



Emission Profiles of PM₁₀-Bound Polycyclic Aromatic Hydrocarbons from Biomass Burning Determined in Chamber for Assessment of Air Pollutants from Open Burning

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ABSTRACT

In order to estimate emission factors (EFs) of air pollutants, three types of biomass (rice straw, maize residue and leaf litter) were collected and burnt in a self-designed stainless steel chamber. The EFs of PM₁₀ from biomass burning were leaf litter ($1.22 \pm 0.29 \text{ g kg}_{\text{dry}}^{-1}$) > rice straw ($0.89 \pm 0.25 \text{ g kg}_{\text{dry}}^{-1}$) > maize residue ($0.59 \pm 0.13 \text{ g kg}_{\text{dry}}^{-1}$), while those of PM₁₀-bound polycyclic aromatic hydrocarbons (PAHs) were leaf litter ($0.91 \pm 0.28 \text{ mg kg}_{\text{dry}}^{-1}$) > maize residue ($0.47 \pm 0.11 \text{ mg kg}_{\text{dry}}^{-1}$) ~ rice straw ($0.46 \pm 0.21 \text{ mg kg}_{\text{dry}}^{-1}$). The results revealed that burning of forest leaf litter emitted higher amounts of particulate pollutants than the agricultural residue burning. New values of diagnostic ratios of some PAHs, including FLA/(FLA + PYR), BaA/(BaA + CHR) and IND/(IND + BPER), were proposed to identify biomass burning sources. Emission rates (ERs) of PM₁₀ and PAHs from biomass burning in Chiang Mai, Thailand were estimated based on the EFs and burning areas recorded in the dry season of 2010 and 2011. The ERs of pollutants from forest burning were found to be much higher than those from agricultural field burning, mainly due to larger burnt areas in the forest. In 2010, PM₁₀ was mainly emitted from the forest fire (2,250 tons), followed by crop burning (133 tons) and paddy field burning (66.9 tons). The same trend was found in 2011 but with much lower emission rates. The ERs of PAHs from biomass burning were 1,815 kg in 2010 and 416 kg in 2011. The ERs of PM₁₀ and PAHs in 2011 were 77% decreased from those in 2010 due to unusually high precipitation in the dry season, causing relatively smaller burnt areas and lower pollutant emissions from open burning. It is expected that the results from this study will be significant information for regulatory actions of air quality management in the northern part of Thailand.

Keywords: Aerosol chemistry; Air pollution; Biomass burning; PM₁₀.

INTRODUCTION

Biomass burning is the burning of living or dead vegetation such as forest, savannas and agricultural lands (Koppmann *et al.*, 2005). Biomass burning has generated global concerns because it emits particles and vapor pollutants into the air, affecting visibility, human health and the global climate (Fang *et al.*, 1999; Yang *et al.*, 2007). Biomass burning is a common method for agricultural residue disposal. This method represents a considerable source of atmospheric pollutants (Korenaga *et al.*, 2001; Chang *et al.*, 2015; Lyu *et al.*, 2015). The principal pollutants emitted by biomass

burning are carbon monoxide (CO), hydrocarbons and particles, with smaller amounts of nitrogen oxide (NO_x) and sulfur dioxide (SO₂). In addition, biomass burning emits volatile organic compounds (VOCs), including polycyclic aromatic hydrocarbons (PAHs), many of which have carcinogenic properties to humans (Liu *et al.*, 2001). During the dry season in Chiang Mai, Thailand (latitude 18°N and longitude 98°E), the combination of biomass burning, especially forest fires and the burning of agricultural residuals, low airflow and temperature inversion, routinely causes a high accumulation of air pollutants (Chantara *et al.*, 2010; Kim Oanh *et al.*, 2011).

PAHs are formed predominantly in the gas phase, whilst those with four or more rings are found mainly in the particulate phase. The PAHs recognized as carcinogenic are mostly associated with particulate matter. Atmospheric PAH concentrations are strongly dependent upon the size of airborne particulate matter (PM) (Lodovici *et al.*, 2003;

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Sevimoglu and Rogge, 2015). The particles with a diameter of less than 10 μm (PM_{10}) can deposit themselves and thereby accumulate in the respiratory system, representing a significant threat to human health (WHO, 2000; Armstrong *et al.*, 2004; US EPA, 2010; Khan *et al.*, 2015; Zhang *et al.*, 2015). The toxic agents are products of incomplete combustion released from both natural and anthropogenic sources. Domestic and industrial coal, wood and biomass combustion, and emissions from road vehicles are the major sources of PAH contamination (Heywood *et al.*, 2006; Wang *et al.*, 2007; Cheruiyot *et al.*, 2015). In addition, there are some emission sources of PAHs, which have large seasonal variations, including residential biofuel and open burning of agricultural wastes and wild fires (Zhang and Tao, 2008).

Other studies have looked at biomass burning and used varying types of chambers to better understand the impact on the health and the environment (Popovicheva *et al.*, 2015; Tian *et al.*, 2015). In India, a stimulation fire test chamber was constructed and used for testing the emissions from various types of garden biomasses, namely grass, leaves, twigs and their mixtures (Kannan *et al.*, 2005). In the United States, the two common agricultural waste residues (almond pruning and rice straw) were burnt in a combustion chamber in order to study the particle size distribution in the smoke emissions (Keshtkar and Ashbaug, 2007). In China, researchers investigated the particle size distribution and PAHs emissions from the burning of rice, wheat and corn straws, which are the three major agricultural crop residues produced in that country, by using a self-designed combustion stove and a stainless-steel environmental aerosol chamber (Zhang *et al.*, 2011). Each study provided critical information on the emission factors and emissions rates of pollutants from biomass burning and helped to inform our tests.

In this study, a chamber was constructed for a biomass burning experiment in order to highlight the impact of open burning on air quality. The primary purposes were (i) to estimate emission factors of PM_{10} and PM_{10} -bound PAHs from biomass burning; (ii) to create the diagnostic ratios to

classify PAH source from biomass burning; and (iii) to provide basic and important emission information to the Seven South East Asian Studies (7-SEAS) network as well as to the region.

MATERIAL AND METHODS

The Self-Designed Stainless Steel Chamber

The chamber was made from stainless steel and designed by the Environmental Chemistry Research Laboratory (ECRL), Faculty of Science, Chiang Mai University. The schematic sketch of the chamber is shown in Fig. 1. It comprises two main parts: a burning chamber and a storage chamber. The burning chamber has a diameter of 0.50 m and 1.23 m long. The storage chamber has a diameter of 0.76 m and 1.50 m long. The burning chamber contains an air inlet, an ignition point equipped with liquefied petroleum gas, a biomass basket and three units of temperature sensor. The storage chamber was set up with a gas analyzer (Testo 350 XL, UK), a mini volume air sampler and a vacuum pump to evacuate the chamber before the beginning of each experiment. Sillapapiromsuk *et al.* (2013) also used this self-designed stainless steel chamber in their work to calculate emission factors and assess the emission rates of PM_{10} from biomass burning.

Collecting and Preparation of Biomass Samples

Three types of biomass were selected based on data of the land used and burning areas in Chiang Mai Province (Office of Chiang Mai Agriculture, 2014). The selected biomass types were rice straw and maize residue, representing the agricultural area, and leaf litter from mixed deciduous forest, representing the forest area. The samples were collected from 4 districts of Chiang Mai Province, where these three types of biomass were dominant. The sampling was randomly carried out by using a 1 m^2 grid. Three times of sampling were performed at each place (Fig. 2). The samples were air dried and stored at room temperature. The

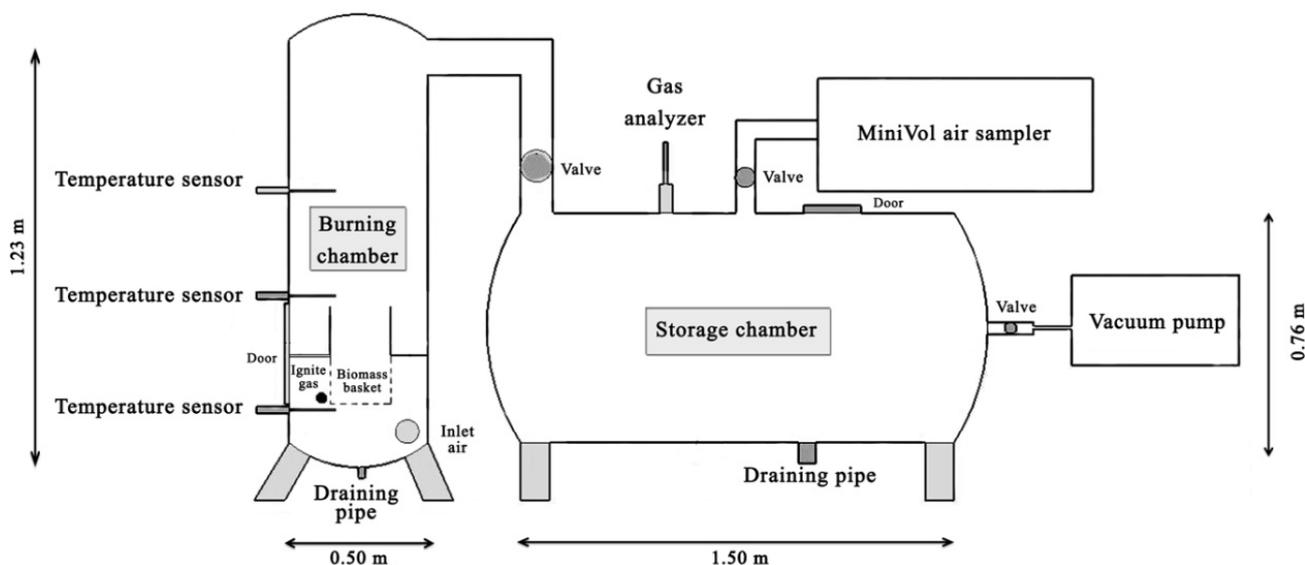


Fig. 1. Schematic diagram of the self-designed stainless steel chamber.

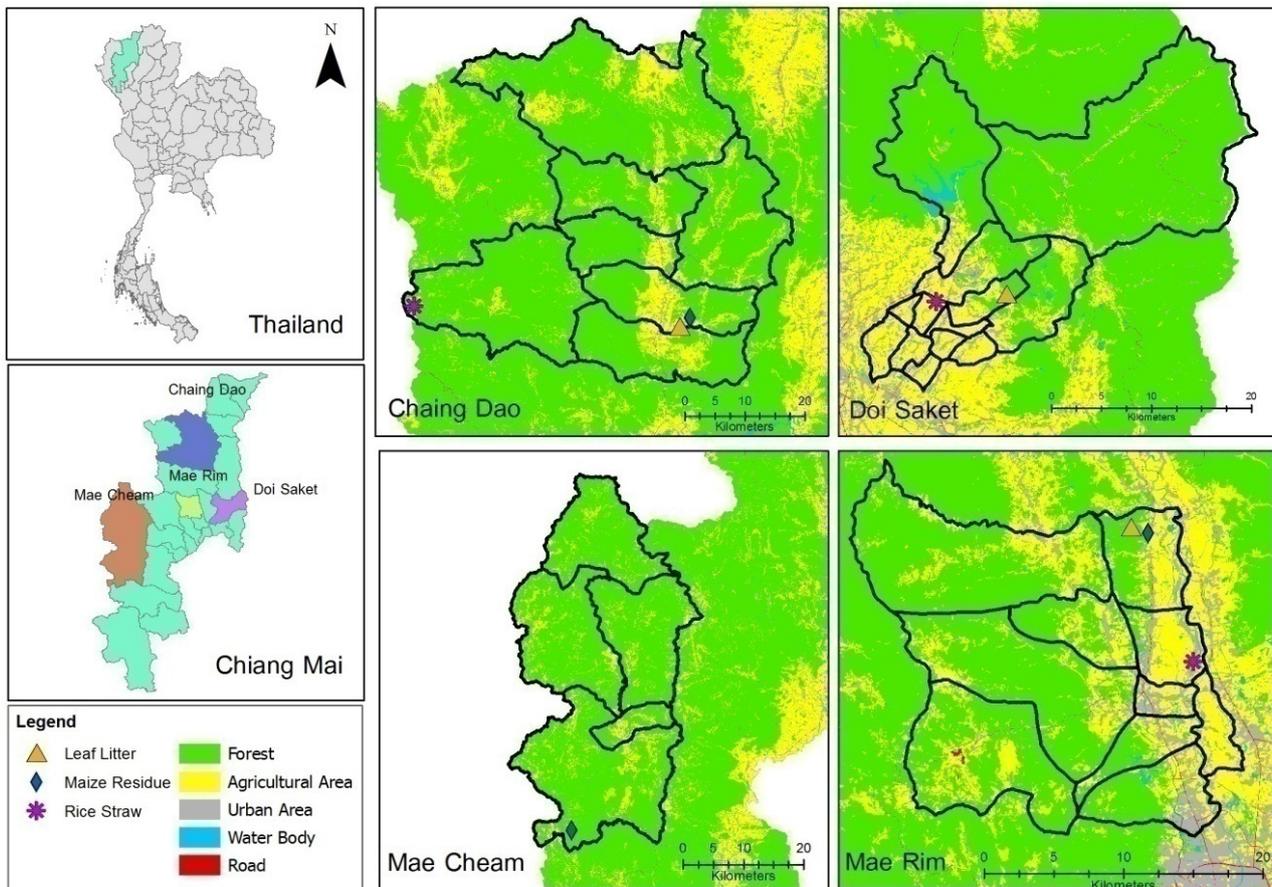


Fig. 2. Mapping of collecting biomass sample in Chiang Mai.

moisture content was measured in 2 g samples using a moisture analyzer (MA50, Germany). The sample taken from each grid was weighed on an analytical balance (AFP-2100L, Adam, Germany). Biomass samples were dried in an oven at a temperature of 80°C for 24 hrs. The samples were cut into small pieces using stainless scissors and blended to powder using a blender. Powder samples were analyzed for the carbon (C), hydrogen (H) and nitrogen (N) contents using a CHNS/O analyzer (PE2400 Series II, Perkin Elmer, Germany) at the Scientific and Technological Research Equipment Center, Chulalongkorn University, Thailand. The samples for burning test were cut into small pieces and homogenized. Approximately 20 g for rice straw and 10 g for maize residue and leaf litter were used for burning experiments (Sillapapiromsuk *et al.*, 2013).

Operation of the Chamber and Collection of Gases and PM_{10}

Operation of the chamber was initiated by drawing the air out of the storage chamber until the vacuum state was reached. Each of the biomass samples (rice straw, leaf litter and maize residue) was then put into a basket inside the burning chamber. Liquefied petroleum gas (LPG) was used for ignition, providing temperatures between 500–700°C. Burning process took about one minute. Concentrations of gases including CO, NO, NO₂ and SO₂ were continuously measured throughout the burning experiment using a gas

analyzer (350-XL, Testo, Germany). The stabilization of pollutant gas concentrations emitted from the burning took approximately five minutes. The PM₁₀ collection began with opening the valve between the storage chamber and the air sampler. The temperature and pressure in the storage tank were measured using a sensor equipped with a gas analyzer and a pressure gauge, respectively. The PM₁₀ sampling started when the pressure in the storage tank was zero and the temperature was about 30°C (or room temperature). Each PM₁₀ sample was collected for five hours with flow rate of 5.0 L min⁻¹ from the storage chamber using the mini volume air sampler (Air metric, USA). The samples were collected on quartz fiber filters (Ø 47 mm) from QM-A Whatman (Maidstone, UK). The filters were weighed before and after using a microbalance (MX5, Mettler Toledo, Switzerland). After PM₁₀ collection, the filters were kept in a plate covered with aluminum foil, and then transferred into a desiccator for 24 hours before being weighed and stored in a freezer until analysis. The chamber was cleaned each time prior to the next experiment. Nine replications for each type of biomass were burnt, therefore 27 PM₁₀ samples were collected. Blank samples were also collected every after three times of sample burning. Blank was done by burning of LPG without biomass in the chamber and collecting the PM₁₀ sample emitted from the burning. The PM₁₀ and PAHs concentrations measured from blank samples were averaged and subtracted from each sample concentration.

Extraction of PM₁₀ Samples and Analysis of 16 PAHs by GC-MS

The PM₁₀ samples were extracted in 25 mL acetonitrile using an ultrasonicator (Elma, Germany) for 30 minutes under controlled temperature (~10°C). The extracted solutions were filtered through 45 µm nylon. The mixture of internal standards (acenaphthene-d₁₀ and pyrene-d₁₂, Supelco, Germany) was spiked into the extracted solution and the volume was adjusted to 2 mL by acetonitrile. The aliquot was analyzed using a gas chromatograph-mass spectrometer (GC-MS) (Agilent 6890, USA) with a 30 m HP-5MS capillary column connected to a mass selective detector (MSD, Agilent 5973, USA). The column was temperature-programmed from 60°C to 290°C with a rate of 6 °C min⁻¹ and then held for 20 minutes. The detection mode was operated in the selective ion monitoring (SIM) mode. Sixteen polycyclic aromatic hydrocarbons (16-PAHs), including naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACE), fluorene (FLU), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYR), benz[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (IND), dibenz[a,h]anthracene (DBA) and benzo[g,h,i]perylene (BPER), were identified. The mixed PAHs standard was obtained from Restek, USA. Internal standards (acenaphthene-d₁₀ and pyrene-d₁₂) from Supelco, Germany were used.

Emission Factors (EFs) and Emission Rates (ERs)

Emission factors (EFs) of PM₁₀ and 16-PAHs from the burning experiment were calculated based on the flow rate of the mini volume air sampler and pollutant concentrations using Eq. (1) (Kim Oanh *et al.*, 2000).

EF =

$$\frac{\text{Conc (ng m}^{-3}\text{)} \times \text{flow rate (m}^2 \text{ h}^{-1}\text{)} \times \text{sampling time (h)}}{\text{biomass (kg)}} \quad (1)$$

EFs of PM₁₀ and PAHs were used to estimate their emission rates (ERs) based on the area of burning converted from hotspot data in each year. A hotspot is a mark on a satellite image indicating a heat source typical of vegetation burning (Davies *et al.*, 2009). Hotspots in this work were identified by the MODIS (Moderate Resolution Imaging Spectroradiometer), on board NASA's Aqua and Terra satellites, using a specific fire detection algorithm that makes use of the thermal band detection characteristics of the sensor (NASA/University of Maryland, 2002). The ER was calculated using Eq. (2).

$$\text{ER} = \text{EF} \times \text{Area of burning (km}^2\text{)} \times \text{biomass (kg km}^{-2}\text{)} \quad (2)$$

Quality Control of PAHs Analysis

Standard reference materials (SRM; Urban Dust 1649b, USA) for PAHs analysis provided by the National Institute of Standards and Technology (NIST) and quartz fiber filter spiked with mixed PAHs standard (Restex, USA) were tested for their recoveries. The optimum extraction condition was used. Recoveries of quartz fiber filter spiked with mixed PAHs standard were 80% to 104%, while the recoveries of 14 PAHs in the SRM 1649 are presented in Table 1 (see supporting information). Detection limits of GC-MS for 16 PAHs were 0.03–0.72 ng m⁻³.

Data Analysis

Multivariate analysis of variance (MANOVA) is simply an analysis of variance (ANOVA) with several dependent variables. It tests for the difference in two or more vectors of means. This study used MANOVA to analyze the differences among biomass types based on their pollutant emissions (PM₁₀ and PAHs) from burning. Pearson correlation (r) was implemented to identify the relationships between the total PAHs (tPAHs), carcinogenic PAHs (cPAHs), non-carcinogenic PAHs (ncPAHs) and PM₁₀. Diagnostic ratios (DRs) were used to determine the possible sources of PAHs in each biomass. PAHs are always emitted as a mixture and the relative molecular concentration ratios are considered (often only as an assumption) to be characteristic of a given emission source (Tobiszewski and Namiesnik 2012). Most DRs involve pairs of PAHs with the same molar mass and similar physicochemical properties, so they undergo similar environmental fate processes. FLA/(FLA + PYR), B(a)A/(B(a)A + CHR) and IND/(IND + BPER) were the conventional DRs to characterize the sources of PAHs in this study due to a number of references providing such ratios.

RESULTS AND DISCUSSION

Characteristics of Biomass and Emission of Pollutant Gases

Table 1 shows the moisture content and content of C, H and N of the biomass samples. Pollutant gases emitted from the biomass burning experiments in the chamber were also presented. The moisture content of leaf litter (12.20%) was higher than rice straw (9.32%) and maize residues (6.51%). The number is apparently depend on the characteristic of the area samples are taken. Leaf litter was collected in a shaded area under tree canopies, while rice straw and maize residues were collected in an open area. Biomass burning emissions play an important role in the biogeochemical cycles of carbon

Table 1. Moisture content, C, H, N content and pollutant gases emitted from burning experiments in the chamber.

Biomass types	%Moisture	%C	%H	%N	Pollutant gases (g kg _{dry} ⁻¹)				
					CO	NO	SO ₂	NO ₂	NO _x
Leaf litter (n = 9)	12.20	46.43	5.84	0.47	71.99	0.85	0.01	1.76	2.61
Maize residue (n = 9)	6.51	40.51	5.67	1.41	58.30	2.10	0.11	3.27	5.37
Rice straw (n = 9)	9.32	36.19	5.17	0.55	44.54	2.67	0.00	2.80	5.48

(Koppmann *et al.*, 2005). Carbon and hydrogen contents in descending order were leaf litter > maize residue > rice straw. Nitrogen content of maize residue was higher than that of rice straw and leaf litters. It is mainly correlated with the amount of fertilizer used in planting areas (Sillapapiromsuk *et al.*, 2013). Similarly, crops contain higher N than leaf litter from the forest area. Pollutant gases were measured during burning time (for duration of approximately 10 minutes at the beginning of the burning process of the chamber). Afterward, the values from the equilibrium phase (constant value) were averaged. The results showed that leaf litter emitted the highest number of CO (72 g kg_{dry}⁻¹) followed by maize residue (58 g kg_{dry}⁻¹) and rice straw (44 g kg_{dry}⁻¹). The CO emissions from each type of biomass were found to be correlated to their carbon content. Concentrations of SO₂ and NO_x (NO + NO₂) emitted from the burning of agriculture waste (maize residues and rice straw) were higher than those of forest fire or leaf litter burning. Agricultural burning emitted many types of gas such as CO₂, CO, CH₄, N₂O, NO_x and SO₂, while forest fire emitted only CO₂, CO, CH₄ and non-methane volatile organic carbon (NMVOC). The results suggest that the agriculture process has more influence than the forest fire (Estrellan and Iino, 2010).

Emission Factors of PM₁₀ and PAHs from Biomass Burning

Nine samples of PM₁₀ collected from the burning of biomass (leaf litter, maize residue and rice straw) in the chamber were analyzed for 16-PAHs. The average and

standard deviation (SD) values of emissions factor (EF) of PM₁₀ and PAHs are shown in Table 2. The EF of PM₁₀ from the burning of the leaf litter (1.22 ± 0.29 g kg_{dry}⁻¹) was significantly ($p < 0.05$) higher than the EF of PM₁₀ from rice straw (0.89 ± 0.25 g kg_{dry}⁻¹) and maize residue (0.59 ± 0.13 g kg_{dry}⁻¹). However, there was no significant difference ($p > 0.05$) between the EF of PM₁₀ from rice straw and maize residue. Similarly, the EF of PM₁₀ from leaf litter burning in this study (1.22 ± 0.29 g kg_{dry}⁻¹) was slightly lower than that reported by Sillapapiromsuk *et al.* (2013) (1.52 ± 0.65 g kg_{dry}⁻¹). The EFs of total PAHs from the biomass burning in descending order were leaf litter (0.910 ± 0.277 mg kg_{dry}⁻¹) > maize residue (0.469 ± 0.106 mg kg_{dry}⁻¹) ~rice straw (0.465 ± 0.213 mg kg_{dry}⁻¹). The non-carcinogenic PAHs (ncPAHs) and carcinogenic PAHs (cPAHs) were classified based on the criteria of the United States Environmental Protection Agency (US EPA, 1993). The emission of tPAHs, ncPAHs and cPAHs from the burning of each type of biomass had the same trend, which was leaf litter > maize residue ~rice straw (Table 2). Moreover, the emissions of ncPAHs and cPAHs were almost equal (50:50). The dominant PAHs emitted from three types of biomass burning were FLA, CHR and BbF. The MANOVA test revealed significant difference ($p < 0.05$) of the EFs of PM₁₀ as well as individual PAH from the burning of leaf litter and the agricultural residues. The test also demonstrated there was no significant difference between the EFs of rice straw and maize residue.

Estimation of occupational and environmental health

Table 2. Emission Factors of PM₁₀ and PAHs from leaf litter, maize residue and rice straw burning in the chamber.

Parameter	Leaf litter ^a (n = 9)			Maize residue ^b (n = 9)			Rice straw ^b (n = 9)		
	Mean	±	SD	Mean	±	SD	Mean	±	SD
PM ₁₀ (g kg _{dry} ⁻¹)	1.22	±	0.29	0.59	±	0.13	0.89	±	0.25
PAHs (mg kg _{dry} ⁻¹)									
NAP	0.005	±	0.001	0.005	±	0.001	0.003	±	0.000
ACY	0.006	±	0.000	0.005	±	0.000	0.003	±	0.000
ACE	0.000	±	0.000	0.000	±	0.000	0.002	±	0.005
FLU	0.011	±	0.002	0.005	±	0.005	0.005	±	0.002
PHE	0.073	±	0.033	0.030	±	0.006	0.025	±	0.012
ANT	0.037	±	0.004	0.034	±	0.001	0.023	±	0.005
FLA	0.118	±	0.047	0.040	±	0.007	0.055	±	0.033
PYR	0.100	±	0.039	0.037	±	0.006	0.053	±	0.032
BaA	0.076	±	0.020	0.040	±	0.009	0.040	±	0.022
CHR	0.117	±	0.033	0.050	±	0.010	0.044	±	0.018
BbF	0.115	±	0.036	0.055	±	0.019	0.068	±	0.033
BkF	0.069	±	0.020	0.041	±	0.008	0.038	±	0.014
BaP	0.045	±	0.011	0.032	±	0.006	0.030	±	0.012
IND	0.055	±	0.014	0.041	±	0.007	0.034	±	0.010
DbA	0.034	±	0.002	0.022	±	0.016	0.017	±	0.007
BPER	0.047	±	0.013	0.032	±	0.006	0.026	±	0.008
tPAHs	0.910	±	0.277	0.469	±	0.106	0.465	±	0.213
ncPAHs	0.475	±	0.137	0.228	±	0.029	0.234	±	0.105
cPAHs	0.435	±	0.103	0.241	±	0.061	0.231	±	0.089
TEQs	0.160	±	0.035	0.105	±	0.031	0.091	±	0.034

The superscripts of a and b represented the difference between groups of biomass. The same superscript of those data means no significant difference ($p > 0.05$), while the different superscript means significant difference ($p < 0.05$) among groups.

^{a, b} Significant difference ($p < 0.05$) from MANOVA test.

risks associated with the exposure of PM₁₀-bound PAHs can be calculated for toxicity equivalent concentrations (TEQs) (Nisbet and LaGoy, 1992; Cecinato, 1997; Wiriya *et al.*, 2013). TEQs calculation is based on the toxic equivalent factors (TEFs) proposed by Nisbet and Lagoy (Nisbet and LaGoy, 1992) (Eq. (3)). In this formula the symbols for the chemicals denote their concentrations.

$$\text{TEQs} = 0.001(\text{NAP} + \text{ACY} + \text{ACE} + \text{FLU} + \text{PHE} + \text{FLA} + \text{PYR}) + 0.01(\text{ANT} + \text{BPER} + \text{CHR}) + 0.1(\text{BaA} + \text{BbF} + \text{BkF} + \text{IND}) + \text{BaP} + \text{DBA} \quad (3)$$

Based on the PAHs emissions from the burning test, the TEQ value of leaf litter ($0.160 \pm 0.036 \text{ g kg}_{\text{dry}}^{-1}$) was higher than maize residues ($0.105 \pm 0.031 \text{ g kg}_{\text{dry}}^{-1}$) and rice straw ($0.095 \pm 0.034 \text{ g kg}_{\text{dry}}^{-1}$). The results showed that the burning of leaf litter emitted higher pollutants and can be more risky to human health. PM₁₀ is particle matter composing of various organic and inorganic compounds, whereas PAHs is a group of organic compounds. Burning of leaf litter showed higher emissions of those pollutants than the agricultural residues. The results revealed that carbon content in the biomass plays an important role in the emission of PM₁₀ and PAHs.

The Profile of PAHs

The profiles of PAHs emitted from biomass burning in the chamber are shown in Fig. 3. The concentrations of the high molecular weight PAHs (4–6 rings) were generally higher than those of low molecular weight. According to Ma *et al.* (2010) and Conde *et al.* (2005), concentrations of low molecular weight of PAHs are high in the gas phase. FLA, CHR and BbF were the dominant PAHs emitted from the burning of all biomass types (Table 2). A study

from Kim Oanh *et al.* (2000) showed that BbF and FLA were the main components of PAHs in solid particle matter emitted from rice straw burning in the hood. The study from Zhang *et al.* (2011) found that the dominant PAHs released from rice straw burning were ANT, FLA and PYR. The burning of wheat straw released ACE, FLA and PYR and corn straw burning released ANT, FLA and CHR. Schmidl *et al.* (2008) reported that BbF was the dominant PAH in PM₁₀ samples collected from burning test heaps of a mixture of pear (*Pyrus communis*), walnut (*Juglans regia*) and birch (*Betulus pendula*) leaves. Shen *et al.* (2011) studied the emission of particulate-bound PAHs from crop residue burning in the brick cooking stove and found that PHE, FLA, FLU and PYR were the major PAHs. According to all studies mentioned above, FLA was the major PAH found in biomass burning emission. CHR was found in corn straw burning (Kim Oanh *et al.*, 2000; Shen *et al.*, 2011; Zhang *et al.*, 2011) and BbF was found in rice straw burning (Kim Oanh *et al.*, 2000) and mixture of tree (Schmidl *et al.*, 2008).

The 16 PAHs were grouped based on the number of aromatic rings in their structures (2-, 3-, 4-, 5- and 6-rings) and plotted as the percentage of compounds emitted from the burning of each biomass type (Fig. 4). It was found that 4 and 5-ring PAHs were dominant (68–74%) in all three biomass types. These values were quite high compared to Atkins *et al.* (2010), in which the 5-ring PAHs contributed between 40% and 60% of total emissions in particulate phase from biomass pellets. The emission from biomass combustion in a wood-fire boiler presented PAH distribution in the particulate phase more than gas phase. It was dominantly high molecular mass compounds (Bignal *et al.*, 2008).

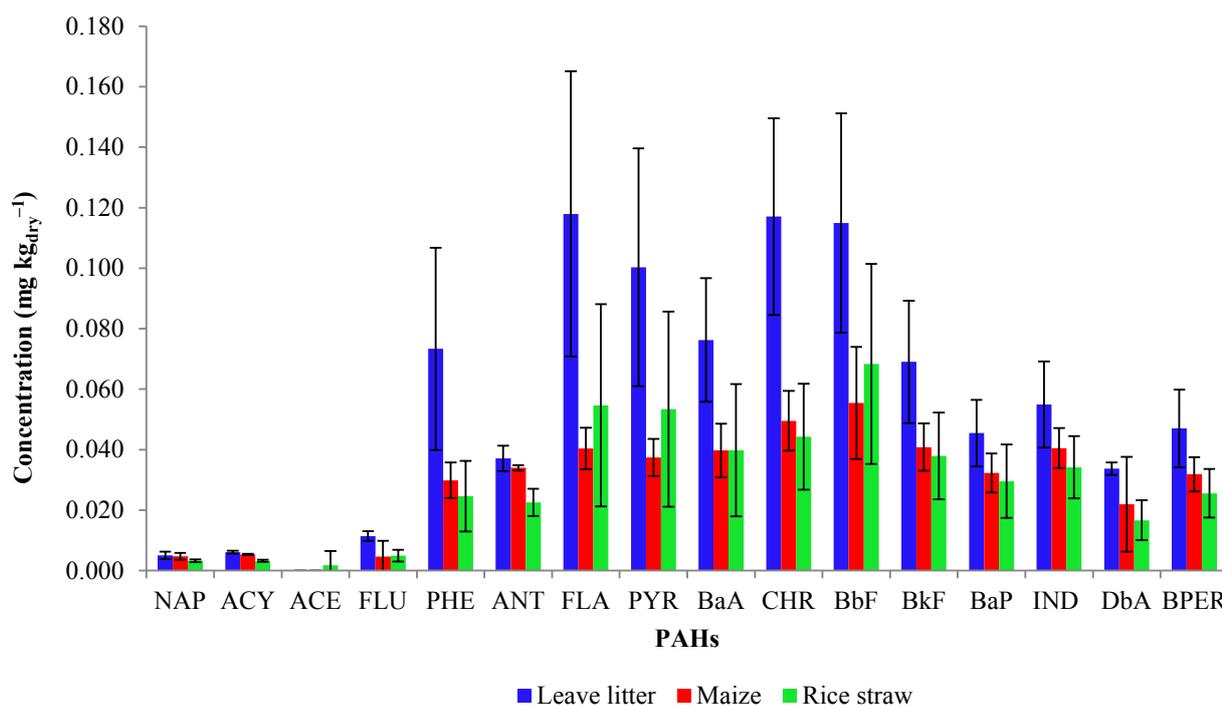


Fig. 3. The profiles of PAHs emitted from the burning of leaf litter, maize residues and rice straw in the chamber.

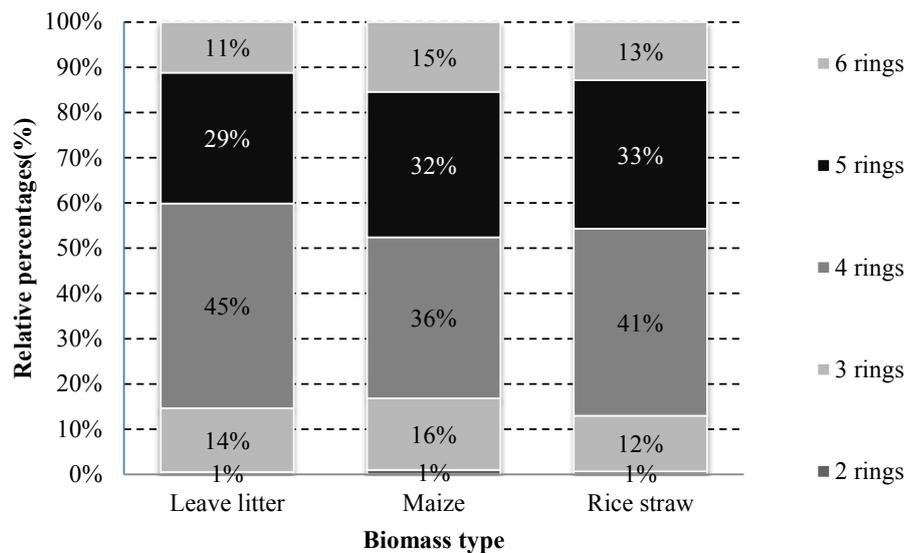


Fig. 4. Distribution of PAHs emitted from leaf litter, maize residues and rice straw burning.

Correlations of PM_{10} and PAHs Species Emitted from Biomass Burning

Correlations of PM_{10} and PAHs emitted from the burning of three types of biomass in the chamber were analyzed (Table 3). The concentrations of PM_{10} and each group of PAHs (tPAHs, ncPAHs and cPAHs) were moderately significantly correlated ($r = 0.598$, 0.582 and 0.416 , respectively) for leaf litter burning. For maize residues, strong correlation was also found between PM_{10} and ncPAHs ($r = 0.819$) while fair correlations were found between PM_{10} and tPAHs ($r = 0.601$) and PM_{10} and cPAHs ($r = 0.428$). The correlation of PM_{10} and each group of PAHs (tPAHs, ncPAHs and cPAHs) were strongly significantly correlated ($r = 0.935$, 0.849 and 0.857 , respectively) for rice straw burning.

In addition, the highest correlation coefficient ($r = 1.000$) between each pair of PAHs emitted from biomass burning was considered for diagnostic ratios in order to identify the PAHs source from biomass burning. The first pair was IND and BPER emitted from leaf litter burning. The second and third pairs were IND and BPER and CHR and BaA emitted from maize residues burning. The last pair was FLA and PYR from rice straw burning. This pair had a low correlation coefficient ($r = 0.998$) than the three couples.

PAH Diagnostic Ratios for Source Identification of Biomass Burning

PAHs diagnostic ratios have been used worldwide to determine their sources (Yunker et al., 2002; Galarneau et al., 2008; Ravindra et al., 2008a; Orechio et al., 2009; Tobiszewski et al., 2012; Da Limu et al., 2013). The ratio value of ANT/(ANT + PHE) lower than 0.1 was recognized as petrogenic sources, while the value above 0.1 was classified as pyrogenic source (Pies et al., 2008). The ratio of BaP/(BeP + BaP) can be used for identification of fresh particles (~ 0.5) and photolysis of particles (< 0.5) (Oliveira et al., 2011). The ratio of FLU/(FLU + PYR) has been used for petrol emissions (< 0.5) and diesel emissions (> 0.5) (Ravindra et al., 2008b). FLA/(FLA + PYR), BaA/(BaA +

CHR) and IND/(IND + BPER) ratios have been used to indicate for many sources (e.g., gasoline engine, diesel engine, wood combustion, coal combustion, cooking smoke and biomass burning) in different range values (Schauer et al., 1996; Yunker et al., 2002; He et al., 2004; Wang et al., 2009a; Ma et al., 2010; Dvorská et al., 2011). Therefore, their ratios were considered and proposed with a narrower range in order to specify a marker for biomass burning in this study. According to the strong correlations between FLA and PYR, BaA and CHR and IND and BPER ($r \sim 1.000$), the new ratios of FLA/(FLA + PYR), BaA/(BaA + CHR) and IND/(IND + BPER) were proposed to identify the biomass burning source for leaf litter, maize residue and rice straw. The proposed values were compared with other studies (Table 4). The FLA/(FLA + PYR) ratio was proposed for leaf litter, maize residue and rice straw burnings separately. The BaA/(BaA + CHR) ratio values as well as the values of IND/(IND + BPER) were also proposed. In order to be practical, all three values for three types of biomass were overlaid and only one set of the values was proposed for burning of mixed biomass. The values of 0.50–0.55 are specified for FLA/(FLA + PYR), 0.40–0.55 for BaA/(BaA + CHR) and 0.50–0.60 for IND/(IND + BPER).

Assessment of Emission Rates (ERs) of Pollutant Gases, PM_{10} and PAHs from Open Burning in Chiang Mai Province

The burnt areas of rice straw, maize residue and leaf litter (forest fire) in Chiang Mai Province were estimated by MODIS Landsat 5 TM (Dontree et al., 2011). The burning periods between December 2009 to April 2010 and December 2010 to April 2011 were considered as the dry season for the years 2010 and 2011, respectively. Approximately 3,073 km² of forest, 326 km² of crop planting and 111 km² rice field were burnt in 2010 while about 615, 173 and 78 km², respectively were burnt in 2011. The percentages of burnt area in descending order were crop (50%) > forest (28%) > rice field (11%) in 2010, while in 2011 they were crop

Table 3. Correlation between PAHs and PM₁₀ from biomass burning.

Biomass types	tPAHs	cPAHs	ncPAHs	PM ₁₀
Leaf litter				
tPAHs	1.000			
cPAHs	0.695**	1.000		
ncPAHs	0.881**	0.272*	1.000	
PM ₁₀	0.598**	0.582**	0.416*	1.000
Maize residue				
tPAHs	1.000			
cPAHs	0.966**	1.000		
ncPAHs	0.846**	0.679**	1.000	
PM ₁₀	0.601**	0.428*	0.819**	1.000
Rice straw				
tPAHs	1.000			
cPAHs	0.903**	1.000		
ncPAHs	0.921**	0.664**	1.000	
PM ₁₀	0.935**	0.849**	0.857**	1.000

** Correlation is significant at the 0.01 level (2-tailed), * Correlation is significant at the 0.05 level (2-tailed).

Table 4. Comparison diagnostics ratio form biomass burning from this study and other study.

Ratio	Sources	References
FLA/(FLA + PYR)	0.50–0.55	Broad-leaf tree
	0.54–0.60	shrub
	0.43–0.53	grass
	> 0.60	biomass burning
	> 0.50	grass, wood, coal combustion
	0.45–0.56	rice straw burning
	0.53–0.55	leaf litter burning
	0.51–0.53	maize burning
	0.49–0.52	rice straw burning
	0.49–0.55	biomass burning
BaA/(BaA + CHR)	> 0.34	biomass burning
	0.37–0.44	leaf litter burning
	0.43–0.46	maize burning
	0.43–0.55	rice straw burning
	0.34–0.55	biomass burning
IND/(IND + BPER)	0.60–0.64	rice straw burning
	0.50	grass, wood, coal combustion
	0.51–0.56	leaf litter burning
	0.55–0.59	maize burning
	0.56–0.59	rice straw burning
	0.51–0.59	biomass burning

(27%) > rice field (8%) > forest (6%). The burnt area in 2010 was obviously higher than that in 2011 due to the rain precipitation effect. Both the annual rain frequency and the amount of rain was less in 2010 (112 days and 1,156 mm) than in 2011 (144 days and 1,308 mm). There were only nine rainy days with 37.4 mm precipitation in dry season of 2010 (December 2009 to April 2010), while higher frequency of rain (28 days) and higher amount of precipitation (163 mm) were detected in the dry season of 2011 (December 2010 to April 2011).

Emission Rates (ERs) of gas pollutant, PM₁₀ and PAHs calculated from burnt area in Chiang Mai Province (Eq. (2)) in years 2010 and 2011 are illustrated in Table 5. Pollutant gases (CO, SO₂ and NO_x) emitted from the three types of

biomass burning in the chamber can be used to estimate for emission of open burning in Chiang Mai. CO was the major gas emitted from all types of biomass burning (Table 1). The emission rate depends on the burnt area. Different types of land use resulted in the same emission trends of CO and NO_x. Forest burning emitted higher CO and NO_x than crop and rice field burning (Table 5). The rate of SO₂ emissions from biomass burning was much lower than CO and NO_x.

The ERs of PM₁₀ in 2010 from the burning of leaf litter, maize residue and rice straw were 2,250, 133 and 67 tons, respectively. Approximately 90% of PM₁₀ were emitted from forest fire in year 2010, while only 80% were emitted in 2011. The ERs of PM₁₀ in 2011 were computed for leaf litter burning (451 tons), maize residue burning (71 tons)

Table 5. PM₁₀ and PAH emission rate from leaf litter, maize and rice straw in year of 2010 and 2011.

Parameter	2010			2011		
	Leave litter	Maize	Rice straw	Leave litter	Maize	Rice straw
PM ₁₀ (mg year ⁻¹)	2,250	133.2	66.90	450.7	70.81	46.92
Gas (mg year ⁻¹)						
CO	132,574	13,202	3,344	26,554	7,018	2,345
SO ₂	18.42	24.91	0.00	3.69	13.24	0.00
NO _x	4,806	1,216	411	963	646	289
PAHs (kg year ⁻¹)						
NAP	9.35	1.08	0.25	1.87	0.57	0.17
ACY	11.2	1.22	0.24	2.25	0.65	0.17
ACE	0.00	0.00	0.13	0.00	0.00	0.09
FLU	21.1	1.06	0.38	4.22	0.56	0.26
PHE	135	6.76	1.85	27.0	3.60	1.29
ANT	68.4	7.70	1.69	13.7	4.09	1.19
FLA	217	9.14	4.10	43.5	4.86	2.88
PYR	185	8.47	4.01	37.0	4.50	2.81
BaA	140	9.00	2.99	28.1	4.78	2.10
CHR	216	11.2	3.32	43.2	5.96	2.33
BbF	212	12.6	5.13	42.4	6.67	3.60
BkF	127	9.24	2.85	25.5	4.91	2.00
BaP	83.8	7.32	2.22	16.8	3.89	1.56
IND	101	9.18	2.56	20.3	4.88	1.80
DbA	62.1	4.96	1.25	12.4	2.64	0.88
BPER	86.5	7.21	1.92	17.3	3.83	1.35
tPAHs	1675	106	34.9	336	56.4	24.5
ncPAHs	874	51.6	17.6	175	27.5	12.3
cPAHs	801	54.5	17.3	161	29.0	12.2
TEQ	294	23.7	6.80	58.9	12.6	4.77

and rice straw burning (47 tons), respectively. The ERs of PM₁₀ from leaf litter burning in the study of Sillapapiromsuk *et al.* (2013) in 2010 and 2011 were 2,794 and 560 tons, respectively. This data was slightly higher than what was revealed in 2010 and 2011 (2,250 and 451 tons, respectively). These differences were from the different EF values of PM₁₀ deployed in each calculation. Kim Oanh *et al.* (2000) reported that 80% of burning area in the northern part of Thailand was in the forest area and only 20% was in agriculture area.

The ERs of tPAHs from open burning in 2010 were forest (1,675 kg) > crop (106 kg) > rice field (35 kg) while the ERs in 2011 were forest (336 kg) > crop (56 kg) > rice field (24 kg). The dominant PAHs emitted from forest and agricultural area was BbF. The ERs of ncPAHs and cPAHs were equally about half of the tPAHs. Furthermore, the TEQs values followed the same trend as tPAHs, ncPAHs and cPAHs. The ERs of all pollutants showed that forest burning emitted higher pollutants than the burning of agricultural waste. It was related to the size of the burnt area. The ERs of open burning in Chiang Mai Province for PM₁₀ and PAHs in both years were almost the same pattern. However, the ERs of PM₁₀ and PAHs in 2010 were 2-5 times higher than those in 2011 due to sources emission and meteorological condition.

CONCLUSIONS

This investigation revealed important information on the

emissions of PM₁₀ and PAHs resulting from the biomass burning of leaf litter, maize residue and rice straw. The mean EFs of PM₁₀ from the burning of leaf litter, maize residue and rice straw species were 1.22, 0.59 and 0.89 g kg_{dry}⁻¹ respectively while those of tPAHs were 0.91, 0.47 and 0.46 mg kg_{dry}⁻¹ respectively. The PAHs with relatively high emission, according to the burning of all three types of biomass, were FLA, CHR and BbF. There are various possible factors, which could affect the emission factors, such as type of combustion chamber used, specific burning conditions and relevant biomass properties (moisture content, elemental composition, planting location, agricultural chemicals used, etc.). Moisture content of biomass has an influence on the net calorific value, the combustion efficiency and the temperature of the actual combustion. On the other hand, it was very difficult to indicate how each factor can directly affect the EFs.

Furthermore, the very strong correlations between IND and BPER, CHR and BaA, and FLA and PYR were considered for calculation of diagnostic ratio to identify PAHs sources. The values of FLA/(FLA + PYR) (0.50–0.55), BaA/(BaA + CHR) (0.40–0.55) and IND/(IND + BPER) (0.50–0.60) were proposed for recognition of biomass burning.

The EF data gained from the burning experiment in the chamber, was used to estimate the emissions of pollutant gases, PM₁₀ and PAHs, resulting from open burning in Chiang Mai Province. The ERs of CO was higher than NO_x (NO + NO₂) and SO₂ in both of 2010 and 2011,

respectively. The ERs of PM₁₀ from open burning in 2010 in descending order were forest (2,250 tons, 92%) > crop (133 tons, 5%) > rice field (67 tons, 3%), while those in 2011 were forest (450 tons, 80%) > crop (71 tons, 12%) > rice field (47 tons, 8%). The ERs of tPAHs from the burning of three types of biomass were 1,815 kg in 2010 and 416 kg in 2011. In 2011, the ERs of PM₁₀ and PAHs decreased by 77% from 2010 levels due to high precipitation and low burning activity in the dry season of 2011.

This study developed emission factors of the PM₁₀ and PAHs resulting from biomass burning, which will likely be useful as an input for other modeling studies in the future. The results of this study will also be important to air pollution and climate change studies for the Southeast Asian region.

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