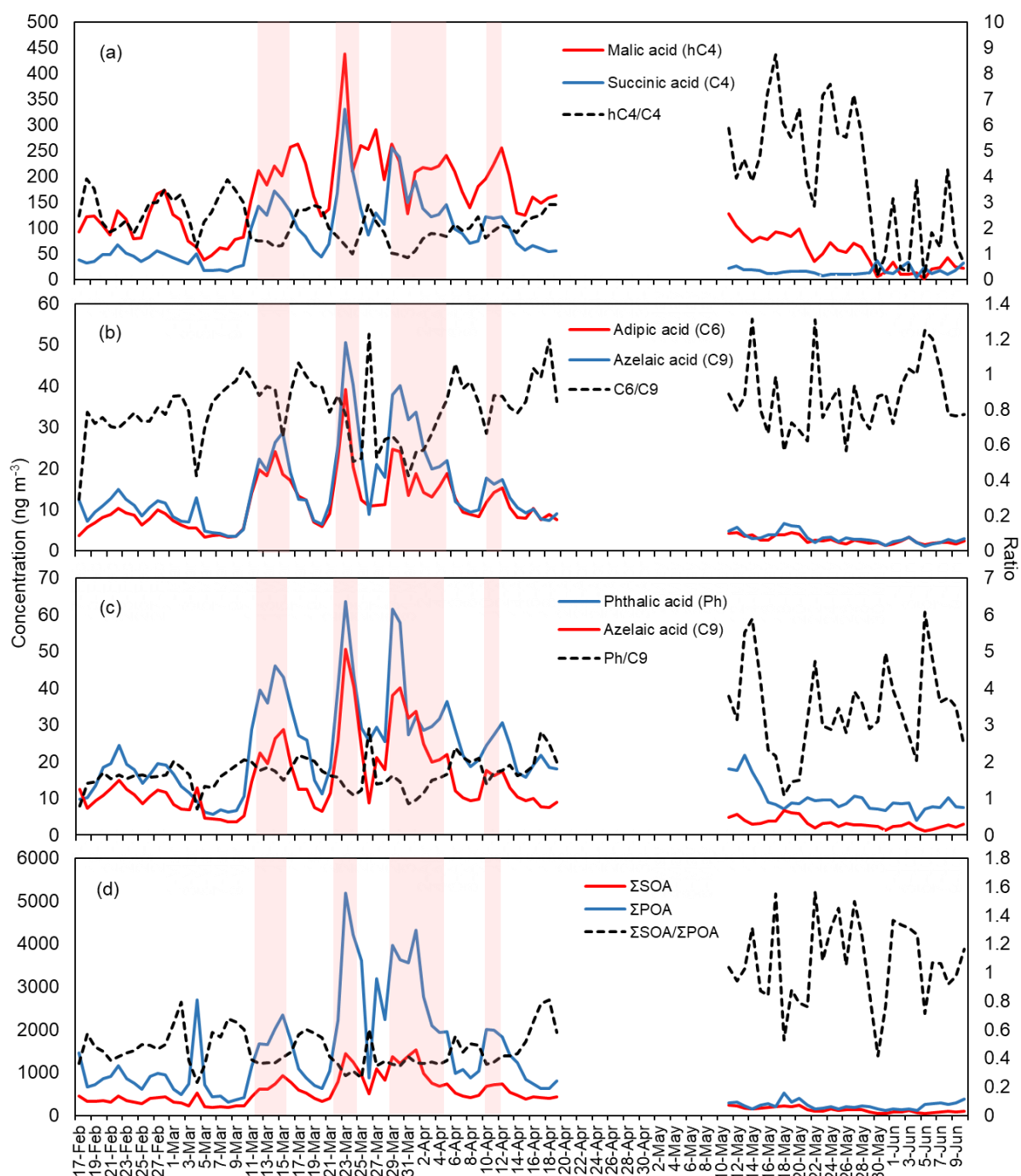


Fig. 6-6 Temporal distributions of selected compounds and their ratio (a) malic and succinic acids, (b) adipic and azelaic acids, (c) phthalic and azelaic acids and (d) ΣSOA and ΣPOA. Light pink date represents episodic haze events (PM_{2.5} > 100 μg m⁻³)

6.5 Conclusion

Fine particulate matter ($PM_{2.5}$) were collected during haze (Feb 17th to Apr 19th) and post-haze (11th May to 10th Jun), 2019 at a rural site in Chiang Mai, northern Thailand. Polar (PAHs and alkanes) and nonpolar (saccharides, nitrate aromatic phenols, carboxylic acids and others) organic compounds were comprehensively analyzed. The measured compounds contributed 3 – 4% of OM mass. HMW PAHs were strongly associated with biomass burning in haze period especially BkF and BghiP. For alkanes, ΣC_{odd} was higher than ΣC_{even} during haze period, while ΣC_{odd} was lower than ΣC_{even} during post-haze period. Average CPI index from alkanes > 1 during haze and < 1 during post-haze, implying the emission of biogenic source and anthropogenic source during haze and post-haze, respectively. Nonpolar compounds did not display significant change in contribution between episodic haze, low haze and post-haze. Saccharide was the major contributor followed by carboxylic acid and nitrate aromatic phenol. The most abundant compounds in saccharides, nitrate aromatic phenol and carboxylic acid were levoglucosan, 4-nitrocatechol and malic acid. According to incremental ratios and studies published previously, nonpolar organic compounds, anhydrosugars were the important compounds of POA content while dicarboxylic acids and nitrated aromatic phenols were abundant group of compounds of SOA followed by monocarboxylic acids and tricarboxylic acids. The ratios of adipic acid to azelaic acid (C6/C9) and phthalic to azelaic acid (o-Ph/C9) indicated anthropogenic source dominant during post-haze. The ratio of malic acid to succinic acid (hC4/C4) displayed a higher photooxidation during post-haze than haze period, episodic haze also slightly suppressed photooxidation, consistent with the ratio of $\Sigma SOA/\Sigma POA$ which was higher in post-haze than haze. This study provides a first comprehensive information on characteristics of polar and nonpolar organic compounds and a broad picture of POA and SOA in different period of haze and post-haze in northern Thailand, which are useful for future study and provides valuable information for policymakers to develop evidence-based policies to mitigate the impacts of SOA and their adverse effects on human health.

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CHAPTER SEVEN:

CONCLUSIONS AND RECOMMENDATIONS

FOR FUTURE WORK

7.1 Conclusions

The northern Thailand has been suffering from recurrent haze event every year during local summer between February to April. During such period, haze in northern Thailand is generally from anthropogenic activities, particularly biomass burning from agricultural practices since Thailand is known as an agrarian country, which contribute significantly to the haze problem. The practice of open burning agricultural fields, crop residues, and forested areas is a common method employed by farmers to clear land for cultivation or manage agricultural waste. This study present i) characteristics of chemical components of PM_{2.5} and source appointments by PMF model using comprehensive set of bulk chemical analysis coupled with source-specific organic tracers, ii) characteristics of polycyclic aromatic hydrocarbons (PAHs) and source-specific lifetime cancer risk (PMF-LCR) from exposure to PAHs and heavy metals, and iii) characteristics of polar and nonpolar organic compounds involving with primary (POA) and secondary organic aerosols (SOA) during haze and post-haze periods in year 2019, in northern Thailand.

Daily PM_{2.5} was collected on quartz fiber filter at a rural site in Chiang Mai, northern Thailand, between haze (17 February to 19 April) and post-haze (11 May to 10 June) in year 2019 by high volume sample, total 92 samples, then the collected samples were kept in a freezer at -20 °C prior to the bulk chemical analysis. Organic carbon (OC) and elemental carbon (EC) were analyzed by DRI model 2015 OC/EC carbon analyzer using IMPROV_A protocol. Water soluble inorganic ions (WSIIs) including NH_4^+ , K^+ , Mg^{2+} , Ca^{2+} , Cl^- , SO_4^{2-} , NO_3^- were analyzed using ion chromatography (IC) by Dionex Integrion HPIC and Dionex ICS 1100 for cation and anion, respectively. Crustal elements and trace elements (total 22 elements including Mg, Al, K, Ca, Ti, Fe, V, Cr, Mn, Co, Ni, Cu, Zn, As, Rb, Sr, Cd, Sb, Ba, Ce, W, and Pb) were analyzed by Inductively Coupled Mass Spectrometry (ICP-MS). Organic compounds, namely, n-alkanes, PAHs, carboxylic acids, nitro phenols, saccharides, fatty acids were analyzed by using GC/MS (Agilent 7890A GC coupled

to 5975C MS) in electron ionization mode (EI, 70 eV). Compounds were separated on a Restek RXxi-5MS column (30 m × 0.25 mm × 0.25 μm).

Daily average PM_{2.5} concentrations were exceeded 24hr average PM_{2.5} for Thailand National Ambient Air Quality standard (37.5 μg m⁻³) and the World Health Organization (WHO) 24hr average standard (15 μg m⁻³) throughout haze period, while during post-haze period, the daily PM_{2.5} were generally lower than Thailand's standard for the most. Average mass concentration of PM_{2.5} during haze and post-haze were 86.6 ± 36.2 and 20.8 ± 11.5 μg m⁻³, respectively. Duration with PM_{2.5} above 100 and below 100 μg m⁻³ were identified as episodic haze and low haze, respectively. Correlation coefficient (*R*²) of study site were strong with other sites across northern Thailand suggesting the haze event was regional scale. OC was the most contributor (28.5 – 42.8%) in PM_{2.5} mass in every period while EC contributed less than 5% of PM_{2.5} mass. The ratio of char-EC/soot-EC indicates biomass burning during haze and vehicular emission during post-haze. SO₄²⁻, NO₃⁻, and NH₄⁺ were the most abundant ions in WSIs. Strong correlation coefficients between NH₄⁺ and SO₄²⁻ indicate secondary sulfate for whole study period while NO₃⁻ and Ca²⁺ during episodic and low haze indicates secondary nitrate. Ratio NO₃⁻/SO₄²⁻ during haze were lower than post-haze suggests that stationary source dominate in haze period. Crustal elements (Mg, Al, K, Ca, Ti and Fe) had higher mass concentration than trace elements. The ratio between levoglucosan/K⁺, levoglucosan/dehydroabietic acid and Cl-/4-nitrocatechol reveal characteristics of episodic haze and low haze; episodic had more contribution of softwood and aged aerosols while low haze had more contribution of hardwood and agricultural waste burning. Mass reconstruction during haze were OM (62%) > WSIs (17%) > mineral dusts (14%) > EC (5%) > other (2%) > tracer elements (0.2%) > sea salt (0.8%) while post-haze was OM (46%) > WSIs (25%) > mineral dusts (16%) > other (11%) > EC (4%) > sea salt (2%) > tracer elements (0.4%).

Source appointment performed by PMF model using comprehensive set of bulk chemical components coupled with organic tracers, dehydroabietic acid and 4-nitrocatechol. 6 main factors

were identified namely general biomass burning, nearby biomass burning (fresh aerosol), softwood burning (aged aerosols), secondary sulfate, mixed dust and traffic related emissions, and secondary nitrate. General biomass burning identified by significant contribution to levoglucosan, OC, and K^+ , this source associate with air mass arriving from northwest direction through continent area and contributed 31% and 5% during haze and post-haze. Nearby biomass burning was identified by significant contribution to Cl^- and moderate contribution to OC, levoglucosan, this source associate with air mass circulating within Chiang Mai and contributed 19% and 32% during haze and post-haze. Softwood burning (aged aerosols) was identified by significant contribution to levoglucosan, OC, dehydroabietic acid (softwood marker), and 4-nitrocetachol (aged aerosol marker), this source associated with air mass arriving from south to southwest directions and contributed 31% and 1% during haze and post-haze. Secondary sulfate was identified by substantial contribution to NH_4^+ and SO_4^{2-} , this source did not show a similar trend with haze and had contribution of 17% and 34% during haze and post-haze. Mixed dusts and traffic related emissions identified by significant contribution to crustal elements (Al, Ti and Fe) and heavy metals that relate to exhaust and non-exhaust emissions (Ni, Ce, Ba, V and Mn) and higher contribution to EC than OC, this source contributed 15% and 26% during haze and post-haze. Secondary nitrate identified by high contribution to NO_3^- and Ca^{2+} which was first time identified in northern Thailand, contribution of this source was 5% and 2% during haze and post-haze.

Polycyclic aromatic hydrocarbons (PAHs) show similar trend with $PM_{2.5}$ mass but the ratio between $\Sigma PAHs/PM_{2.5}$ dropped as $PM_{2.5}$ mass increase, suggesting haze contributed to PAHs in some extent. $\Sigma PAHs$ mass during episodic haze, low haze and post-haze were 13.2 ± 3.6 , 9.7 ± 1.3 and 7.9 ± 2.2 ng m^{-3} , respectively which were higher than previous studies but lower than of those studies in China and India. 4-rings PAHs were the most abundant group followed by 5- and 6-rings PAHs. However, the contributions of 5- and 6-rings PAHs increased with haze events while other number of ring-PAHs decreased with haze, specifically, only the contribution of

benzo(b)fluoranthene and benzo(ghi)pyrene increased with haze events. The correlation of high molecular weight PAHs had strong correlation with levoglucosan during haze while lower molecular weight PAHs had moderate correlation with PM_{2.5} implying mixed source contributed to PAHs. Overall lifetime cancer risk was in moderate range of 4.21×10^{-5} and 0.67×10^{-5} for haze and post-haze, respectively. Source specific lifetime cancer risk obtained from PMF-LCR 6 sources including gasoline, diesel, biomass burning, softwoods burning, mixed dusts and secondary sulfate. LCR during haze and post-haze for each source were gasoline (0.26×10^{-5} and 0.28×10^{-5}), diesel (1.35×10^{-5} and 0.47×10^{-5}), biomass burning (0.6×10^{-5} and 0.21×10^{-5}), softwood burning (1.23×10^{-5} and 0.16×10^{-5}), mixed dusts (0.51×10^{-5} and 0.04×10^{-5}), and secondary sulfate (0.19×10^{-5} and 0.25×10^{-5}). By mitigating 3 main contributors of source specific LCR during haze (biomass burning, softwood burning, and diesel emissions) and 2 main contributors during post-haze (diesel and gasoline emissions) by 50% could result in 41% and 21% reduction of lifetime cancer risk during haze and post-haze, respectively.

Average concentrations during episodic haze, low haze, and post-haze of nonpolar organic compounds were 121.1 ± 34.2 , 83.6 ± 27.1 and 53.1 ± 12.0 ng m⁻³, and polar organic compounds were 3563.0 ± 1337.8 , 1278.6 ± 693.5 and 313.3 ± 133.1 ng m⁻³, respectively. Contribution of both polar and nonpolar organic compounds ranged between 3 – 4% of organic matter (OM). For nonpolar organic compounds, the concentrations of LMW PAHs and HMW PAHs were 1.4 ± 0.4 , 1.1 ± 0.4 , 0.5 ± 0.3 and 11.7 ± 3.3 , 8.4 ± 1.1 , 4.7 ± 2.0 ng m⁻³, average concentrations of total alkanes were 107.9 ± 31.4 , 74.1 ± 26.6 , 48.8 ± 10.7 ng m⁻³, in episodic haze, low haze and post-haze, respectively. Total concentration of odd number alkanes (ΣC_{odd}) was higher than even number alkanes (ΣC_{even}) during episodic and low haze while ΣC_{odd} was lower than ΣC_{even} during post-haze. Carbon preference index (CPI) higher than 1 during haze indicates combustion from biogenic source while CPI lower than 1 during post-haze indicates anthropogenic emissions. For polar organic compounds, total saccharide was 2561.7 ± 1026.7 , 873.1 ± 533.4 , and 147.2 ± 86.6 ng m⁻³ for episodic haze, low haze and post-haze, respectively, and levoglucosan was the most abundant

of saccharides and all identified compounds. Total concentration of total nitrated aromatic phenol was 315.2 ± 227.4 , 69.1 ± 75.8 and 8.4 ± 8.2 ng m⁻³ during episodic haze, low haze and post-haze, respectively, with the most abundance of 4-nitrocetachol. Strong correlation of nitrated aromatic phenol, dicarboxylic acid with anhyrosugars suggests SOA contributions from these group. Total concentration of carboxylic acids during episodic haze, low haze and post-haze were 661.6 ± 163.6 , 325.6 ± 116.2 and 153.5 ± 63.5 ng m⁻³, respectively, with the highest abundance of malic acid. Incremental ratios of nonpolar organic compounds were lower than 2 while polar organic compounds were mostly higher than the incremental ratios of PM_{2.5}, implying SOA formation in polar organic compounds especially dicarboxylic acids and nitrate aromatic phenols. The ratio between malic acid to succinic acid (hC4/C4) demonstrated that photooxidation was proportionally higher during post-haze than haze period with a slight suppression during episodic haze due to reduced solar radiation from intense haze.

7.2 Recommendations for future work

Finally, as the perspective of overall outcome from this study, haze events in northern Thailand compared to previous studies has not successfully been mitigated and become more complex in terms of biomass burning source. To improve feasible implication on targeted source mitigation, the following points should be taken into account:

- This study has shown that biomass burning in northern Thailand is a highly complex source, since Thailand is an agricultural county where open burning of biomass is difficult to identify. Therefore, a better separation in source apportionment is required by using a comprehensive set of chemical species and additional source specific organic markers such as markers for plastic burning, diesel or gasoline combustions, and different type of biomasses.

- Studies regarding source appointment in Thailand by the proper receptor model, PMF, are scarce, resulting in a lack of reliable comparison between studies. Therefore, rather than PCA model, PMF is strongly recommended as a robust receptor model.
- According to benefit from the use of PMF, it allows to perform a method in which incorporate with back trajectory analysis called potential source contribution function (PSCF), it is recommended to employ this method to display specific source associate with air mass direction and contribution.
- Since haze events in Thailand are generally in regional scale cover a vast area, to enable spatiotemporal and interdisciplinary research, besides ground-based experiments, integration by multiple types of information such as remote sensing from satellite, emission inventories and modeling forecasting is recommended.
- Assessment of increment lifetime cancer risk is crucial for public health aspect, as the results shown in this study, estimated lifetime cancer risks were substantially lower than the study where estimated both PAHs from gas phase and particulate phase. It is worth noting that estimated risk only from PM_{2.5} may be underestimated. Future study on lifetime cancer risk then should consider both gas and particulate bound PAHs.
- Biomass burning might not be a sole source that policymakers should consider on, conversely, as this study has shown that PAHs and heavy metals had a proportionally higher contribution in certain sources rather than biomass burning, such as diesel and gasoline emissions. It is recommended that longer periods of study during non-haze, and additional multiple receptor sites near traffic should be performed to receive an accurate cancer risk estimation.
- Study on SOA in Thailand is scarce, especially on derivatives PAHs, nitro-PAHs and oxy-PAHs, which can cause more harm than their parents PAHs. Understanding on SOA formation pathway provides valuable information for local authorities to inhibit their production such as mitigating the precursor gases emission.

APPENDICES

Supplement A

Table SA-1 Contribution of OC and EC (%) to PM mass in SEA countries

Country	Season	Site	PM size	OC	EC	TC	OC/EC	Ref
Thailand								
Phimai	dry	Rural	PM10	28.8	6.1	34.9	4.8	(Li <i>et al.</i> , 2013)
Bangkok	dry	traffic	PM2.5			55.8		(Chuersuwan <i>et al.</i> , 2008)
	dry	traffic	PM10			42.7		
Chiang Mai	haze	Rural	PM2.5	40.5	6.0	46.5	6.7	(Pani <i>et al.</i> , 2019a)
	haze	Urban	PM2.5	41.2	8.4	49.7	4.9	
Chiang Mai	LBB	urban	PM2.5	40.2	7.7	47.9	5.2	(Pani <i>et al.</i> , 2018)
	MBB	urban	PM2.5	31.4	5.9	37.3	5.3	
	HBB	urban	PM2.5	26.5	4.9	31.4	5.4	
	EBB	urban	PM2.5	32.3	6.0	38.3	5.4	
Chiang Mai	haze	urban	PM2.5	36.7	4.4	41.1	8.3	(Thepnuan <i>et al.</i> , 2019a)
Chiang Mai	haze	rural	PM2.5	40.9	7.3	48.2	5.6	(Chuang <i>et al.</i> , 2013)
Phuket	annual	suburban	PM2.5	7.2	1.5	8.7	4.8	(Choochuay <i>et al.</i> , 2020)
Vietnam								
Hanoi	wet	traffic	PM2.5	30.6	8.9	39.6	3.4	(Thuy <i>et al.</i> , 2018)
	dry	traffic	PM2.5	37.4	10.1	47.5	3.7	
	wet	mixed	PM2.5	40.2	9.7	49.9	4.2	
	dry	mixed	PM2.5	31.5	8.1	39.6	3.9	
	wet	traffic	PM10	36.6	10.1	46.6	3.6	
	dry	traffic	PM10	73.3	18.3	91.6	4.0	
	wet	mixed	PM10	42.2	8.8	51.0	4.8	
	dry	mixed	PM10	29.2	7.1	36.2	4.1	
Tamdao	wet	rural	PM2.5		8.8			(Co <i>et al.</i> , 2014)
	dry	rural	PM2.5		8.0			
Hanoi	dry	urban	PM2.5	19.2	6.2	25.4	3.1	(Hai & Kim Oanh, 2013)
	dry	urban	PM10-2.5	7.2	1.7	9.0	4.2	
Hanoi	annual	suburban	PM2.5		8.1			(Cohen <i>et al.</i> , 2010a)
Mong Duong	wet	Industrial	PM2.5	9.4	5.7	15.1	1.7	(Hang & Kim Oanh, 2014)
	dry	rural	PM2.5	19.0	3.7	22.7	5.2	
Cam Hai	wet	rural	PM2.5	10.7	3.0	13.7	3.6	
	dry	rural	PM2.5	20.9	2.5	23.4	8.5	
Sonla	BBSC	rural	PM2.5	35.2	5.1	40.3	7.0	(C. Te Lee <i>et al.</i> , 2016)
	BBIC	rural	PM2.5	46.5	7.5	54.0	6.2	
	BBSS	rural	PM2.5	38.9	6.1	45.0	6.4	
Malaysia								
Kuala Lumpur	haze	suburban	PM2.5	25.3	8.3	33.6	3.1	(Khan, Sulong, <i>et al.</i> , 2016)
Kuala Lumpur	annual	urban	PM2.5		15.8			(Rahman <i>et al.</i> , 2011)
	annual	urban	PM10		2.6			
Selangor, Malaysia	haze	urban	PM2.5	24.0	9.2	33.3	2.6	(Fujii <i>et al.</i> , 2016)
Singapore								
Singapore	non	urban	PM2.5	16.9	8.2	25.1	2.1	(Tham <i>et al.</i> , 2019)
	haze	urban	PM2.5	20.1	4.4	24.5	4.6	
	episodic	urban	PM2.5	46.3	3.0	49.4	15.4	
Singapore	haze	urban	PM2.5	52.1	11.2	63.3	4.7	(Hapsari Budisulistiorini <i>et al.</i> , 2018)
Singapore	haze	urban	PM2.5	19.7	7.1	26.8	2.8	(See <i>et al.</i> , 2006)
	non	urban	PM2.5	17.7	9.1	26.8	1.9	
Indonesia								
Serpong	Aug – Nov 2008	Industrial	PM2.5		17.2			(Santoso <i>et al.</i> , 2011)
		Residential	PM2.5		12.9			
Avg SEA				30.7	7.5	38.3	4.9	

Table SA-2 Contribution of OC and EC (%) to PM mass in other countries

Country	Season	Site	PM size	OC	EC	TC	OC/EC	Ref
China								
Beijing	haze	urban	PM2.5	14.2	3.4	17.6	4.2	(Zhang <i>et al.</i> , 2016)
	non-haze	urban	PM2.5	24.6	7.4	32.0	3.3	
Beijing	haze	urban	PM2.5	10.1	2.3	12.4	4.4	(R. Shen <i>et al.</i> , 2017)
	haze	urban	PM2.5	12.1	3.7	15.7	3.3	
	haze	urban	PM2.5	10.1	4.0	14.1	2.5	
	non-haze	urban	PM2.5	17.7	4.3	22.0	4.1	
Beijing	heavy-polluted	urban	PM2.5	15.8	4.3	20.1	3.7	(Tao <i>et al.</i> , 2015)
	clean	urban	PM2.5	25.0	6.4	31.4	3.9	
Beijing	haze	urban	PM2.5	18.6	2.4	21.0	7.6	(K. F. Ho <i>et al.</i> , 2016)
Xi'an	haze	urban	PM2.5	17.5	3.3	20.8	5.4	
Xiamen	haze	urban	PM2.5	20.0	7.0	27.0	2.9	
Hong Kong	haze	urban	PM2.5	16.5	9.8	26.3	1.7	
Beijing	annual	urban	PM2.5	14.9	4.4	19.3	3.4	(Ji <i>et al.</i> , 2016)
Brijing	haze	urban	PM2.5	36.5	15.6	52.1	2.3	(X. Xu <i>et al.</i> , 2018)
Beijing	haze	urban	PM2.5	21.7	5.6	27.3	3.9	(Shao <i>et al.</i> , 2018)
	non-haze	urban	PM2.5	13.4	4.1	17.6	3.2	
	avg	urban	PM2.5	19.8	5.3	25.0	3.7	
Nanjing	annual	industrial	PM2.5	19.2	6.4	25.6	3.0	(Li <i>et al.</i> , 2016)
	annual	urban	PM2.5	13.0	4.8	17.8	2.7	
Handan	winter 2013	urban	PM2.5	16.0	6.5	22.5	2.5	(Zhao <i>et al.</i> , 2019)
	summer 2013	urban	PM2.5	8.5	3.9	12.4	2.2	
	annual 2013	urban	PM2.5	14.2	5.5	19.7	2.6	
Handan	winter 2017	urban	PM2.5	33.6	6.6	40.2	5.1	
	summer 2017	urban	PM2.5	9.3	4.2	13.4	2.2	
	annual 2017	urban	PM2.5	21.0	5.4	26.4	3.9	(J. Tan <i>et al.</i> , 2017)
Lanzhou	winter	residential	PM2.5	29.4	11.5	40.8	2.6	
	summer	residential	PM2.5	28.6	13.0	41.6	2.2	
	annual	residential	PM2.5	29.2	11.8	41.0	2.5	(X. Liang <i>et al.</i> , 2019)
Ningdong (Yinglite)	winter	industrial	PM2.5	7.0	4.7	11.7	1.5	
	summer	industrial	PM2.5	6.1	3.2	9.4	1.9	(Lu <i>et al.</i> , 2020)
Cixi	haze	mixed	PM2.5	16.8	6.1	22.9	2.8	
	non-haze	mixed	PM2.5	18.9	7.4	26.3	2.6	(Niu <i>et al.</i> , 2016)
Xi'an	spring	urban	PM2.5	14.8	3.8	18.5	3.9	
	summer	urban	PM2.5	14.4	3.3	17.7	4.3	
	autumn	urban	PM2.5	18.1	5.1	23.2	3.5	
	winter	urban	PM2.5	13.7	3.0	16.7	4.6	
	annual	urban	PM2.5	16.9	4.2	21.1	4.0	(Jiang <i>et al.</i> , 2018)
Zhengzhou	heavy polluted	urban	PM2.5	16.9	3.5	20.4	4.8	
	other days	urban	PM2.5	14.3	4.7	19.0	3.0	(Tao <i>et al.</i> , 2013)
Chengdu	non-haze	urban	PM2.5	16.6	4.8	21.4	3.4	
	dust	urban	PM2.5	8.4	2.1	10.5	4.0	
	haze	urban	PM2.5	21.0	4.5	25.4	4.7	
	annual	urban	PM2.5	15.5	4.3	19.8	3.6	
Avg China				17.4	5.5	23.0	3.4	

Table SA-2 (continue)

Country	Season	Site	PM size	OC	EC	TC	OC/EC	Ref
India								
Delhi	annual	industrial	PM2.5	28.2	9.1	37.3	3.1	(Mandal <i>et al.</i> , 2014)
Delhi	summer	traffic	PM2.5	30.2	13.4	43.6	2.3	(Pant <i>et al.</i> , 2015)
	winter	traffic	PM2.5	37.7	16.7	54.4	2.3	
Delhi	annual	urban	PM2.5	14.1	8.2	22.3	1.7	(Sharma & Mandal, 2017)
	winter	urban	PM2.5	14.7	8.5	23.2	1.7	
	summer	urban	PM2.5	13.8	8.4	22.2	1.6	
Delhi	winter	urban	PM2.5	12.9	5.8	18.7	2.2	(Jain <i>et al.</i> , 2020)
	summer	urban	PM2.5	9.5	5.1	14.6	1.9	
	monsoon	urban	PM2.5	9.8	4.4	14.2	2.2	
	annual	urban	PM2.5	12.0	5.6	17.6	2.2	
Pune	May – Oct 2013	urban	PM2.5	30.5	2.3	32.9	13.1	(Pipal & Satsangi, 2015)
		urban	Pm10	20.0	1.5	21.5	13.8	
Trombay	annual	industrial	PM2.5		12.5			(Police, Kumar, et al., 2018)
	annual	industrial	PM2.5		14.2			
Ahmedabad	winter	urban	PM2.5	32.9	5.4	38.2	6.1	(Rengarajan <i>et al.</i> , 2011)
Van Vihar National Park	annual		PM2.5	30.0	13.0	43.0	2.3	(Kumar & Sunder Raman, 2020)
Avg India				21.2	8.4	26.9	3.5	
UK								
Birmingham	11-21 Sep 2012	Traffic (Tunnel)	PM2.5	30.1	30.0	60.1	1.0	(Pant <i>et al.</i> , 2017)
Birmingham	summer 2-18 June 2011	Urbanbg	PM2.5	26.0	9.0	35.1	2.9	(Taiwo, 2016)
			PM2.5-10	13.5	0.3	13.8	42.9	
London	winter	urban	PM2.5	17.5	7.5	25.0	2.3	(Crilley <i>et al.</i> , 2015)
Detling, Kent	winter	rural	PM2.5	13.8	3.8	17.5	3.7	
Harwell, Oxfordshire	winter	rural	PM2.5	16.4	5.0	21.4	3.3	
London	winter	urban	PM2.5	22.3	9.6	31.9	2.3	(J. Yin et al., 2015)
Harwell, Oxfordshire	winter	rural	PM2.5	20.9	6.2	27.1	3.4	
Europe								
Barcelona	annual	urban back	PM2.5	16.1	6.5	22.6	2.5	(Salameh <i>et al.</i> , 2015)
Marseille	annual	urban back	PM2.5	31.6	9.2	40.8	3.4	
Genoa	annual	urban back	PM2.5	19.3	10.0	29.3	1.9	
Thessaloniki	annual	urban back	PM2.5	17.7	3.5	21.2	5.1	
Paris, France	annual	suburban	PM2.5	21.1	8.6	29.6	2.5	(Bressi <i>et al.</i> , 2013)
	annual	urban	PM2.5	20.3	9.5	29.7	2.1	
	annual	rural	PM2.5	23.0	9.5	32.5	2.4	
America/Canada								
Toronto, Canada	Annual	urban	PM2.5	7.5	7.0	14.5	1.1	(Jeong <i>et al.</i> , 2020)
Alberta, Canada	annual	urban	PM2.5	23.3	9.0	32.3	2.6	(Bari & Kindzierski, 2016)
Los Angeles, US	annual	urban	PM2.5-10	8.2	0.7	8.9	11.6	(Cheung <i>et al.</i> , 2011)
	annual	rural	PM2.5-10	5.7	0.4	6.1	15.4	

Table SA-3 Contribution of WSIs (%) to PM mass in SEA countries

Country	Season	Site	PM size	Ca ²⁺	Na ⁺	K ⁺	Mg ²⁺	NH4 ⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	NO ₃ ⁻ /SO ₄ ²⁻	Ref
Thailand													
Chiang Mai	haze	Rural	PM2.5	0.4	1.0	1.6		3.1	0.9	6.2	4.4	1.4	(Pani et al., 2019a)
	haze	Urban	PM2.5	0.5	1.2	1.6		2.0	1.2	3.5	4.3	0.8	
Chiang Mai	LBB	urban	PM2.5	0.7	4.3	2.5		5.0	1.8	2.5	15.5	0.2	(Pani <i>et al.</i> , 2018)
	MBB	urban	PM2.5	0.5	3.6	2.3		3.3	1.1	2.9	8.1	0.4	
	HBB	urban	PM2.5	0.3	2.6	2.0		2.6	0.6	3.0	4.1	0.7	
	EBB	urban	PM2.5	0.3	2.4	1.6		2.4	0.7	2.7	4.5	0.6	
Chiang Mai	haze	urban	PM2.5	1.2		2.0	0.2	5.2	0.2	4.2	13.6	0.3	(Thepnuan et al., 2019a)
Chiang Mai	haze	Rural	PM2.5	0.4	0.3	2.6		2.9	0.5	1.3	9.7	0.1	(Chuang <i>et al.</i> , 2013)
Chiang Mai	haze	urban	PM2.5	0.6	3.5	2.2		3.3	1.1	2.9	8.4	0.3	(Khamkaew <i>et al.</i> , 2016)
	haze	rural	PM2.5	0.5	4.9	2.2		1.2	1.2	5.0	3.0	1.6	
Chiang Mai	hot dry	urban	PM2.5	0.4	0.1	1.3	0.0	2.6	0.1	2.3	8.2	0.3	(Chansuebsri <i>et al.</i> , 2022)
	wet	urban	PM2.5		0.8	1.0		2.4	0.2	2.3	9.1	0.3	
	cold dry	urban	PM2.5		0.0	1.8		2.3	0.1	3.3	8.0	0.4	
	hot dry	Rural near source	PM2.5	0.1	0.2	1.2		1.9	0.1	2.4	6.1	0.4	
	wet	Rural near source	PM2.5		0.8	0.8		2.1	0.3	2.1	8.2	0.3	
	cold dry	Rural near source	PM2.5	0.1	0.5	1.1		1.5	0.4	1.1	5.7	0.2	
Phimai	dry	Rural	PM10	1.2	1.1	1.7	0.5	6.7	0.2	3.0	19.4	0.2	(Li <i>et al.</i> , 2013)
Bangkok	dry	traffic	PM2.5										(Chuersuwan <i>et al.</i> , 2008)
	dry	traffic	PM10					0.6	1.9	1.3	2.1	0.6	
Phuket	annual	suburban	PM2.5	2.3	3.5	0.7	0.3	0.7	1.3	1.3	5.5	0.2	(Choochuay <i>et al.</i> , 2020)
Vietnam													
Tamdao	wet	Rural	PM2.5	0.4	0.3	1.6	0.0	14.0	0.1	0.4	30.4	0.0	(Co <i>et al.</i> , 2014)
	dry	Rural	PM2.5	0.6	1.0	2.2	0.2	7.7	0.8	3.1	19.4	0.2	
Hanoi	dry	Urban	PM2.5		0.6	1.2		10.1		8.9	21.8	0.4	(Hai & Kim Oanh, 2013)
	dry	Urban	PM10-2.5		2.5	0.3		3.1		5.9	4.1	1.4	
Mong Duong	wet	indust	PM2.5	0.9	0.3	0.3	0.2	2.3	0.2	0.3	7.1	0.0	(Hang & Kim Oanh, 2014)
	dry		PM2.5	0.8	0.3	0.8	0.2	4.5	0.5	2.7	11.8	0.2	
Cam Hai	wet	Rural	PM2.5	0.4	0.4	0.4	0.0	4.1	0.2	0.5	10.0	0.1	
	dry		PM2.5	0.4	0.2	1.1	0.0	6.2	0.4	2.8	17.4	0.2	
Sonla	BBSX	Rural	PM2.5	0.2	0.2	1.4		5.2	0.2	4.0	14.4	0.3	(C. Te Lee et al., 2016)
	BBIC	Rural	PM2.5	0.2	0.0	1.7		2.8	0.2	1.7	7.9	0.2	
	BBSS	Rural	PM2.5	0.3	0.0	1.3		5.1	0.3	1.9	15.1	0.1	
Malaysia													
Bangi, Selangor	haze	suburban	PM2.5	0.2	0.3	0.8	0.6		0.2	1.3	3.9	0.3	(Jaafar <i>et al.</i> , 2018)
Kuala Lumpur	haze	suburban	PM2.5	1.1	0.8	1.2	0.3	3.0	0.1	1.5	10.8	0.1	(Khan, Sulong, et al., 2016)
Kuala Lumpur	wet	urban	PM2.5	1.4	1.2	0.6	0.3	2.0	0.2	3.7	8.3	0.5	(Khan <i>et al.</i> , 2017)
Bachok	wet	Rural	PM2.5	0.6	7.0	2.4	0.6	10.4	4.1	3.8	66.1	0.1	(Farren <i>et al.</i> , 2019)

Table SA-3 (continue)

Country	Season	Site	PM size	Ca ²⁺	Na ⁺	K ⁺	Mg ²⁺	NH ₄ ⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	NO ₃ ⁻ /SO ₄ ²⁻	Ref
Singapore													
Singapore	non	urban	PM2.5			1.3					19.5		(Tham <i>et al.</i> , 2019)
	haze	urban	PM2.5			1.1					13.2		
	episodic	urban	PM2.5			0.5					2.9		
Singapore	haze	urban	PM2.5	0.9	0.4	0.9		5.4	0.6	4.1	11.2	0.4	(Hapsari Budisulistiorini <i>et al.</i> , 2018)
Singapore	haze	urban	PM2.5		2.8	2.8		5.0	4.1	3.9	20.9	0.2	(See <i>et al.</i> , 2006)
	non	urban	PM2.5		4.1	2.2		4.9	3.2	3.1	22.8	0.1	
Indonesia													
Riau	haze	Near source	PM2.5					1.0	1.1	0.1	0.5	0.2	(Fujii <i>et al.</i> , 2019)
	haze	background	PM2.5					0.8	0.1	0.7	1.6	0.4	
Avg SEA				0.6	1.6	1.4	0.3	3.8	0.8	2.7	11.4	0.4	

Table SA-4 Contribution of WSIs (%) to PM mass in other countries

Country	Season	Site	PM size	Ca ²⁺	Na ⁺	K ⁺	Mg ²⁺	NH ₄ ⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	NO ₃ ⁻ /SO ₄ ²⁻	Ref
China													
Beijing	haze	urban	PM2.5	0.8	0.4	1.0	0.3	14.5	0.8	18.4	40.3	0.5	(Zhang <i>et al.</i> , 2016)
	non-haze	urban	PM2.5	2.4	0.7	1.3	0.4	10.5	0.7	14.4	26.4	0.5	
Beijing	haze	urban	PM2.5	0.6	0.3	0.7	0.1	12.4	2.1	24.1	19.7	1.2	(R. Shen <i>et al.</i> , 2017)
	haze	urban	PM2.5	0.7	0.3	0.7	0.1	13.3	0.6	16.2	24.5	0.7	
	haze	urban	PM2.5	0.4	0.3	1.0	0.1	17.2	2.3	17.4	26.1	0.7	
	non-haze	urban	PM2.5	2.9	0.5	2.0	0.2	6.3	1.4	8.2	11.2	0.7	
Beijing	Heavy-polluted	urban	PM2.5	0.2	0.5	1.2		12.5	3.4	14.9	18.6	0.8	(Tao <i>et al.</i> , 2015)
	Clean	urban	PM2.5	0.9	0.7	1.1		8.4	4.8	10.7	12.3	0.9	
Beijing	haze	urban	PM2.5					9.5		16.2	19.5	0.8	(K. F. Ho <i>et al.</i> , 2016)
Xi'an	haze	urban	PM2.5					11.0		22.4	20.6	1.1	
Xiamen	haze	urban	PM2.5					11.6		22.7	31.6	0.7	
Hong Kong	haze	urban	PM2.5					0.6		4.6	11.0	0.4	
Beijing	annual	urban	PM2.5										(Ji <i>et al.</i> , 2016)
Beijing	haze	urban	PM2.5	0.3	0.6	2.2	0.4	8.8	5.0	18.2	14.6	1.2	(Zhang <i>et al.</i> , 2018)
	non-haze	urban	PM2.5	1.2	0.9	0.3	0.6	0.1	6.2	26.9	14.0	1.9	
Beijing	haze	urban	PM2.5	2.1	1.5	1.9	0.3	8.1	4.9	4.3	9.5	0.4	(X. Xu <i>et al.</i> , 2018)
Beijing	haze	urban	PM2.5	0.3	0.3	0.7		8.3	2.2	15.7	11.3	1.4	(Shao <i>et al.</i> , 2018)
	non-haze	urban	PM2.5	0.6	0.3	0.5		4.6	1.3	8.0	5.8	1.4	
	avg	urban	PM2.5	0.3	0.3	0.7		7.4	2.0	14.3	10.0	1.4	
Nanjing	annual	industrial	PM2.5	1.2	0.9	0.5		3.8	2.6	9.7	20.9	0.5	(Li <i>et al.</i> , 2016)
	annual	urban	PM2.5	1.1	0.9	0.6		4.9	2.1	12.4	18.3	0.7	
Taiyuan	annual	urban	PM2.5	1.3	2.4	0.8	0.4	4.9	3.1	5.6	14.3	0.4	(Q. He <i>et al.</i> , 2017)
Shenzhen	winter	suburban	PM2.5	3.4	4.8			9.7	2.5	8.1	20.0	0.4	(Dai <i>et al.</i> , 2013)
	summer	suburban	PM2.5	4.0	9.3			4.2	3.4	6.1	11.4	0.5	
Handan 2013	winter	urban	PM2.5	0.3		1.0		9.0	4.6	10.0	15.0	0.7	(Zhao <i>et al.</i> , 2019)
	Summer	urban	PM2.5	0.4		0.7		7.5	1.1	9.3	22.4	0.4	
	annual	urban	PM2.5	0.6		0.9		8.4	3.7	7.4	16.5	0.5	
Handan 2017	winter	urban	PM2.5	0.5		1.5		10.4	6.6	16.3	16.0	1.0	
	summer	urban	PM2.5	0.5		0.5		9.6	1.1	7.7	20.7	0.4	
	annual	urban	PM2.5	0.6		0.9		9.9	0.9	15.3	15.1	1.0	
Lanzhou	winter	residential	PM2.5	1.8	0.7	1.0		5.6	4.3	6.0	9.8	0.6	(J. Tan <i>et al.</i> , 2017)
	summer	residential	PM2.5	3.8	0.8	1.1		5.7	2.0	5.5	12.6	0.4	
	annual	residential	PM2.5	2.3	0.8	1.0		5.6	3.8	5.9	10.4	0.6	
Ningdong (Yinglite)	winter	industrial	PM2.5	4.1	0.6	0.4		2.9	1.1	4.7	5.9	0.8	(X. Liang <i>et al.</i> , 2019)
	summer	industrial	PM2.5	6.5	0.4	0.2		1.0	0.3	1.6	5.7	0.3	

Table SA-4 (continue)

Country	Season	Site	PM size	Ca ²⁺	Na ⁺	K ⁺	Mg ²⁺	NH ₄ ⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	NO ₃ ⁻ /SO ₄ ²⁻	Ref
Zhengzhou	haze	urban	PM2.5					11.4		32.6	10.3	3.2	(Dong <i>et al.</i> , 2020)
	sandy haze	urban	PM2.5					10.3		29.9	13.7	2.2	
	non-haze	urban	PM2.5					13.0		26.9	12.7	2.1	
Cixi	haze	mixed	PM2.5					0.8	0.3	1.1	0.8	1.3	(Lu <i>et al.</i> , 2020)
	non-haze	mixed	PM2.5					0.3	0.4	0.2	0.2	0.8	
Xi'an	spring	urban	PM2.5	2.2	1.1	0.6	0.2	4.9	2.0	10.6	12.1	0.9	(Niu <i>et al.</i> , 2016)
	summer	urban	PM2.5	1.9	0.9	0.6	0.1	6.7	0.6	9.1	21.6	0.4	
	autumn	urban	PM2.5	1.1	0.6	0.8	0.1	4.9	2.1	9.5	10.8	0.9	
	winter	urban	PM2.5	1.0	1.1	1.1	0.2	6.5	3.2	11.1	12.0	0.9	
	annual	urban	PM2.5	1.5	1.0	0.8	0.2	5.7	2.2	10.1	13.1	0.8	
Zhengzhou	heavy polluted	urban	PM2.5					8.3		11.5	15.1	0.8	(Jiang <i>et al.</i> , 2018)
	other days	urban	PM2.5					8.9		12.1	16.6	0.7	
Chengdu	non-haze	urban	PM2.5	1.6	0.4	2.3		4.2	1.9	8.6	13.3	0.6	(Tao <i>et al.</i> , 2013)
	dust	urban	PM2.5	3.0	0.7	0.8		0.3	1.2	2.4	5.2	0.5	
	haze	urban	PM2.5	1.3	0.2	4.1		4.7	4.2	7.9	11.8	0.7	
	annual	urban	PM2.5	1.8	0.4	2.3	0.0	3.5	2.0	7.3	11.6	0.6	
Avg China				1.6	1.1	1.1	0.2	7.2	2.5	12.0	14.9	0.9	
India													
Delhi	summer	traffic	PM2.5		0.7	1.5		8.5	3.7	7.5	17.1	0.4	(Pant <i>et al.</i> , 2015)
	winter	traffic	PM2.5		0.2	1.4		12.4	10.0	11.9	9.4	1.3	
Delhi	annual	urban	PM2.5		3.6	3.9		8.7	5.8	8.6	10.4	0.8	(Sharma & Mandal, 2017)
	winter	urban	PM2.5		2.5	2.9		8.1	5.3	9.0	8.2	1.1	
	summer	urban	PM2.5		4.8	5.3		10.1	6.6	7.1	12.3	0.6	
Delhi	summer	urban	PM2.5	3.9	4.1	3.9	0.5	8.5	8.3	4.5	10.0	0.4	(Saxena <i>et al.</i> , 2017)
	monsoon	urban	PM2.5	5.1	17.5	5.9	2.5	6.9	15.6	8.6	23.8	0.4	
	winter	urban	PM2.5	1.6	3.2	2.8	0.5	6.8	6.6	8.7	8.5	1.0	
	spring	urban	PM2.5	2.2	3.6	3.0	0.3	8.9	6.8	7.4	10.2	0.7	
	annual	urban	PM2.5	2.5	5.1	3.4	0.8	8.2	8.8	8.8	11.7	0.8	
Delhi	winter	urban	PM2.5	1.6	3.1	2.8	0.2	9.1	6.0	11.0	10.2	1.1	(Jain <i>et al.</i> , 2020)
	summer	urban	PM2.5	3.1	3.7	3.6	0.4	6.5	6.9	6.3	10.1	0.6	
	monsoon	urban	PM2.5	4.2	5.9	3.9	0.9	4.4	6.0	5.1	14.2	0.4	
	annual	urban	PM2.5	2.3	3.7	3.3	0.4	7.5	6.3	8.9	10.6	0.8	
Ahmedabad	winter	urban	PM2.5	0.9	0.8	1.6	0.1	5.8	0.0	2.2	17.4	0.1	(Rengarajan <i>et al.</i> , 2011)
Van Vihar National Park	annual		PM2.5	1.8	3.6	2.2	0.4	4.7	3.3	6.9	7.6	0.9	(Kumar and Sunder Raman, 2020)
Avg India				2.6	4.1	3.2	0.6	7.8	6.6	7.6	12.0	0.7	

Table SA-4 (continue)

Country	Season	Site	PM size	Ca ²⁺	Na ⁺	K ⁺	Mg ²⁺	NH ₄ ⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	NO ₃ ⁻ /SO ₄ ²⁻	Ref
UK													
Birmingham	summer 2-18 June 2011	urban background	PM2.5	0.4	4.4			11.8	4.2	10.9	24.5	0.4	(Taiwo, 2016)
			PM2.5-10	1.9	5.7			17.0	15.0	26.8	3.9	6.9	
London	Winter	urban	PM2.5			0.4							(Crilley <i>et al.</i> , 2015)
Detling, Kent	winter	rural	PM2.5										
Harwell, Oxfordshire	winter	rural	PM2.5			0.5							
London	winter	urban	PM2.5						4.3	22.3	11.5	1.9	(J. Yin <i>et al.</i> , 2015)
Harwell, Oxfordshire	winter	rural	PM2.5						4.6	29.1	15.5	1.9	
Europe													
Barcelona	annual	urban back	PM2.5		1.6			5.4		5.4	15.1	0.4	(Salameh <i>et al.</i> , 2015)
Marseille	annual	urban back	PM2.5		0.5	0.5		8.7		8.7	11.2	0.8	
Genoa	annual	urban back	PM2.5		0.7	0.4		3.6		3.6	25.7	0.1	
Thessaloniki	annual	urban back	PM2.5		0.5	0.3		5.6		6.5	10.5	0.6	
Paris, France	annual	suburban	PM2.5	0.5	1.1	0.9	0.2	9.9	1.2	19.1	13.8	1.4	(Bressi <i>et al.</i> , 2013)
	annual	urban	PM2.5	0.8	1.2	0.8	0.2	9.5	1.3	19.6	13.5	1.5	
	annual	rural	PM2.5	0.4	1.1	1.0	0.2	9.5	1.3	17.5	15.1	1.2	
America													
Toronto, Canada	Annual	urban	PM2.5		0.4		0.1	9.0	0.3	16.8	18.6	0.9	(Jeong <i>et al.</i> , 2020)
Alberta, Canada	annual	urban	PM2.5	0.1	0.4	0.4	0.6	4.4	0.3	4.4	8.4	0.5	(Bari & Kindzierski, 2016)
Los Angeles, US	annual	urban	PM2.5-10					0.4	4.8	16.2	5.4	3.0	(Cheung <i>et al.</i> , 2011)
	annual	rural	PM2.5-10					0.2	2.1	4.8	2.2	2.2	

Table SA-5 Contribution of crustal and trace elements (%) to PM in SEA countries

Country	Season	Site	PM size	Na	K	Mg	Al	Si	Ca	Ti	V	Mn	Fe	Ni	Cu	Zn	Pb	As	Ref
Thailand																			
Bangkok	dry	traffic	PM2.5	2.116	1.420	0.681	2.768		4.319		1.609	0.072	2.072	0.377	0.116	1.130	0.261	0.449	(Chuersuwan <i>et al.</i> , 2008)
	dry	traffic	PM10	1.739	1.119	0.768	2.794		4.431		1.045	0.083	1.970	0.028	0.083	0.777	0.167	0.296	
Chiang Mai	LBB	urban	PM2.5		3.929	1.071	2.679									0.893			(Pani <i>et al.</i> , 2018)
	MBB	urban	PM2.5		3.548	1.075	1.828									0.430			
	HBB	urban	PM2.5		3.269	1.538	1.218									0.064			
	EBB	urban	PM2.5		3.139	1.211	1.704					0.045		0.045		0.448			
Chiang Mai	haze	urban	PM2.5		3.545	1.128	1.869					0.021		0.032	0.021	0.494			(Khamkaew, Chantara, Janta, <i>et al.</i> , 2016b)
	haze	rural	PM2.5		3.217	1.020	2.005					0.024		0.036	0.012	0.240			
Vietnam																			
Hanoi	annual	suburban	PM2.5	0.228	1.804		0.415	2.000	0.850	0.057	0.006	0.113	0.730	0.007	0.019	0.902	0.437		(Cohen <i>et al.</i> , 2010a)
Mong Duong	wet	indust	PM2.5			0.171	0.143	0.571	1.143	0.029	0.011	0.086	0.857	0.029	0.086	0.286	0.200		(Hang & Kim Oanh, 2014)
	dry		PM2.5			0.167	0.333	1.333	0.833	0.033	0.013	0.050	0.667	0.050	0.033	0.333	0.167		
Cam Hai	wet	Rural	PM2.5			0.011	0.148	0.741	0.370	0.074	0.026	0.074	0.741	0.074	0.011	0.370	0.370		
	dry		PM2.5			0.002	0.189	0.566	0.755	0.038	0.019	0.094	0.377	0.038	0.038	0.377	0.377		
Tamdao, Veit	wet	Rural	PM2.5				1.600	4.800	1.200	0.400		0.160	1.200		0.008	0.400	0.400		(Co <i>et al.</i> , 2014)
	dry	Rural	PM2.5			0.196	1.961	4.902	1.176	0.008	0.196	0.020	0.392	0.004	0.020	0.098	0.039		
Malaysia																			
Kuala Lumpur	haze	suburban	PM2.5		0.656	0.046	0.065				0.007	0.007	0.031	0.004	0.013	0.086	0.025	0.018	(Khan, Sulong, <i>et al.</i> , 2016)
Kuala	wet	urban	PM2.5	1.435	0.435	0.391	0.609		0.348		0.024	0.022		0.007	0.038	0.046	0.043	0.005	(Khan <i>et al.</i> , 2017)
Kuala lumpur	annual	urban	PM2.5	1.043	1.453	0.447	0.559	1.192	0.484	0.030	0.015	0.022	0.410	0.011	0.063	0.145	0.119	0.034	(Rahman <i>et al.</i> , 2011)
	annual	urban	PM10	1.602	1.825	1.024	3.649	7.566	4.406	0.196	0.022	0.040	1.958	0.018	0.049	0.134	0.098	0.036	
Selangor	haze	urban	PM2.5		1.971		0.400	0.953	0.252	0.029			0.229			0.132	0.084		(Fujii <i>et al.</i> , 2016)
Singapore																			
Singapore	haze	urban	PM2.5			0.236	2.683	0.301	1.534	0.034	0.071	0.034	1.242	0.282	0.902	0.753	0.278		(See <i>et al.</i> , 2006)
	non	urban	PM2.5			0.439	2.076	0.443	1.031	0.064	0.124	0.059	1.465	0.544	1.457	1.191	0.300		
Avg SEA				1.360	2.238	0.612	1.441	2.114	1.542	0.083	0.228	0.057	0.956	0.093	0.175	0.442	0.210	0.140	

Table SA-6 Contribution of crustal and trace elements (%) to PM in other countries

Country	Season	Site	PM size	Na	K	Mg	Al	Si	Ca	Ti	V	Mn	Fe	Ni	Cu	Zn	Pb	As	Ref
China																			
Beijing	Haze	urban	PM2.5	0.549	1.037	0.305	0.579		1.037		0.001	0.049	0.671	0.001	0.030	0.395	0.258	0.055	(R. Shen et al., 2017)
	Haze	urban	PM2.5	0.976	1.037	0.451	1.116		1.280		0.004	0.051	0.854	0.005	0.024	0.237	0.182	0.020	
	Haze	urban	PM2.5	0.560	1.120	0.184	0.464		1.360		0.007	0.042	0.800	0.006	0.021	0.332	0.236	0.014	
	Non-haze	urban	PM2.5	0.980	1.569	1.333	2.941		3.922		0.006	0.100	2.549	0.005	0.041	0.229	0.176	0.020	
Nanjing	annual	industrial	PM2.5				0.600			0.050		0.051	0.650	0.011	0.048	0.341	0.159	0.009	(Li et al., 2016)
	annual	urban	PM2.5				0.547			0.052		0.052	0.619	0.012	0.053	0.360	0.171	0.009	
Ji'nan, Shangdong		urban	PM2.5				0.644			0.040		0.059	1.030	0.010	0.030	0.347	0.158	0.040	(S. Zhou et al., 2014)
		industrial	PM2.5				0.515			0.038		0.085	1.854	0.008	0.031	0.338	0.154	0.046	
Lanzhou	Winter	residential	PM2.5							0.119	0.010	0.178		0.012	0.057	0.412	0.876	0.028	(J. Tan et al., 2017)
	summer	residential	PM2.5							0.269	0.011	0.182		0.013	0.069	0.393	0.416	0.029	
	annual	residential	PM2.5							0.153	0.010	0.182		0.012	0.061	0.417	0.796	0.029	
Ningdong (Yinglite)	winter	industrial	PM2.5	0.314	0.333	0.574	2.065		4.832	0.061		0.051	0.988	0.003	0.003	0.051	0.035	0.013	(X. Liang et al., 2019)
	summer	industrial	PM2.5	0.337	0.516	1.274	4.611		12.434	0.121		0.058	2.027	0.005	0.005	0.058	0.016	0.005	
Zhengzhou	haze	urban	PM2.5		1.094		1.356	1.481	1.663			0.090	1.179	0.002	0.034	0.224	0.078	0.026	(Dong et al., 2020)
	Sandy haze	urban	PM2.5		1.680		3.569	8.227	2.541			0.083	2.522	0.003	0.014	0.137	0.040	0.014	
	Non-haze	urban	PM2.5		1.700		3.277	5.356	6.617			0.182	3.119	0.004	0.028	0.320	0.060	0.053	
Cixi	Haze	mixed	PM2.5	0.485	0.351	0.152	0.285	0.971	1.036	0.009	0.006	0.025	0.313	0.004	0.021	0.315	0.054		(Lu et al., 2020)
	Non-haze	mixed	PM2.5	0.674	0.399	0.155	0.517	1.118	0.685	0.013	0.009	0.031	0.541	0.006	0.039	0.373	0.062		
Zhengzhou	Heavy polluted	urban	PM2.5			0.138	0.416					0.033	0.361	0.002	0.014	0.293	0.080	0.010	(Jiang et al., 2018)
	Other days	urban	PM2.5			0.364	0.735					0.053	0.730	0.003	0.023	0.415	0.172	0.013	
Chengdu	Non-haze	urban	PM2.5				1.097	0.907	0.898	0.104			1.287						(Tao et al., 2013)
	Dust	urban	PM2.5				5.361	5.567	3.353	0.363			4.412						
	haze	urban	PM2.5				0.351	0.339	0.300	0.045			0.581						
	annual	urban	PM2.5				1.809	1.712	1.291	0.143			1.794						
Avg China				0.609	0.985	0.493	1.565	2.853	2.883	0.105	0.007	0.082	1.375	0.006	0.032	0.299	0.209	0.024	

Table SA-6 (Continue)

Country	Season	Site	PM size	Na	K	Mg	Al	Si	Ca	Ti	V	Mn	Fe	Ni	Cu	Zn	Pb	As	Ref
India																			
Delhi	annual	urban	SPM	0.698	0.867	1.162	2.444		3.356		0.097	0.136	3.010	0.027	0.676	0.856	0.081		(Shridhar <i>et al.</i> , 2010)
	annual	rural	SPM	0.670	0.925	1.027	2.678		3.077		0.065	0.075	2.190	0.015	0.211	0.386	0.052		
Delhi	summer	traffic	PM2.5				1.392	3.230	1.340	0.120	0.017	0.052	1.220	0.007	0.034	0.344	0.137		(Pant <i>et al.</i> , 2015)
	winter	traffic	PM2.5				0.625	0.646	0.401	0.025	0.004	0.029	0.415	0.004	0.025	0.231	0.217		
Delhi	annual	urban	PM2.5			0.773	2.351	1.371	2.932	0.088		0.016	0.207		0.032	0.104	0.016	0.056	(Sharma & Mandal, 2017)
	winter	urban	PM2.5			0.903	1.704	1.036	2.179	0.112		0.015	0.026		0.046	0.133	0.026	0.077	
	summer	urban	PM2.5			0.554	3.036	1.855	3.831	0.060		0.012	0.145		0.012	0.036	0.012	0.012	
Delhi	Winter	urban	PM2.5				1.164			0.049		0.011	0.464		0.066	0.295	0.224	0.093	(Jain <i>et al.</i> , 2020)
	summer	urban	PM2.5				1.573			0.087		0.039	0.767		0.107	0.311	0.223	0.049	
	Monsoon	urban	PM2.5				1.928			0.101		0.029	1.000		0.116	0.319	0.290	0.029	
	annual	urban	PM2.5				1.427			0.069		0.008	0.595		0.076	0.290	0.229	0.069	
Trombay	2010	industrial	PM2.5	1.261	0.830	0.704	2.355	2.767	1.154	0.123	0.063	0.038	1.164	0.044	0.031	0.270	0.459	0.006	(Police, Kumar, et al., 2018)
	2011	industrial	PM2.5	1.917	1.141	1.202	2.404	3.646	1.531	0.130	0.112	0.065	1.477	0.087	0.047	0.191	0.365	0.007	
Ahmedabad	winter	urban	PM2.5				1.257												(Rengarajan <i>et al.</i> , 2011)
Van Vihar National Park	annual		PM2.5		2.307		2.573	3.050	0.900	0.098	0.011	0.041	1.100	0.007	0.041	0.559	0.150	0.005	(Kumar & Sunder Raman, 2020)
Avg India				1.137	1.214	0.904	1.927	2.200	2.070	0.089	0.053	0.040	0.984	0.027	0.109	0.309	0.177	0.040	
UK																			
Birmingham	11-21 Sep 2012	Traffic (Tunnel)	PM2.5						0.003	0.030	0.002	0.050			0.170	0.166			(Pant <i>et al.</i> , 2017)
Birmingham	summer 2-18 June 2011	Urban background	PM2.5									0.738	0.148	0.148	0.148	0.240			(Taiwo, 2016)
			PM2.5-10									0.739	0.092	0.370	0.333	0.037			
London	winter	urban	PM2.5				0.280	0.892											(J. Yin et al., 2015)
Harwell, Oxfordshire	winter	rural	PM2.5				0.245	0.700											

Table SA-6 (Continue)

Country	Season	Site	PM size	Na	K	Mg	Al	Si	Ca	Ti	V	Mn	Fe	Ni	Cu	Zn	Pb	As	Ref
Europe																			
Barcelona	annual	urban back	PM2.5		0.688	0.253			0.780		0.032	0.027	0.833	0.016	0.038	0.226	0.032		(Salameh <i>et al.</i> , 2015)
Marseille	annual	urban back	PM2.5		1.148	0.735			5.571		0.031	0.056	1.352	0.020	0.087	0.122	0.041		
Genoa	annual	urban back	PM2.5		0.714	0.257			0.786		0.100	0.029	1.014	0.050	0.043	0.136	0.043		
Thessaloniki	annual	urban back	PM2.5		1.164	1.186			5.495		0.030	0.245	2.441	0.070	0.226	0.207	0.110		
America																			
Toronto, Canada	Annual	urban	PM2.5		0.418		0.201	0.490	0.552	0.025	0.002	0.017	0.543	0.002	0.025	0.090	0.021	0.005	(Jeong <i>et al.</i> , 2020)
Alberta, Canada	annual	urban	PM2.5				0.045		0.000	0.004	0.001	0.034	0.176	0.005	0.021	0.080	0.005	0.002	(Bari & Kindzierski, 2016)
Los Angeles, US	annual	urban	PM2.5-10			1.332	2.550		2.855	0.250			4.239		0.199	0.078			(Cheung <i>et al.</i> , 2011)
	annual	rural	PM2.5-10			0.910	3.574		2.273	0.321			3.275		0.229	0.055			

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Supplement B

Table SB-1 Correlation coefficients (R^2) of average 24hr PM_{2.5} concentration between Mae Hia site and other neighboring provinces in Northern Thailand

	Mae Hia	35t	68t	58t	37t	70t	67t	69t
Mae Hia	-	-	-	-	-	-	-	-
35t Chiang Mai City center	0.98	-	-	-	-	-	-	-
68t Lamphun	0.94	0.95	-	-	-	-	-	-
58t Mae Hong Son	0.83	0.85	0.78	-	-	-	-	-
37t Lam Pang	0.88	0.87	0.91	0.63	-	-	-	-
70t Phra Yao	0.90	0.89	0.90	0.71	0.89	-	-	-
67t Nan	0.82	0.81	0.86	0.57	0.91	0.92	-	-
69t Phrae	0.79	0.80	0.83	0.52	0.94	0.78	0.86	-
57t Chiang Rai	0.81	0.83	0.81	0.77	0.71	0.88	0.76	0.61

Table SB-2 Correlation coefficient (R^2) between cations and anions during episodic haze, low haze and post-haze

	Episodic haze			Low haze			Post-haze		
	Cl ⁻	SO ₄ ²⁻	NO ₃ ⁻	Cl ⁻	SO ₄ ²⁻	NO ₃ ⁻	Cl ⁻	SO ₄ ²⁻	NO ₃ ⁻
NH ₄ ⁺	0.31	0.97	0.35	0.55	0.98	0.55	-0.45	0.98	-0.06
K ⁺	0.79	0.24	0.64	0.85	0.60	0.78	-0.31	0.87	0.33
Mg ²⁺	0.93	0.25	0.76	0.95	0.51	0.80	-0.28	0.71	0.58
Ca ²⁺	0.74	0.43	0.90	0.77	0.46	0.91	-0.39	0.79	0.51

Table SB-3 Correlation matrix of crustal and trace elements in haze and post-haze episodes, strong correlation coefficients ($R^2 > 0.8$) are labelled with pink color. Crustal elements: Mg, Al, Ca, Fe and Ti. Trace elements: V, Cr, Mn, Co, Ni, Cu, Zn, As, Rb, Sr, Cd, Sb, Ba, Ce, W and Pb

	OC	Levo	K ⁺	Mg	Al	Ca	Fe	Ti	V	Cr	Mn	Co	Ni	Cu	Zn	As	Rb	Sr	Cd	Sb	Ba	Ce	W	Pb
OC		0.87	0.92	0.86	0.81	0.76	0.89	0.81	0.90	0.44	0.89	0.85	0.83	0.40	-0.33	0.08	0.89	0.76	0.11	0.60	0.66	0.59	-0.42	0.84
Levo	0.91		0.80	0.80	0.78	0.68	0.80	0.71	0.80	0.34	0.77	0.75	0.88	0.34	-0.09	0.19	0.77	0.69	0.16	0.47	0.60	0.54	-0.37	0.76
K ⁺	0.95	0.84		0.83	0.70	0.77	0.88	0.75	0.89	0.50	0.89	0.83	0.72	0.39	-0.31	0.14	0.91	0.78	0.28	0.61	0.68	0.55	-0.35	0.89
Mg	0.46	0.30	0.53		0.95	0.78	0.95	0.94	0.93	0.58	0.94	0.94	0.88	0.40	-0.28	0.20	0.91	0.97	0.04	0.62	0.89	0.56	-0.25	0.80
Al	0.45	0.27	0.53	0.77		0.67	0.90	0.92	0.88	0.48	0.89	0.87	0.90	0.40	-0.25	0.22	0.84	0.85	-0.08	0.56	0.84	0.58	-0.29	0.73
Ca	0.46	0.27	0.45	0.71	0.63		0.69	0.63	0.69	0.21	0.71	0.68	0.62	0.05	-0.41	0.32	0.72	0.70	0.15	0.29	0.62	0.35	-0.32	0.54
Fe	0.50	0.29	0.57	0.83	0.84	0.68		0.96	0.98	0.62	0.99	0.97	0.87	0.49	-0.28	0.21	0.95	0.96	0.04	0.68	0.87	0.59	-0.29	0.88
Ti	0.51	0.32	0.57	0.70	0.90	0.61	0.87		0.93	0.60	0.95	0.93	0.86	0.48	-0.29	0.16	0.92	0.88	-0.11	0.64	0.88	0.55	-0.30	0.76
V	0.35	0.15	0.45	0.64	0.76	0.47	0.84	0.81		0.59	0.96	0.96	0.87	0.45	-0.30	0.21	0.94	0.92	0.00	0.60	0.82	0.60	-0.35	0.88
Cr	0.41	0.33	0.43	0.43	0.34	0.30	0.42	0.41	0.32		0.62	0.65	0.49	0.76	-0.09	0.60	0.58	0.64	0.16	0.50	0.62	0.07	0.17	0.55
Mn	0.56	0.38	0.60	0.86	0.76	0.69	0.86	0.73	0.68	0.41		0.96	0.83	0.46	-0.32	0.17	0.95	0.95	0.05	0.73	0.87	0.56	-0.30	0.87
Co	0.26	0.10	0.31	0.58	0.62	0.47	0.69	0.58	0.59	0.33	0.64		0.85	0.42	-0.28	0.31	0.90	0.98	0.04	0.61	0.84	0.52	-0.23	0.81
Ni	0.50	0.38	0.48	0.52	0.43	0.37	0.49	0.49	0.43	0.66	0.51	0.36		0.46	-0.15	0.21	0.83	0.80	0.04	0.46	0.73	0.58	-0.22	0.76
Cu	0.13	0.09	0.20	0.31	0.43	0.29	0.34	0.31	0.31	0.15	0.31	0.51	0.34		0.02	0.17	0.43	0.37	0.10	0.47	0.42	0.16	-0.12	0.53
Zn	-0.07	-0.11	0.02	0.14	0.42	0.13	0.20	0.18	0.28	0.01	0.17	0.42	0.10	0.73		0.16	-0.31	0.04	0.73	-0.09	-0.15	-0.01	0.25	-0.14
As	0.67	0.47	0.73	0.36	0.46	0.43	0.51	0.54	0.45	0.34	0.47	0.31	0.41	0.15	0.05		0.22	0.71	0.15	-0.01	0.43	-0.08	0.51	0.20
Rb	0.83	0.76	0.90	0.62	0.64	0.47	0.63	0.64	0.49	0.36	0.68	0.36	0.42	0.30	0.10	0.57		0.87	0.08	0.62	0.86	0.59	-0.26	0.84
Sr	0.06	-0.03	0.09	0.49	0.40	0.56	0.36	0.39	0.30	0.28	0.28	0.39	0.27	0.29	0.14	0.15	0.11		0.30	0.55	0.92	0.41	0.19	0.81
Cd	0.21	0.18	0.16	0.01	0.04	0.31	0.07	0.01	0.07	0.04	0.12	0.15	0.36	0.24	0.20	0.25	0.07	-0.06		0.14	0.03	0.05	0.28	0.30
Sb	0.05	0.01	0.01	-0.17	-0.04	0.05	-0.13	0.07	0.00	-0.10	-0.23	0.00	0.17	0.12	-0.02	0.17	-0.13	0.24	0.29		0.64	0.28	-0.32	0.73
Ba	0.57	0.41	0.59	0.77	0.78	0.79	0.79	0.76	0.62	0.37	0.84	0.59	0.46	0.44	0.29	0.43	0.70	0.42	0.29	-0.06		0.44	-0.10	0.74
Ce	0.39	0.27	0.37	0.41	0.51	0.37	0.38	0.42	0.40	0.05	0.33	0.22	0.22	0.08	0.16	0.34	0.33	0.19	0.04	0.18	0.38		-0.29	0.58
W	0.01	0.02	-0.01	-0.05	-0.08	-0.05	-0.06	-0.06	-0.08	0.03	0.00	0.55	-0.05	0.01	-0.05	-0.03	-0.05	0.16	-0.04	0.13	0.01	-0.05		-0.23
Pb	0.07	0.00	0.06	0.11	0.00	0.16	0.18	0.13	0.22	0.06	0.04	0.22	0.32	0.21	-0.02	0.20	-0.05	0.23	0.24	0.67	0.00	0.13	0.09	

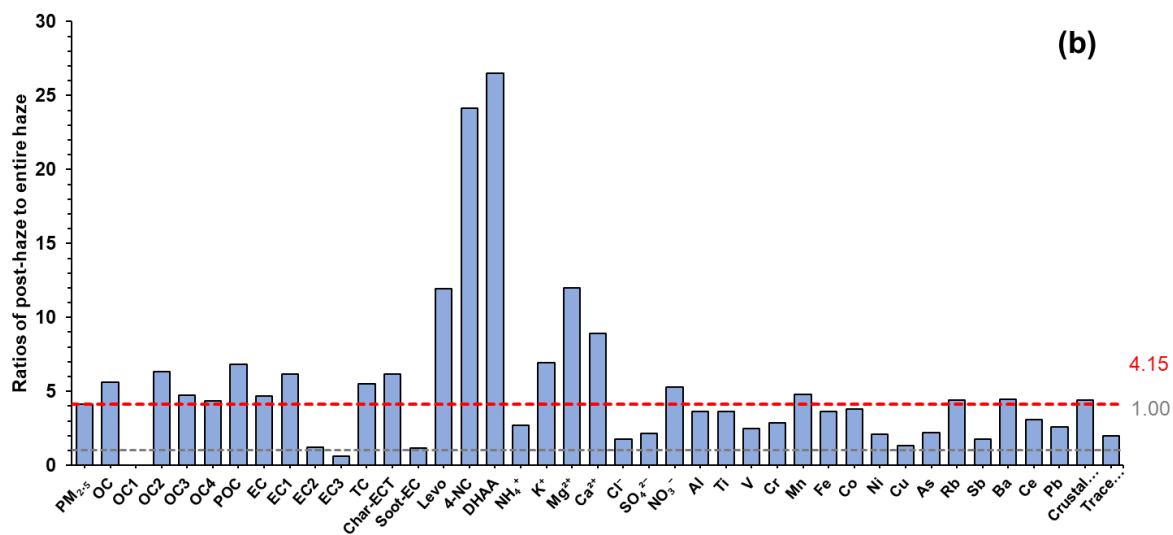
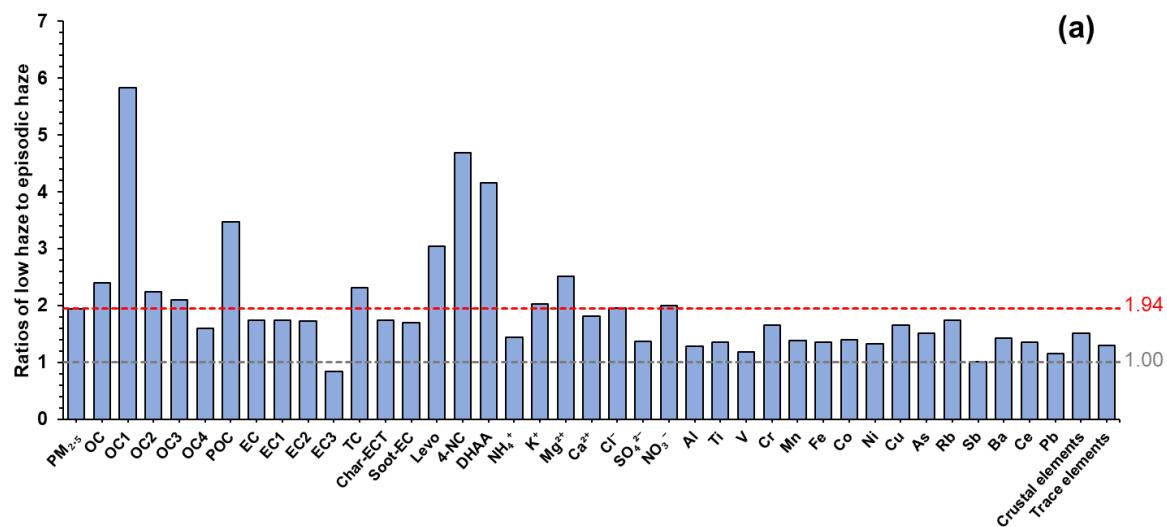


Fig. SB-1 Mass ratios of PM_{2.5} and components during low haze to episodic haze (a) and pot-haze to entire haze (b)

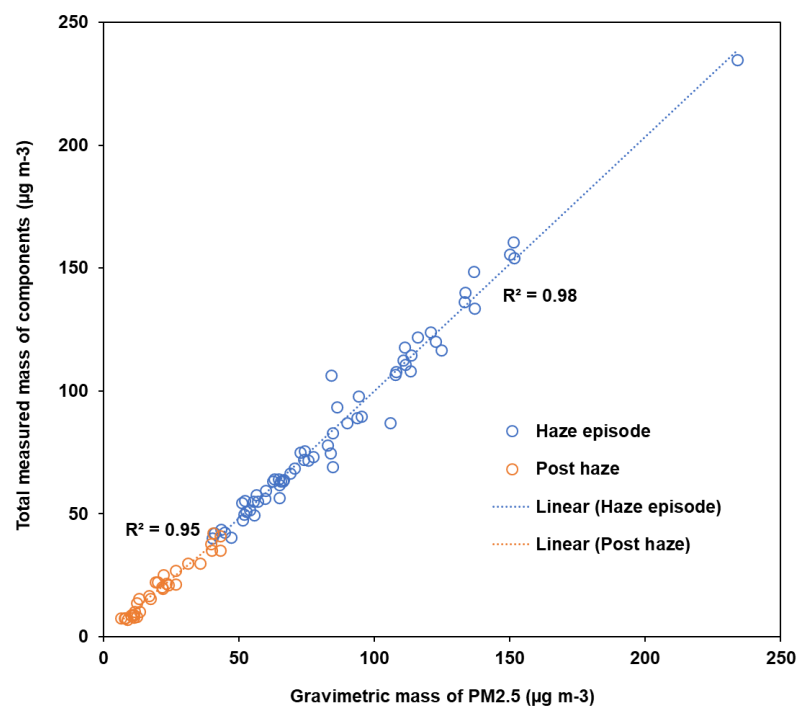
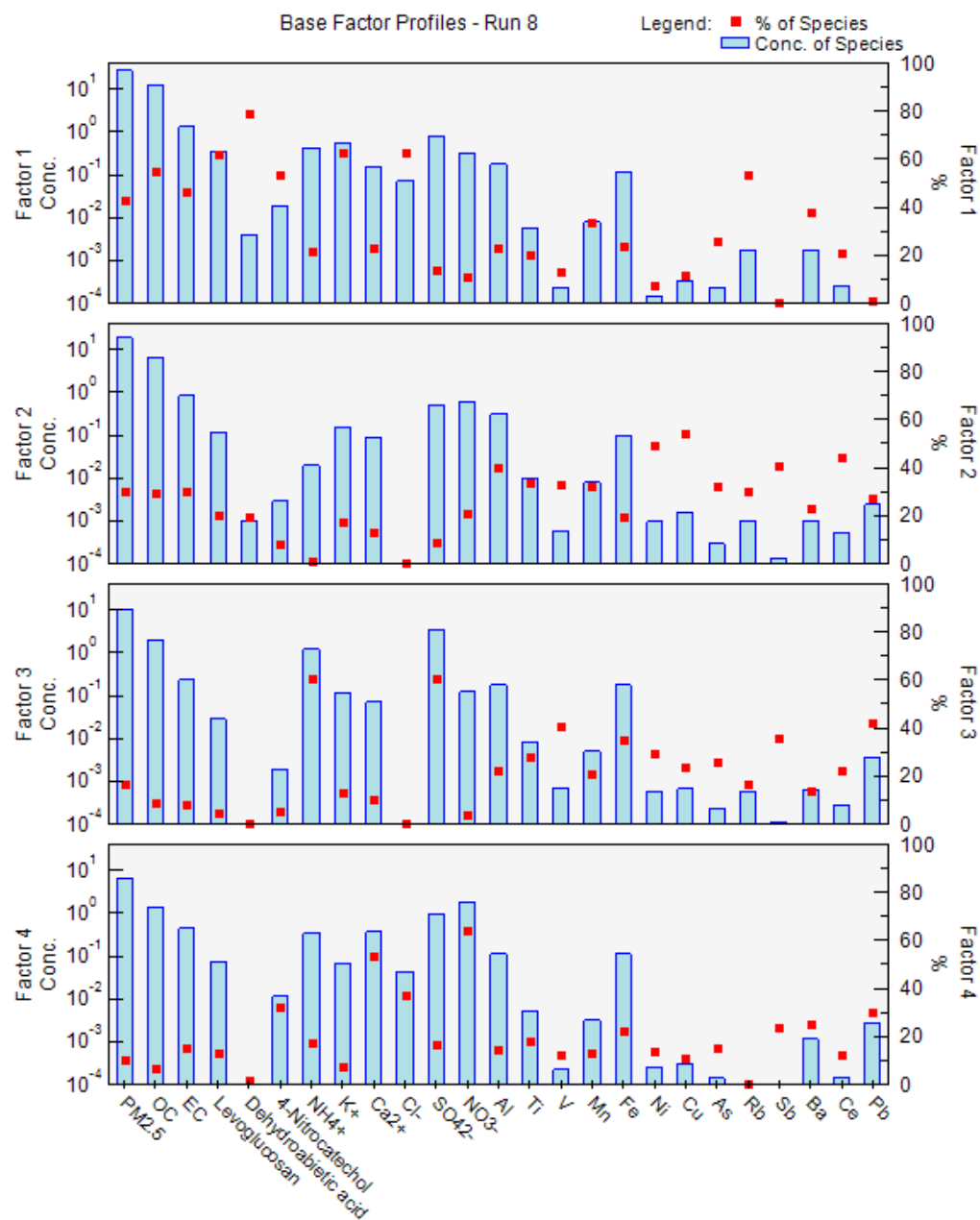


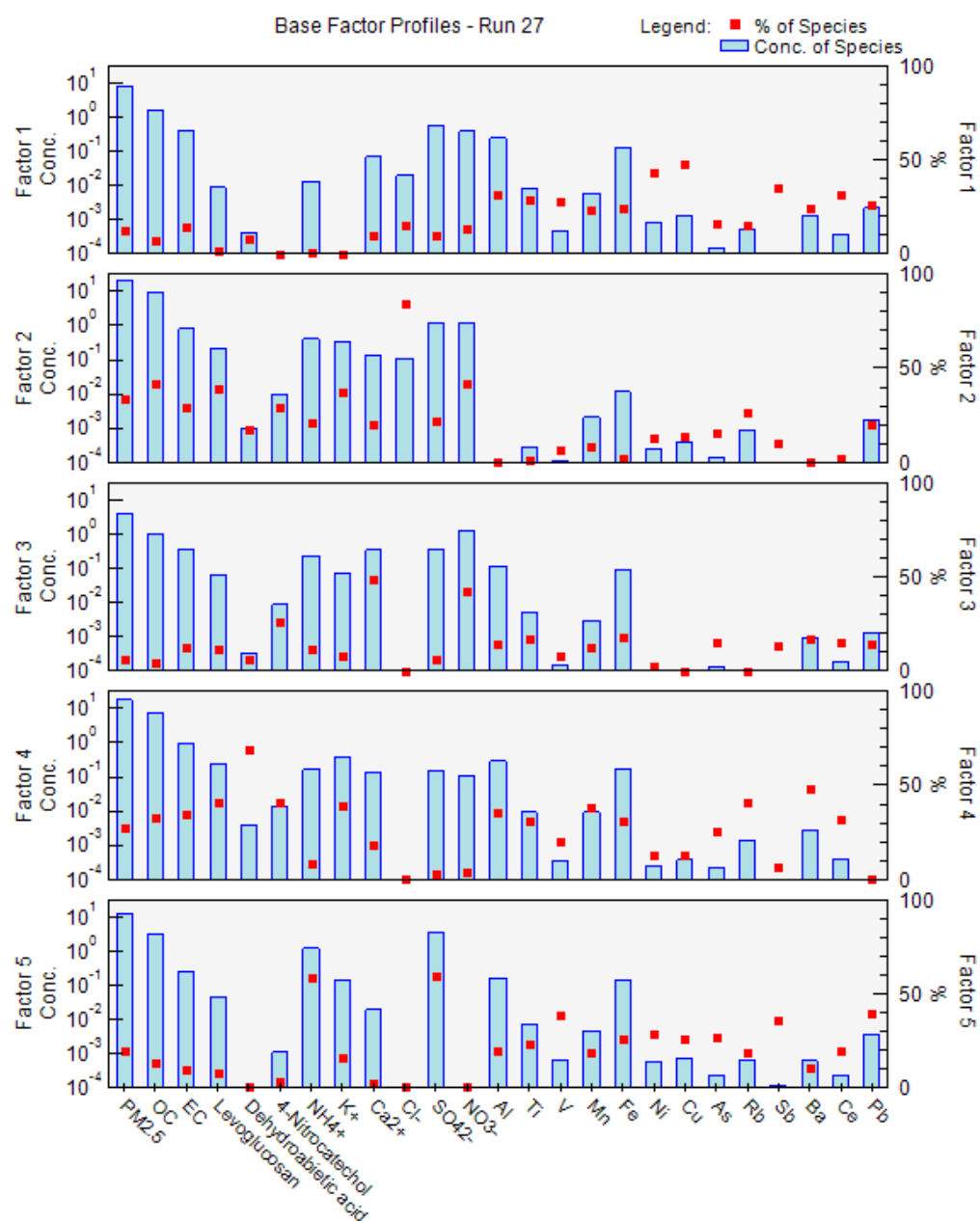
Fig. SB-2 Correlation between gravimetric mass of PM_{2.5} and total measured mass of all components in haze and post-haze episodes.



	Factor 1	Factor 2	Factor 3	Factor 4	Unmapped
Boot Factor 1	95	5	0	0	0
Boot Factor 2	14	72	1	13	0
Boot Factor 3	0	0	99	1	0
Boot Factor 4	5	4	0	91	0

Fig. SB-3 Chemical source profiles of 4 factors solution and bootstrap run

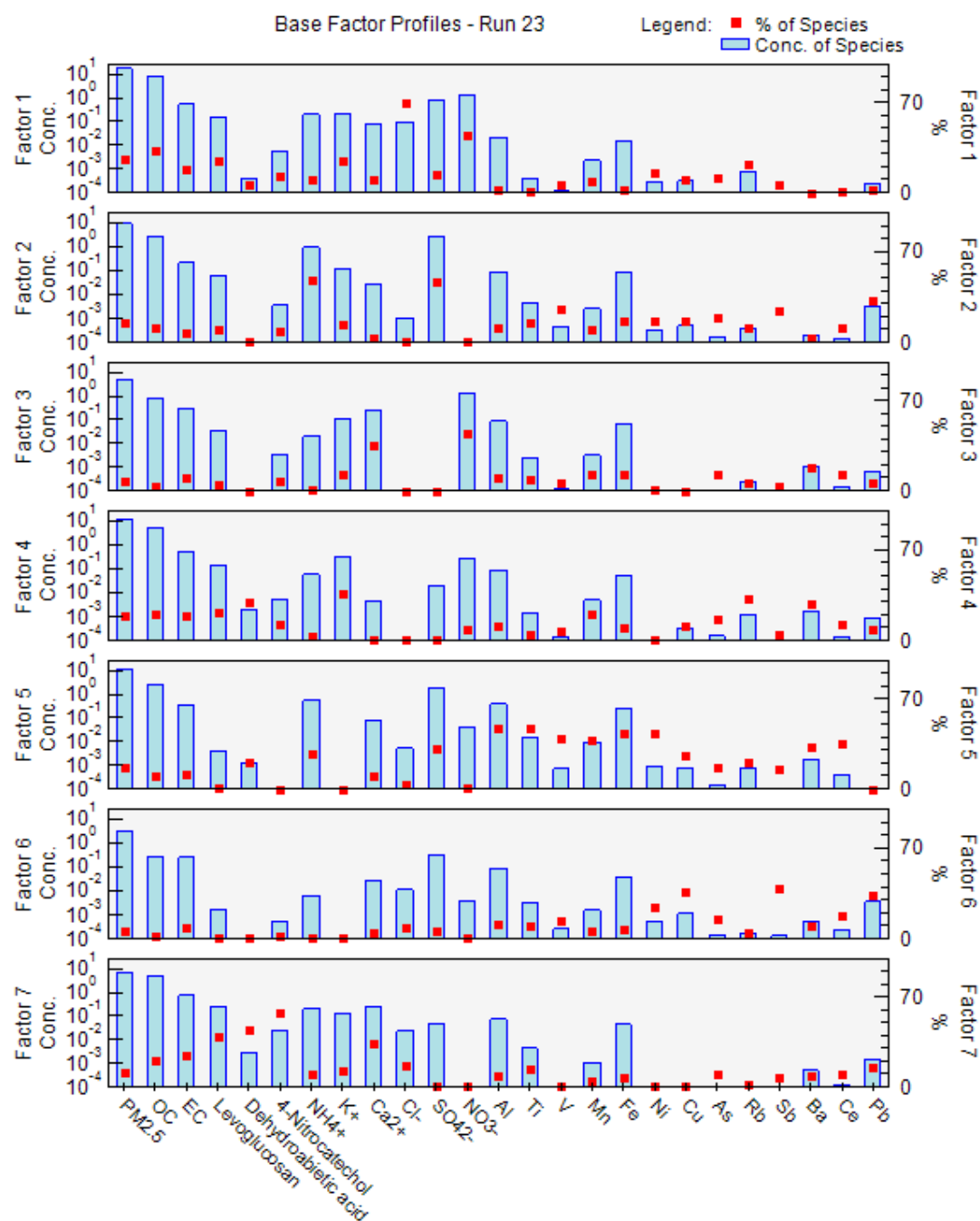
Factors were not separated well, factor 1 was a mix of types of biomass burnings. Bootstrap factor 2 was below 80.



	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Unmapped
Boot Factor 1	90	2	1	4	3	0
Boot Factor 2	2	94	0	4	0	0
Boot Factor 3	1	2	96	1	0	0
Boot Factor 4	0	3	0	97	0	0
Boot Factor 5	0	0	1	0	99	0

Fig. SB-4 Chemical source profiles of 5 factors solution and bootstrap run

Nearby biomass burning separated out from biomass burning source, but factor 4 still shows mixed types of biomass burnings.



	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6	Factor 7	Unmapped
Boot Factor 1	99	0	0	0	0	0	1	0
Boot Factor 2	0	98	0	1	1	0	0	0
Boot Factor 3	5	0	89	0	2	1	3	0
Boot Factor 4	7	2	2	82	5	0	2	0
Boot Factor 5	4	3	2	3	86	2	0	0
Boot Factor 6	3	2	1	2	14	75	3	0
Boot Factor 7	12	1	4	2	0	2	79	0

Fig. SB-5 Chemical source profiles of 7 factors solution and bootstrap run

3 different types of biomasses burning, secondary nitrate and sulfate were well separated, but factor 6 cannot be firmly explained to the original of the source.

Supplement C

Table SC-1 Reference concentration (RfC), inhalation unit risk (IUR), cancer slope factor (CSF), and toxic equivalent factor of PAHs (TEFs) used in this study.

	RfC (mg/m ³)	IUR (µg/m ³) ⁻¹	CSF (mg/kg bw/day) ⁻¹	TEFs
Cr		0.012	4.87E-05	
Co		0.009	3.65E-05	
Ni		0.00024	9.73E-07	
As		0.0043	1.74E-05	
Cd		0.0018	7.3E-06	
Benzo(a)anthracene				0.1
Chrysene				0.01
Benzo(b)fluoranthene				0.1
Benzo(k)fluoranthene				0.1
Benzo(a)pyrene	0.000002		2.43*	1
Dibenzo(a,h)anthracene				1
Indeno(1,2,3-cd)pyrene				0.1
Napthalene				0.001
Acenaphthylene				0.001
Anthracene				0.01
Phenanthrene				0.001
Fluoranthene				0.001
Pyrene				0.001
Benzo(ghi)perylene				0.01

RfC and IUR obtain from USEPA website (Basic Information about the Integrated Risk Information System (IRIS)) (<https://www.epa.gov/iris/basic-information-about-integrated-risk-information-system>)

Cancer slope factor is calculated as:

$$CDI_j \text{ (mg kg}^{-1} \text{ day}^{-1})^{-1} = \frac{IUR \times BW}{IR}$$

Where BW is body weight (average weight for adult is 65.7 kg) and IR is inhalation rate (average for adult is 16.2 m³ day⁻¹)

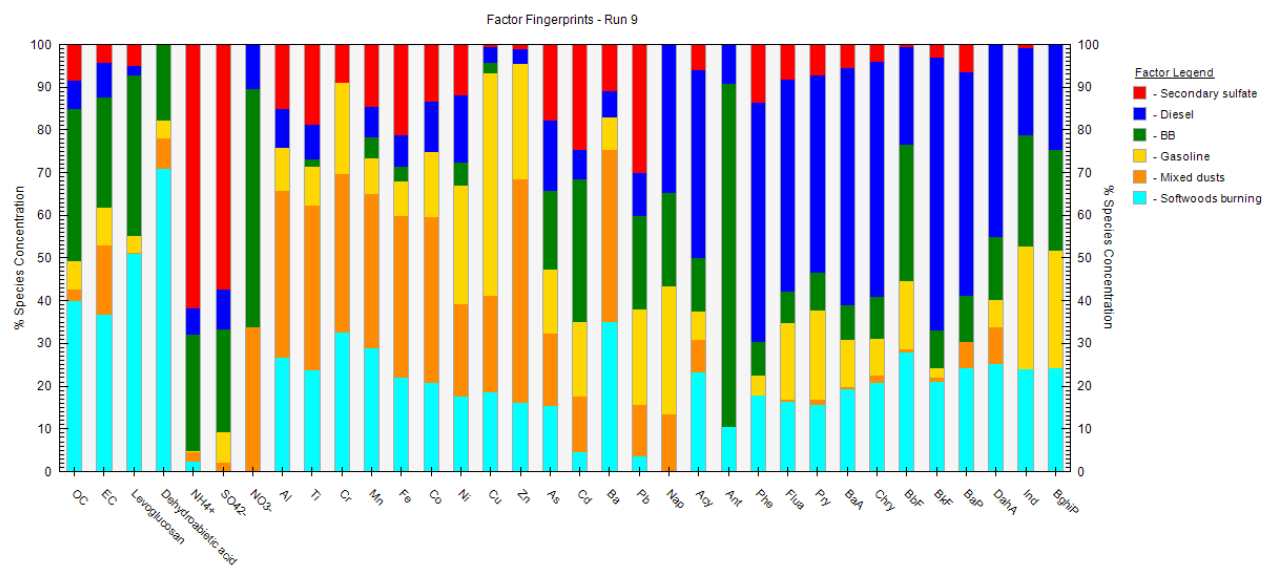


Fig. SC-1 factor fingerprint of identified sources

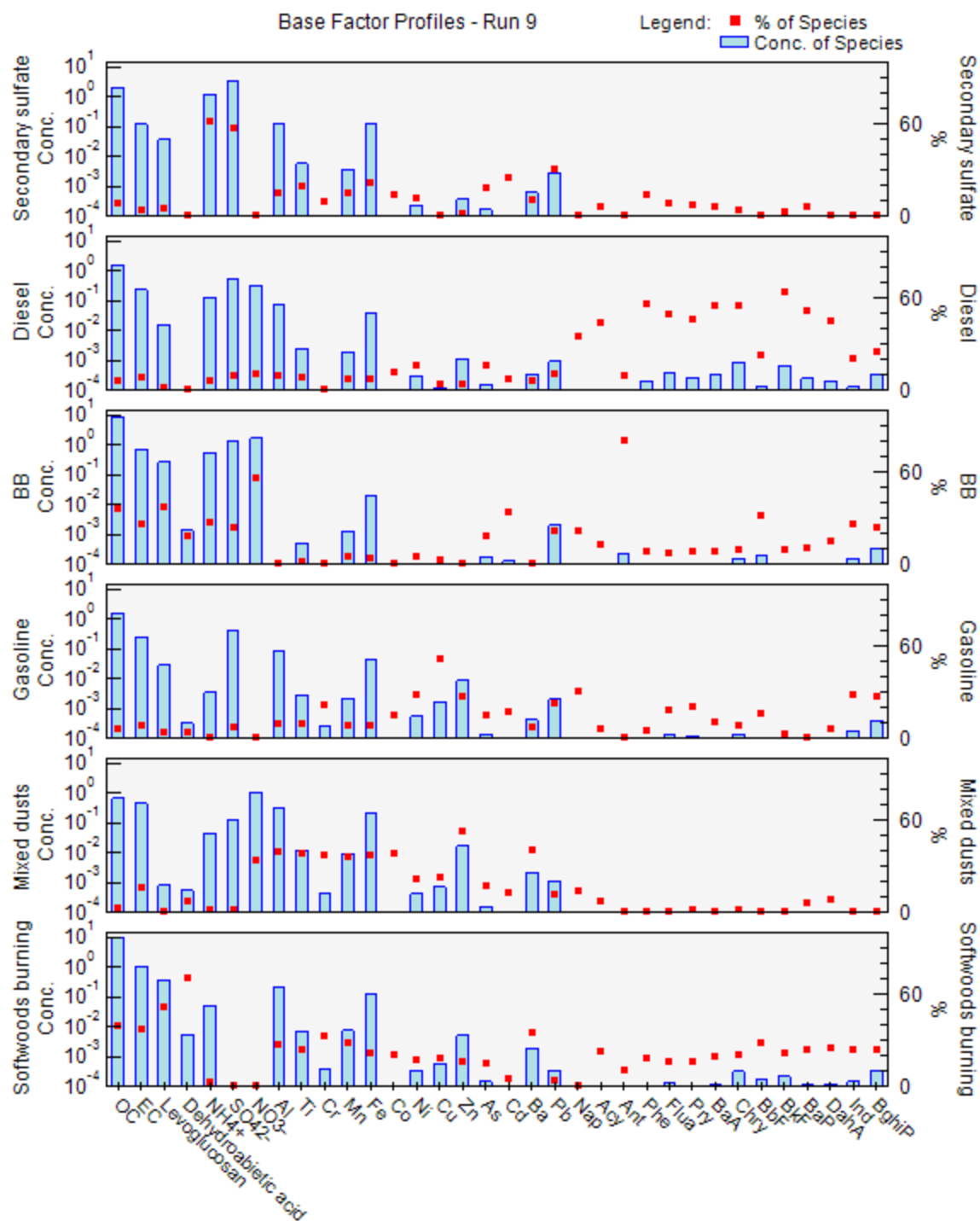


Fig. SC-2 Chemical source profiles of 6 identified sources

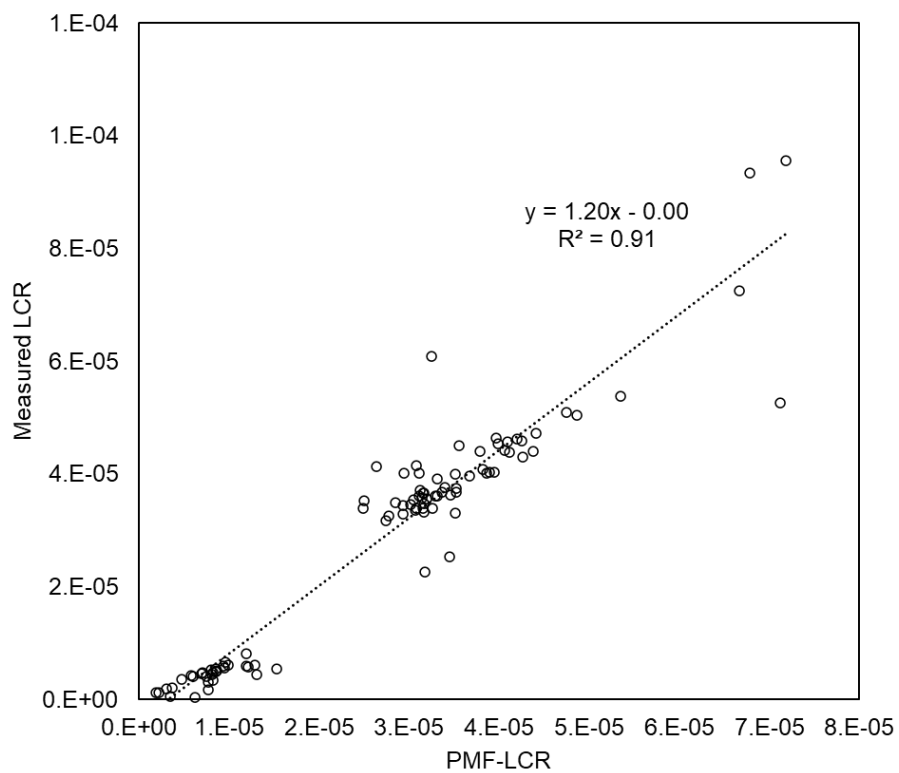


Fig. SC-3 scatter plots between measured LCR and PMF-LCR

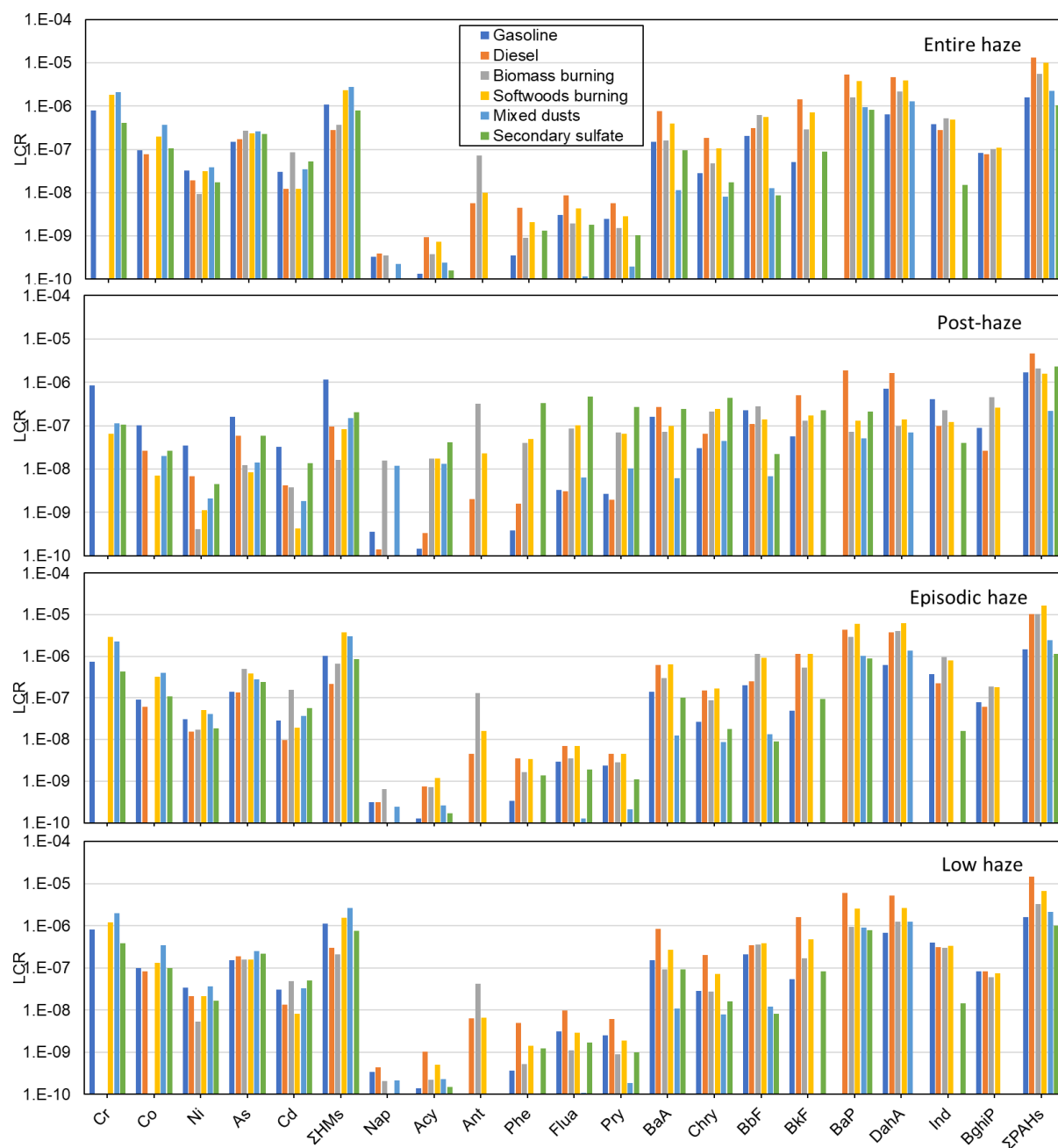


Fig. SC-4 LCR of individual heavy metals and PAHs by specific source during haze, post-haze, episodic haze and low haze

Supplement D

Table SD-1 Concentration and SD of nonpolar organic compounds (ng m⁻³) during episodic, low haze and post-haze and their incremental ratios between entire haze to post-haze.

	Episodic	SD	Low haze	SD	Post-haze	SD	Incremental ratio
Alkanes							
C16	2.3	5.2	2.3	2.3	1.0	1.7	2.4
C17	2.6	3.0	5.6	6.1	2.5	4.1	1.9
C18	0.4	0.2	0.5	0.1	0.2	0.1	1.9
C19	0.6	0.2	0.6	0.1	0.4	0.2	1.6
C20	0.4	0.1	0.3	0.0	0.2	0.1	1.7
C21	0.4	0.1	0.2	0.1	0.1	0.1	3.1
C22	0.5	0.2	0.3	0.2	0.2	0.2	2.4
C23	1.2	0.5	0.7	0.5	0.2	0.2	4.1
C24	4.5	1.5	3.3	1.3	2.2	0.3	1.7
C25	7.9	2.6	5.1	3.0	2.9	0.3	2.1
C26	9.0	3.0	5.9	2.8	3.8	0.6	1.8
C27	14.5	4.5	8.5	4.3	4.1	0.8	2.5
C28	12.3	4.1	8.7	4.0	6.2	2.4	1.6
C29	15.7	5.2	9.0	4.6	4.5	1.7	2.5
C30	7.0	2.2	4.4	1.6	3.5	1.0	1.5
C31	8.9	2.8	5.9	1.7	4.8	1.0	1.5
C32	6.1	1.9	5.0	0.5	4.8	0.3	1.1
C33	6.9	2.2	5.7	0.4	5.2	0.3	1.2
C34	7.4	1.9	6.1	0.8	5.4	0.2	1.2
ΣCodd	58.7	17.6	39.9	15.4	23.6	6.2	
ΣCeven	49.2	14.2	34.1	11.9	25.2	5.5	
ΣC≤25	20.9	9.2	18.9	9.4	9.8	5.6	
ΣC≥26	87.0	25.0	55.2	22.6	39.0	7.7	
ΣAlkanes	107.9	31.4	74.1	26.6	48.8	10.7	
CPI	1.2	0.1	1.2	0.2	0.9	0.2	
PAHs							
Napthalene (Nap)	0.1	0.0	0.1	0.1	N.D.	N.D.	-
Acenaphthylene (Ace)	0.1	0.0	0.1	0.0	0.1	0.0	1.1
Anthracene (Ant)	0.8	0.2	0.7	0.1	0.7	0.0	1.0
Phenanthrene (Phe)	0.5	0.1	0.4	0.1	0.4	0.1	1.1
Fluoranthene (Flua)	1.0	0.3	0.8	0.0	0.8	0.0	1.2
Pyrene (Pyr)	0.7	0.2	0.6	0.1	0.5	0.0	1.2
Benz(a)anthrazene (BaA)	0.8	0.2	0.7	0.0	0.6	0.0	1.2
Chrysene (Chy)	2.1	0.6	1.6	0.1	1.4	0.0	1.2
Benz(b)fluoranthene (BbF)	1.2	0.4	0.6	0.2	0.3	0.1	3.0
Benz(k)fluoranthene (BkF)	1.4	0.4	1.1	0.1	1.0	0.0	1.2
Benz(a)pyrene (BaP)	0.7	0.2	0.5	0.0	0.5	0.0	1.2
Dibenz(a,h)anthracene (DahA)	0.7	0.2	0.5	0.0	0.5	0.0	1.3
Indeno(1,2,3-cd)pyrene (Ind)	1.0	0.3	0.6	0.2	0.4	0.1	2.1
Benz(ghi)perylene (BghiP)	2.3	0.7	1.4	0.4	0.8	0.2	2.0
2 rings	0.1	0.0	0.1	0.1	N.D.	N.D.	
3 rings	1.4	0.4	1.1	0.4	0.4	0.3	
4 rings	4.5	1.3	3.7	0.2	2.5	1.3	
5 rings	3.9	1.1	2.7	0.4	1.4	0.8	
6 rings	3.3	1.0	2.0	0.5	1.1	0.3	
ΣPAHs	13.2	3.6	9.5	1.3	4.8	2.3	
Low molecular weight PAHs	1.4	0.4	1.1	0.4	0.5	0.3	
High molecular weight PAHs	11.7	3.3	8.4	1.1	4.7	2.0	
Total non - polar organic compounds	121.1	34.2	83.6	27.1	53.1	12.0	
N.D. – not detected							

Table SD-2 Concentration and SD of polar organic compounds (ng m⁻³) during episodic, low haze and post-haze and their incremental ratios between entire haze to post-haze.

	Episodic	±SD	Low haze	±SD	Post-haze	±SD	Incremental ratio
<i>Saccharides</i>							
Levoglucozan	2223.1	917.7	717.3	479.2	100.7	73.1	11.9
Mannosan	119.3	51.3	37.0	22.8	5.7	3.8	11.2
Galactosan	51.6	22.9	15.5	9.7	2.8	2.1	9.7
ΣAnhydrosugars	2398.6	991.9	772.5	511.0	111.8	79.1	
D-threitol	3.8	1.2	2.1	2.2	0.6	0.1	4.2
Meso-erythritol	163.9	42.6	101.3	29.7	37.4	16.4	3.3
ΣSugar Alcohols	187.6	47.0	111.3	31.7	39.5	16.7	
ΣSaccharides	2561.7	1026.7	873.1	533.4	147.2	86.6	
<i>Nitrate aromatic phenols</i>							
4-Nitrophenol	14.2	4.8	5.9	2.4	1.2	0.9	7.2
2-Methyl-4-nitrophenol	5.8	2.2	2.0	1.1	0.4	0.3	8.3
4-Nitrocatechol	294.9	222.3	60.8	73.8	5.6	6.5	24.1
Bisphenol A	0.4	0.4	0.4	0.4	1.2	1.6	0.4
ΣNitrate aromatic phenols	315.2	227.4	69.1	75.8	8.4	8.2	
<i>Carboxylic acids</i>							
<i>Dicarboxylic acids</i>							
Succinic acid	164.7	55.6	56.8	27.5	16.0	7.7	5.7
Glutaric acid	4.2	1.8	2.3	1.3	0.5	0.3	5.9
Adipic acid	18.8	6.2	8.0	2.8	2.6	0.9	4.4
Pimelic acid	7.9	3.0	3.0	1.2	1.1	0.3	4.4
Suberic acid	11.0	4.5	5.0	5.3	1.5	0.6	4.7
Azelaic acid	26.9	9.4	9.9	3.7	3.2	1.4	4.9
Malic acid	232.7	58.9	136.3	58.6	52.3	34.1	3.2
Phthalic acid	38.1	11.5	17.2	6.7	9.8	3.9	2.5
Isophthalic acid	9.6	2.9	4.1	1.8	0.6	0.5	9.0
ΣDicarboxylic acids	466.2	124.7	221.3	90.2	76.7	36.5	
<i>Tricarboxylic acids</i>							
Hemimellitic acid	21.0	5.8	11.7	4.5	3.9	3.0	3.8
Trimellitic acid	16.7	4.3	9.3	3.4	3.2	2.4	3.7
Trimesic acid	0.5	0.1	0.3	0.1	0.1	0.1	3.5
ΣTricarboxylic acids	85.9	22.2	42.6	15.9	17.5	8.9	
<i>Monocarboxylic acids</i>							
cis-Pinonic acid	5.4	2.2	4.9	1.3	1.6	0.5	3.2
Dehydroabietic acid	42.4	39.5	9.9	8.8	1.6	1.9	12.8
Pimaric acid	2.8	1.4	1.4	0.5	1.0	0.5	1.9
Isopimaric acid	3.0	3.8	0.6	0.7	0.1	0.0	15.6
DHOPA	19.9	6.2	9.1	3.7	2.8	1.1	4.5
ΣMonocaboxylic acids	73.5	46.3	25.9	11.9	5.8	3.1	
<i>Fatty acids</i>							
Palmitic acid	21.2	9.8	19.0	14.0	25.1	26.5	0.8
Linoleic acid	0.9	1.0	1.8	3.9	1.3	0.9	1.7
Oleic acid	1.9	0.8	3.7	11.1	3.9	3.8	0.8
Stearic acid	12.0	6.4	11.2	8.7	23.8	27.2	0.5
ΣFatty acids	36.0	16.6	35.8	32.2	53.6	57.2	
Σcarboxylic acids	661.6	163.6	325.6	116.2	153.5	63.5	
<i>Others</i>							
Syringaldehyde	19.3	17.5	7.4	13.5	0.8	0.2	14.7
Cholesterol	1.5	0.5	1.7	2.4	1.4	0.5	1.2
1-Pentadecanol	2.9	1.2	3.0	1.7	2.7	2.7	1.1
Total polar organic compounds	3563.0	1337.8	1278.6	693.5	313.3	133.1	

Table SD-3 Concentration and SD of PM_{2.5} and selected chemical components ($\mu\text{g m}^{-3}$), gaseous pollutants and meteorological data (temperature and relative humidity) during episodic, low haze and post-haze and incremental ratios of chemical constituents between entire haze to post-haze.

	Episodic	SD	Low haze	SD	Post-haze	SD	Incremental ratio
Components ($\mu\text{g m}^{-3}$)							
PM _{2.5}	129.6	28.8	66.2	15.4	20.9	11.5	4.2
OM	89.0	26.0	36.8	13.3	9.5	4.5	5.6
OC	55.6	16.2	23.0	8.3	5.9	2.8	5.6
SO ₄ ²⁻	8.7	2.2	6.4	1.9	3.1	2.6	2.3
NO ₃ ⁻	6.5	2.5	3.1	1.3	0.7	0.3	6.4
Other gaseous pollutants							
O ₃ (ppb)	49.3	7.3	43.3	6.7	34.3	10.3	1.3
NO ₂ (ppb)	18.1	3.0	13.0	3.4	6.6	1.7	2.2
Meteorological data							
Temp (°C)	29.2	1.7	28.8	2.3	30.0	1.6	
RH (%)	45.5	3.8	47.5	4.4	67.2	7.3	

Table SD-4 Pearson's correlation (R^2) matrix between polar organic compounds during entire haze and post-haze. Correlations with $R^2 \geq 0.8$ are in bold red.

Entire haze

	Galactosan	Mannosan	Levoglucosan	D-threitol	Meso-erythritol	4-Nitrophenol	2-Methyl-4-nitrophenol	4-Nitrocatechol	Bisphenol A	Succinic acid	Glutaric acid	Adipic acid	Pimelic acid	Suberic acid	Azelaic acid	Malic acid	phthalic acid	Isophthalic acid	Hemimellic acid	Trimelitic acid	Trimesic acid	cis-Pinonic acid	Dehydroabietic acid	Pimaric acid	Isopimaric acid	DHOPA	Palmitic acid	Linoleic acid	Oleic acid	Stearic acid	Syringaldehyde	Cholesterol	1-Pentadecanol	
Galactosan		.97**	.97**	.67**	0.34	.51**	.38*	.95**	0.33	0.02	.69*	.57**	.75**	.83**	.85**	.62**	0.18	.64**	.64**	.62**	0.43	.46**	.56*	.53*	-.052	.48**	-.01	0.22	-.014	-.017	0.61	-.011	-.014	
Mannosan	.99**		.98**	.67**	.36*	.51**	.38*	.91**	0.31	-0.05	.75*	.63**	.81**	.88**	.88**	.68**	0.21	.68**	.70**	.67**	0.48	.48**	.69**	.62**	-.041	.56**	-.012	0.18	-.017	-.02	0.76	-.009	-.017	
Levoglucosan	.99**	.99**		.72**	.38*	.58**	.44*	.95**	.36*	-0.05	0.64	.65**	.84**	.91**	.91**	.74**	0.23	.72**	.75**	.72**	.51*	.49**	.63*	.58*	-.055	.60**	-.016	0.29	-.02	-.023	0.62	-.016	-.021	
D-threitol	.60**	.63**	.65**		.68**	.40*	0.28	.65**	.45*	0.32	.73*	.79**	.74**	.74**	.86**	.65**	.38*	.72**	.69**	.67**	.72**	.49**	.67**	.64**	.084	.59**	-.012	0.18	-.014	-.015	0.74	-.023	-.011	
Meso-erythritol	.85**	.84**	.87**	.56**		0.13	0.06	0.3	0.16	0.18	0.22	.50**	0.34	0.35	.54**	0.18	-0.2	0.18	0.2	0.14	0.27	.73**	.59*	.49*	-.027	0.15	0.06	-.013	0.07	0.03	-.062	-.002	0.06	
4-Nitrophenol	.88**	.87**	.85**	.43**	.69**		.91**	.64**	.43*	0.06	0.27	.43*	.55**	.53**	.52**	.48**	0.2	.53**	.55**	.55**	.67**	0.24	0.24	0.06	-.073	0.33	0.02	0.33	-.012	-.005	-.025	0.05	0.01	
2-Methyl-4-nitrophenol	.88**	.88**	.88**	.76**	.68**	.94**		.53**	.41*	0.03	0.14	0.26	.37*	0.36	.37*	0.3	0.14	.37*	0.34	0.36	0.46	0.16	0.06	-.003	-.058	0.16	-.001	0.35	-.015	-.008	-.043	0.05	0	
4-Nitrocatechol	.86**	.86**	.88**	.44**	.68**	.77**	.91**		.45*	-0.07	0.47	.54**	.74**	.81**	.82**	.65**	0.19	.65**	.66**	.65**	0.4	.44*	0.41	0.45	-.071	.49**	-.017	0.33	-.02	-.024	0.34	-.016	-.021	
Bisphenol A	0.18	0.16	0.17	0.15	0.07	0.19	0.2	0.19		0.15	0.02	.41*	.44*	.40*	.47**	0.32	0.23	.52**	.36*	.38*	0.34	0.04	0.09	0.02	-.006	0.26	0.03	-.009	0.04	0	-1.00**	0.01	0.02	
Succinic acid	.90**	.91**	.89**	.51**	.82**	.90**	.83**	.71**	0.06		0.11	0.21	-0.08	-0.11	0.09	-0.18	0.21	0.06	-0.07	-0.06	.72**	0.03	0.2	-.01	0.39	-.017	0.23	-.023	0.16	0.23	-.074	0.06	0.29	
Glutaric acid	.82**	.81**	.81**	.65**	.61**	.61**	.75**	.78**	.27*	.58**		.68*	.74*	.78*	.80**	0.46	0.15	0.58	0.55	0.5	0.5	0.15	0.7	0.79	0.51	0.4	-.008	-.048	-.021	-.015	1.00**	0.17	-.013	
Adipic acid	.80**	.81**	.77**	.44**	.69**	.85**	.72**	.54**	0.03	.96**	.47**		.86**	.81**	.85**	.76**	.57**	.86**	.82**	.81**	.85**	.44*	.53*	.52*	0.24	.71**	-.023	-.001	-.025	-.028	-0.91	-.026	-.025	
Pimelic acid	.84**	.86**	.81**	.91**	.72**	.85**	.72**	.57**	0.09	.95**	.59**	.99**		.98**	.91**	.92**	.54**	.94**	.95**	.93**	.82**	.36*	.62*	.56*	0.53	.88**	-.025	0.06	-.028	-.031	.99	-.025	-.03	
Suberic acid	.68**	.71**	.69**	.95**	.55**	.57**	.73**	.47**	0.18	.64**	.67**	.63**	.99**		.94**	.91**	.47**	.90**	.92**	.90**	.77**	.39*	.62*	.58*	-.05	.83**	-.027	0.18	-.03	-.033	0.62	-.028	-.032	
Azelaic acid	.96**	.96**	.94**	.53**	.81**	.91**	.87**	.80**	0.14	.96**	.73**	.91**	.92**	.67**		.78**	.36*	.82**	.81**	.78**	.65**	.58**	.67**	.64**	-0.84	.69**	-.02	0.14	-.024	-.026	0.14	-.025	-.022	
Malic acid	.66**	.68**	.67**	.36**	.72**	.66**	.54**	.40**	-0.11	.83**	.26*	.83**	.81**	.45**	.71**		.63**	.91**	.98**	.97**	.94**	0.2	0.43	0.45	0.8	.94**	-.03	0.3	-.031	-.034	.086	-.031	-.035	
phthalic acid	.79**	.80**	.77**	.41**	.71**	.84**	.74**	.58**	-0.03	.96**	.45**	.96**	.95**	.58**	.91**	.83**		.73**	.66**	.70**	0.46	-.031	-.009	-.002	0.46	.69**	-.023	0.03	-.022	-.024	-.045	-.024	-.023	
Isophthalic acid	.77**	.78**	.74**	.43**	.67**	.83**	.71**	.54**	0.01	.93**	.47**	.96**	.94**	.60**	.88**	.83**	.97**		.95**	.97**	.88**	0.15	0.41	0.48	0.36	.86**	-.029	0.06	-.031	-.033	-0.92	-.03	-.032	
Hemimellic acid	.58**	.58**	.56**	.32*	.65**	.63**	.47**	.27*	-0.09	.80**	0.21	.86**	.83**	.45**	.68**	.92**	.84**	.87**		.99**	.96**	0.2	0.48	0.48	0.77	.92**	-.03	0.21	-.033	-.035	0.78	-.031	-.034	
Trimelitic acid	.65**	.66**	.64**	.37**	.70**	.69**	.53**	.35**	-0.06	.85**	.29*	.88**	.86**	.50**	.74**	.94**	.88**	.91**	.99**		.97**	0.14	0.41	0.45	0.6	.90**	-.031	0.22	-.033	-.036*	0.9	-.031	-.035	
Trimesic acid	.63**	.65**	.64**	.77**	.69**	.60**	.47**	.36**	-0.01	.75**	.38**	.76**	.74**	.73**	.67**	.93**	.75**	.81**	.90**	.93**		0.11	0.4	0.24	0.62	.77**	0.09	-.001	-.015	-.022	.97	0.02	0.08	
cis-Pinonic acid	0.25	.26*	0.2	0.25	0.08	.32*	0.23	0.12	0.12	.34**	.31*	.40**	.40**	.39**	.37**	0.09	.41**	.38**	0.21	0.2	0.05		0.45	.49*	-.068	0.13	0.01	0.13	-.005	-.002	-.051	-.009	0.01	
Dehydroabietic acid	.70**	.74**	.74**	.36**	.56**	.60**	.76**	.83**	0.04	.63**	.58**	.49**	.50**	.38**	.69**	.37**	.52**	.41**	0.22	.26*	0.25	0.15		.95**	0.65	0.36	0.19	-.01	0.02	0.07	0.6	0.24	0.12	
Pimaric acid	.71**	.75**	.75**	.65**	.60**	.66**	.80**	.81**	0.04	.67**	.64**	.52**	.53**	.55**	.72**	.39**	.59**	.47**	.27*	.31*	.27*	.27*	.94**		.099	0.37	-.044	0.37	-.046	-.046	0.58	-.037	-.046	
Isopimaric acid	.58**	.61**	.62**	.34**	.41**	.48**	.68**	.79**	0.15	.45**	.57**	.30*	.33*	.32*	.54**	0.17	.31*	0.24	0.02	0.06	0.07	0.03	.93**	.82**		.09	0.48	-.047	0.51	0.66	0.44	0.64	0.63	
DHOPA	.81**	.83**	.81**	.62**	.76**	.80**	.70**	.55**	0	.94**	.49**	.95**	.96**	.74**	.87**	.87**	.91**	.89**	.85**	.87**	.79**	.27*	.55**	.57**	.37**		-.022	0.08	-.021	-.025	-.004	-.023	-.026	
Palmitic acid	0.24	0.23	.26*	.43**	.40**	0.03	0.11	.26*	0.08	0.08	.42**	-0.03	0.04	.32*	0.15	-.005	-.003	-.006	-.006	-.004	0	-.013	0.2	0.23	0.21	0.1		-.028	.95**	.99**	.091	.90**	.99**	
Linoleic acid	-.001	-.003	-.006	-.01	0.07	-.018	-.02	-.008	0.09	-.016	0.18	-.017	-.011	-.008	-.007	-.017	-.019	-.017	-.015	-.015	-.009	-.004	-.01	-.013	-.011	-.018	.63**			-.025	-.028	0.68	-.031	-.026
Oleic acid	0.06	0.08	0.11	.79**	0.05	-.01	-.012	0.04	0.14	-.009	.36**	-.014	-.009	.66**	-.004	-.02	-.016	-.012	-.019	-.017	-.011	-.012	-.002	0	0.09	0.05	.52**	.78**		.95**	.086	.90**	.93**	
Stearic acid	0.18	0.17	0.21	.40**	.37**	-.002	0.04	0.19	0.09	0.05	.35**	-.004	0.02	.29*	0.11	-.004	-.003	-.006	-.004	-.002	0	-.011	0.16	0.2	0.16	0.09	.97**	.55**	.49**		.083	.86**	.99**	
Syringaldehyde	.63**	.61**	.66**	.73**	.50**	.43**	.75**	.70**	.28*	.36**	.83**	0.2	.36**	.64**	.49**	0.03	0.21	0.23	-.002	0.06	0.17	0.15	.46**	.61**	.54**	.28*	.50**	0.06	.62**	.43**		0.77	0.9	
Cholesterol	0.09	0.12	0.16	.80**	0.08	-.007	0.14	0.09	0.15	-.005	.38**	-.01	0.08	.68**	0.01	-.018	-.012	-.008	-.017	-.014	-.006	0.18	0.03	.31*	0.13	0.08	.52**	0.23	.99**	.50**	.65**		.87**	
1-Pentadecanol	0.14	0.13	0.14	.38**	0.2	-.006	0.02	0.16	0.13	-.004	.42**	-.013	-.005	.30*	0.06	-.25*	-.011	-.014	-.022	-.019	-.022	0.15	0.1	0.17	0.12	-.005	.90**	.64**	.57**	.86**	.54**	.58**		

Post-haze

** Correlation is significant at the 0.01 level (2-tailed).

* Correlation is significant at the 0.05 level (2

Post-haze