ELEMENTAL COMPOSITION OF PM2.5 EMITTED FROM RICE STRAW BURNING IN COMBUSTION CHAMBER

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Abstract- Biomass open burning emits significant amount of atmospheric pollutants to the atmosphere which affects air quality and can be with numerous negative health impacts. This study aims to determine particulate matter with diameter less than 2.5 µm ($PM_{2.5}$) and its elemental composition emitted from rice straw burning. Agricultural residues burning are a common practice in Southeast Asia (SEA) and rice straw is one of the major part of residues. Rice straw samples were collected during harvest season from five locations (n=15) in Northern Thailand where air pollution is critical during the dry season due to open burning. The average emission factor (EF) of $PM_{2.5}$ of rice straw burning was 3.7 ± 2.0 $g/kg_{(dry\ biomass)}$. Overall 11 elements were detected in composition of $PM_{2.5}$. EF of K emitted from rice straw was 514 ± 249 $mg/kg_{(dry\ biomass)}$ which makes up 66% of all measured elements in the composition of $PM_{2.5}$. The EF of elements in descending order were K>Na>Ca>Mg>Fe> Al>Sb>Cr >Sn>Zn and Si. $PM_{2.5}$ EF was strongly correlated (r=0.62) to EF of K, but with same strength and negative correlation showed with EFs of Sb ($p < 0.05$) and strong adverse relation with EF of Cr ($r = -0.64$, $p < 0.01$), and aggregate mass of measured elements consisting 18% mass of emitted $PM_{2.5}$ from rice straw. Presence of such complex and toxic matter in atmospheric air increases the toxicity of emission from biomass open burning of rice straw, and has potential environmental and health effects.

Keywords- Air Pollution, Open Burning, $PM_{2.5}$, Emission Factor.

I. INTRODUCTION

Rice is cultivated globally and is by far the most popular crop in Asia[1]. Rice cultivation covers large area of southeast Asian countries [2]. Thailand is an agricultural based country and the main crop is rice [3], which produces plenty of stubbles remaining after harvesting. Following harvesting of rice there are various practices to clear the land before next cultivation. The common practice is burning, which is the most convenient, cheapest and fastest way to eliminate rice straw [4], therefore, Open burning is frequently used to remove agro-residues because of its simplicity and economical sound and for controlling of crop residues and weeds in the field after harvesting is completed [5].

Crop residue burning represents an important source of air pollutants in many countries, especially in developing countries [6]. In Asia open burning activities commonly take place during the dry season when stagnant atmospheric conditions are normally observe hence can lead to high air pollution build-up in adjacent urban areas [7]. For this reason, the smoke-haze episodes in the Southeast Asia (SEA) region for the past three decades has been an almost annual transbounadry pollution problem affecting several countries in the region [8]. Atmospheric aerosols emitted by biomass burning in SEA is the dominant pollutant causing exceedances of ambient air quality threshold on a regional scale [8]. Biomass burning due to forest fire and agricultural waste burning has been a major concern in SEA affecting human health, atmospheric visibility, land use, and aquatic ecosystems [9].

Open burning of biomass is a serious problem in Northern Thailand, especially in dry season [4], [6], [10], [11]. Biomass burning includes the open burning of crop residues, forest fire and grass land fire[6], [12], and biomass burning refers to the burning of existing and dead vegetation [6], [13], which is a significant source of fine particulate matter [14]–[17].

Burning of rice straw emitting large amount of toxic pollutants [18], [19], such as atmospheric particulate matter which itself is a complex mixture and its potential environmental and health effects depend on particle size and composition [20]. $PM_{2.5}$ also known as find particles (generally defined as those particles with an aerodynamic diameter of 2.5 µm or less) is the major cause of haze in most areas [21]. Emission of air pollutants such as atmospheric particles in the Asian region are increasing rapidly [22]. Trace elements are

one of the important constituents of atmospheric particles [23] emitted from biomass burning, including heavy metals (Pb, Cu, Fe, Zn, and Hg, etc), which can adversely affect human health and have effects on the climate change [25]. Biogeochemical cycles of trace elements in tropical regions may also be changing due to biomass burning emissions. Emission of heavy metals can cause local and regional pollution of the atmosphere [24].

The $PM_{2.5}$ particles are likely to penetrate deep into alveolar sacks of lung. The particles can accumulate in the respiratory system and are associated with numerous negative health impacts. A number of studies have shown that exposure to find particles matter ($PM_{2.5}$) is most closely associated with increased hospital admission and emergency room visit for heart and lung diseases [26]. However, the effect of $PM_{2.5}$ goes beyond risk to human health, it affects weather and reduces visibility [27].

This research aims to evaluate the $PM_{2.5}$ emission factor (EF) and its elemental composition that emitted from rice straw open burning over the northern Thailand, where agricultural related biomass disposal is intensive during the dry season.

II. MATERIALS AND METHODOLOGY

A. Reagents

All the reagents used during the experiment were chemically pure, analytical reagent grade and were used without further purification. Elements standard stock solution of (1000 µg/ml) (BDH, England), 65% extra pure nitric acid (Merck, Germany) and 37% extra pure hydrochloric acid (Merck, Germany) were used.

B. Biomass Sampling and burning experiment

B.I. Field sampling

Biomass samples (rice straw) were collected using square grid (2m x 2m) from five locations (3 replications) in four provinces, namely Chiang Mai (CM), Chiang Rai (CR), Phayao (PY), and Phyrae (PH) in Upper Northern Thailand (UNT) during harvesting season except sample from PH that was collected later, details of sampling given in Table I. Prior to burning experiment all sampled biomass residues were weighed, air dried and homogenized.

Sample location	Sampling date	Biomass mass (g)	Remained ash (g)	$PM_{2.5}$ EF ($g/kg_{dry\ biomass}$)
CR1	28-Nov-15	1009±5	186±3	6.6±1.8
PY	28-Nov-15	1005±3	198±14	4.7±1.0
CR2	29-Nov-15	1014±3	170±6	3.7±0.7
CM	15-Dec-15	1009±7	228±11	2.1±0.7
PH	4-Jun-16	1009±5	200±15	1.5±0.2

Table I. Details of biomass sampling and open burning experiment

B.II. Open burning experiment

Biomass open burning simulation performed using the constructed open system combustion chamber. The chamber details as (Amaral, et al., 2014) [28] was designed and built by Environmental Chemistry Research Laboratory (ECRL) for simulation of biomass open burning, that is equipped with a gas analyzer (350-XL, Testo), PM samplers [29], flow meter (TSI, 4000) and temperature sensor, that provide opportunity to collect $PM_{2.5}$ samples and analysis the gasses simultaneously. Each replication of each biomass sample was burned separately in the open-system combustion chamber. $PM_{2.5}$ samples were collected by $PM_{2.5}$ sampler [29] on quartz fiber filters (Whatman's, UK, Ø 47 mm) during the biomass open burning simulation. Blank samples were also collected on quartz fiber filters by air sampling for about 5 minutes without fire (no burning process) for background subtraction. Analytical microbalance (MX5, Mettler Toledo, Switzerland) is used to weigh quartz fiber filter before and after collecting $PM_{2.5}$ samples to determine amount $PM_{2.5}$ samples during each experiment.

C. Elemental analysis of $PM_{2.5}$ samples

C. I . Analysis of elemental composition of $PM_{2.5}$

$PM_{2.5}$ collected on quartz fiber filter was extracted in 4 ml of aqua-regia ($HCl:HNO_3$, v/v, 3:1) solution using double layer Teflon digestion bomb at a temperature of 140°C for 4 hours. Extracted solutions was cooled down to room temperature, then the volume was adjusted to 25 ml with 2% HNO_3 in a volumetric flask, filtered with a nylon membrane (0.45 µm pore size, 13 mm diameter) and stored at 4°C prior to analysis [30].

Elemental analyses were performed by inductively coupled plasma optical emission spectroscopy (ICP-OES). The sample solutions were measured in triple replications, while instrument performance stability was tested using of a mixed standard solution (1 µg/ml) for peak area comparison at every 9 sample injections. Blank samples were also extracted and analyzed using the same method as for the samples. A medium solution was used for background subtraction from each sample. for accuracy of measurement percent recovery of all elements obtained by using certified reference material (CRM) of pond sediment (No.2, 300.21, Japan) and spiked concentration that ranged from 90% to 101% except for Al which showed a recovery of 80%.

C.II. Emission Factors calculation

Emission factor (EF) is a representative value that attempts to relate the quantity of a pollutant released to the atmosphere with an activity associated with the release of that pollutant. These factors are usually expressed in terms of the mass of pollutant emitted per unit mass of dry mass used (g or $mg/kg_{\ dry\ mass}$)[11].

$PM_{2.5}$ EF of each biomass type (EF_X) was measured from the net weight (m) of $PM_{2.5}$ (g), dilution factor (DF using (1)) and dry biomass (kg).

$$DF = \frac{V_{(air)}}{V_{(sampling)}} \quad (1)$$

Volume of air ($V_{(air)}$) is directly proportional to the height of smoke produced during the burning experiment and square of stack's radius, while volume of sampling ($V_{(sampling)}$) is measured from flow rate of Mini volume sampler and sampling time. Therefore the estimated EF will represent amount of $PM_{2.5}$ in all smoke produced during the open burning simulation in open system combustion chamber, (2) applied to calculate EF of $PM_{2.5}$.

$$EF_X = \frac{m_{(PM\,2.5)} \times DF_X}{m_{dry-biomass}} \quad (2)$$

Where $m_{(PM2.5)}$ is mass of $PM_{2.5}$ (g).

In this study EF of elements (EF_E) from $PM_{2.5}$ is calculated using the ratio of DF and mass of dry biomass and mass of elements calculated in the samples solution, where C is concentration (µg/ml) of each element in solution and V is the volume of solution (25 ml) using (3).

$$EF_E = \frac{CV \times DF_X}{m_{dry-biomass}} \quad (3)$$

D. Method validation

Accuracy of the method was tested by using certified reference material (CRM) and spiked method to calculate the percent recovery (%R) of the method for each targeted element. Limit of detection (LOD) and limit of quantification (LOQ) were calculated solution sued for calibration curve construction three and ten times of standard deviation (SD) of lowest concentration of mixed standard. Additionally, linearity of was also confirmed for sensitivity of determination of elements, selecting range for calibration curve with a linear interval.

E. Data analysis

Multivariate analysis of variance (MANOVA) and analysis of variance (ANOVA) method is performed in this study to find out and analyze the differences and correlations (r) among EFs $PM_{2.5}$ its elemental composition.

III. RESULTS AND DISCUSSIONS

A. Emission factor $PM_{2.5}$

Open burning simulation performed for each replication of biomass from each location separately and time in-between of each pairs of burning experiment was respected to ensure that there is no effect from previous burning experiment to the new. EFs of all five locations were measured by burning approximately one kilogram (Table I) of air dried and homogenized biomass sample of rice straw for each burning experiment. Average $PM_{2.5}$ EF of rice straw was 3.7 ± 2.0 ($g/kg_{dry\ biomass}$) (Fig. 1), though this EF for $PM_{2.5}$ from rice straw is lower than some previous literatures (8.3 g/kg) [27], [31] and (12.95 g/kg) [32] and very similar to some earlier studies [33], but $PM_{2.5}$ EF from RS is even higher than the PM_{10} EF (0.69 g/kg) of rice straw studied for Chiang Mai [13].

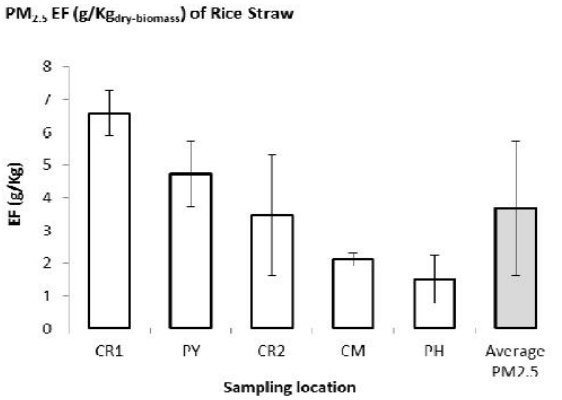


Fig. 1. $PM_{2.5}$ emission factor, from rice straw burning collected from 5 locations of four provinces in UNT.

Emission factors of $PM_{2.5}$ fluctuated considerably among biomass samples from all 5 locations despite using almost the same amount of dry biomass (Table I) for each open burning simulation. Rice straw biomass samples from CR_1, PY and CR_2 were collected late November 2015 (Table I), immediately after crop harvested areas and showed high $PM_{2.5}$ emission factor while rice straw biomass samples from PH was gathered from fields which were harvested a period of time ago (June, 2016) and emitted lower $PM_{2.5,}$ an effect is observed for late collected samples, despite this fluctuation very smaller than some previous studies [31], though this fluctuation and decline did not impacted concentration of most of trace elements except K and Na. Furthermore, meteorological condition such as wind during burning simulation can impact the EFs, while composition, type, and form of elements in soil can impact uptake and concentration of elements in a plant (Fig.1).

B. $PM_{2.5}$ composition

*E*lements in the extracted solution of $PM_{2.5}$ samples were investigated and determined by using ICP-OES. Composition of $PM_{2.5}$ emitted from rice straw is shown in Table II. Potassium (K) as biomass burning tracer [30] is the major element detected, this trend is similar but higher in respect to some earlier literatures [34].

	$PM_{2.5}$ EF (g/Kg)	Elements EF (mg/kg $_{dry\text{-}biomass}$) ± SD											Total EFs/E
		Al	Ca	Cr	Fe	K	Mg	Na	Sb	Si	Sn	Zn	
CR_1	$6.6±1.8^{a}$	$5.4±2^{a}$	$102±9^{a}$	$2.7±1^{a}$	$10.8±3^{a}$	$681±77^{a}$	$34±10^{a}$	$125±18^{a}$	$3.4±0.5^{a}$	$1.2±0.2^{a}$	$2.9±1.4^{a}$	$1.9±0.8^{a}$	$950±148^{bc}$
PY	$4.7±1.0^{ab}$	$7.8±5^{a}$	$72±13^{a}$	$4.7±3^{ab}$	$10.8±2^{a}$	$387±57^{bc}$	$40±15^{a}$	$139±32^{a}$	$5.8±3.8^{ab}$	$1.6±0.8^{a}$	$2.7±1.1^{a}$	$1.4±0.3^{a}$	$690±168^{bc}$
CR_2	$3.5±0.7^{bc}$	$7.2±3^{a}$	$55±1^{a}$	$2.7±1^{a}$	$12±6^{a}$	$858±103^{a}$	$28±3^{a}$	$108±9^{a}$	$3.3±0.1^{a}$	$1.1±0.1^{a}$	$2.9±0.9^{a}$	$1.3±0.6^{a}$	$1079±127^{a}$
CM	$2.1±0.7^{bc}$	$7.8±1^{a}$	$78±19^{a}$	$9.8±1^{c}$	$16.2±5^{a}$	$445±136^{b}$	$36±2^{a}$	$137±9^{a}$	$11±1.4^{c}$	$0.8±0.4^{a}$	$3.2±1.1^{a}$	$2.5±2.1^{a}$	$745±179^{ab}$
PH	$1.5±0.2^{c}$	$5.2±2^{a}$	$53±3^{a}$	$7.9±1^{bc}$	$10±2^{a}$	$203±10^{c}$	$20±2^{a}$	$12±4^{a}$	$8.9±0.2^{bc}$	$1.4±0.4^{a}$	$2.7±0.3^{a}$	$1.6±0.5^{a}$	$434±23^{c}$
Ave	3.7±2	6.7±1.3	72±17	5.6±3.2	12±2.4	514±257	34±5	124±14	6.4±3	1.1±0.3	2.7±0.5	1.6±0.6	780±247

Table II. Emission factors (EFs) of $PM_{2.5}$ and its elemental composition

[a,b & c] Same superscripts of each group are showing similarity while different superscripts indicate significantly ($p < 0.05$) difference between means of group.

The highest EF of K (858 ± 103 g/kg$_{dry\ biomass}$) is found in biomass from CR province in the second sampling location (CR_2), while the highest $PM_{2.5}$ EF observed in the first sampling location of CR (CR_1) biomass samples. $PM_{2.5}$ of the rice straw from PH produced the least EF of K. The trend was similar to the $PM_{2.5}$ EF comparing to biomass from other locations, though other elements did not show the same trend. In composition of $PM_{2.5}$ emitted from PH samples, EF of K was high in RS since rice crops uptake large amounts of potassium (K), which is mainly supplied from inorganic fertilizer [35], therefore K contain rice straw samples of PH possibly reduced in the open area and resulted to lower $PM_{2.5}$ EF as well. EFs of potassium from all location in descending were CR_2 > CR_1 > Cm > PY > PH while $PM_{2.5}$ EFs were CR_1 > PY > CR_2 >Cm > PH (Table II). Decrease in EF of K in all location samples resulted to increase in EF of Na and similar trend exist among K and Magnesium (Mg), while this relation was not observe for other major elements (Ca, Fe and Al).

Emission factor of sodium (Na) was the second foremost EF studied, the highest EF for Na (139±32 g/kg$_{dry\ biomass}$) observed in composition of $PM_{2.5}$ emitted from PY biomass samples which was followed by CM, CR_1, CR_2 and PH provinces respectively, while Similar to K the EF of Na in MHS samples was significantly lower comparing to other samples.

The third leading EF of elements in the samples found was EF of calcium (Ca). The results indicates that sample from CR_1 location in CR province has higher EF of Ca comparing to samples from other locations, and lowest EF was found in PH province samples. After three mentioned elements EF of Mg is the fourth dominant, and it followed the same trend as of EF of Na. In case of heavy metals, EF of Fe is considerably high in biomass samples from all locations while the highest Fe EF (16.2±4.7 g/kg$_{dry\ biomass}$) was found in CM samples, and followed by EFs of Sc, Cr, Sn and Zn respectively, alike to Fe all heavy metals EFs were higher in CM samples . Eventually Si showed the lowest EF almost in all $PM_{2.5}$ samples. Though EFs of measured elements are lower comparing to some previous literatures [31] but consistency of EFs calculated during this study were very high, and low standard deviation from means similarity of EFs testify the accuracy and precision of this work. Most of measured trace elements via this research are higher or similar to similar previous studies while some lower [31]. The EF of traced element we measured are are higher comparing to trace elements studied in composition of PM10 emitted from RS [36].

Comparing total EFs of elements from each location showed that highest cumulative EF is related to CR_2 (1079 mg/kg$_{dry\ biomass}$), while CR_1 average $PM_{2.5}$ EF was as larger as twice of $PM_{2.5}$ EF of CR_2. In addition EFs of other locations from high to low were CR_1 > CM > PY > PH which were 950, 745, 690 and 437 (g/kg$_{dry\ biomass}$), respectively. Ratio of cumulative EFs of elements of each location and EF of $PM_{2.5}$ from RS is higher for CM (36 %) and followed by CR_2 (31 %), PH (29 %), PY (15 %) and CR_1 (14 %). EFs of elements were independent from EF of $PM_{2.5}$ and were not showing the same trend as EF of $PM_{2.5}$ from RS.

The overall element contribution in composition of $PM_{2.5}$ emitted from rice straw and mass percentage of emitted $PM_{2.5}$ and all elements are given in fig. 2).

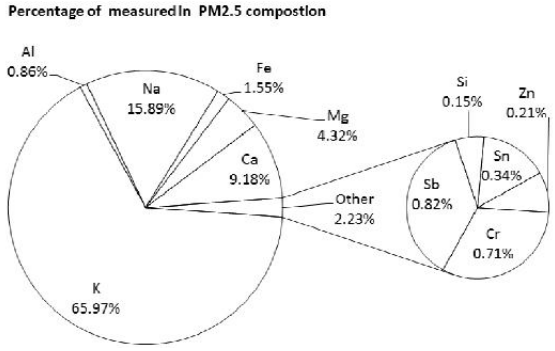


Fig. 2 Percentage of measured elements in $PM_{2.5}$ composition.

The trace elements are categorized into two major and minor groups of elements by two interrelated pie charts. EF of K covered 66% of EFs of all elements in the composition $PM_{2.5}$ which was followed by Na, Ca, Mg, Fe, Al, Sb, Cr, Sn, Zn and Si. EF of heavy metals were considerably high in the samples, that have potential to harm human, affect environment and reduce air quality of atmosphere.

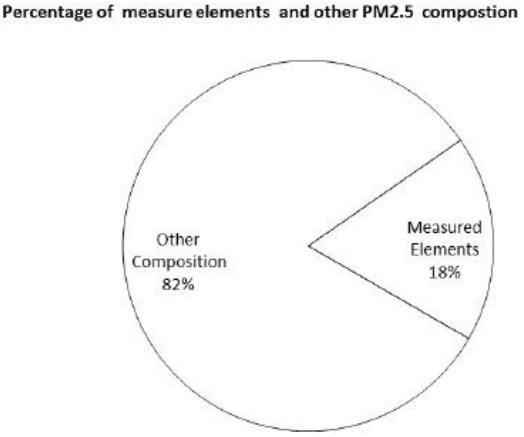


Fig. 3 Contribution of elements mass percentage of $PM_{2.5}$.

Detected and measured elements making 18% of aggregated mass of $PM_{2.5}$ emitted from rice straw, but remaining 82 % $PM_{2.5}$ mass was consist other constituents beyond measured elementst (Fig.3).

C. Statistical analysis

Analysis of variance performed to compare means of EF of $PM_{2.5}$ and EFs of elements. EF of $PM_{2.5}$ from CR_1 was significantly different from CM and PH ($p < 0.01$) and different from CR_2 and PY ($P < 0.05$), and with 95% of confidence level EF of $PM_{2.5}$ from PH and PY were different, while EF of $PM_{2.5}$ of CR_1 with PY, and EF of $PM_{2.5}$ from CR_2, CM and PH were not significantly deferent. With a confidence level of 99%, the EF of K was significantly different between CM and CR_2, CR_2 with PH and PY and PH with CR_1, while other pairs were not significantly different. Furthermore, EF of Cr was significantly ($p < 0.01$) different from Cr emitted from RS of CR_1 , CR_2 and PY, EF of Cr from PH was also different from EF of Cr from CR_1 and CR_2 and with confidence level of 95%. EF of Sb from RS showed exactly the same trend as EF of Cr for all locations, finally EFs of Al, Ca, Fe, Mg, Na, Sb, Si, Sn and Zn were not statistically different from each other between studied locations.

Pearson correlation was used to assess the covariance of EFs after transforming data to normal data. some significant ($P<0.05$ and 0.01) correlation between $PM_{2.5}$ EF and EFs of some elements as well as inter elements correlation observed. A strong correlation observed between EFs of $PM_{2.5}$ and K ($r = 0.62$) EFs $PM_{2.5}$ and K showed negative strong correlation Sb ($r = -0.62$) $p < 0.05$ and similarly with EFs of Cr ($r = -0.64$, and $p <0.01$) therefore emission factor of $PM_{2.5}$ was directly proportional to EF of K in the RS biomass residues and Sb negatively affect emission of $PM_{2.5}$ while Cr had very high adverse impact, as both Sb and Cr reduce EF of K from RS burning, these correlations were significantly shown among K, Sb and Cr with amount of ash in adverse order, further more Cr and Sb were very strongly correlated ($r = 1.00$ and $p < 0.01$), Al had strong correlations with Ca, Cr, Fe, Mg, Na and Si ($r= 0.53$-0.63), furthermore, EF of Ca was strongly correlation with EFs of Mg and Na ($r=0.90$, 0.87 respectively), while Sn EF was moderately correlated to Si ($r=0.52$), and finally EF of Zn was correlated only to EF of Fe ($r=0.47$), despite these elements had very significant ($P<0.5$ and 0.01) and strong correlations but EF of $PM_{2.5}$ is independent from EFs of these elements and have no impact on EFs $PM_{2.5}$ from RS except EFs of K, SB and Cr.

CONCLUSION

This study was designed to evaluate $PM_{2.5}$ EF of rice straw and investigate the composition of sampled $PM_{2.5}$ over Northern Thailand. The $PM_{2.5}$ EF varied considerably between biomass of various location depending on range of factors such as length of crop post-harvest duration for sampling and meteorological condition during open burning experiments that resulted to fluctuation of $PM_{2.5}$ EF between biomass samples from different location. Average $PM_{2.5}$ EF from rice straw was 3.7±2.0 ($g/kg_{dry\ biomass}$) and $PM_{2.5}$ EF was significant ($P<0.05$) correlated to K while negatively correlated to Cr ($P<0.01$) and Sb ($P<0.05$), furthermore EFs of Ca, Na and Mg were strongly correlated to each other ($P < 0.05$ and 0.01) elements. EF of elements from $PM_{2.5}$ details that K was the major element with EF of 514±249 ($mg/kg_{dry\ biomass}$) that comprised 66% of overall detected elements and other elements EFs in descending order are Na (124 ±20) > Ca (67 ± 21) > Mg (38 ± 8) > Fe (12 ± 4) > Al (7 v 3) > Sb (6 ± 3) > Cr (6 ± 3) > Sn (3 ± 1) > Zn (2 ± 1) and Si (1 ± 0.5). additionally in contradict with $PM_{2.5}$ EFs, ratio of measure elements total mass and emitted $PM_{2.5}$ mas was higher for CM (0.36 or 36 %), while accumulative EFs of elements was higher for CR_2 (1079 $mg/kg_{dry\ mass}$) A wide range of EFs obtained by this study suggests that care should be taken to reduce EFs from rice straw open burning since $PM_{2.5}$ EFs of RS was high, and emitted $PM_{2.5}$ was a complex mixture of various metals and potentially harmful to human health and environment and EFs of RS.

ACKNOWLEDGEMENT

The authors would like to thank Thailand International Cooperation Agency (TICA).

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