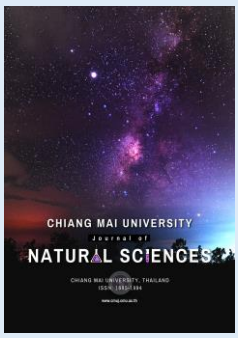

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Research article

Significant Contribution of C_3 - Type Forest Plants' Burning to Airborne $PM_{2.5}$ Pollutions in Chiang Mai Province, Northern Thailand

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Abstract This study aimed to identify the contributing sources of $PM_{2.5}$ in Chiang Mai Province during February to April 2016. We therefore investigated the chemical compositions of $PM_{2.5}$ at two different sites. An urban site is in Chiang Mai University (CMU) while a rural site is in Mae Chaem (MC) District where frequent intensive biomass burning was reported. Thirty pair samples of 24-h $PM_{2.5}$ were analyzed for organic carbon (OC), elemental carbon (EC), levoglucosan and stable carbon isotope ($\delta^{13}C$). The mean concentrations (Mean ± SD) of $PM_{2.5}$, OC and EC at the CMU vs MC sites were not significant different (P >0.05) including 44.5 ± 32.1 vs 40.5 ± 21.2 μg/m^3; 14.9 ± 12.5 vs 14.8 ± 10.0 μg/m^3; and 1.80 ± 1.60 vs 1.62 ± 0.80 μg/m^3, respectively. Levoglucosan concentrations, a tracer of biomass burning from both sites were not significant different (P >0.05) and the mean ± SD concentrations at CMU vs MC sites were 0.46 ± 0.56 μg/m^3 vs 0.55 ± 0.67 μg/m^3, respectively. Meanwhile, the mean values of δ13C in total carbon (TC) at CMU vs MC sites were -27.9 ± 0.68 vs -27.6 ± 0.60‰, respectively which major data (n = 48, 85.4%) fell within the ranged of C_3-type plants and minor data (n = 48, 14.6%) in C_3-type plants and motor vehicle sources. This finding corresponds to the vast biomass burning area from satellite data. Forest plants in northern Thailand, Chiang Mai particular are mostly mixed deciduous forest i.e. C_3-type plants which falling leaves in dry season and easily causing fire. The results of this study therefore strongly suggest that the burning of C_3-type forest plants attribute to airborne $PM_{2.5}$ pollutants in Chiang Mai Province.

Keywords: Air pollution, Biomass burning, C3-type plants, Levoglucosan, Stable carbon isotope

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INTRODUCTION

Keywords: Since 2007, air pollution has become a serious problem in upper northern Thailand. The major sources of air pollution in this area are mostly from open burning of agricultural waste and forest fires from February to April (Pengchai et al., 2008; Chantara et al., 2012; Chuang et al., 2013; Pani et al., 2019; Punsompong and Chantara, 2019). Chiang Mai Province is in the Chiang Mai-Lamphun Basin which is a flat plain surrounded by high mountain ranges. Forest area data from the Royal Forest Department indicate that forested areas are abundant, consisting of mixed deciduous, deciduous dipterocarp, dry evergreen, and hill evergreen forests. The geographical feature of high mountains surrounding a large valley basin often results in limited ventilation and dispersion of air pollution. Biomass burning (BB) can either occur naturally or from human activity. Open burning of forest fires and grassland, agriculture residue and residential combustion of biofuel for cooking is a significant contributor to trace gases and particulate matter (PM) in the troposphere (Chang et al., 2015). In this area, high number of hotspots were reported and the dominant sources of $PM_{2.5}$ were contributed from the biomass burning especially in dry season. According to a previous study in Chiang Mai, the primary sources of air pollution in this area indicated biomass burning emissions (agricultural and forest fires), as well as transportation emissions (Pongpiachan, Pongnailert, Ho, & Cao, 2014; Janta et al., 2019; ChooChuay et al., 2020)

In Southeast Asia, the main source of biomass burning includes forest fires and burning of agricultural waste products (Yadav et al., 2017). A previous study found that large amounts of fine PM or $PM_{2.5}$ (particulate matter with an aerodynamic diameter less than 2.5 μm) in Chiang Mai were emitted from biomass burning, with an average ambient $PM_{2.5}$ concentration of 45.5 ± 8.8 μg/m^3 (Chuang et al., 2013). The daily average values of $PM_{2.5}$ in ambient air in Chiang Mai Province have exceeded Thailand's national ambient air quality standard of 50 μg/m^3 during 2014 - 2016. Khamkaew et al., (2016) reported that the mean $PM_{2.5}$ concentrations collected at Chiang Mai University (CMU) between March and April 2014 were largely attributed to local open burning of agricultural matter and forest fires (Khamkaew et al., 2016). Air pollution in Chiang Mai City studied in the dry season of 2010 suggested that open burning of plants was the predominant contributor to air pollution in this area and was found to be highly correlated with element concentrations in ambient PM10 (particulate matter with an aerodynamic diameter less than 10 μm) and the combustion of teak, yangna, and corn stalks (Kiatwattanacharoen et al., 2017). Mae Chaem (MC) is a rural district in Chiang Mai Province and the terrain consists of mostly foothills and mountain ridges surrounding a small basin approximately 350 m above mean sea level. MC has had the highest number of hotspots and smoke haze pollution during the dry season for more than a decade (Arunrat et al., 2018).

Prapamontol and colleagues reported that in February to March in 2012, the maximum ambient 24-hour average PM10 in MC was 191.5 μg/m^3, about 1.5 times higher than the standard PM_{10}, 24-h average, in Thailand (120 μg/m^3). Moreover, particles of $PM_{2.5}$ often contain a variety of chemical species, organic carbon (OC) and elemental carbon (EC) and numbers of studies regarding the chemical characterization of fine particles were carried out in the southern part of Thailand such as in Hat Yai city (Pongpiachan, 2014; Pongpiachan, Pongnailert, Ho, & Cao, 2014). Carbonaceous particles emitted from biomass burning can be categorized into elemental carbon (EC) and organic carbon (OC) based on their thermal, chemical, and optical properties (Nunes and Pio, 1993; Saarikoski et al., 2008). In general, EC is released from primary combustion, emitting directly from incomplete combustion of fossil combustion (coal, fuel oil and petrol) and biomass burning, agricultural residue, and forest fire (Schwarz et al., 2008). EC is referred to as black carbon (BC) aerosol, and arises from incomplete combustion of biomass, motor vehicle fuel and residential coal (Watson, 2002). Thus, EC is frequently used as a primary tracer element due to its inert physiochemical properties in the atmosphere. In contrast, OC is the result of secondary combustion and is comprised of a complex mixture of many compounds including primary source emissions (biogenic source, biomass burning, traffic, cooking and industry) and

secondary organic carbon (SOC) which is formed by the atmospheric oxidation of gaseous precursors. OC usually consists of a mixture of many organic compounds, such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzo-d-dioxin and dibenzofuran (PCDD/F), which have mutagenic and/or carcinogenic characteristics (Feng et al., 2009; Kanakidou et al., 2012). Levoglucosan (1,6-anhydro-β-D-glucopyranose) is a tracer of biomass burning and one of the most studied sugar compounds produced by thermal decomposition of cellulose and hemicellulose. It is emitted during the combustion of biomass such as that used in residential heating and wild/agriculture fires and has been a useful molecular marker of biomass (Simoneit et al., 1999; Křůmal et al., 2015). Recent studies (Hennigan et al., 2010; Hu et al., 2013; Zhao et al., 2014) have suggested that levoglucosan is prone to degradation by OH radicals in the atmosphere. Thus, its concentration during long-range transport may be underestimated, especially in summer (Fraser and Lakshmanan, 2000; Simoneit, 2002; Simoneit et al., 2004, 1999). Detailed characterization of the proportional yield of levoglucosan to its isomers and particularly to mannosan in fuel source emissions has further permitted discrimination by specific types of fuel combustion (Ward et al., 2006). In addition, studies of stable carbon isotopes in atmospheric particulate matter are potentially useful for identifying sources of carbonaceous particles and this approach may be a useful new tool for studies of air pollution composition (Wang et al., 2013; Mkoma et al., 2014; Bikkina et al., 2016). Stable carbon isotopes ($\delta^{13}C$) measurement values can be used to trace carbon sources in atmospheric studies by taking advantage of the different stable carbon isotope ratios of C_4- and C_3- type plants (Kawashima and Haneishi, 2012; Cao et al., 2017). Due to their different photosynthetic pathways, C_4-type plants, such as corn and warm-climate grasses, are enriched in stable carbon isotopes compared to C_3-type plants, which include most other plants, trees, and cool-climate grasses (DeNiro and Epstein, 1978). Kawashima and Haneishi (2012) employed stable carbon isotopes ($\delta^{13}C$) measurement to study aerosol carbon from various sources such as gasoline and diesel vehicle exhaust, fireplace soot, open biomass burning emissions, street dust, soils, charcoal, and coal. They found that $\delta^{13}C$ values of fuel combustion ranged from −20.6‰ to −20.5‰, whereas, the values obtained from C_3-type plants (-34.7‰ to -28.0‰) were lighter than obtained from C_4-type plants (-19.3‰ to -16.1‰) (Kawashima and Haneishi, 2012).

This study aimed to identify the contributing sources of airborne particulate matter with an aerodynamic diameter less than 2.5 μm ($PM_{2.5}$) in Chiang Mai Province where severe smoke-haze pollution occurred almost every dry season (February to April). So, we determined chemical compositions including carbonaceous carbons (EC and OC), levoglucosan and stable carbon isotopes in ambient $PM_{2.5}$ samples collected from urban and rural sites in Chiang Mai Province during February to August 2016. The chemical compositions were then used to characterize and identify sources contributing to ambient $PM_{2.5}$ in Chiang Mai Province.

MATERIALS AND METHODS

Sampling sits and sample collection

The study had two sampling sites located in Chiang Mai Province as shown in Figure 1. The first sampling site was in urban area located on the rooftop of the four–story building at the Research Institute for Health Science (RIHES), Chiang Mai University (called CMU site; 18°47'43.63''N, 98°57'28.17''E, 331 m mean sea level; MSL). The second sampling site was a rural area located in Mae Chaem District, where PM2.5 was collected at the Debaratana Vejjanukula Hospital (MC site; 18°29'52.94"N, 98°22'45.38"E 535 m MSL). $PM_{2.5}$ samples were collected from February to August 2016 using MiniVol air samplers (Air metric, USA) with a flow rate of 5 L/min. $PM_{2.5}$ samples were collected for 24 hours starting at 9.00 a.m., on quartz fiber filters (Whatman's, UK, Ø 47 mm). The filters were stored in desiccators filled with silica gel before and after sampling for 24 hours prior to being weighed using a 5-place microbalance (Mettler Toledo, AB135-S/FACT, Switzerland). Each filter was weighed three times in a controlled room (25 ± 2°C and 40 ± 5% RH). The collected filters

were covered with aluminum foil to protect the samples from sunlight and kept in a -20 °C freezer until analysis.

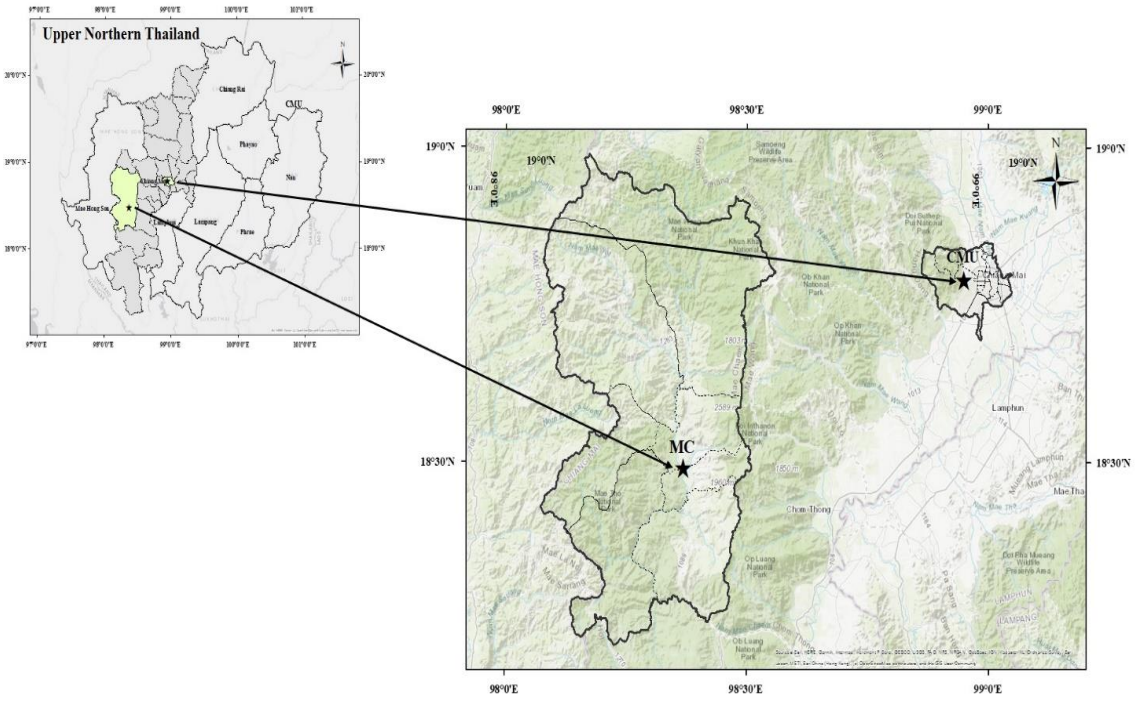

Figure 1. The location of CMU and MC sites.

Carbonaceous aerosol analysis

The concentration of OC and EC on $PM_{2.5}$ samples were analyzed using a thermal optical transitions OC/EC analyzer (Sunset Laboratory, Model - 4, USA) using the thermal-optical transmittance (TOT) method and applying the NIOSH (National Institute for Occupational Safety and Health) 5040 protocol, with improvements from Chow et. al., 2007 (Chow et al., 2007) at Yale-NUIST Center on Atmospheric Environment, Nanjing University of Information Science and Technology (NUIST), Nanjing, China. Briefly, an aliquot of quartz filter disks with a 17 mm diameter were packed in a quartz tube inside the thermal desorption chamber. The pyrolysis products were converted to carbon dioxide (CO_2) by a redox reaction with manganese dioxide. The CO_2 was quantified using a self-contained nondispersive infrared (NDIR) system. At the end of each analysis, a fixed volume of an internal standard containing 5% methane and 95% helium was injected and thus a known carbon mass could be derived. An external sucrose standard (4.2 µg/L) calibration was conducted every week to insure repeatable quantification. Calibration with an instrument blank was conducted every day. Both detection limits for OC and EC of the instrument was 0.5 µg/m^3. A duplicate was run to test for analytical errors in the precision of the measurement. The results showed good reproducibility (reported as percentage relative standard deviation; % RSD), from duplicate analysis of the filter samples at less than 8% for OC and 15% for EC. The sample results were corrected by blank values.

Levoglucosan analysis

Levoglucosan analysis was performed at the Yale-NUIST Center. A filter samples punch 17 mm in diameter was placed in a high-density polyethylene (HDPE) bottle with 3 mL of deionized water and sonicated (PS-D40, China) for 30 minutes at controlled temperature (~10 ºC). Using ice to extract the target compounds, the filter was removed and the extract was then filtered through a 0.22 µm polyether sulfone

(PES) and the aqueous filtrate was characterized by a modified IC method based on Hsieh et al., (2008) and Tsai et al., (2010). The IC (DX-5000+, Dionex) was equipped with pulsed amperometry detection (PAD). The gradient pump (Model GP 5000, a spectra system automated sampler (AS 5000) with 5 mL vials, used a CarboPac MA1 analytical column (250 mm × 4 mm I.D.), and sodium hydroxide solution (480 mM, 0.4 mL/min) as eluent.

Stable carbon isotope analysis

The stable carbon isotopes in $PM_{2.5}$ were determined using an elemental analyzer (EA) coupled with an isotope ratio mass spectrometer (IRMS, Finnigan MAT Delta Plus) at the Yale-NUIST Center on Atmospheric Environment, (Jung and Kawamura, 2011). A filter disk of 14 mm diameter was packed in a tin cup, loaded into the EA by an auto-sampler and then oxidized by chromium (III) oxide at 1020°C. The resulting CO_2 was purified by an online GC column equipped in the EA and then measured with a thermal conductivity detector. A small aliquot of CO_2 gas was introduced to the IRMS through an interface ConFlo II (Thermo Quest), the carbon isotopic composition was expressed as $\delta^{13}C$, which is relative to the Pee Dee Belemnite (PDB). External calibration was conducted using five known amounts (ranging from 0.2 to 0.6 mg) of acetanilide (Thermo Scientific, USA) with known $\delta^{13}C$ of TC (-27.26‰.). The analytical errors of $\delta^{13}C$ based on the duplicate analyses were less than 0.06 ‰. TC concentrations measured with EA agree well (P <0.01). The $\delta^{13}C$ value in a sample was expressed on a per mill (‰, that is, parts per thousand) basis. The isotope ratio of a sample (R_{sample}) was compared with a standard ($R_{standard}$) as follows:

$$\delta^{13}C\ (‰) = (R_{sample}/ R_{standard} - 1) \times 1{,}000 \quad (1)$$

where R is $^{13}C/^{12}C$. Pee Dee Belemnite (PDB) was used as the standard.

Data Analysis

All data from this study were analyzed using the SPSS statistical program. The average value was expressed as Mean ± Standard deviation (SD). T-tests were used for comparisons of pollutants between the two sites and correlation coefficients (r) were calculated. Linear regression analysis was performed in identifying the associations of parameters. The Moderate Resolution Imaging Spectroradiometer (MODIS) on board NASA's Aqua and Terra satellites were used to calculate the number of hotspots during the intensive biomass burning season.

Backward Trajectories Analysis

Three-day back trajectories in samples from the CMU and MC study sites were calculated using the National Oceanic and Atmospheric Administration's Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model version 4 (Draxler & Hess, 1998). Backward trajectories were calculated every day from February to May 2016 starting at 00, 06, 12, and 18 UTC at altitudes 100 m above ground level (AGL) and only one at which the air arrives at the sites. The selection of 100 m arriving height as the lowest level resulted from the orography around the sites which is surrounded by mountain and forest. The clustered to the characterized distribution of fire directions and orientation arriving at the sampling receptors.

RESULTS

General results of $PM_{2.5}$, OC, and EC

The average concentrations of $PM_{2.5}$ at the CMU and MC sites were 44.5 ± 32.1 μg/m^3 and 40.3 ± 21.2 μg/m^3, respectively, which were not significantly different (P >0.05). The highest number of hotspots, 120, was observed on April 18, coinciding with the highest concentration of $PM_{2.5}$ at the CMU site, 129.3 μg/m^3. These

high concentrations were well in accord with the number of hotspots detected in Chiang Mai Province in March and April 2016. Meanwhile $PM_{2.5}$ level was relatively low at MC site at the same day. There are several reasons toward this phenomenon. Chiang Mai Province locates in upper northern part of Thailand and composes of 9 provinces (about 102,259 Square km in total) and mountainous topography. CMU site was designed as a representative Chiang Mai City area which locates in Chiang Mai - Lamphun Basin (large basin approximate ranged 174 km long) while MC site locates in Mae Chaem Basin (approximate ranged 7 km long and about 120 km south-west direction of Chiang Mai City. Moreover, the frequency of biomass burning was also different. Besides forest fire, it is common practices of getting rid of garbage in the rural community i.e. MC site. Furthermore, there are few more reasons for the difference of $PM_{2.5}$ levels such as the meteorological parameters i.e. wind direction, wind speed, rain fall etc. In addition, the dominant of wind direction from southwest direction which passed through the MC site. This phenomenon might disperse $PM_{2.5}$ as well as other pollutants away. The 24-hour average concentrations of OC in $PM_{2.5}$ at the CMU and MC sites during the sampling period were 14.6 and 14.3 μg/m^3, respectively. Meanwhile, the average concentrations of EC at the CMU and MC sites were 1.8 and 1.7 μg/m^3, respectively (Table 1).

Table 1. The concentrations of $PM_{2.5}$, OC, EC, TC, levoglucosan, stable carbon isotope and the ratio of OC/EC, levoglucosan/OC at CMU and MC sites.

Parameter	CMU site				MC site				*P value*
	Min.	Max.	Mean	S.D.	Min.	Max.	Mean	S.D.	
Concentrations (μg/m^3)									
$PM_{2.5}$	7.80	129.30	44.50	32.10	11.90	83.70	40.30	21.20	0.557
OC	0.50	46.10	14.60	11.90	0.84	43.0	14.30	10.20	0.964
EC	0.15	6.81	1.80	1.61	0.33	3.60	1.66	0.76	0.774
TC	0.50	52.9	15.80	13.30	0.84	46.70	15.30	11.20	0.073
Levoglucosan	0.02	2.35	0.46	0.56	0.12	2.34	0.55	0.67	0.565
$\delta^{13}C$ (‰)	-29.30	-26.40	-27.90	0.67	-28.80	-25.80	-27.60	0.60	0.067
OC/EC ratio	6.77	34.00	15.80	8.01	8.26	27.70	13.50	5.22	0.995
Levoglucosan/OC (%)	0.29	6.37	2.67	1.59	0.16	9.02	3.15	2.26	0.304

In addition, the moderate correlations coefficient (r) between TC and $PM_{2.5}$ mass concentrations of 0.67 and 0.57 (P <0.001) were observed at the CMU and MC sites, respectively, suggesting TC and $PM_{2.5}$ have similar sources and formation processes, allowing researchers to predict that the TC and $PM_{2.5}$ possibly originated from same biomass-type burning which came from local emission and transportation sources. Moreover, OC was significantly correlated with levoglucosan (Figure 3.) suggesting that biomass burning emissions contributed to carbonaceous aerosols (Cao, Zhang, Kawamura, & Zhang, 2016). The average values of OC/EC ratios were 15.8 at the CMU and 13.5 at the MC sites, indicating that biomass burning was the likely source of carbonaceous species. Previous studies have shown that high OC/EC ratios were related to biomass burning, with a ratio of OC/EC 14.5 for forest fires (Watson, Chow, & Houck, 2001) and 15.7 for rice straw burning (Engling, Lee, Sie, Wu, & I, 2013). The values of the OC/EC ratio for fossil fuel combustion, mainly vehicular emission was 2.9 ± 0.5 (Safai, Raju, Rao, & Pandithurai, 2014), 1.88 ± 0.24 (Panda et al., 2016) and 0.71 (Saarikoski et al., 2008). A significant correlation coefficients found between OC and EC for the whole period for the CMU and MC sites were 0.876 and 0.832 (P <0.001), and this finding suggested that a large fraction of OC and EC was emitted from the same biomass sources in atmospheric $PM_{2.5}$ (B. Huang et al., 2013; Qi et al., 2018). More specifically, we employed the stable carbon isotopes to clarify emission sources of $PM_{2.5}$. Range for $\delta^{13}C$ were -29.3 to -26.4‰ and -28.8 to -25.8‰ at CMU

and MC, respectively. The mean value of $\delta^{13}C$ was -27.9 ± 0.67‰ at CMU and -27.6 ± 0.60‰ at MC, indicating dominant contribution from combustion of C_3 plants such as rice residues, teak, and grass. It confirmed that the sources of aerosol $PM_{2.5}$ were more influenced by forest fire burning and agricultural residues. C_4 plants burning such as wheat straw as shown in secondary emission sources. C_4 plants such as corn and warm - climate grasses, are enriched in stable carbon isotopes compared to C_3 plants, which include most other plants, trees, and cool-climate grasses. These results were similar to previous study, Cao et al., 2016 have been reported the average $\delta^{13}C$ value (−26.2‰) from biomass burning aerosols in Northeast China was dominant contribution from combustion of C_3 plants. Additionally, the PM ranges for $\delta^{13}C$ values from biomass components burning showed sources differed signally. For C_3 plants burning source, $\delta^{13}C$ ranged from −34.7 to −25.4 ‰ whereas for C_4 plants ranged from −22.2 to −13.0‰ (Aguilera & Whigham, 2018). For C_3 plants burning source, $\delta^{13}C$ ranged from −34.7 to −25.4 ‰ whereas for C4 plants ranged from −22.2 to −13.0‰ (Aguilera & Whigham, 2018).

Identification of biomass burning episodes

Temporal variation of levoglucosan

The average concentrations of levoglucosan observed at the CMU site was 0.46 ± 0.56 μg/m^3, while the concentrations of levoglucosan at the MC site were 0.55 ± 0.67 μg/m^3, with no significant difference (P <0.05) between two sites. The levoglucosan concentrations peaked at the CMU site on 25 March at 0.23 μg/m^3, while the peak at the MC site was 0.23 μg/m^3 on 18 April (Figure 2.).

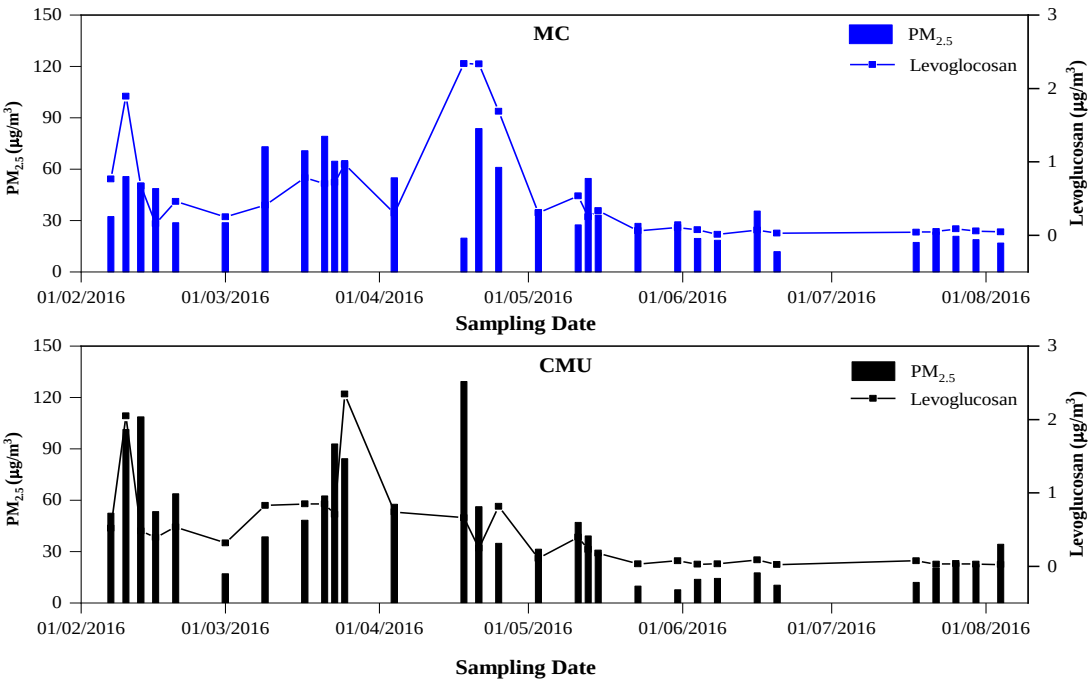

Figure 2. Daily variation in $PM_{2.5}$ and levoglucosan concentrations during the sampling period at the CMU and MC sites.

Based on results from previous studies, it is likely that open burning of agricultural biomass and forest fires were the primary sources of biomass derived particles in our study (Liu et al., 2013; Maenhaut et al., 2016; Thepnuan et al., 2019). Levoglucosan concentration as a specific marker of biomass burning because levoglucosan is the degradation product from cellulose part in some biomass (Klejnowski, Janoszka, & Czaplicka, 2017; Bhattarai et al., 2019; Janoszka, Czaplicka, & Klejnowski, 2020). There is no critical level or range of levoglucosan concentration for proving that the particles were from biomass burning but whatever the level of

levoglucosan detected it will indicate the biomass-burning source. The average levoglucosan concentrations from CMU and MC sites were 0.46 ± 0.56 and 0.55 ± 0.67 μg/m^3, respectively which higher than the report from a rural mountaintop in East China, (Liu et al., 2013) with an average level of 0.064 μg/m^3 and ranged from 0.0028 to 0.11 μg/m^3. Meanwhile, the values from our study were lower than those observed at a Chiang Mai City site with an average 1.22 ± .75 μg/m^3 and 1.13 μg/m^3 (Khamkaew et al., 2016; Thepnuan, Chantara, Lee, Lin, & Tsai, 2019). The variability of levoglucosan levels depending on such biomass containing cellulose component. A moderate correlation between levoglucosan concentrations and $PM_{2.5}$ was found in the CMU site (0.654) but rather low at MC site (0.494) (Figure 3.), suggesting the source of air pollution at both study sites was from open biomass burning during this study period.

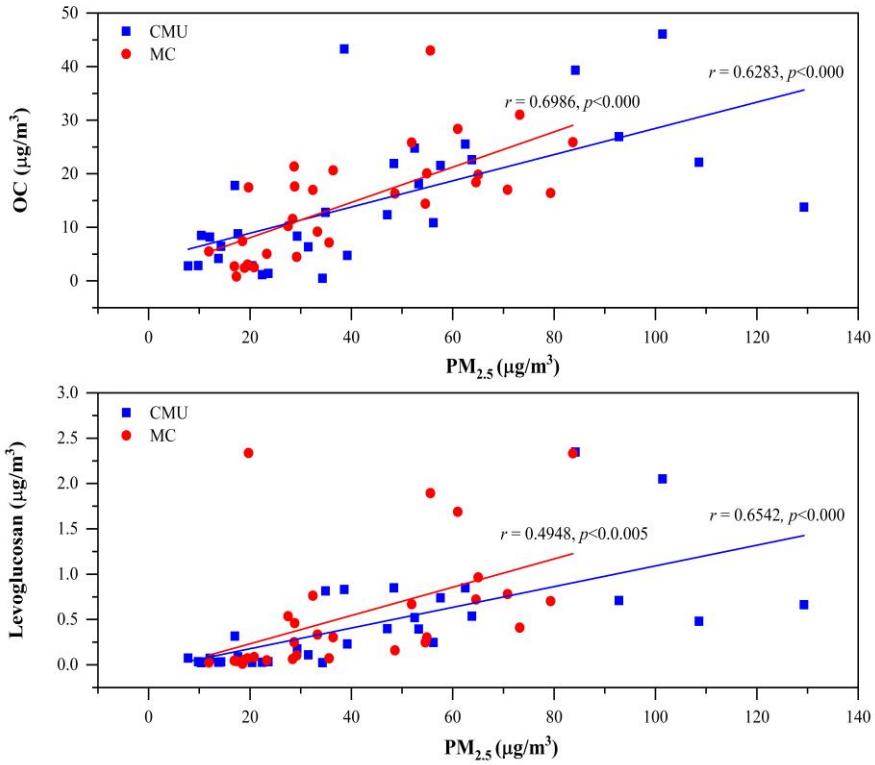

Figure 3. Correlation between $PM_{2.5}$, OC, and levoglucosan at the CMU and MC sites.

This Figure 3. shows concentrations of levoglucosan, a tracer of biomass burning, associated with $PM_{2.5}$ with the r of 0.6542 and 0.4948 in CMU and MC sites, respectively. Meanwhile, OC has association with $PM_{2.5}$ with the r of 0.63 and 0.70, respectively. These two graphs show that OC and levoglucosan attribute to $PM_{2.5}$ in CMU site greater than in the MC site. Interestingly, the backward trajectory indicated that the air mass arrived from southern and southwest of CMU where MC site is in that direction. Though MC has been the intensive area of biomass burning, other meteorological and topographical factors might impact on aerosol plume. To our best knowledge, this study will be the first of its kind from rural area of biomass burning site. Table 1 shows the ratio of OC to levoglucosan, and the mean levoglucosan/OC ratio at the CMU and MC sites were 2.7% and 3.2%, respectively which was similar with previous studies suggesting biomass burning as the source (Sullivan et al., 2008; Ho et al., 2014; Zhang et al., 2014).

Evidence from hotspot maps and trajectory

For the biomass burning season period from February to May 2016 shown in Figure 4. , the HYSPLIT model was used to calculate three-day backward trajectories to evaluate potential sources of air mass flows arriving at CMU and MC. The CMU site is at the altitude of 100 m above ground level (00, 06, 12 and 18 UTM; start time), the data were classified in to 3 clusters. In February, the air masses arrived mainly from the southern direction (56.9%) passing from lower northern Thailand, while the dominant air masses were transported from the southwestern direction in March (49.2%) and April (43.3%). The air mass from the south-west passes through southern Myanmar and some parts of Thailand (Mae Hong Son Province and south-west districts of Chiang Mai Province). In these regions, there was high numbers of fire hotspots during the study period. In May, the bulk of air masses arrived at the study locations from the south-west of Thailand and travelled over the Andaman Sea (60.5%). At the MC site, the direction of air mass flows was largely similar as that found at the CMU site. The primary direction was from south and southwest Myanmar, with higher contribution to overall airflow in February, March, and April at 56.9%, 50.0% and 43.3%, respectively. The result of air mass movement in May was again like the CMU site, the mainly of air masses in these cluster originated from the Andaman Sea was observed for 6 days (in accounts for 47.6% of total). The dominant air masses at CMU and MC sites originated from southern Myanmar and the south of Mae Hong Son Province. The major direction of the air masses at both sites passed through southern Myanmar and the parts of Mae Hong Son. In this region, there was high distribution of fire hotspots during the study period as also shown in previously reports (Khamkaew et al., 2016; Kiatwattanacharoen et al., 2017; Punsompong and Chantara, 2019).

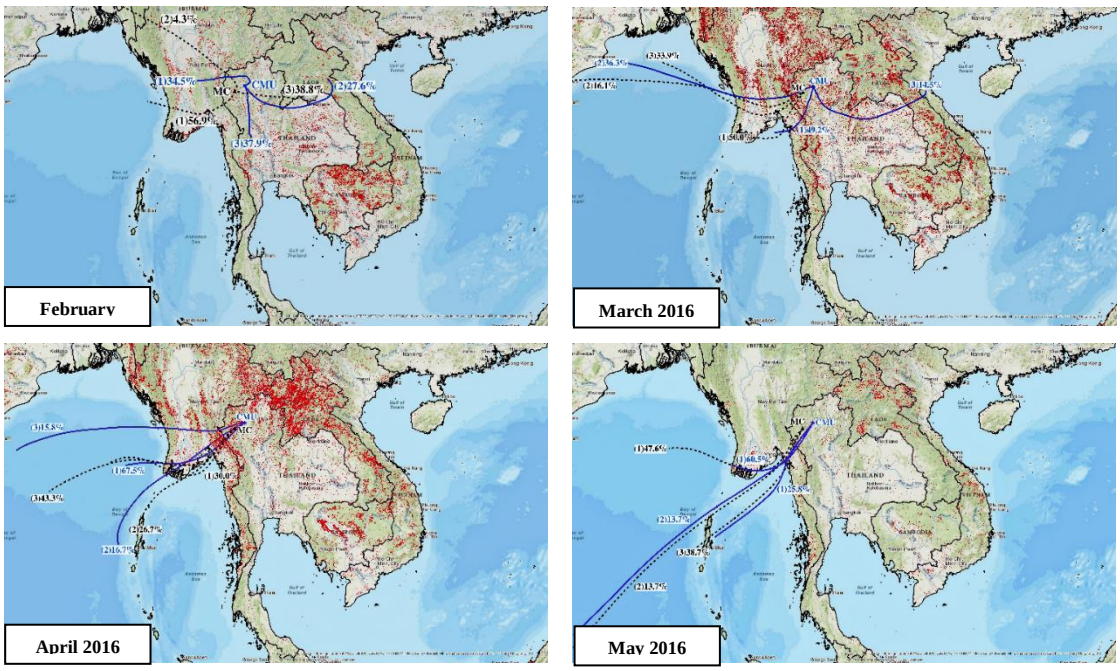

Figure 4. The number of hotspots were obtained from NASA and cluster analysis of three-day BWT arriving at 100 m AGL at both sites.

Major source of biomass type from stable isotope data

The results from the $\delta^{13}C$ measurements of the $PM_{2.5}$ samples are shown in Figure 5. The variation in $\delta^{13}C$ values of TC aerosol from the CMU vs MC sites ranged between -29.3‰ to -26.4‰ vs -28.8‰ to -25.8‰ with a mean of -27.9 ± 0.7‰ vs -27.6 ± 0.6‰, respectively. Our mean values were similar to the values reported for the plant leaf samples from Thailand in 2006 whose mean values in the wet and dry seasons were -29.2‰ and -28.6‰, respectively (Yoneyama et al., 2010). Furthermore, the $\delta^{13}C$ values of C_3-type plants ranged from -30.6‰ to 24.4‰, while

the C_4-type plants ranged from -19.3‰ to 11.6‰ (Table. 2) Therefore, the $\delta^{13}C$ values from both CMU and MC sites indicated the burning of C_3-type plants.

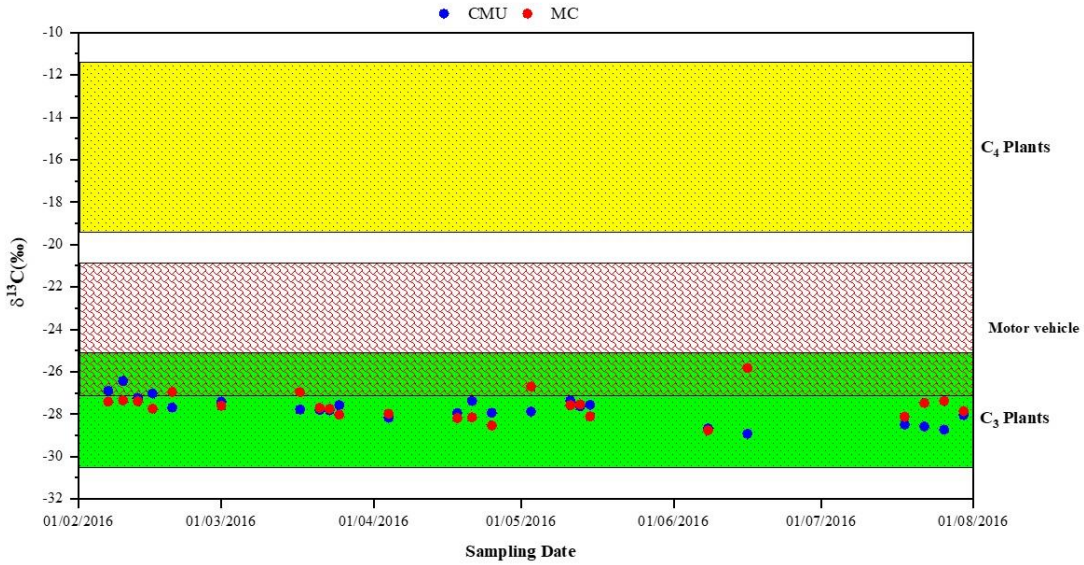

Figure 5. Data plots for $\delta^{13}C$ of TC from CMU and MC sites based on typical value thresholds from various sources such as combustion/emissions from C_3- and C_4-type plants (Turekian et al., 1998; Lü et al., 2000; Yoneyama et al., 2006; Girard et al., 2011; Kawashima and Haneishi, 2012) and motor vehicle emissions (Widory and Javoy, 2003; Widory et al., 2004; López-Veneroni, 2009; Kawashima and Haneishi, 2012)

Table 2. The values of stable carbon isotope in the present study and those reported in the literature.

Plants	Species	$\delta^{13}C$ (‰)	Location	Reference
C4 – type plants	Corn	-16.1	Japan	Kawashima and Haneishi, 2012
	Corn	-13.5 to -11.6	Canada	Girard et al., 2011
	Grass	-19.3	Japan	Kawashima and Haneishi, 2012
	Saccharum	-12.9	South Africa	Turekian et al., 1998
	Saccharum officinarum	-12.1	Thailand and Philippines	Yoneyama et al., 2010
	Zea mays	-15.1	China	Lü et al., 2000
C3 – type plants	Rice plant	-28.5	Japan	Kawashima and Haneishi, 2012
	Dry leaves	-29.4	Japan	Kawashima and Haneishi, 2012
	Mopane (Genus; Colospherum)	-24.4	South Africa	Turekian et al., 1998
	Phyllostachy arcana	-24.8	China	Lü et al. 2000
	Bamboo Indocalamus latifolius	-25.6	China	Lü et al., 2000
	Hay	-29.7 to -27.5	Canada	Kawashima and Haneishi, 2012
	Soybean	-30.6 to -27.7	Canada	Kawashima and Haneishi, 2012
	Dipterocarpus alatus	−28.6 ± 0.4	Thailand and Philippines	Yoneyama et al., 2010
	Tectona grandis	−29.6 ± 0.2	Thailand and Philippines	Yoneyama et al., 2010
	Gigantocha hasskariana	−29.9 ± 0.3	Thailand and Philippines	Yoneyama et al., 2010
C3 – type plants	Mix deciduous forest	−27.9 ± 0.7	CMU, Thailand	This study
	Mix deciduous forest	−29.6 ± 0.6	MC, Thailand	This study

Table 3. Estimated total burned area in Chiang Mai Province, Thailand (hectares).

Year	Forest area	Agriculture area
2015	395,339.8 (90.9%)	39,735.7 (9.1%)
2016	200,974.6 (97.8%)	4,434.1 (2.2%)

DISCUSSION

Our studies suggested the sources of $PM_{2.5}$ in two different sites at Chiang Mai Provinces from February to September 2016 intensive biomass burning period. The chemical composition in airborne PM2.5 were analyzed including levoglucosan, OC, EC, and stable carbon isotope.

Levoglucosan is a specific biomarker of biomass burning marker and derived from cellulose burning (Rushdi et al., 2017; Simpson, Dills, Katz, & Kalman, 2004). The average concentrations of levoglucosan from this study in both sites was significantly higher than the reported value in the Kathmandu Valley, the capital region of Nepal (0.788 ± 0.685 μg/m^3, Xin et.al, 2018) (Xin et al., 2018) and higher than the concentrations of levoglucosan at CMU, Chiang Mai Province, Thailand were 1.13 μg/m3 (Khamkaew et al., 2016). Open burning from forest fire and agricultural were predominant sources of biomass burning in this study. In the present study, the ratio of OC/EC ratio in CMU and MC sites also analyzed. We found that the highest of OC/EC ratios is mostly indicates the $PM_{2.5}$ samples were derived from biomass burning (Zhang et al., 2007). However, the lower ratios of OC/EC were reported in the literature for vehicle fuel formulations in many studies at Beijing, Langfang, and Tianjin (Qi et al., 2018), Wanzhou (Huang, Zhang, Li, Chen, & Yang, 2020), Beijing-Tianjin-Hebei (Ji et al., 2019). Several studies have reported OC/EC ratio for various emission sources which include vehicular exhaust. In addition, we have also use stable carbon isotopic to provide information about the sources of $PM_{2.5}$ and cold be applied in various type of environment studied to identify emission sources in the first time of this area (CMU and MC sites). This finding corresponds to the vast biomass burning area from satellite data (Table 3). The mean value of stable carbon isotope in these studies was suggest that the $PM_{2.5}$ samples were contribution from C_3 plants combustion were similar to the value reported at Doi Ang Kang, Thailand in 2018 during 1 March to 13 April, 2015 intensive biomass burning period (Boreddy, Parvin, Kawamura, Zhu, & Lee, 2018).

However, this study has some limitation represent weakness within a small number of $PM_{2.5}$ samples. There were difficult to find significant relationships from the data such as the correlation between the concentrations of $PM_{2.5}$ and chemical composition. Including the statistical tests normally require a larger sample size to ensure a representative distribution of the samples size to be considered representative of studies area. The authors recommendation the importance of sample size should be greater in quantitative and qualitative studies.

CONCLUSION

In the present study, the chemical composition, and characteristics of $PM_{2.5}$ were investigated at two different sites (CMU and MC) in Chiang Mai Province to identify the contributing sources of airborne $PM_{2.5}$ in Chiang Mai Province. High concentrations of $PM_{2.5}$ were found at both the CMU and MC sites which corresponded with large numbers of hotspots to the southwest and west of the study sites during the study period. The mean values of $\delta^{13}C$ in total carbon (TC) from $PM_{2.5}$ at CMU and MC sites suggest C_3-type plants' burning sources according to major data (n = 48, 85.4%) fell within the range of C_3-type plants and minor data (n = 48, 14.6%) in C_3-type plants and motor vehicle sources. Forest plants in northern Thailand, Chiang Mai particular are mostly mixed deciduous forest i.e. C_3-type plants which falling leaves in dry season and easily causing fire. The results of this study therefore strongly suggest that the burning of C_3-type forest plants attribute to airborne $PM_{2.5}$ pollutants in Chiang Mai Province. As

this study result, control of forest burning on local and regional scales should be seriously considered by the national and local government in reduction of airborne $PM_{2.5}$ in Chiang Mai Province.

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AUTHOR CONTRIBUTIONS

Tippawan Prapamontol: Supervision, Conceptualization, Methodology and Editing. Sawaeng Kawichai: Investigation Writing- Original draft preparation, Writing-Reviewing. Suchart Kiatwattanacharoen: Software, Validation. Fang Cao: Supervision and Methodology. Xiao - Yan Liu: Investigation. Wen - Huai Song: Investigation. Yan-Lin Zhang: Supervision, Conceptualization, Methodology and Editing.

CONFLICT OF INTEREST

The authors declare that they hold no competing interests.

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