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Original Article

Emissions Factors of Air Pollutants from Rice Straw Burning-hood Experiments

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Abstract: The burning of rice straw produces a significant amount of particulate matter (PM) and gaseous air pollutants on a regional and global scale. In this study, the hood experiments were conducted to investigate the emission of air pollutants from rice straw burning (RSB). Samples of PM were collected isokinetically following the U.S. EPA methods 1, 1A, and 5. Gaseous pollutants were directly measured using a flue gas analyzer (Testo 350 XL). Emission factors (EF) ($g.kg^{-1}$) determined for PM, CO_2, CO, and SO_2 were 17 ± 3.8, 1399 ± 228, 68 ± 22, and 1.5 ± 0.4, respectively meanwhile NO and NO_2 were not detected in the flue gas. It was observed that flaming is the main phase in the process of RSB. The total emission from rice straw burning of 13 provinces in the Mekong River Delta of Vietnam in 2020 was estimated using the EF obtained from the hood experiments. The result shows that Kien Giang, An Giang, and Dong Thap were the high emission group in the Mekong River Delta, contributing 19%, 17%, and 14% to total emissions. The results of this study provide a scientific basis for further studies to determine EF from rice straw open burning in the fields and find sustainable measures to control this activity.

Keywords: Rice straw burning, emission factor, hood experiment, Mekong River Delta, emission estimation.

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1. Introduction

Global rice production is mainly concentrated in Asia, accounting for nearly 90%, of which Vietnam is the fifth largest rice-producing country in the world [1]. According to the General Statistics Office, in 2020, rice production of 42.8 million tonnes would produce about 55.6 million tonnes of rice straw (ratio 1:1.3), 80-90% of which was burnt [1, 2]. The Mekong River Delta is a significant rice-producing area in Vietnam (23.8M tonnes in 2020) and is the region with the highest percentage of rice straw burning in the country. Rice straw burning (RSB) emits large amounts of air pollutants, including particulate matter (PM), gaseous pollutants such as CO_2, CO, NO, NO_2, and SO_2, which can cause significant impacts on the air quality and human health [3]. To assess the impacts of RSB, the emission factor (EF) must be determined. EF is the mass of a pollutant emitted per unit of mass or volume of the emission activities [4]. EF depends on many factors, such as fuel properties, combustion facilities, burning conditions, and experimental methods [5].

Currently, there are some methods to determine the EF of air pollutants emitted from RSB, in which carbon mass balance and laboratory measurement are the most popular methods [5]. The carbon mass balance method is usually conducted with uncontrolled burning in the field experiments done in Thailand, China, India, Nepal, Bangladesh, and North Vietnam [5-9]. The advantage of this approach is to provide a set of EF that reflect actual conditions for emission inventories more closely. However, this method requires a suitable emission sampling system and good monitoring techniques [5]. Therefore, studies in laboratory measurement are performed more commonly and accessible due to lower costs and available sampling techniques. In addition, laboratory measurements are essential for studying the formation of pollutants and emission mechanisms. They can validate influencing factors to the combustion process or provide a scientific basis for further determinations of EFs on-field scales.

In Vietnam, studies to build the EF database from biomass burning were carried out in the laboratory measurement in the Northern of Vietnam [8-12]. In contrast, such studies in the Mekong River Delta are scarce, except the study of Arai et al., 2014, which determined the EFs of CH_4 and N_2O [13]. Therefore, this study conducted experiments in the laboratory to calculate EFs of selected air pollutants with rice straw (RS) samples collected from An Giang and Hau Giang in 2018.

2. Materials and Methods

2.1. Fuel and Equipment

2.1.1. Fuel - Rice Straw

In this study, twelve samples of rice straw were taken from An Giang (six samples, coded from H.AG1 to H.AG6) and Hau Giang (six samples, coded from H.HG1 to H.HG6) in the winter-spring crop in 2018 (Figure 1). Samples of rice straw were collected 3 to 5 days after harvest. Then, the samples were kept in plastic bags, vacuumed to minimize moisture loss, and transported by plane to the School of Environmental Science and Technology (INEST) laboratory, Hanoi University of Science and Technology, where the hood experiments were conducted.

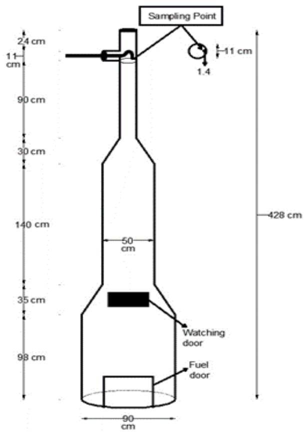


Figure 1. Hood and sampling port.

2.1.2. Specifications of the Burning Hood

A hood was designed and installed in INEST elaborate tory following the method presented in the previous studies from Thailand and Vietnam [10, 14]. A sampling of PM was done at a port in the hood determined according to the US.EPA method 1A ensures the flue gas's uniform flow and representative sampling [15]. The set-up was made to imitate as closely as possible the in-situ field burning. The description of the hood is shown in Figure 2.

2.2. Monitoring

2.2.1. Before burning

Before burning, all target parameters, including PM, CO_2, CO, NO, NO_2, and SO_2 in the background, were determined to calculate the net contribution of the emission from RSB. While PM was sampled by a sampler (C5000, Thermo Andersen), CO_2 was directly measured by Lutron GCH–2018, and other gasses were directly measured using the Multiwwarn II monitor. In addition, the carbon (C) content of RS was determined by the ASTM E777 method (Standard Test Method for Carbon and Hydrogen in the Analysis Sample of Refuse-Derived Fuel) [8], and S content in RS was determined by ASTM-E775–87: 2004 and EPA 5050. At the same time, the moisture of RS was analyzed by the oven-drying method.

2.2.2. Burning

Each RS sample of 5-7 kg was divided into a smaller amount of 0.7–1 kg (sub-windrow) for each hood sampling batch. Each sub-windrow was placed in a tray of 20×30 cm^2, and the trays were continuously fed. The burning of rice straw was conducted with a natural air supply through an opening door of the hood. The ignition was done from the bottom of the fuel bed. PM was collected isokinetically using the sampler (C5000, Thermo Andersen, ESC-American) according to the U.S. EPA method 5 [16]. During the sampling, the filter box and the sampling probe were heated at 120 ± 14°C. After sampling, the mass of PM was determined by the gravimetric method. Gaseous pollutants (CO, CO_2, NO, NO_2 and SO_2) were directly measured using a flue gas analyzer (Testo 350 XL) every 7 mins during burning. Before sampling, all the sampler parts in contact with the flue gas were properly cleaned and rinsed with acetone twice. The sampling time started from the moment of stable flame to the end of the burning process. And then, the ash and unburnt rice straw were collected to determine their carbon content. The procedure of experiments in the laboratory is presented in Figure 3.

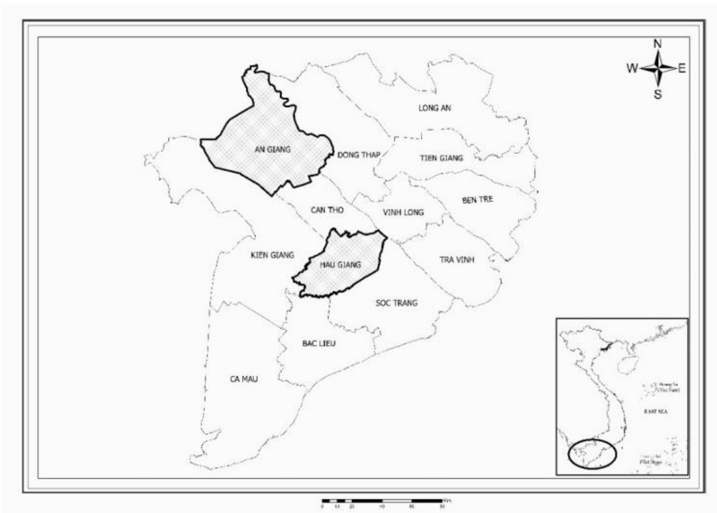


Figure 2. Map of provinces collected rice straw samples.

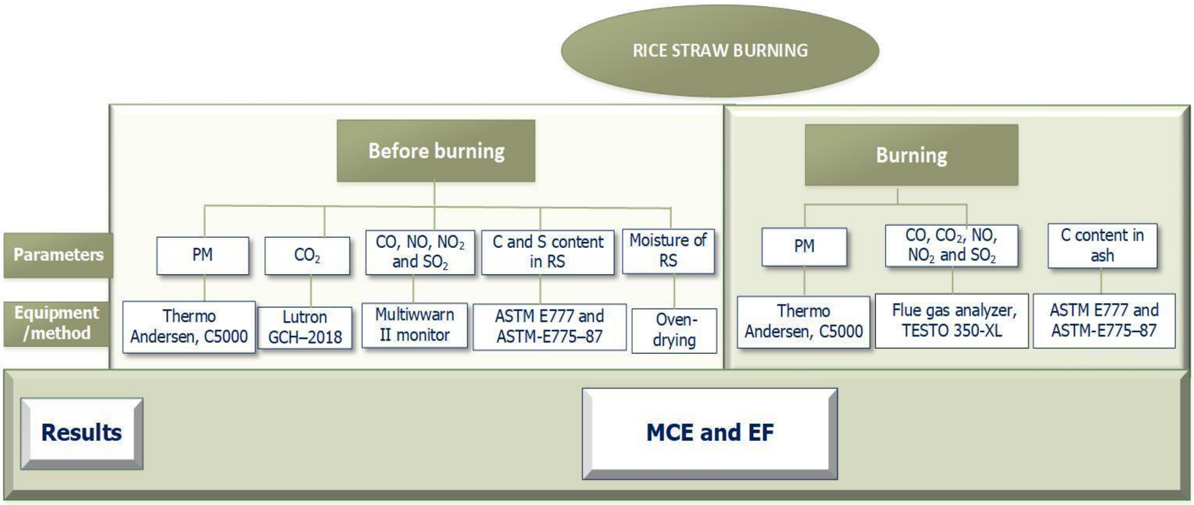


Figure 3. The procedure of experiments in the laboratory.

2.3. Method for the Determination of Emission Factors

Emission factors of air pollutants were calculated using equation (1) [17, 18]

$$EFi = \frac{Mi}{M} \quad (1)$$

Where EF_i is the emission factor of pollutant i (g/kg or mg/kg); M_i is the total mass of pollutant i emitted during burning (g or mg); M is the total mass of RS (dry basis) *burnt in each burning experiment (kg).*

Combustion efficiency (CE) is the ratio of carbon mass released in terms of CO_2 to the total mass of carbon released during combustion [19]. Therefore, it may be used to determine the completeness of carbon oxidation during the combustion of biomass fuels. Alternatively, if only CO_2 and CO are measured, the modified combustion efficiency (MCE) was used to distinguish the flaming phase from the smoldering stage during burning. The MCE can be calculated using the equation (2):

$$MCE = C_{CO2}/(C_{CO2}+C_{CO}) \quad (2)$$

2.4. Emissions Inventory

The annual emission of any air pollutant from RSB is estimated as follows [12, 20]

$$E_i = [P \times N \times B \times D \times C] \times EF_i \quad (3)$$

Where, E_i is the annual emission of pollutant i (Gg/year); P is the yearly paddy production (Gg/year); N is the rice straw-to production ratio; B is the fraction of RS that is burned in the field (0 -1); D is the dry matter-to-RS ratio (fraction 0-1); C is the combustion efficiency (the fraction oxidized during combustion); EF_i is the emission factor of pollutant species i (g kg^{-1} dry rice straw).

2.5. Statistical Analysis

The primary data was processed and analyzed using Sigma Plot 14 to obtain their average value, range, standard deviation, and standard error. All experimental data were expressed as means ± standard deviations. The relation between emission factors of air pollutants and moisture contents of RS was determined by Pearson's correlation analysis using the SPSS software package (IBM SPSS Statistics V20) and expressed at $p < 0.05$ and $p < 0$.

3. Results and Discussion

3.1. Burning Characteristic

RS varieties in the hood experiments in this study were currently prevalent in the Mekong River Delta of Vietnam, such as OM42128 variety at Hau Giang provinces and IR50404 variety at An Giang province. The carbon content in dry RS ranged from 34.3 to 48.2%, in which the IR50404 at H.AG3 contained the highest C content of 48.2% (Table 1). The average carbon content found in this study was 41.7 ± 5.5%. These results are similar to those reported by Jenkins et al. (1996) (38%) but lower than that of Kim Oanh et al., (2011) (49.0 ± 2.7%). Differences in carbon content in combustion samples can affect the emissions factor of carbon-containing compounds such as CO, CO_2, organic compounds [9].

Otherwise, the average sulfur content of RS in this study was 0.19 ± 0.05%, which is higher than that in California (0.09%) [21] and China (0.17%) [22]. The higher sulfur content in RS in this study may be due to using high-dose fertilizers with high sulfur content, which is commonly used in Vietnam [8]. In addition, machine harvesters, which lead to leakage of diesel oil with 0.005% of sulfur content for the harvest, may lead to higher S content in rice straw compared with other studies. However, this point needs to be confirmed in further studies.

The moisture content of fuel influences the flame residence time, the duration of smoldering combustion and thus affects MCE and emission factors of pollutants [23]. In this study, the moisture of RS ranged from 8.5 to 16.5% (Table 1). As a result, MCE in all experiments was higher than 0.9 (from 0.92 – 0.97), demonstrating that all invested experiments' combustion stage is flaming. However, it is noted that the ratios of CO_2 to (CO+ CO_2) in real-time emission fluctuate, including values < 0.9 and > 0.9. Therefore, the combustion state of burning is both flaming and smoldering in which the flaming is dominated.

Table 1. Rice straw characteristics and burning condition

Samples ID (n = 12)	C content of RS (%)	S content of RS (%)	Moisture of RS (%)	Burning duration (min)	Burning rate (kg/h)	MCE
H.HG1	34.3	0.27	13.5	73	5.7	0.94
H.HG2	34.5	0.27	14.0	73	5.7	0.94
H.HG3	35.9	0.16	9.0	57	6.0	0.97
H.HG4	35.9	0.12	9.5	65	5.5	0.96
H.HG5	40.2	0.15	9.0	60	6.0	0.96
H.HG6	40.2	0.15	8.5	57	6.2	0.97
H.AG1	47.4	0.11	14.0	73	6.0	0.96
H.AG2	47.4	0.16	14.0	77	5.8	0.96
H.AG3	48.2	0.29	16.0	82	5.8	0.95
H.AG4	47.8	0.29	16.5	85	5.6	0.92
H.AG5	43.3	0.17	11.5	72	5.8	0.96
H.AG6	45.1	0.10	10.5	61	6.0	0.96
Mean ± STD	41.7 ± 5.5	0.19 ± 0.05	12.2 ± 2.9	70.2 ± 9.1	5.9 ± 0.2	0.95 ± 0.01

3.2. Emission Factors of Particulate Matter and Gaseous Pollutants

The EFs of PM from the RSB are presented in Table 2. The mean value of EF_{PM} was 17 ± 3.1 g.kg-1, while EF_{PM} in An Giang (18.6 ± 2.9 g.kg-1) is higher than that in Hau Giang (15.4 ± 2.7 g.kg-1). Table 1 shows that the moisture of RS in An Giang (13.8%) with IR50404 variety is higher than in Hau Giang (10.6%) with OM4218 variety. The results also revealed the difference of varieties and moisture of RS lead to the difference in EF_{PM} (Table 2). These results are close to previous studies in America [8, 21] and the North of Vietnam [8, 21].

Table 2 shows that EFCO2 is the primary carbon compound emitted by rice straw burning, at the burning rate of 5.9 ± 0.2 kg/h (Table 1). Overall average $EFCO_2$ (1399.1 ± 227.8 g kg-1, n=12) from this study (Table 2) are consistent with previous research in America, China, and the North of Vietnam [9, 10, 24]. However, it noted that CO2 formed from the burning of rice straw is the prompt CO2. In the fact that CO2 emissions represent the immediate release of CO2 to the atmosphere, the net release is about one-third to one-half of the former due to the uptake by plants covering the post-burned areas. Therefore, it was interesting that CO2 was the main combustion product from rice straw. Still, the CO2 was considered neutral to the greenhouse gas effect, and this is regarded as the environmental benefit of rice straw burning [25]. This study's range of CO/CO2 emission ratios covers most of the previously published results. Most of these ratio values are between 4% and 15% [8, 9], so this study means the value of 5 ± 2%, derived from complete combustion, is a highly reliable value. Our results show that CO is the second but most crucial carbon emission with the mean of EFCO was 68.2 ± 22.1 g.kg-1 (Table 2). This EF is in reasonable agreement with published values which were ranged from 53.3 to 92 g.kg-1 for CO [8, 9, 19, 24].

.Table 2. EFs of PM and gaseous pollutants from *RSB* and comparison with other studies

Samples ID (n = 12)	EFs of pollutants ($g.kg^{-1}$ dry RS)			
	PM	CO	CO_2	SO_2
H.HG1	18.5	77.3	1315.3	1.2
H.HG2	18.7	88.8	1410.5	1.7
H.HG3	12.6	43.9	1299.1	1.6
H.HG4	15.4	56.8	1493.9	1.1
H.HG5	14.6	46.1	1155.2	1.1
H.HG6	12.9	37.4	1238.4	1.3
H.AG1	17.0	71.2	1578.8	1.5
H.AG2	20.5	78.9	1823.0	2.0
H.AG3	22.2	94.6	1790.3	2.2
H.AG4	20.5	108.5	1171.8	2.2
H.AG5	16.7	58.4	1287.3	1.4
H.AG6	14.6	55.8	1225.5	1.0
Mean ± STD	17.0 ± 3.1	68.2 ± 22.1	1399.1 ± 227.8	1.5 ± 0.4
Other studies	10.2 ± 8.5 [8]; 17 ± 0.65 [21]; 13 [9].	53.3 ± 16.6 [8]; 64.2 ± 4.9 [19]; 67.98 ± 25.58 [24]; 92 ± 84 [9]; 72 ± 12 [10].	1233.8 ± 185.9 [8]; 791.3 ± 12.5 [19]; 1674.12 ± 452.26 [24]; 1515 ± 117 [9]; 1465 ± 261 [10].	1.82 ± 1.77 [8]; 0.18 ± 0.31 [24]; 0.25 ± 0.045 [10].

Sulfur dioxide was also emitted with significant emission factors in these experiments, and it was a product of flaming combustion [9]. The mean of EF_{SO2} observed in this study is 1.5 ± 0.4, which is comparable to those compiled by Andreae and Merlet (2019) (0.88 ± 0.92 $g.kg^{-1}$) [23]. However, EF_{SO2} depends on the fuel S content in RS and combustion behavior [23]. In this research, H.AG3 and H.AG4 experiments with EF_{SO2} are the highest (2.2 $g.kg^{-1}$), corresponding to the highest S content (0.29%). In contrast, the H.AG6 experiment has EF_{SO2} is the lowest (1 $g.kg^{-1}$) corresponding to S accounting for the lowest percentage in combustion experiments (0.1%). Otherwise, EF_{SO2} from these experiments is also associated with the flaming phase. Furthermore, negative correlations between EF_{SO2} and MCE (r=-0.457, Table 3) with more SO_2 emitted from burns with the smoldering phase.

Besides, the concentration of NO and NO_2 was also measured to determine the corresponding EFs. However, NO and NO_2 in the flue gas were not detected. It means that the concentration of NO and NO_2 was lower than the limit of detection (1 ppm). Nitrogen oxides in combustion gases are usually thermal, prompt, and fuel nitrogen oxides, while thermal nitrogen oxides are the most significant. However, based on estimates on the thermal mechanism alone, zero NO_x would be produced at temperatures below 1300°C [26]. It notes that the temperatures are barely high enough without preheating, so a mechanism for the formation of the thermal NO is negligible. This reason agrees with the results of Dung, N.T and Thang, N.V, (2011), which uses the same hood and combustion method. Otherwise, the RS mainly consists of cellulose, which has little nitrogen in the fuel (0.71 – 0.87% N content of RS) [21, 27]. So, fuel NO and prompt NO formation are also trivial, which may cause the concentration of NO in the flue gas to be deficient. The NO_2 in the flue gas would be much lower than 1ppm because it only accounts for 10-20 % in a total concentration of NO_x (NO + NO_2) [25].

The result from Table 3 shows the negative correlation between MCE and EF_{PM} (r=-0.686, p<0.05), similar to findings of other studies on emissions of rice straw. A laboratory-based study by Hosseini *et al.* (2013) also observed a strong negative relationship (R^2 =-0.8) between EF_{PM} and MCE [28]. Strong positive correlations between emissions of particulates and other substances, such as EF_{CO} and EF_{SO2}, were also found in this study. The results of correlation analysis between emission factors of air pollutants and moisture content of RS are presented in Table 3. The strong positive correlations between moisture content of RS and EF_{CO} (r=0.967, p<0.01), EF_{PM} (r=0.938, p<0.01), and EF_{SO2} (r=0.768, p<0.01). These results are in good agreement with the previous reports [8, 29]. In addition, the significantly positive correlations between EF_{PM} and EF_{CO} (r=0.926, p<0.01), EF_{CO2} (r=0.589, p<0.05) and EF_{SO2} (r=0.753, p<0.01) were also observed in this study.

Table 3. Correlation of emission factors of air pollutants and moisture content of RS

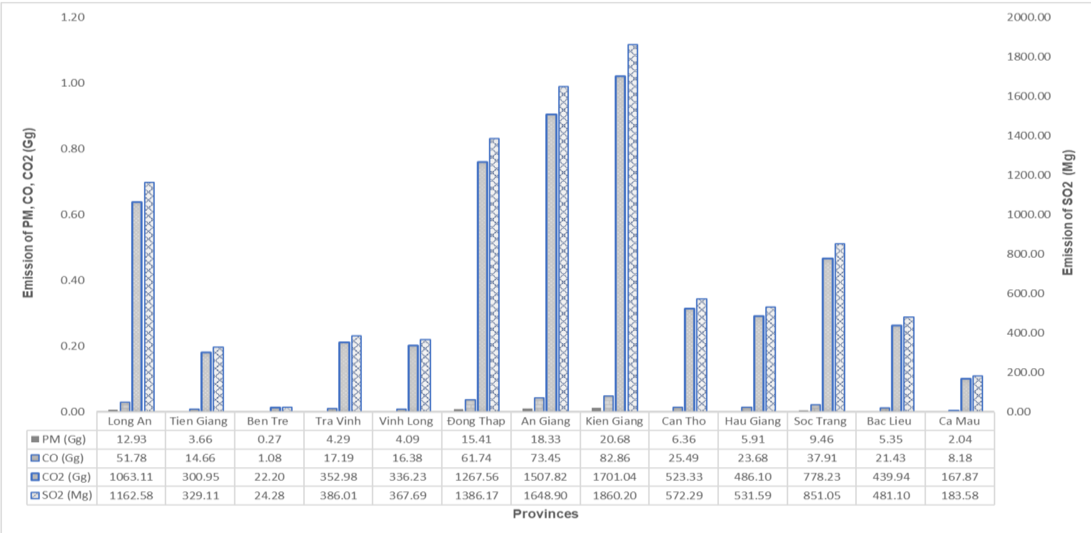
	The moisture content	MCE	EFs			
			PM	CO	CO_2	SO_2
The moisture content	1					
MCE	-0.76^{**}	1				
EF_{PM}	0.94^{**}	-0.69^{*}	1			
EF_{CO}	0.97^{**}	-0.87^{**}	0.92^{**}	1		
EF_{CO2}	0.47	0.11	0.59^{*}	0.37	1	
EF_{SO2}	0.77^{**}	-0.46	0.75^{**}	0.75^{**}	0.52	1

3.3. Emission Estimation

Figure 4 presents the emission estimate for different pollutants from rice straw burning in the 13 provinces in the Mekong River Delta of Vietnam for 2020. According to Equation (3), the annual paddy production (Gg/year) was extracted from the statistical yearbook 2020 [30], N (rice straw-to-production ratio) is 1.3, C (combustion efficiency) is 0.95; D (dry matter-to-RS ratio) is 0.23, and EF_i of pollutant species are from above findings (Table 2). B could be determined by using a top-down approach, which refers to the use of satellite observations, often combined with the models to estimate emissions) or a bottom-up approach, which is based on emissions calculations from all the individual sources (for example interview method...). In this study, the bottom-up approach was applied; B is inherited from the data of published studies, B has an average value of 0.85 [1, 31, 32].

The result shows the ranking of provinces in the Mekong River Delta that encountered emissions from rice straw burning is ranking from 22.2 to 1701.04 Gg for CO_2, 1.08 to 82.86 Gg for CO, 0.27 to 20.68 Gg for PM, and 0.02 to 0.53 Gg for SO_2. The percentage of emissions by region found that Kien Giang, An Giang, and Dong Thap were the high emission group of Mekong River Delta, contributing 19, 17, and 14% of all emissions, respectively. These provinces have the most significant rice production in the Mekong River Delta.

According to (Eq. (3)), air pollutant emissions are dependent on the amount of rice straw open burning. We, therefore, calculated based on the average burning ratio of previous studies in the Mekong River Delta and the survey results of farmers that were published in the ministerial project titled "Determination of the greenhouse gas emission factors from open burning of agricultural by-products (rice husks and rice straw) in Southwest Vietnam" Code number: TNMT.2017.05.18. The second factor that can cause uncertainty in the emission calculation is the rice straw-to-production ratio (N). Therefore, the determined amount of rice straw generated per 1 m^2 was determined by repeating three times at three different sampling locations).



	Long An	Tien Giang	Ben Tre	Tra Vinh	Vinh Long	Đong Thap	An Giang	Kien Giang	Can Tho	Hau Giang	Soc Trang	Bac Lieu	Ca Mau
PM (Gg)	12.93	3.66	0.27	4.29	4.09	15.41	18.33	20.68	6.36	5.91	9.46	5.35	2.04
CO (Gg)	51.78	14.66	1.08	17.19	16.38	61.74	73.45	82.86	25.49	23.68	37.91	21.43	8.18
CO2 (Gg)	1063.11	300.95	22.20	352.98	336.23	1267.56	1507.82	1701.04	523.33	486.10	778.23	439.94	167.87
SO2 (Mg)	1162.58	329.11	24.28	386.01	367.69	1386.17	1648.90	1860.20	572.29	531.59	851.05	481.10	183.58

Figure 4. Emission of the pollutants at provinces in the Mekong River Delta in 2020

4. Conclusion

The EFs of PM and pollutant gasses (CO_2, CO, NO_x, and SO_2) from rice straw burning in the Mekong River Delta were investigated using the burning hood in the laboratory. The results show the average EFs of PM, CO_2, CO, and SO_2 were 17 ± 3.8, 1399 ± 228, 68 ± 22, and 1.5 ± 0.4 $g.kg^{-1}$, respectively. NO, and NO_2 are not detected in the flue gas. MCE of all burning experiments was higher than 0.9, so flaming is the main phase during RSB. The strong positive correlations between the moisture content of RS and EF_{CO}, EF_{PM}, and EF_{SO2} were found in this study. From EFs obtained in this study, the estimated total emission from rice straw burning of 13 provinces in the Mekong River Delta of Vietnam in 2020 was conducted. The result shows that Kien Giang, An Giang, and Dong Thap have a high emission contribution in the Mekong River Delta. The results would provide the necessary scientific basis for determining the emission factor in the field.

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