



Molecular markers for biomass burning associated with the characterization of PM_{2.5} and component sources during dry season haze episodes in Upper South East Asia

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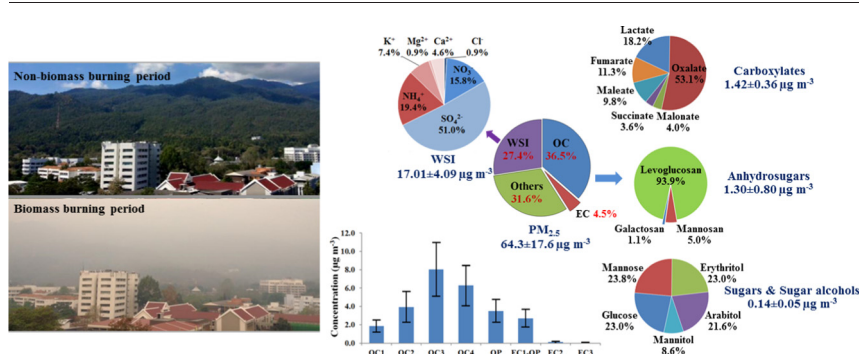
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HIGHLIGHTS

- Combustion of biomass was the major source of organic compounds in the PM_{2.5} aerosols.
- Forest and agri-waste burning were identified as major contributors to the aerosols.
- Oxalate originated mainly from primary emissions from biomass burning.
- Long-range transport of PM_{2.5} into Northern Thailand was from the west and southwest.

GRAPHICAL ABSTRACT



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ABSTRACT

Severe air pollution in the form of smoke haze in the northern part of Southeast Asia (SEA) occurs annually in the dry season due to huge open area burning. Molecular markers of biomass burning were investigated by characterization of fine particles (PM_{2.5}) collected in the dry season (23 February–28 April 2016). The average PM_{2.5}, organic carbon (OC) and elemental carbon (EC) concentrations were $64.3 \pm 17.6 \mu\text{g m}^{-3}$, $23.6 \pm 8.1 \mu\text{g m}^{-3}$ and $2.85 \pm 0.98 \mu\text{g m}^{-3}$, respectively. SO_4^{2-} was the dominant species ($8.73 \pm 2.88 \mu\text{g m}^{-3}$) of water-soluble ion, followed by NH_4^+ ($3.32 \pm 1.01 \mu\text{g m}^{-3}$) and NO_3^- ($2.70 \pm 0.51 \mu\text{g m}^{-3}$). High concentrations of the biomass burning tracers K^+ ($1.27 \pm 0.38 \mu\text{g m}^{-3}$) and levoglucosan ($1.22 \pm 0.75 \mu\text{g m}^{-3}$) were observed. The ratios of levoglucosan/ K^+ (0.92 ± 0.35) and levoglucosan/mannosan (20.4 ± 4.1) identified forest and agricultural waste burning as major contributors to the aerosol. Strong correlations ($r > 0.800$) between levoglucosan and OC, K^+ , anhydrosugar isomer (mannosan and galactosan) and other saccharides (mannose, arabinol and mannitol) verified that combustion of biomass was the major source of organic compounds associated with PM_{2.5} aerosols. Oxalate was the most abundant ($0.75 \pm 0.17 \mu\text{g m}^{-3}$; 53%) of the carboxylates. The concentration of oxalate was strongly correlated to that of PM_{2.5} ($r = 0.799$) and levoglucosan ($r = 0.615$), indicating that oxalate originates mainly from primary emissions from biomass burning rather than secondary formation from

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photochemical processes. Backward trajectories indicated that long-range transport air masses influencing air quality in Northern Thailand originated to the west and southwest.

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1. Introduction

“Smoke-haze” episodes, characterized by high concentrations of PM_{2.5} in the dry season, occur annually with increasing severity in Southeast Asia (SEA) (Marlier et al., 2013). The main cause is periodic biomass burning (BB), including forest fires and agricultural burning (Pengchai et al., 2009; Kim Oanh et al., 2011; Chantara et al., 2012), which produces large amounts of PM_{2.5} emissions (van der Werf et al., 2010; Chang et al., 2015; Lyu et al., 2015; Shi et al., 2015; Wiriya et al., 2016). Emissions of PM_{2.5} are increasing rapidly in SEA (Shi et al., 2018). These emissions have local, regional and global impacts on air quality, public health and climate (Dai et al., 2013; Pani et al., 2016a, 2016b; Tsay et al., 2016; Chen et al., 2017; Pani et al., 2018). For decades, SEA has been a source of pollution affecting countries within the region (Dotse et al., 2016) and also outside of it, in East Asia, as the aerosol plume is transported eastward by the prevailing westerly winds. Regional transport of SEA BB aerosols have been investigated over locations in East Asia including southeastern China, the South China Sea and central/southern Taiwan, most commonly in the dry season (e.g., Lee et al., 2011; Lin et al., 2013; Chuang et al., 2014; Tsai et al., 2015; Pani et al., 2016a).

To estimate the contribution of different sources to a given ambient particulate sample, a characteristic compound in each of the potential source materials is considered. Chemical analysis of smoke aerosols during forest fires showed that potassium (K⁺) and ammonium ions (NH₄⁺) are the dominant cations and that sulfate ions (SO₄²⁻) are the most important anions (Pio et al., 2008). A study of agricultural burning, meanwhile, reported a four- to five-fold increase in OC, EC, NO₃⁻, NH₄⁺, SO₄²⁻ and K⁺ (Niemi et al., 2004). Multiple studies confirm the role of K⁺ as a biosmoke tracer (Duan et al., 2004; Zhang et al., 2010; Lee et al., 2011; Cheng et al., 2013).

Biomass burning produces many biomarkers in the transported air and burnt soil which can be used as species for source tracing, including the three isomeric anhydrous sugars levoglucosan, mannosan, and galactosan (Simoneit et al., 1999; Elias et al., 2001; Simoneit, 2002). These sugars are formed during pyrolysis of cellulose, and are therefore emitted in large quantities during the burning of biomass but are not emitted from burning other materials such as fossil fuels (Elias et al., 2001; Simoneit et al., 1999; Caseiro et al., 2009). Several studies have used the levoglucosan to K⁺ ratio and the levoglucosan to PM ratio to estimate the contribution of biomass burning emissions to ambient aerosols (Duan et al., 2004; Schmidl et al., 2008a, 2008b; Zhang et al., 2008; Tsai et al., 2013; Yu et al., 2018). In an attempt to specify the type of biomass burning, the levoglucosan to mannosan ratio (Schmidl et al., 2008a, 2008b) and the levoglucosan to OC ratio (Pio et al., 2008; Sullivan et al., 2008) have also been used in analyses.

In addition to the above, other sugars and dicarboxylic acids have been utilized as biomass-burning tracers (Tsai et al., 2013; Nirmalkar et al., 2015). Sugar alcohols originate from the metabolism of fungi (Bauer et al., 2008) often found on the bark and leaves of trees (Medeiros et al., 2006), and these sugar alcohols are emitted through biomass burning (Simoneit et al., 1999). Therefore, the presence of sugar alcohols in combination with high-levels of levoglucosan and its isomer specifically indicates that biomass burning has contributed to the aerosol content. Carboxylates account for a small but important fraction of the organics. Their sources include direct release into the atmosphere from traffic emissions, wood burning, biogenic activities and as secondary by-products of photochemical oxidation of organic precursors (Khawaja, 1995; Kawamura et al., 1996; Hsieh et al., 2008; Wang and Shooter, 2004; Yao et al., 2004; Tsai et al., 2008, 2010, 2013).

However, only a few studies have been focused on sugars and carboxylates in PM_{2.5} aerosols in SEA (Tsai et al., 2013).

A number of studies have focused on the apportionment of biomass-burning emissions to ambient aerosols over the American and Asian continents, especially in China and India (Duan et al., 2004; Zheng et al., 2005; Zhang et al., 2008, 2010, 2013; Cheng et al., 2013; Chuang et al., 2013a, 2013b; Bae et al., 2014; Cheng et al., 2014; Kusumaningtyas et al., 2016; Du et al., 2017; Li et al., 2017; Tian et al., 2017; Martins et al., 2018; Pani et al., 2018; Shahid et al., 2018; Thuy et al., 2018). However, information on the quality and quantity of biomass-burning aerosols in SEA is limited. Near-source investigations of the influence of forest fires and agricultural waste burning on SEA aerosols are few in number (Chuang et al., 2013b; Tsai et al., 2013; Khamkaew et al., 2016a).

Northern Thailand has been experiencing the impacts of air pollution from biomass burning for over 10 years. The biggest city, Chiang Mai, and its environs suffer particularly badly because of their location in an inter-mountain basin, together with meteorological conditions involving temperature inversion and low wind speed in the dry season that limit air dispersion and ventilation (Pani et al., 2018). Backward trajectory (BWT) studies have shown that haze episodes and the concomitant increase in PM_{2.5} concentrations (Shi et al., 2018) in Chiang Mai are typically associated with a main air mass movement into the region from the southwest through India and Myanmar (Chantara et al., 2012; Wiriya et al., 2013; Punsompong and Chantara, 2018). Areas downwind of the prevailing westerlies in SEA are frequently influenced by the long-range transport of aerosols. All this implies that haze events in Chiang Mai may be caused by forest fires and agricultural waste burning from both the local area and from regions some distance to the west/southwest. The purpose of this study is (1) to investigate the level of PM_{2.5} and its chemical composition, including anhydrosugars, water-soluble ions and carboxylates, during the open burning season in near source areas, (2) to determine the potential sources of PM_{2.5} by an investigation of bound chemical compounds within the PM_{2.5}, and (3) to identify the source(s) of pollutants in Chiang Mai based on cluster analysis of backward trajectories.

2. Materials and methods

2.1. Sampling site

The sampling site was located in Chiang Mai Province, Thailand. Chiang Mai is the largest metropolitan area in northern Thailand. It is the second largest city in Thailand with a population of 1.73 million in a total area of 20,107 km² (Chiang Mai, 2017). The region has mountains running in a north-south direction, covered in deciduous forest, and borders Myanmar to the north and Mae Hong Son Province to the west. The city of Chiang Mai is surrounded by forest (3665 km² or 61% of the area) and agricultural land (1459 km² or 24% of the area) (Land Development Department, 2015). In general, trees annually lose some or all of their leaves in the dry season, and fires usually occur from December to March every year (Kim Oanh et al., 2011; Chantara et al., 2012; Junpen et al., 2013; Punsompong and Chantara, 2018). Chiang Mai University (CMU), in which the sampling site was located, sits at a height of 373 m above sea level, 18° 48' 5.40"N, 98° 57' 12.18"E, in the Chiang Mai-Lamphun basin, and is surrounded by high mountains (Fig. 1). There is an industrial estate 15 km to the southeast. In order to minimize effects from traffic emissions directly, the rooftop of a tall building (~30 m above ground) was selected as the sampling site: the air samplers were set on the rooftop of the nine-story Science Complex

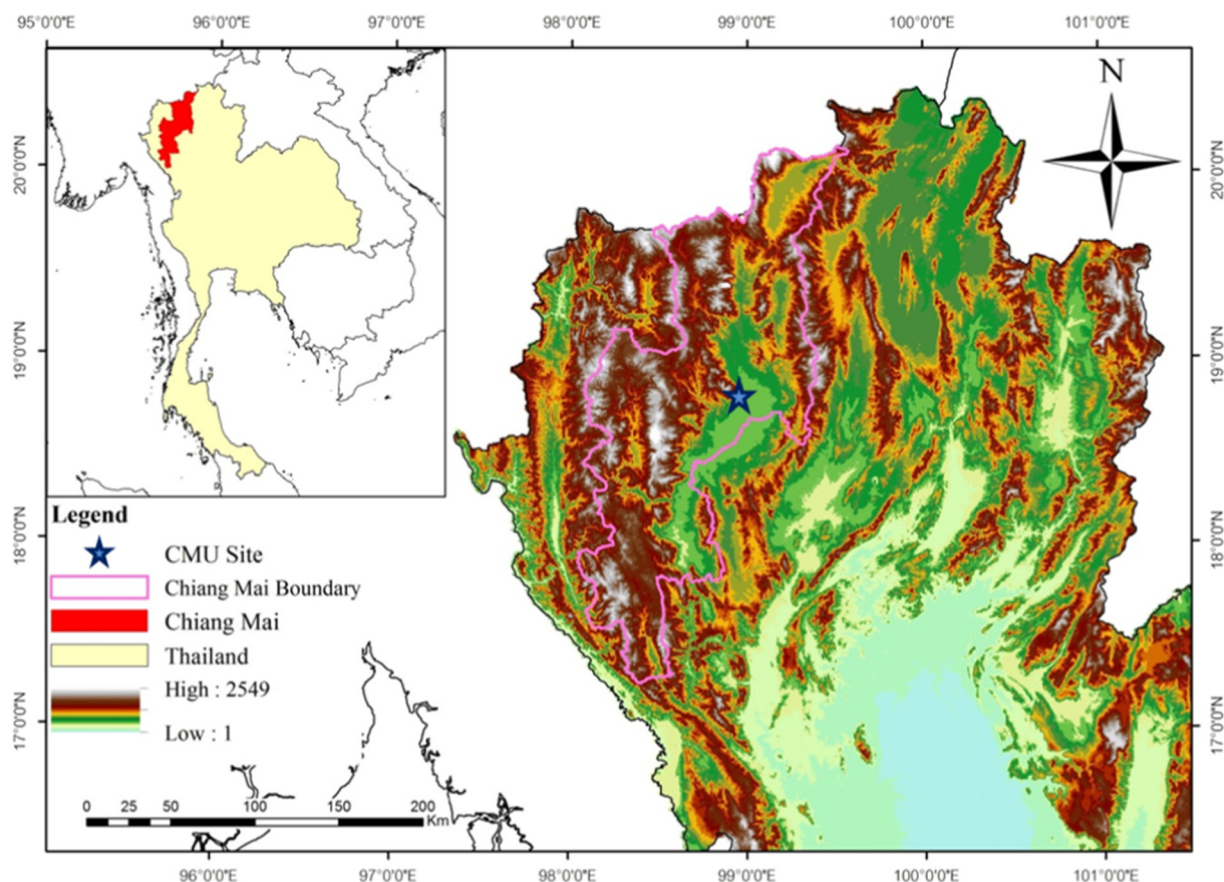


Fig. 1. Topographic map of Northern Thailand and location of Chiang Mai University (CMU) air quality monitoring site (blue star), Chiang Mai Province. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Building 1 in CMU, which is situated adjacent to the Suthep mountain foothill.

2.2. Sample collection

Sampling was carried out for two months (23 February–28 April) during the dry season of 2016. One $PM_{2.5}$ sample was collected daily (24 h) using a mini volume air sampler (Air metric, USA) with a flow rate of 5 L/min. The samples were collected on quartz fiber filters (\varnothing 47 mm) purchased from QM-A Whatman (Maidstone, Kent, UK). There were a total of 60 samples.

Prior to sampling, all filters were stored in a desiccator for 24 h to remove moisture and were then pre-weighed three times on an MX-5 microbalance (Mettler Toledo, Switzerland) under controlled temperature ($25 \pm 2^\circ\text{C}$) and relative humidity ($41 \pm 5\%$). After sample collection, the filters were stored individually in plastic boxes wrapped with aluminum foil to avoid photodegradation until transfer to a desiccator for 24 h, after which they were re-weighed. The filter boxes were stored in sealed plastic bags and kept refrigerated until analysis.

2.3. OC/EC analysis

OC and EC content were analyzed using a DRI Model 2001A Carbon Analyzer (Atmoslytic Inc., CA, USA) employing the Thermal Optical Reflectance method. The Interagency Monitoring of Protected Visual Environments (IMPROVE) protocol was adopted for this analysis (Chow et al., 2005; Han et al., 2007; Cheng et al., 2015).

The IMPROVE protocol for analyzing OC and EC fractions as a function of temperature is described in detail by Chuang et al. (2013a). This protocol requires a gradual increase in temperature from room temperature to 840°C . OC and EC are separated based on the capacity

of OC to volatilize from the sample punch in a pure helium (He) atmosphere at low temperatures while EC is oxidized with the introduction of a small fraction of oxygen in the He flow and removed at high temperatures. OC is defined as the sum of the carbon fractions that evolved from the 0.526 cm^2 filter sample punch in a pure He atmosphere ending at 120°C (OC1), 280°C (OC2), 480°C (OC3), and 580°C (OC4) plus the pyrolyzed OC (OP). EC is the sum of carbonaceous fractions that evolved in a 98% helium/2% oxygen atmosphere ending at 580°C minus OP (EC1-OP), 740°C (EC2), and 840°C (EC3). OP is defined as the carbonaceous fraction that evolved between the moment at which oxygen was introduced into the He atmosphere at 580°C and that at which the surface reflectance of a laser on the filter punch returned to its value at the beginning of the analysis.

2.4. Sample extraction and chemical analysis

The sample filters were cut into halves using stainless steel scissors. The extraction and filtration procedure was similar to that described by Tsai et al. (2013). Briefly, one half of each sample filter was taken and placed in a PE bottle, 5.0 mL of deionized water (resistivity $> 18.2\text{ M}\Omega\text{-cm}$ at 25°C , Milli-Q Direct 8/16 System, Germany) was added, and the contents were shaken (Yihder TS-500 Shaker) in an unlit refrigerator at 4°C for 90 min to extract the sample. The liquid was then filtered through an ester acetate syringe membrane (C020A025A, 25 mm, pore size $0.2\text{ }\mu\text{m}$, Advantec, Japan).

Cations, including sodium (Na^+), ammonium (NH_4^+), potassium (K^+), magnesium (Mg^{2+}) and calcium (Ca^{2+}), were determined using a Dionex ICS-1000 equipped with a gradient pump (ICS-3000 Model SP-1), Dionex CSRS Ultra 4 mm Self-regenerating suppressor and conductivity detector and an Ion Pac CS12A (250 mm length \times 4 mm I.D.) analytical column. An autosampler (AS3000) with an injection volume

of 25 μL and isocratic 20 mM Methanesulfonic acid (MSA) as eluent was used with a flow rate of 1.0 mL min^{-1} .

Anhydrosugars (levoglucosan, mannosan and galactosan), sugars (glucose and mannose) and sugar alcohols (erythritol, arabitrol and mannitol) were determined using a Dionex ICS-5000+ IC equipped with pulsed amperometric detection, a CarboPac MA1 guard column (50 mm length \times 4 mm I.D.), an anion-exchange analytical column (250 mm length \times 4 mm I.D.), and a Dionex ICS-5000 Electrochemical Detector with a gold working electrode and a Dionex pH-Ag/AgCl electrode as reference. An autosampler (AS-AP) was used for the injection; 200 μL of sample was introduced into the system using a gradient pump (ICS-3000 Model SP-1) coupled to a Teflon injection valve. The eluent was 250 mM NaOH and the flow rate was maintained at 0.5 mL min^{-1} .

Anions, including chloride (Cl^-), nitrate (NO_3^-), sulfate (SO_4^{2-}), and carboxylates (lactate, succinate, malonate, maleate, fumarate and oxalate) were analyzed using a Dionex ICS-5000+ IC, which was equipped with a gradient pump (ICS-3000 Model SP-1) and AMMS 300 MicromembraneTH suppressor and conductivity detector. An autosampler (AS-AP) with 2 mL vials and a Teflon injection valve used a 1000 μL sample loop. The analytical columns and guard columns were Ion Pac AG11-HC RFICTM (250 mm length \times 4 mm I.D.) and AS-11-HC RFICTM (50 mm length \times 4 mm I.D.), respectively. The gradient elution was at a flow rate of 0.5 mL min^{-1} with DI water, 5 mM sodium hydroxide (NaOH), 100 mM NaOH, and 100% methanol. In addition, 100 mM sulfuric acid solution was used as a regenerate suppressor.

2.5. Quality assurance and quality control for IC and OC/EC analysis

The method detection limit (MDL) for individual species was defined as a signal-to-noise ratio from multiple injections of a lowest concentration standard solution corresponding to three times the standard deviation of the species. Accuracy was tested by spiking half of the quartz filter with the mixed standards before extraction. MDLs and percentage recovery of the four chemical compound groups measured are listed in Table 1. MDL values were 0.025–0.063 $\mu\text{g m}^{-3}$ for cations, 0.025–0.061 $\mu\text{g m}^{-3}$ for anions, 2.75–12.3 ng m^{-3} for carboxylates, 1.95–2.77 ng m^{-3} for anhydrosugars and 1.76–7.02 ng m^{-3} for sugar alcohols and sugars. High-percentage recoveries were obtained for all

Table 1
Method detection limits (MDLs) and percentage extraction recovery for four chemical groups measured using IC systems.

| Compound | MDL | Recoveries (%) (n = 3) | RSD (%) (n = 3) |
|--|-------|---------------------------|--------------------|
| Water soluble ions ($\mu\text{g m}^{-3}$) | | | |
| Na^+ | 0.063 | 102.3 | 4.65 |
| NH_4^+ | 0.025 | 96.9 | 2.42 |
| K^+ | 0.027 | 98.5 | 2.58 |
| Mg^{2+} | 0.043 | 101.4 | 3.05 |
| Ca^{2+} | 0.052 | 103.1 | 4.12 |
| Cl^- | 0.025 | 102.1 | 2.55 |
| NO_3^- | 0.045 | 97.6 | 3.48 |
| SO_4^{2-} | 0.061 | 100.4 | 4.98 |
| Carboxylates (ng m^{-3}) | | | |
| Lactate | 12.3 | 108.7 | 8.34 |
| Succinate | 2.75 | 103.1 | 5.16 |
| Malonate | 6.68 | 105.1 | 5.35 |
| Maleate | 3.58 | 98.2 | 4.20 |
| Fumarate | 7.70 | 103.3 | 3.15 |
| Oxalate | 10.3 | 99.8 | 5.74 |
| Anhydrosugars (ng m^{-3}) | | | |
| Levoglucosan | 2.32 | 102.5 | 4.76 |
| Mannosan | 1.95 | 98.9 | 2.87 |
| Galactosan | 2.77 | 104.4 | 3.85 |
| Sugars and sugar alcohols (ng m^{-3}) | | | |
| Glucose | 7.02 | 103.1 | 4.38 |
| Mannose | 5.04 | 104.1 | 3.97 |
| Erythritol | 1.76 | 99.5 | 2.94 |
| Arabitrol | 3.04 | 103.6 | 4.19 |
| Mannitol | 3.02 | 99.8 | 4.02 |

species (96.9–108.7%) with low percentage RSD (2.42–8.34), which confirmed the relatively high degree of accuracy and reproducibility of the analysis. Quality control checks were performed on the IC system every 10–12 sample injections by the use of three levels of mixed standard solution for peak area or peak height comparison. Blank filters were also extracted and analyzed using the same method as for the samples. A mean value was used for background subtraction from each sample. The detection limit of the carbon analyzer used in this study was 0.05 $\mu\text{g C cm}^{-2}$ for both OC and EC. The carbon fractions were corrected using the averaged blank concentrations.

2.6. Air mass trajectories and air pollutants analysis

Backward trajectories (BWT) of air masses during the sampling period (March to April 2016) were determined in order to identify the origin and transport paths of air masses and their arrival at the sampling site. Two-day BWT were obtained using the TrajStat-Trajectory Statistics program version 1.2.2.6 developed by Wang et al. (2009). This program uses the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT, Draxler and Rolph, 2003) model as an external process to calculate trajectories, and Euclidean distance clustering as well as a geographic information systems (GIS) technique for spatial data management, visualization and analyses. The meteorological input for the trajectory model was the GDAS (Global Data Assimilation System) and meteorological data ($1^\circ \times 1^\circ$). In this study, BWT were calculated four times per day, at 0:00, 6:00, 12:00 and 18:00 UTC, for each day of the sampling period. Daily concentrations of individual pollutants were compared against BWT to assess potential pollutant origins.

3. Results and discussion

3.1. $\text{PM}_{2.5}$ mass concentrations

The daily $\text{PM}_{2.5}$ concentration at the CMU site obtained via the gravimetric method was compared to the values obtained from an automatic active sampler (Taper Element Oscillation Microbalance, TEOM) at the Chiang Mai Air Quality Monitoring (CM-AQM) station belonging to the Pollution Control Department located at the center of Chiang Mai City. The patterns of average daily $\text{PM}_{2.5}$ concentration from 23 February to 28 April 2016 are shown in Fig. 2. Daily mean $\text{PM}_{2.5}$ concentrations at the CMU site were strongly correlated ($r = 0.9434$) with those at the CM-AQM station. The average $\text{PM}_{2.5}$ mass concentration during the two months of sampling was $64.3 \pm 17.6 \mu\text{g m}^{-3}$. This value was in good agreement with previous studies at the same sampling site, i.e. $74.5 \pm 43.5 \mu\text{g m}^{-3}$ in March 2013 (Khamkaew et al., 2016b) and $93.1 \pm 37.3 \mu\text{g m}^{-3}$ in March to Mid-April 2014 (Khamkaew et al., 2016a). More than 82% (49/60 days) of the 24-h $\text{PM}_{2.5}$ concentrations exceeded the Thailand National Ambient Air Quality Standard ($50 \mu\text{g m}^{-3}$).

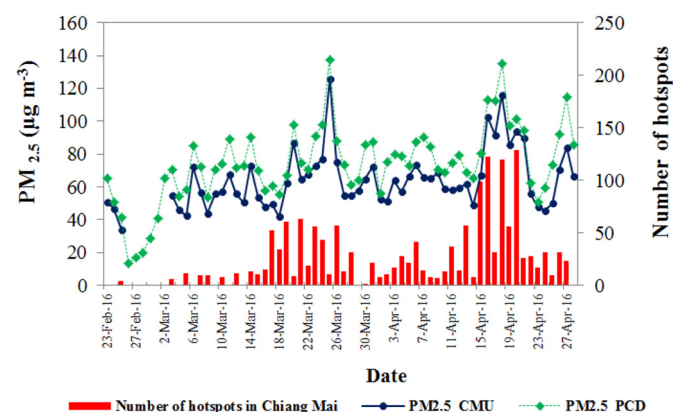


Fig. 2. Daily $\text{PM}_{2.5}$ concentrations ($\mu\text{g m}^{-3}$) at CMU sites obtained via the gravimetric method compared to the values from TEOM and number of hotspots in Chiang Mai.

Punsompong and Chantara (2018) reported high correlations between PM₁₀ concentration and number of active fires during 3-day BWT studies over six years (2010–2015). They concluded that the major open burning source during March and April was in the agricultural and forest areas of Myanmar, although the agricultural areas of northern Thailand also contributed to the PM₁₀ concentration in Chiang Mai. In the dry period, fires are set in Northern Thailand mostly to burn agricultural wastes in preparation for subsequent cultivation. For comparison, the active fire data (hotspots) for nine provinces in Northern Thailand, detected by satellite remote sensing imagery from the Moderate Resolution Imaging Spectroradiometer (MODIS) and available on Web Fire Mapper (<http://earthdata.nasa.gov/data/near-real-time-data/firms>), were added to the average daily PM_{2.5} concentration graph (Fig. 2). The meteorological and topographical conditions in the dry season affect the air pollutants in this area, and those pollutants accumulate in the cities located in the mountain valleys of Northern Thailand.

3.2. Chemical components and their contribution to PM_{2.5} during smoke haze period

The concentration of chemical components and their contribution to PM_{2.5} mass concentration are shown in Table 2. Fig. 3 shows the percentages of various chemical species detected in PM_{2.5}. OC, EC and water-soluble ions comprised 36.5, 4.5 and 27.4% of the PM_{2.5} mass concentration, respectively, while the remainder of the PM_{2.5} mass concentration included unidentified species, namely trace metals, some ionic species (Na⁺, PO₄³⁻ and NO₃⁻) and other organic matter. Water soluble organic compounds (carboxylates, anhydrosugars, and sugars & sugar alcohols) represented a small but nevertheless important component. These were analyzed and their percent contributions to PM_{2.5} are shown in Fig. 4.

Of the four OC fractions, OC3 was the most abundant, followed by OC4 and OC2. Within EC, EC1-OP was the major fraction. EC1-OP and OC3 are also good biomass burning tracers near sources (Chuang et al., 2013b), while OC2 is the most abundant fraction of OC in coal combustion and motor vehicle exhausts (Cheng et al., 2015). Biomass burning combustion sources are considered as the largest contributors to primary carbonaceous particles in the atmosphere (Zheng et al., 2005; Cheng et al., 2013; Chen et al., 2017) and many studies have used the above-mentioned components for source apportionment of biomass burning aerosols (Chow et al., 2004; Cao et al., 2005; Han et al., 2007). Results in this study indicate that the aerosol carbonaceous content was influenced by biomass burning.

Rajput et al. (2013, 2014) linked variability in the OC/EC ratio with emission sources such as biomass burning, fossil fuel combustion and secondary formation, and attributed an elevated OC/EC ratio to biomass burning emission as well as to secondary organic aerosol (SOA) formation. In this study, the OC/EC ratio in PM_{2.5} also varied, being at 3.6 initially and gradually increasing to 11 (Fig. 5). These ratios are similar to those observed at Xi'an, which varied from 2.2 to 15.2 (Shen et al., 2010) and reflected emission from coal combustion and biomass burning. Sorathia et al. (2018) have also assessed the OC/EC ratio and found a large daytime and nighttime variability of 10.8–14.7 and 7.6–10.6, respectively. Moreover, the biomass burning tracers, levoglucosan and K⁺ also exhibited variability patterns quite similar to that of the OC/EC ratio. Meanwhile, Lee et al. (2006) reported very low OC/EC ratios for vehicular emissions in Hong Kong. The OC/EC ratios for motor vehicle exhaust, coal combustion, residential wood combustion and forest fires were reported as 1.4, 2.7, 4.3 and 15.7, respectively (Watson et al., 2001). In this study, a higher OC/EC ratio indicated that biomass burning plays an important role in carbonaceous aerosol in Chiang Mai, which corresponds with the studies at the same site in 2014 (Pani et al., 2018).

Of water soluble ions, SO₄²⁻ was the dominant species, followed by NH₄⁺ and NO₃⁻, representing 51.0, 19.4 and 15.8% of total ion mass concentration, respectively. These ions are secondary inorganic aerosols

Table 2

Concentrations of PM_{2.5} and their water-soluble ions, carboxylates, anhydrosugars, sugars and sugar alcohols collected during smoke haze period in 2016.

| Parameter | Concentration (μg m ⁻³) | | Contribution in PM _{2.5} (%) |
|---|-------------------------------------|-------------------|---------------------------------------|
| | Mean ± sd (n = 60) | Min–max | Mean ± sd |
| PM _{2.5} (μg m ⁻³) | 64.3 ± 17.6 | 34.1–126 | |
| OC | 23.6 ± 8.1 | 8.1–48.8 | 36.5 ± 5.3 |
| OC1 | 1.88 ± 0.64 | 0.48–3.72 | 2.92 ± 0.64 |
| OC2 | 3.94 ± 1.66 | 1.42–9.00 | 6.02 ± 1.31 |
| OC3 | 8.03 ± 2.91 | 3.87–17.29 | 12.35 ± 1.91 |
| OC4 | 6.28 ± 2.19 | 2.46–12.96 | 9.66 ± 1.65 |
| OP | 3.52 ± 1.21 | 1.52–7.96 | 5.53 ± 1.29 |
| EC | 2.85 ± 0.98 | 1.59–7.22 | 4.48 ± 1.04 |
| EC1-OP | 2.72 ± 0.94 | 1.47–7.13 | 4.25 ± 0.89 |
| EC2 | 0.12 ± 0.09 | ND–0.37 | 0.19 ± 0.16 |
| EC3 | 0.02 ± 0.06 | ND–0.30 | 0.03 ± 0.11 |
| Water-soluble ions (μg m ⁻³) | 17.01 ± 4.09 | 8.48–24.4 | 27.4 ± 7.0 |
| Total cations | 5.44 ± 1.33 | 2.53–8.11 | 8.66 ± 1.85 |
| NH ₄ ⁺ | 3.32 ± 1.01 | 1.24–5.29 | 5.31 ± 1.62 |
| K ⁺ | 1.27 ± 0.38 | 0.57–2.25 | 1.88 ± 0.57 |
| Mg ²⁺ | 0.15 ± 0.05 | 0.08–0.28 | 0.23 ± 0.08 |
| Ca ²⁺ | 0.79 ± 0.16 | 0.56–1.22 | 1.24 ± 0.36 |
| Total anions | 11.56 ± 2.88 | 5.94–16.98 | 18.7 ± 5.2 |
| Cl ⁻ | 0.14 ± 0.14 | ND–0.61 | 0.25 ± 0.28 |
| NO ₃ ⁻ | 2.70 ± 0.51 | 1.43–3.79 | 4.34 ± 0.84 |
| SO ₄ ²⁻ | 8.73 ± 2.88 | 3.37–14.28 | 14.11 ± 5.10 |
| Carboxylates (ng m ⁻³) | 1421 ± 365 | 845–2786 | 2.28 ± 0.63 |
| Lactate | 259 ± 220 | ND–1709 | 0.43 ± 0.45 |
| Succinate | 51 ± 62 | 3–350 | 0.08 ± 0.09 |
| Malonate | 57 ± 30 | 12–125 | 0.09 ± 0.05 |
| Maleate | 139 ± 34 | 52–218 | 0.22 ± 0.06 |
| Fumarate | 160 ± 24 | 124–240 | 0.26 ± 0.04 |
| Oxalate | 754 ± 167 | 510–1347 | 1.20 ± 0.19 |
| Anhydrosugars (ng m ⁻³) | 1300 ± 804 | 357–4098 | 1.94 ± 0.77 |
| Levoglucosan | 1222 ± 751 | 336–3853 | 1.82 ± 0.721 |
| Mannosan | 65 ± 48 | 16–217 | 0.10 ± 0.05 |
| Galactosan | 14 ± 7 | 3–37 | 0.02 ± 0.01 |
| Sugars and sugar alcohols (ng m ⁻³) | 138 ± 53 | 79–318 | 0.22 ± 0.05 |
| Glucose | 32 ± 14 | 16–108 | 0.05 ± 0.02 |
| Mannose | 33 ± 20 | 8–98 | 0.05 ± 0.02 |
| Erythritol | 32 ± 8 | 17–52 | 0.05 ± 0.01 |
| Arabitol | 30 ± 13 | 14–86 | 0.05 ± 0.01 |
| Mannitol | 12 ± 7 | ND–39 | 0.02 ± 0.01 |

(SIA), which were reported by Yao et al. (2016) as the dominant fractions of PM_{2.5}, especially in the summer period. SO₄²⁻ is usually generated mainly from fossil fuel combustion by the photochemical formation of SO₂ from traffic and/or industrial activity (Kalabokas et al., 1999). With proximity to the city of Chiang Mai and to industrial estates located in the Chiang Mai-Lamphun basin, the CMU site may be affected by traffic emission as well as industrial emissions (petrochemical, cement, ceramic and metal industries) and hence fossil fuel combustion is a potential source of SO₄²⁻. However, Sillapapiromsuk et al. (2013) revealed that SO₄²⁻ was the major ion found in particulate matters emitted from biomass burning, while Pengchai et al. (2009) described a source-apportionment model in which SO₄²⁻ formation was from vehicle exhaust gases and biomass burning. Hence, SO₄²⁻ found at the CMU site could be from mixed sources of fossil fuel combustion and biomass burning. Potential sources of NO₃⁻ are traffic emission (Minguillon et al., 2014), biomass burning (Ryu et al., 2007; Chantara et al., 2012) and agricultural activities (Gurjar et al., 2015). The abundant NO_x emitted from biomass burning activity (Lobert et al., 1991) is oxidized by OH radicals and forms HNO₃ in the transported air masses (Gupta et al., 2003; Gobbi et al., 2007; Karar and Gupta, 2007). Therefore, NO₃⁻ was reported as the most prominent component enhanced during biomass burning (Fabian et al., 2005; Lee et al., 2011). Wang et al. (2016) reported that high emissions of NO_x from biomass burning and their long/medium-range transport, especially when air masses pass over open biomass burning areas, has a significant impact on NO₃⁻ in an aerosol sample.

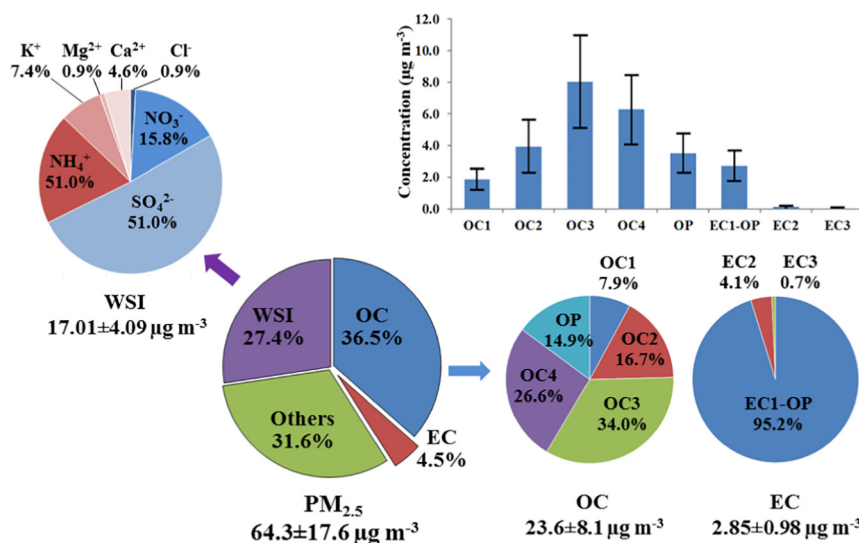


Fig. 3. Percentages of OC, EC, WSI and others species as composition of PM_{2.5} collected during smoke haze period in 2016.

Comparison of concentrations of PM mass and water-soluble ions in this study with those reported in the literature is provided in Table 3. In this study, the concentrations of SO_4^{2-} were $8.73 \pm 2.88 \mu\text{g m}^{-3}$, which is similar to those reported at the same site in 2014 ($7.81 \pm 2.60 \mu\text{g m}^{-3}$) (Khamkaew et al., 2016a), but lower than reported in Taiwan during the agricultural waste burning period ($16.9 \pm 5.8 \mu\text{g m}^{-3}$) (Cheng et al., 2009). The concentration of NO_3^- ($2.70 \pm 0.51 \mu\text{g m}^{-3}$) in this study was similar to those reported in previous studies in the same location, with an average $2.34 \pm 1.95 \mu\text{g m}^{-3}$ (Tsai et al., 2013) and $2.65 \pm 1.13 \mu\text{g m}^{-3}$ (Khamkaew et al., 2016a) during the dry seasons of 2010 and 2014, respectively, while it was lower than that reported in a near-source site located at a higher altitude near the Myanmar border ($4.13 \pm 2.86 \mu\text{g m}^{-3}$) (Khamkaew et al., 2016a).

Ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$) and ureas are the recommended and the most widely used nitrogen (N) fertilizers for crop production (Bufogle et al., 1998) and because of this NH_4^+ in aerosols mainly originate from agricultural activity. High concentration of NH_4^+ ($3.32 \pm 1.01 \mu\text{g m}^{-3}$) and K^+ ($1.27 \pm 0.38 \mu\text{g m}^{-3}$) indicate significant inputs from agricultural activity and biomass burning. In this study, K^+ recorded higher concentrations than have been reported in previous studies in non-episodic periods in Chiang Mai, Thailand (average $0.89 \pm 0.57 \mu\text{g m}^{-3}$, Tsai et al., 2013), as well as in non-agricultural waste burning in Taiwan (average $0.4 \pm 0.2 \mu\text{g m}^{-3}$, Cheng et al., 2009). Ca^{2+} and

Mg^{2+} originate from roadside soil (Tsai and Chen, 2006), demonstrating that crustal matter (i.e. mineral dust) may be a potential contributor to PM_{2.5}. In this study, Ca^{2+} ($1.24 \pm 0.36\%$) and Mg^{2+} ($0.23 \pm 0.08\%$) were found in quite low percentages.

Carboxylates were found in a small fraction, comprising $2.28 \pm 0.63\%$ of PM_{2.5} mass concentration. Oxalate, the final product of photochemical decomposition of other dicarboxylic acids in atmospheric aerosol (Kawamura et al., 1996), was the most abundant carboxylate in PM_{2.5}, followed by lactate, fumarate and maleate, at 53.1, 18.2, 11.3 and 9.8% of total carboxylate mass concentration, respectively. Carboxylate sources include direct release into the atmosphere via anthropogenic and/or biogenic activities and as secondary byproducts of photochemical oxidation of organic precursors (Khwaja, 1995; Kawamura et al., 1996; Tsai et al., 2013).

Succinate and malonate were small components of total carboxylate, at 3.6% and 4.0%, respectively. These are important dicarboxylic acid species and are noted as traffic-related emission species and photochemical species in the literature (Wang and Shooter, 2004; Yao et al., 2004; Hsieh et al., 2008; Tsai et al., 2008; Ho et al., 2010). Malonic acid is produced by the photochemical breakdown of succinic acid (Kawamura and Bikkina, 2016). The malonate/succinate ratios have been used as an indicator of enhanced photochemical aging of organic aerosols in the atmosphere. Lower malonate/succinate ratios in the

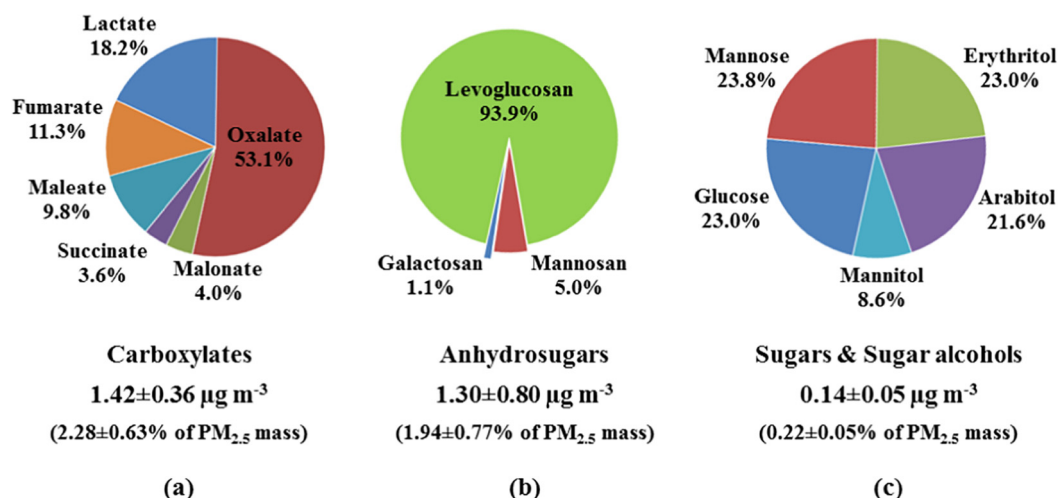


Fig. 4. Percentages of water soluble organic compounds (carboxylates, anhydrosugars and sugars & sugar alcohols) detected in PM_{2.5} samples during smoke haze period in 2016.

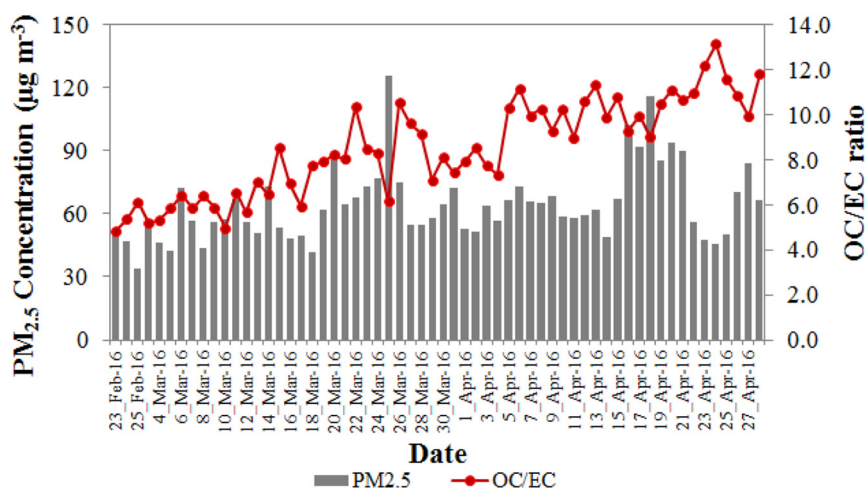


Fig. 5. Comparison of daily $PM_{2.5}$ concentration and OC/EC ratio in $PM_{2.5}$ during smoke haze period in 2016.

range 0.3–0.6 have been reported for fossil fuel combustion while ratios in the range 1.5–2.7 (Yao et al., 2004) and >3 (Kawamura et al., 1996; Kawamura and Bikkina, 2016) have been reported for secondary formation. In this study the ratio of malonate/succinate ranged from 0.32 to 7.41 with a mean value of 2.05 ± 1.37 , suggesting that secondary oxidation and subsequent transformation in aerosols is more significant than direct emission of malonate from biomass and wood burning and fossil fuel combustion.

During the study period, anhydrosugars comprised $1.94 \pm 0.77\%$ of the $PM_{2.5}$ mass concentration, indicating that during smoke haze periods more pollutants from biomass burning are emitted into the ambient environment. Levoglucosan, the most useful marker for burning of both natural forest and agricultural waste, clearly dominated, with an average concentration of $1.22 \pm 0.75 \mu\text{g m}^{-3}$, followed by mannosan ($0.065 \pm 0.048 \mu\text{g m}^{-3}$) and galactosan ($0.014 \pm 0.007 \mu\text{g m}^{-3}$), equating to 93.9%, 5.0% and 1.1% of total anhydrosugars, respectively. A comparison of anhydrosugars in this work with those reported elsewhere is presented in Table 3. Levoglucosan concentrations have been quantified in a few previous studies conducted in Chiang Mai. The levoglucosan levels reported by Khamkaew et al. (2016a) averaged $1.13 \pm 0.70 \mu\text{g m}^{-3}$ during the dry season of 2014 with comparable values observed in 2010 ($1.18 \pm 0.80 \mu\text{g m}^{-3}$) by Tsai et al. (2013). Compared with annual average levoglucosan concentrations in Beijing ($0.43 \pm 0.44 \mu\text{g m}^{-3}$) (Cheng et al., 2013), Chiang Mai was characterized by rather high levoglucosan concentrations, especially during a smoke-haze period in the dry season. However, the levoglucosan concentrations measured in Chiang Mai were half of those observed in India during a biomass burning period, which produced an average value of $2.26 \pm 0.73 \mu\text{g m}^{-3}$ (Nirmalkar et al., 2015).

Sugars and sugar alcohols were found to comprise $0.22 \pm 0.05\%$ of the $PM_{2.5}$ mass concentration. Fig. 4 shows the percentage contribution of individual sugar and sugar alcohol to the total. The concentrations of glucose, mannose, erythritol and arabitol were approximately the same, averaging $30\text{--}33 \text{ ng m}^{-3}$, while mannitol was the lowest. Sugar alcohols originate from the metabolism of fungi (Bauer et al., 2008), often found on the bark and leaves of trees (Medeiros et al., 2006). Erythritol, from soil microbial metabolites, is produced when the burning of agricultural waste in the field also causes the soil to be burnt at the same time (Tsai et al., 2015). The concentration of erythritol in this study ($32 \pm 8 \text{ ng m}^{-3}$) was similar to that at the same site ($42 \pm 18 \text{ ng m}^{-3}$) during the biomass burning season in 2010 (Tsai et al., 2013) but only a quarter of that observed in India ($135 \pm 98 \text{ ng m}^{-3}$) during three episodic periods from October to November 2011 (Nirmalkar et al., 2015). The presence of sugar alcohol and high-levels of levoglucosan and its isomer specifically indicated that biomass burning contributed to atmospheric aerosols in the Chiang Mai Basin.

3.3. Correlations of $PM_{2.5}$ and its chemical composition in relation to source identification

Pearson correlations of $PM_{2.5}$ with OC, EC and associated chemical components were investigated and results are shown in Table 4. The $PM_{2.5}$ concentrations were strongly correlated with OC ($r = 0.911$) (Fig. 6a) and EC ($r = 0.747$) (Fig. 6b). A strong correlation ($r = 0.914$) between OC and levoglucosan and a high slope ($m = 9.83$) (Fig. 6c) suggested that a large fraction of OC may have originated from biomass burning during the study period. The positive offset of OC ($\sim 11.6 \mu\text{g m}^{-3}$) as levoglucosan concentrations decreased to zero can be explained by direct emissions of OC from fossil fuel combustion, other primary organic aerosol sources, and secondary organic aerosol formation via a gas-to-particle conversion in the atmosphere (Jung et al., 2014). Moreover, because most of the OC was generated from biomass burning and levoglucosan is an organic compound, a strong correlation was observed for levoglucosan and OC. The EC values were moderately correlated with levoglucosan ($r = 0.597$) (Fig. 6d). This result agreed well with Jung et al. (2014), in which EC correlation with levoglucosan was 0.447. EC is inorganic and only a small portion of EC may be attributed to biomass burning. The larger portion may be emitted from other sources such as fossil fuel combustion.

Strong correlations between $PM_{2.5}$ and K^+ were obtained ($r = 0.763$). K^+ is often used as a tracer for biomass burning (Duan et al., 2004; Zhang et al., 2010; Lee et al., 2011; Cheng et al., 2013). Chantara et al. (2012) found strong correlations between K^+ and PM_{10} concentrations during the dry season (December–May) and the transition period (March–April) in Chiang Mai, Thailand. In this study, a strong correlation ($r = 0.819$) between biomass tracers (K^+ with levoglucosan) was found, which confirmed significant inputs from agricultural biomass combustion. Moreover, a moderate correlation between $PM_{2.5}$ and NO_3^- ($r = 0.659$) was obtained. Surprisingly, good correlations between NO_3^- and levoglucosan ($r = 0.738$) (Fig. 7a) and NO_3^- and K^+ ($r = 0.686$) (Fig. 7c), were also obtained, while no correlation was found between NO_3^- and SO_4^{2-} ($r = -0.020$) (Fig. 7e), although both are photochemical products in fine particulate matters (Kalabokas et al., 1999; Yao et al., 2016). The result was similar to a previous study (Khamkaew et al., 2016a) at the same sampling site in which a low correlation ($r = -0.242$) between NO_3^- and SO_4^{2-} was reported during a biomass burning period in 2014, revealing that NO_3^- was generated from biomass burning activity rather than fossil fuel combustion from traffic and related photochemical reactions.

Strong correlation was also found between SO_4^{2-} and NH_4^+ ($r = 0.954$), with an equivalent mole ratio of SO_4^{2-}/NH_4^+ equal to 1.02 ± 0.01 . The result agrees well with Khamkaew et al. (2016a), who reported high correlation ($r = 0.905$) between SO_4^{2-} and NH_4^+ . In general,

Table 3
Comparison of concentrations of PM, water-soluble ions and anhydrosugars in the present study with those reported in the literature.

| Location | Period | Site | PM | Concentration ($\mu\text{g m}^{-3}$) | | | | | | Levo/K ⁺ | Levo/Man | Ref. |
|-----------------------|------------------|-------------|-------------------|--|----------------|-----------------|------------------------------|-------------------------------|-------------|---------------------|-------------|-------------------------|
| | | | | PM | K ⁺ | Cl ⁻ | NO ₃ ⁻ | SO ₄ ²⁻ | Levo | Man | | |
| Chiang Mai, Thailand | March–April 2016 | CMU | PM _{2.5} | 64.3 ± 17.6 | 1.27 ± 0.38 | 0.14 ± 0.14 | 2.70 ± 0.51 | 8.73 ± 2.88 | 1.22 ± 0.75 | 0.07 ± 0.05 | 20.4 ± 4.13 | This study |
| Chiang Mai, Thailand | March–April 2014 | CMU | PM _{2.5} | 90.7 ± 27.9 | 2.06 ± 0.60 | 1.02 ± 0.42 | 2.65 ± 1.13 | 7.81 ± 2.60 | 1.13 ± 0.70 | | | Khamkaew et al. (2016a) |
| Chiang Mai, Thailand | Feb–April 2010 | Non-episode | PM _{2.5} | 80.8 ± 33.1 | 1.82 ± 0.68 | 1.02 ± 0.22 | 4.13 ± 2.86 | 6.20 ± 2.51 | 1.38 ± 0.60 | | | Tsai et al. (2013) |
| | | Episode | PM ₁₀ | 58.1 ± 30.9 | 0.89 ± 0.57 | 0.25 ± 0.18 | 1.22 ± 0.62 | 1.99 ± 1.46 | 0.33 ± 0.17 | 0.06 ± 0.02 | 5.73–7.69 | |
| | | AWB | PM ₁₀ | 139.6 ± 14.0 | 2.31 ± 0.56 | 0.59 ± 0.92 | 2.34 ± 1.95 | 3.86 ± 1.08 | 1.18 ± 0.80 | 0.08 ± 0.02 | 14.1–14.9 | |
| Taichung, Taiwan | | NAWB | PM _{2.5} | 12.4 ± 6.1 | 2.8 ± 1.6 | 3.1 ± 3.8 | 9.8 ± 6.0 | 16.9 ± 5.8 | | | | Cheng et al. (2009) |
| | | NAWB | PM _{2.5} | 32.6 ± 9.9 | 0.4 ± 0.2 | 0.3 ± 0.3 | 1.8 ± 1.8 | 5.7 ± 2.0 | | | | Zhang et al. (2010) |
| Guangzhou, China | July 2006 | Summer | PM ₁₀ | 87.0 ± 38.5 | 0.81 ± 0.60 | 1.94 ± 1.00 | 3.59 ± 1.50 | 21.4 ± 12.2 | | | | Nirmalkar et al. (2015) |
| Eastern central India | Oct–Nov, 2011 | | PM _{2.5} | 200 ± 127 | 10.2 ± 2.16 | | | | 2.26 ± 0.73 | | | Jung et al. (2014) |
| Daejeon, Korea | Oct–Dec, 2012 | Fall | | | 0.21 ± 0.08 | | | | 0.18 ± 0.11 | | | |
| Beijing, China | BB Episode | | | | | | | | | | | |
| | Typical summer | | | | | | | | | | | |
| | Typical winter | | | | | | | | | | | Cheng et al. (2013) |

Table 4
Correlation coefficients between concentrations of PM_{2.5} and associated chemical components.

| | PM _{2.5} | OC | EC | NH ₄ ⁺ | K ⁺ | Mg ²⁺ | Ca ²⁺ | Cl ⁻ | NO ₃ | SO ₄ ²⁻ | Oxalate | Levoglucon | Mannosan | Galactosan | Glucose | Mannose | Erythritol | Arabitol |
|-------------------------------|-------------------|--------------|--------------|------------------------------|----------------|------------------|------------------|-----------------|-----------------|-------------------------------|--------------|--------------|--------------|--------------|--------------|--------------|--------------|--------------|
| OC | 0.911 | | | | | | | | | | | | | | | | | |
| EC | 0.747 | 0.727 | | | | | | | | | | | | | | | | |
| NH ₄ ⁺ | 0.440 | 0.264 | 0.219 | | | | | | | | | | | | | | | |
| K ⁺ | 0.763 | 0.799 | 0.477 | 0.458 | | | | | | | | | | | | | | |
| Mg ²⁺ | 0.492 | 0.417 | 0.224 | 0.355 | 0.638 | | | | | | | | | | | | | |
| Ca ²⁺ | 0.579 | 0.544 | 0.325 | 0.304 | 0.726 | 0.756 | | | | | | | | | | | | |
| Cl ⁻ | -0.210 | -0.118 | -0.080 | -0.304 | -0.248 | 0.107 | 0.050 | | | | | | | | | | | |
| NO ₃ | 0.659 | 0.762 | 0.427 | 0.142 | 0.686 | 0.417 | 0.574 | 0.163 | | | | | | | | | | |
| SO ₄ ²⁻ | 0.235 | 0.057 | 0.027 | 0.954 | 0.303 | 0.393 | 0.493 | -0.006 | -0.020 | 0.011 | | | | | | | | |
| Oxalate | 0.799 | 0.797 | 0.743 | 0.254 | 0.579 | 0.393 | 0.477 | -0.038 | 0.738 | -0.035 | 0.615 | | | | | | | |
| Levoglucon | 0.800 | 0.914 | 0.597 | 0.125 | 0.819 | 0.477 | 0.527 | -0.010 | 0.742 | -0.002 | 0.609 | 0.985 | | | | | | |
| Mannosan | 0.790 | 0.912 | 0.558 | 0.148 | 0.821 | 0.496 | 0.525 | -0.104 | 0.730 | -0.030 | 0.770 | 0.869 | 0.856 | | | | | |
| Galactosan | 0.845 | 0.899 | 0.742 | 0.188 | 0.748 | 0.381 | 0.482 | -0.106 | 0.740 | -0.016 | 0.594 | 0.575 | 0.549 | 0.549 | | | | |
| Glucose | 0.618 | 0.587 | 0.724 | 0.153 | 0.438 | 0.202 | 0.292 | 0.106 | 0.470 | -0.055 | 0.613 | 0.966 | 0.976 | 0.516 | 0.528 | | | |
| Mannose | 0.753 | 0.884 | 0.518 | 0.093 | 0.781 | 0.457 | 0.518 | 0.045 | 0.774 | -0.052 | 0.457 | 0.465 | 0.871 | 0.871 | 0.549 | 0.453 | | |
| Erythritol | 0.504 | 0.571 | 0.542 | 0.364 | 0.399 | 0.221 | 0.194 | 0.008 | 0.290 | -0.052 | 0.597 | 0.832 | 0.976 | 0.822 | 0.549 | 0.837 | 0.513 | |
| Arabitol | 0.733 | 0.837 | 0.567 | 0.119 | 0.769 | 0.461 | 0.522 | -0.148 | 0.657 | -0.052 | 0.597 | 0.871 | 0.871 | 0.810 | 0.598 | 0.868 | 0.500 | |
| Mannitol | 0.799 | 0.863 | 0.621 | 0.196 | 0.762 | 0.429 | 0.503 | -0.155 | 0.647 | 0.023 | 0.597 | 0.906 | 0.891 | 0.810 | 0.598 | 0.868 | 0.500 | 0.909 |

Bold indicates correlation is significant at the 0.01 level (2-tailed)

Bold and Italic indicates correlation is significant at the 0.05 level (2-tailed)

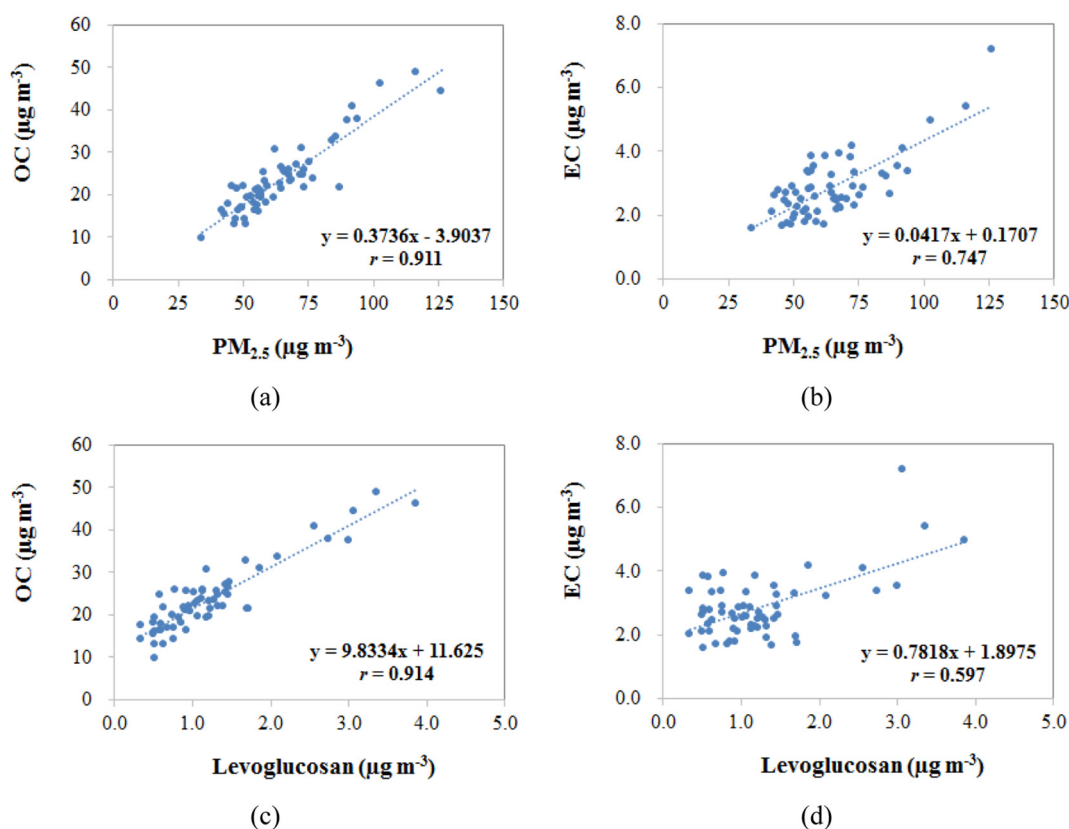


Fig. 6. Correlation between the concentration of OC and EC versus $\text{PM}_{2.5}$ and levoglucosan in $\text{PM}_{2.5}$ during smoke haze period in 2016.

a correlation between NH_4^+ and SO_4^{2-} is usually observed, due to their high affinity (Gupta et al., 2003; Tsai et al., 2013; Seinfeld and Pandis, 2016). NH_3 , the only common alkaline gas in the atmosphere, plays a major part as a neutralizing agent of atmospheric acids. This neutralization occurs predominantly in aerosols via a complicated system that includes ammonia, sulfuric acid, nitric acid and water (Gupta et al., 2003; Karar and Gupta, 2007; Seinfeld and Pandis, 2016). NH_3 is emitted from a large number of sources, such as volatilization from animal waste, agricultural processes including fertilizers and biomass and fossil fuel burning (Krupa, 2003). Chantara et al. (2012) reported that NH_3 was a major gas found in Chiang Mai, mainly emitted from agricultural activities.

Among carboxylate compounds, oxalate was found to be correlated with $\text{PM}_{2.5}$ ($r = 0.799$), OC ($r = 0.797$), and EC ($r = 0.743$). Oxalate, the smallest dicarboxylic acid, is present in atmospheric aerosol as direct emissions from biogenic sources, biomass burning, vehicles and motor exhaust (Chebbi and Carlier, 1996; Huang and Yu, 2007; Wang et al., 2007). Secondary formation of oxalate is from the end products of photochemical formation of organic species (Martinelango et al., 2007). In this study, a moderate correlation was found between oxalate and levoglucosan ($r = 0.615$) (Fig. 7b) and oxalate and K^+ ($r = 0.579$) (Fig. 7d), while no correlation was found between oxalate and SO_4^{2-} ($r = 0.011$) (Fig. 7f). This result implies that oxalate mainly originated from primary biomass burning emissions rather than secondary formation from photochemical processes.

All the anhydrosugars, sugars and sugar alcohols were correlated well. Levoglucosan had the highest concentration among anhydrosugar species and was highly correlated with its isomer, mannosan ($r = 0.985$) and galactosan ($r = 0.869$). The considerable amount of levoglucosan and its good correlation with $\text{PM}_{2.5}$ ($r = 0.800$) and K^+ ($r = 0.819$) suggested that biomass burning is a significant source of ambient aerosols over the region during the study period. A strong correlation (>0.80) was also found between levoglucosan and mannose, mannitol and arabitol (Table 4).

There was a moderate correlation between levoglucosan and erythritol ($r = 0.465$). As an indicator for soil microbial metabolism, this correlation verified that during the burning of agricultural wastes in the field, soil microbial metabolites were also burnt and released into the atmosphere. A strong correlation, $r = 0.917$, was reported in Taiwan during a period of moderate air quality by Tsai et al., 2015. They suggested that the aerosols produced by agricultural waste burning in China were transported to Tainan through air currents. The more moderate correlation in this study suggests that emission from forest fires was more significant than from agricultural waste burning.

3.4. The relationships between biomass burning tracers: levoglucosan, mannosan and K^+

Previous studies, listed in Table 3, have used the levoglucosan/potassium (Levo/ K^+) or levoglucosan/mannosan (Levo/Man) ratio to characterize particular types of biomass burning in atmospheric aerosols. A higher Levo/ K^+ ratio is usually found for forest fire aerosol, examples including 4.35–58.8 for fires in the US (Fine et al., 2002, 2004) and 0.38–22.0 in forest fire smoke studied by Ward et al. (2006). Because K^+ are contributed from potassium fertilizer and hence agricultural waste burning, a lower Levo/ K^+ ratio is usually connected with agricultural burning. Biomass burning aerosols were studied in Beijing by Cheng et al. (2013). They reported that the combustion of crop residuals, i.e. rice straw, wheat straw and corn straw, was significant and that the Levo/ K^+ ratio was lower during the typical summer period (averaging 0.21 ± 0.16) compared with the typical winter period (averaging 0.51 ± 0.15), while during the biomass burning period the Levo/ K^+ ratio averaged 0.11 ± 0.06 . Several studies of Asian crop residue burning have reported a Levo/ K^+ ratio in the range of 0.1–1.2 (Sheesley et al., 2003; Sullivan et al., 2008; Engling et al., 2009; Kim Oanh et al., 2011; Cheng et al., 2013). In a recent study conducted in the Chiang Mai basin, the Levo/ K^+ ratio during a PM_{10} episode occurring in the

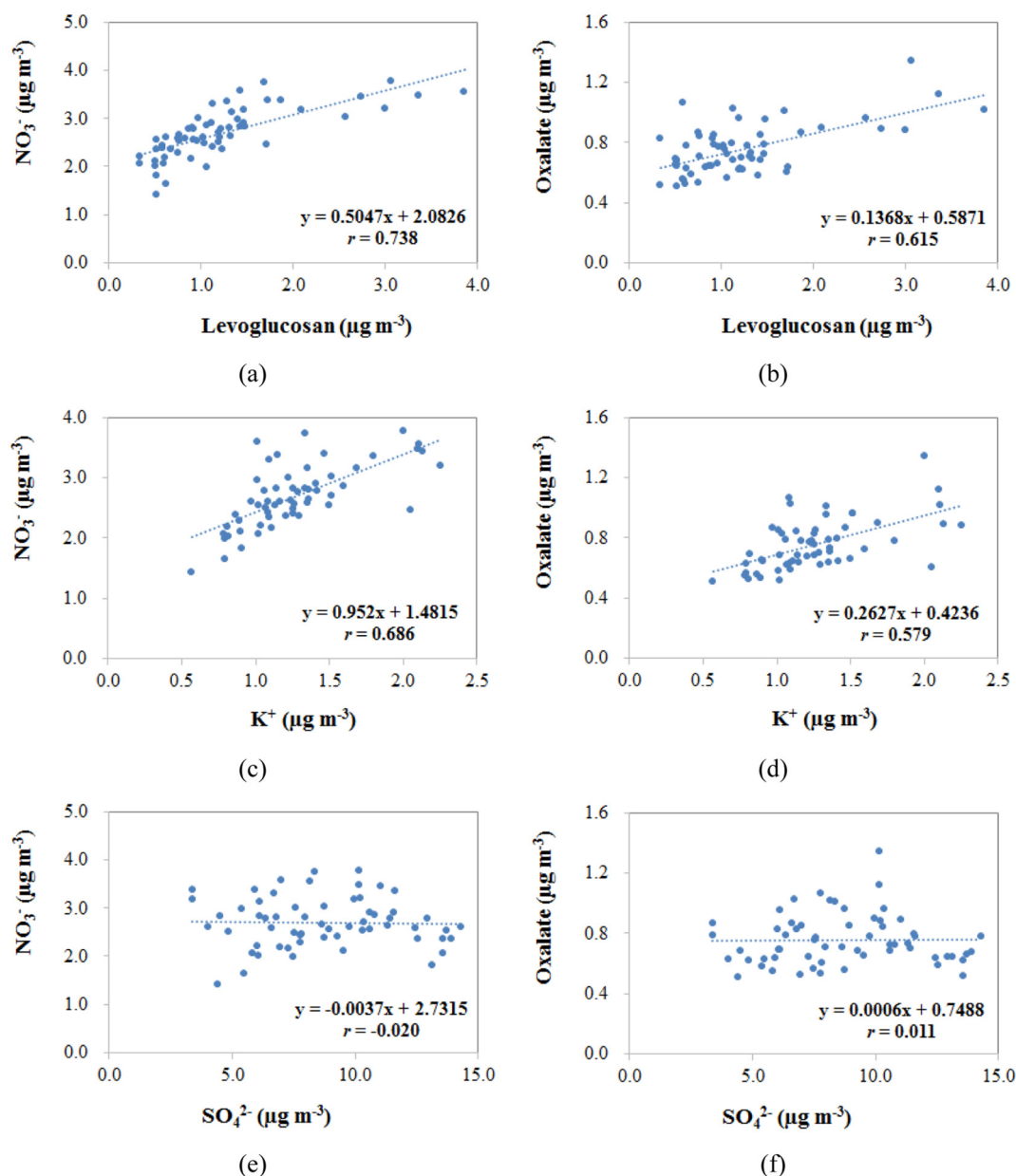


Fig. 7. Correlation between the concentration of NO_3^- , oxalate, levoglucosan, K^+ and SO_4^{2-} in $\text{PM}_{2.5}$ during smoke haze period in 2016.

dry season ranged from 0.51–0.56, due to the burning of crop residuals (Tsai et al., 2013). In the present study, the Levo/ K^+ ratio was 0.92 ± 0.35 (0.33–1.83), suggesting that the ambient environment of Chiang Mai during the dry season in 2016 was impacted by burning of forest combined with burning of agricultural crop residuals, i.e. rice straw, maize residue and sugar cane, in the field.

The ratio of Levo/Man in PM can be used to identify the relative contribution from hardwood or softwood burning. The Levo/Man ratios in the present study were in the range of 15.7–37.6 with a mean of 20.4 ± 4.1 , much higher than those obtained from softwood burning, (typically 2.6–5.0) (Schmidl et al., 2008a; Engling et al., 2009), and similar to those obtained from hardwood burning, (typically 13–32) (Fine et al., 2004; Engling et al., 2009), corn straw burning (19.5 ± 3.4) and Asian crop residue burning (12–55) (Sheesley et al., 2003; Sullivan et al., 2008; Engling et al., 2009; Kim Oanh et al., 2011; Cheng et al., 2013). The use of Levo/ K^+ and Levo/Man ratios to track possible sources of biomass burning, as suggested by Cheng et al. (2013), indicates that biomass-burning tracers from aerosols collected in this study could be from crop residues.

Fig. 8 shows a scatter plot of Levo/ K^+ and Levo/Man of $\text{PM}_{2.5}$ obtained from the burning of four biomass types collected in Northern Thailand and combusted in the laboratory in an open-system

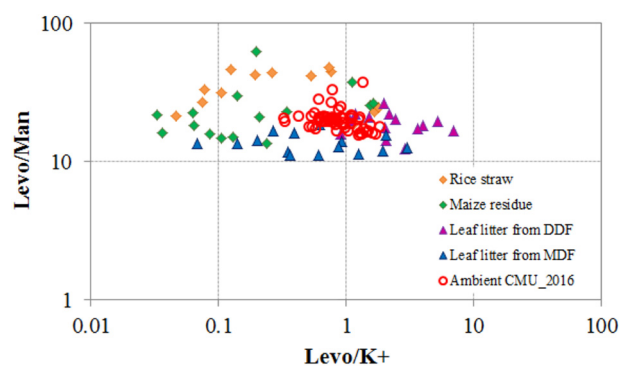


Fig. 8. Mass ratio of levoglucosan to K^+ versus mass ratio of levoglucosan to mannosan in the different types of biomass burning emission.

combustion chamber (Chantara et al., 2018) along with the Levo/ K^+ and Levo/Man ratios from the present study, indicated by red circles. The Levo/Man ratios were similar to those obtained from leaf litter of dry dipterocarp forest and from maize residue with a mean of 19.0 ± 3.5 and 24.2 ± 12.3 , respectively, while the Levo/ K^+ ratios were similar to those obtained from leaf litter of mixed deciduous forest, with a mean of 0.90 ± 0.88 . These results suggest an origin from forest fires and agricultural wastes burning.

3.5. Use of principal component analysis for air pollution source identification based on $PM_{2.5}$ and chemical compositions

The data sets of measured water-soluble ions, carboxylates, sugars, anhydrosugars and sugar alcohols in $PM_{2.5}$ aerosol samples were subjected to a factor analysis with Varimax rotation using SPSS to further investigate air pollution sources in the atmosphere of Chiang Mai, Thailand. Five principal components (PCs) were found to account for 84.2% of the variance in the data set of $PM_{2.5}$ samples (Table 5). PC-1 accounted for 45.8% of the variance with high loading of OC, K^+ , NO_3^- , anhydrosugar, dicarboxylate and sugar alcohol. It seems to be related mostly to aerosols from combustion sources. Levoglucosan and its isomer are considered as excellent tracers for biomass burning (Simoneit et al., 1999). Dicarboxylate also originates primarily from biomass burning aerosols (Narukawa et al., 1999; Falkovich et al., 2005; Decesari et al., 2006), while NO_3^- was reported as the most prominent component enhanced during biomass burning (Fabian et al., 2005; Lee et al., 2011). The high loading of these species, including arabitol, mannitol, mannose and glucose, in PC-1 indicated a biomass burning source. Moderate loading of erythritol was found in PC-1, revealing a contribution from soil microbial metabolites that is usually associated with emissions from biomass burning of agricultural waste (Tsai et al., 2015).

The high loading of succinate and malonate in PC-2 suggests sources from traffic-related emission. Moderate loading of EC was also found in PC-2, attributed to vehicle exhaust emission. The loading of SO_4^{2-} in PC-3 suggests sources from the photochemical formation of SO_2 from both traffic/industrial activity and biomass burning. Because of the high affinity of SO_4^{2-} to NH_4^+ , a high loading of NH_4^+ was also obtained in PC-3. The high loading of Mg^{2+} and Ca^{2+} in PC-4 suggests an origin from

crustal matter as resuspension dust. Although Cl^- occurs as the only high loading in PC-5, there is no clear evidence suggestive of a specific source for this pollutant. Moreover, Na^+ analysis was not possible due to its high concentration in blank filters and we, therefore, cannot discuss any potential contribution from sea salts.

3.6. $PM_{2.5}$ and chemical concentrations associated with clustering trajectories

Average 48-hour BWT to CMU in March–April 2016 (60 days) were calculated to classify air mass movements. BWT calculated at 0:00, 6:00, 12:00 and 18:00 UTC for each day gave 4 arriving times \times 60 days = 240 trajectories. These were grouped into three clusters based on their major air mass directions, as shown in Fig. 9a. The main air mass movement to the CMU site originated from northern India and passed over the Bay of Bengal and Myanmar through Chiang Mai in a northwest direction (cluster 1; 37.3%). Approximately 34.8% originated from the Indian Ocean and moved directly east passing the Myanmar continental area before arriving at CMU from the west (cluster 2). About 27.9% was a short distance movement (cluster 3) from the southwest near the border between Thailand and Myanmar. Both cluster 1 and 2 paths represented long-range transport of air mass whereas cluster 3 was a regional air mass. The results of the present study were in good agreement with a previous study (Chantara et al., 2012) conducted at the same study site, in which it was reported that the main air mass of 3-day backward trajectories in the dry season from 2005 to 2009 came from the southwest of Chiang Mai. These results show that Chiang Mai aerosols during the study period were influenced by activities to the southwest through the Indian continental area. Maps of monthly hotspots together with clustering BWT are shown for March and April 2016 in Fig. 9b and c. The major air mass movements were similar for both months and were from the west of Chiang Mai.

Table 6 shows the mean concentrations of $PM_{2.5}$ and their chemical compositions associated with each of the three air mass clusters derived from data shown in Fig. 9a. It was found that there was no significant difference between clusters for observed $PM_{2.5}$, EC and most of the ions, but there was a significant difference ($p < 0.05$) between clusters for observed OC, NO_3^- , levoglucosan, mannosan, succinate and malonate. The concentration of these species, with the exception of succinate and malonate, was low and approximately equal in clusters 2 and 3, but higher in cluster 1. This implies that the high concentration of biomass burning tracers in Chiang Mai aerosols came from long-range transport of air mass from India and Myanmar to the west and southwest of the CMU site. Succinate and malonate, noted as traffic-related emission species, were observed in the highest concentration in cluster 3, the regional air mass. It has been noted that the sampling site is located in an urban area with potential impacts from traffic and industrial activity. The air cluster results suggest that in addition to long-range transport, local traffic and industrial sources also contributed to the concentration of ambient aerosols in Chiang Mai.

4. Conclusions

During a two-month biomass burning period, >80% of the 24-h $PM_{2.5}$ levels in Chiang Mai exceeded the Thailand National Ambient Air Quality Standard ($50 \mu g m^{-3}$). OC was the largest component of $PM_{2.5}$ and the dominant fraction of OC3 and OC4. The OC/EC ratio indicated that OC was mainly from biomass burning. Water-soluble ions were the second most abundant compound in $PM_{2.5}$, with SO_4^{2-} , NH_4^+ , NO_3^- and K^+ the major species. The source identification of chemical species was mainly based on their correlation coefficients under a single component obtained from PCA. A high correlation between SO_4^{2-} and NH_4^+ confirmed the secondary formation of these species. A good correlation between NO_3^- and biomass burning tracers (K^+ and levoglucosan) confirmed that NO_3^- was generated from biomass burning activity rather than fossil fuel combustion emitted from traffic and/or related

Table 5
Principal component analysis (PCA) for measured chemical species in $PM_{2.5}$ at CMU site.

| Species | PC-1 | PC-2 | PC-3 | PC-4 | PC-5 |
|---------------------------|--------------|--------------|--------------|--------------|--------------|
| OC | 0.944 | 0.079 | 0.120 | 0.195 | 0.020 |
| EC | 0.698 | 0.565 | 0.057 | 0.033 | 0.068 |
| NH_4^+ | 0.117 | 0.163 | 0.901 | 0.214 | −0.170 |
| K^+ | 0.756 | −0.098 | 0.341 | 0.382 | −0.202 |
| Mg^{2+} | 0.282 | −0.105 | 0.353 | 0.733 | 0.250 |
| Ca^{2+} | 0.512 | 0.089 | 0.261 | 0.692 | −0.033 |
| Cl^- | −0.018 | 0.081 | −0.222 | 0.138 | 0.926 |
| NO_3^- | 0.761 | −0.027 | −0.009 | 0.427 | 0.056 |
| SO_4^{2-} | −0.077 | 0.048 | 0.947 | 0.130 | −0.077 |
| Lactate | 0.363 | 0.759 | −0.228 | 0.021 | −0.060 |
| Succinate | −0.102 | 0.746 | 0.274 | −0.085 | 0.108 |
| Malonate | −0.040 | 0.897 | 0.128 | 0.096 | 0.034 |
| Maleate | 0.714 | 0.218 | −0.269 | 0.295 | 0.004 |
| Fumarate | 0.705 | 0.324 | −0.127 | 0.435 | −0.032 |
| Oxalate | 0.682 | 0.378 | 0.008 | 0.445 | 0.050 |
| Levoglucosan | 0.961 | −0.065 | 0.060 | 0.117 | 0.030 |
| Mannosan | 0.955 | −0.086 | 0.107 | 0.133 | 0.094 |
| Galactosan | 0.897 | 0.181 | 0.024 | 0.182 | −0.024 |
| Erythritol | 0.561 | 0.281 | 0.449 | −0.214 | 0.409 |
| Arabitol | 0.930 | 0.076 | −0.050 | 0.117 | −0.051 |
| Mannitol | 0.926 | 0.127 | 0.061 | 0.067 | −0.067 |
| Mannose | 0.937 | −0.078 | 0.031 | 0.165 | 0.106 |
| Glucose | 0.693 | 0.416 | 0.000 | −0.031 | 0.087 |
| Eigenvalues | 10.5 | 2.9 | 2.5 | 2.1 | 1.2 |
| Cumulative eigenvalues | 10.5 | 13.5 | 16.0 | 18.1 | 19.4 |
| % of variances | 45.8 | 12.8 | 11.0 | 9.2 | 5.3 |
| Cumulative % of variances | 45.8 | 58.6 | 69.7 | 78.9 | 84.2 |

Bold indicates species loading >0.70.

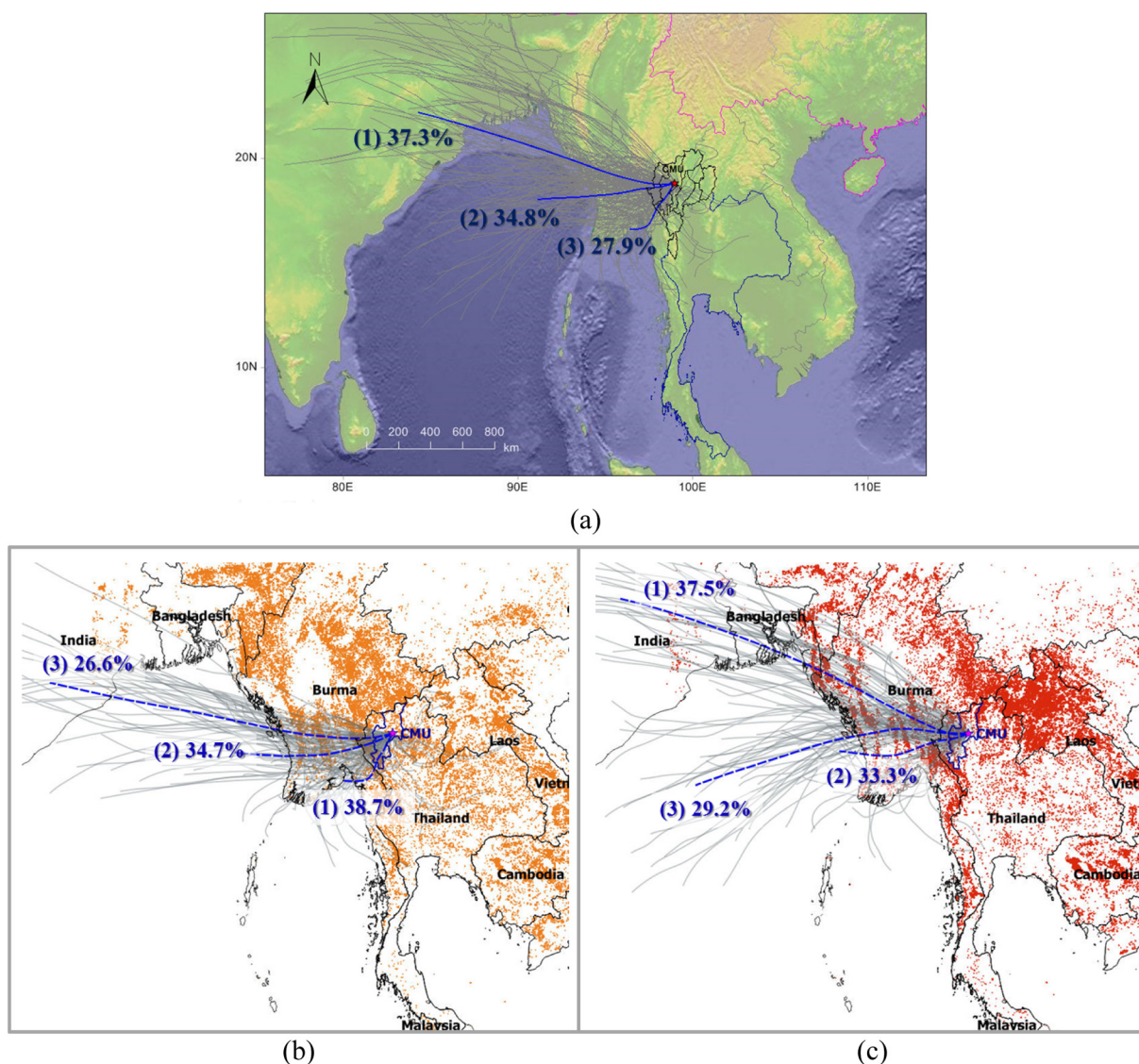


Fig. 9. Hotspots and clusters of 48-hour BWT arriving at 1500 m AGL (endpoints: 00, 06, 12 and 18 UTC) at CMU site during (a) March–April 2016, (b) March 2016 and (c) April 2016.

photochemical reactions. A high concentration of oxalate was observed and its correlations with OC and biomass burning tracers were strong. This data indicated that oxalate mainly originated from primary

Table 6

Concentrations of PM_{2.5}, OC, EC and water-soluble ions, carboxylates and anhydrosugars classified by clusters of air mass movements.

| Parameters | Concentration ($\mu\text{g m}^{-3}$) | | |
|-------------------------------|--|---------------------------------|--------------------------------|
| | Cluster 1 | Cluster 2 | Cluster 3 |
| PM _{2.5} | 71.0 \pm 19.9 ^a | 59.8 \pm 9.8 ^a | 61.7 \pm 19.9 ^a |
| OC | 27.2 \pm 9.5 ^b | 22.0 \pm 4.3 ^{ab} | 21.6 \pm 8.6 ^a |
| EC | 2.95 \pm 1.02 ^a | 2.64 \pm 0.61 ^a | 2.96 \pm 1.25 ^a |
| NH ₄ ⁺ | 3.44 \pm 0.84 ^a | 3.33 \pm 1.01 ^a | 3.19 \pm 1.19 ^a |
| K ⁺ | 1.39 \pm 0.43 ^a | 1.24 \pm 0.33 ^a | 1.15 \pm 0.33 ^a |
| NO ₃ ⁻ | 2.89 \pm 0.49 ^b | 2.65 \pm 0.42 ^{ab} | 2.54 \pm 0.58 ^a |
| SO ₄ ²⁻ | 9.14 \pm 2.27 ^a | 8.87 \pm 3.08 ^a | 8.12 \pm 3.28 ^a |
| Succinate | 0.038 \pm 0.038 ^{ab} | 0.037 \pm 0.046 ^a | 0.080 \pm 0.085 ^b |
| Malonate | 0.050 \pm 0.020 ^{ab} | 0.049 \pm 0.028 ^a | 0.073 \pm 0.036 ^b |
| Levoglucosan | 1.56 \pm 0.95 ^b | 1.06 \pm 0.37 ^{ab} | 1.02 \pm 0.70 ^a |
| Mannosan | 0.088 \pm 0.063 ^b | 0.055 \pm 0.024 ^{ab} | 0.051 \pm 0.040 ^a |
| Galactosan | 0.016 \pm 0.009 ^a | 0.012 \pm 0.005 ^a | 0.014 \pm 0.010 ^a |

^{a,b,c} Same superscripts express similarity ($p > 0.05$) between means of the individual parameter for three clusters. Different superscripts show a significant difference ($p < 0.05$) between means of the individual parameter for three clusters.

biomass burning emissions rather than secondary formation from photochemical processes. The presence of succinate and malonate confirmed that traffic-related emission sources also contributed to aerosol in this area.

Levoglucosan, the biomass tracer, was the most abundant compound among anhydrosugar species. It was strongly correlated with PM_{2.5}, OC and K⁺ during the study period, indicating that the contribution of aerosols from biomass burning was greater than other sources during this smoke-haze period and that the biomass burning was specifically forest fires and agricultural waste burning. This was verified by Levo/K⁺ and Levo/Man ratios.

Investigation of BWT of air mass movement at the CMU site indicated that the major air movement path originated from India and Myanmar from the west and southwest. Moreover, the open burning in the border area between Thailand and Myanmar also contributed to air quality in Northern Thailand.

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