



Emissions of air pollutants from indoor charcoal barbecue



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HIGHLIGHTS

- Emission inventory of air pollutants established for charcoals used in Taiwan.
- High-risk air pollutants emitted from charcoal briquette combustion.
- Harmful PM_{2.5} was main particulate generated during charcoal combustion.
- More HCHO and C₂H₄O produced at flaming stage of charcoal combustion.

ARTICLE INFO

Article history:

Received 7 June 2015

Received in revised form

19 September 2015

Accepted 22 September 2015

Available online 26 September 2015

Keywords:

Indoor air quality

Barbecue

Formaldehyde

Acetaldehyde

Particulate matter

ABSTRACT

Ten types of commercial charcoal commonly used in Taiwan were investigated to study the potential health effects of air pollutants generated during charcoal combustion in barbecue restaurants. The charcoal samples were combusted in a tubular high-temperature furnace to simulate the high-temperature charcoal combustion in barbecue restaurants. The results indicated that traditional charcoal has higher heating value than green synthetic charcoal. The amount of PM₁₀ and PM_{2.5} emitted during the smoldering stage increased when the burning temperature was raised. The EF for CO and CO₂ fell within the range of 68–300 and 644–1225 g/kg, respectively. Among the charcoals, the lowest EF for PM_{2.5} and PM₁₀ were found in Binchōtan (B1). Sawdust briquette charcoal (I1S) emitted the smallest amount of carbonyl compounds. Charcoal briquettes (C2S) emitted the largest amount of air pollutants during burning, with the EF for HC, PM_{2.5}, PM₁₀, formaldehyde, and acetaldehyde being the highest among the charcoals studied. The emission of PM_{2.5}, PM₁₀, formaldehyde, and acetaldehyde were 5–10 times those of the second highest charcoal. The results suggest that the adverse effects of the large amounts of air pollutants generated during indoor charcoal combustion on health and indoor air quality must not be ignored.

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1. Introduction

Because of social change and urban crowding, people spend over 80% of their time indoors [1]. Under the fast development of industry and commerce, lifestyle and dietary habits of people also change quickly. Eating out has become quite common catering pattern in Taiwan. The self-service dining restaurants of buffet barbecue bloom in Taiwan in recent years [2]. Charcoal is made by slow thermal pyrolysis process, which burning wood or other materials in the absence of oxygen. Because charcoal has many advantages including higher energy content, easier to handle, burning evenly for a long time, easily to extinguish and reheat, and not liable to damage by insects and fungi, it has been widely used











in barbecue [3]. However, people will probably expose to emitted air pollutants directly from charcoal-fired appliance during barbecue in indoor. It was noted that combustion pollutants, particularly carcinogenic compounds, could cause health effects on barbecue customers under insufficient ventilation. Formaldehyde, related to nasopharyngeal tumors [4], classifying as carcinogenic to humans (Group 1) by International Agency for Research on Cancer [5]. Acetaldehyde was classified as possibly carcinogenic to humans in 1987 and confirmed in 1999 by IARC [6,7]. The levels of airborne formaldehyde are often 2~10-fold higher than those in outdoor air [8–10]. The common indoor noncombustion sources of formaldehyde and acetaldehyde are released from many industrial and consumer products; otherwise, they are also produced as byproducts from various combustion activities [11–13] such as cooking, smoking, heating, and incense burning.

Lee and Wang [14] burned mosquito coils in a chamber and found formaldehyde and acetaldehyde were the major carbonyls

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Table 1
Commonly used charcoals in Taiwan.

Charcoal ID	Charcoal name	Photo	Country source	Approximately profile
C1S	Eco-friendly charcoal		China	Brick shape
C2S	Charcoal briquettes		China	Oblate spheroid shape 4 cm in diameter
C3S	Charcoal briquettes		China	Oblate spheroid shape 4 cm in diameter
I1S	Sawdust briquette charcoal		Indonesia	Hexagon 4 cm in diameter and 1 cm diameter hollow core
I2	Mangrove charcoal		Indonesia	15–20-cm long and 3–6-cm in diameter
I3S	Charcoal briquettes		Indonesia	Oblate spheroid shape 4 cm in diameter
T1S	Eco-friendly charcoal		Taiwan	Quasi rectangular pyramid(4 × 4 × 7 cm)
T2	Acacia charcoal		Taiwan	Irregular shape 15 cm in diameter
T3	Longan charcoal		Taiwan	Irregular shape 15 cm in diameter
B1	Binchōtan		Southeast Asia	10–15 cm long and 2–4 cm in diameter

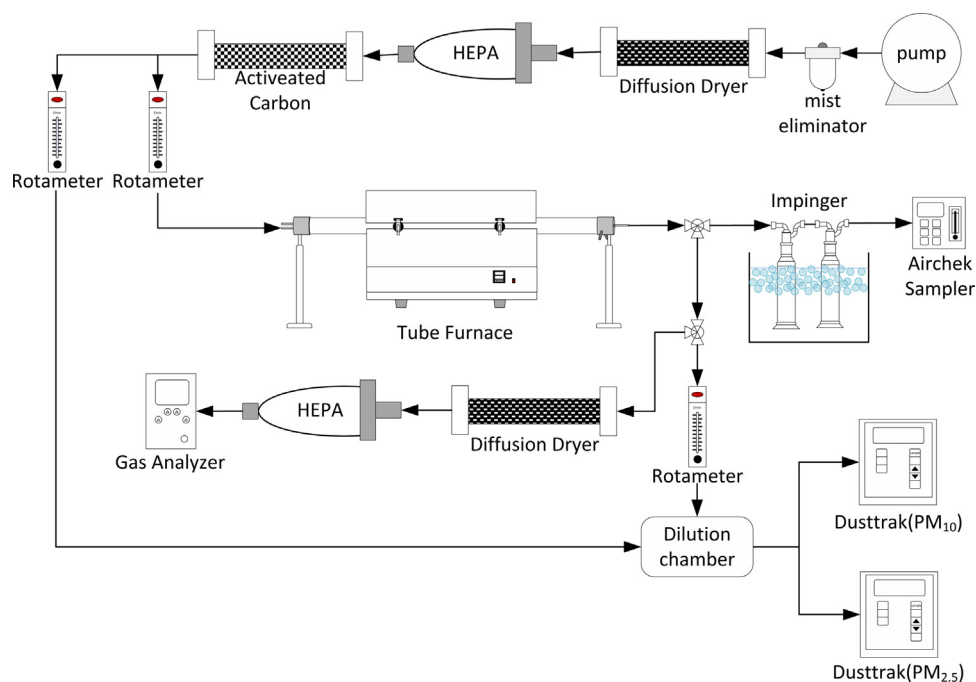


Fig. 1. Experimental system of charcoal combustion.

species presented in smoke. Manoukian et al. [15] indicated that the maximum concentrations of formaldehyde and acetaldehyde in an experimental room were produced immediately after burning of incense sticks. Huang et al. [16] found that the generated formaldehyde were almost the same by using towngas and liquefied petroleum gas as cooking fuels. However, acetaldehyde, the highest emitted carbonyl species, was only observed at the kitchen using towngas as cooking fuel. Carter et al. [17] investigated average emission factors of formaldehyde were 14.4 mg/kg and 8.0 mg/kg from wick and injection heater with five kinds of fuel respectively. The emission factors of formaldehyde were higher

than those of acetaldehyde for two kinds of heater. Cerqueira et al. [18] characterized the average concentration ratio of formaldehyde to acetaldehyde in the stove with burning of 5 kinds of wood was in the range of 2.1–2.9. Pyrenean oak produced the highest emissions for both formaldehyde and acetaldehyde and maritime pine produced the lowest emissions. Kabir et al. [19] showed that formaldehyde and acetaldehyde were the abundant among emitted carbonyl compounds from the combustion of commonly used barbecue charcoals produced from 4 countries in Korea. Rahman and Kim [20] also indicated that aldehydes were the most dominant odorant released during combustion of barbecue charcoals

produced from five countries. Their measured mean concentrations of emitted formaldehyde and acetaldehyde from charcoal combustion exceeded guidelines of Korean Ministry of Environment.

Carbon dioxide, monoxide, nitrogen oxide, hydrocarbons, metals, and particulate matter of different sizes are also commonly generated from indoor combustion activities. Many studies investigated emission factors of these combustion pollutants from fuel/stove combinations [21], biomass burning activity [22] wood and charcoal-fired cookstoves [23], trees-fired stove [24], residential wood combustion appliances [25], and wood-burning fireplace and stove [26]. These studies showed that emission factors of airborne pollutants during combustion distributed in a wide range because of different burning fuels, combustion appliances, combustion conditions, and experimental methods. Only few studies explored combustion pollutants emitted from burning of charcoals used for barbecue. Most barbecue restaurants are airtight with air-conditioners in Taiwan. A variety of air pollutants emitted during charcoal barbecue could be accumulated in indoor, worse indoor air quality, and harm diners under inadequate ventilation. In order to understand the potential hazard to people resulted from air pollutants produced in charcoal BBQ restaurants, air pollutants released from combustion of commonly available charcoals in Taiwan under different temperatures and airflow conditions were explored in this study.

2. Material and methods

2.1. Experimental design

To understand the influence of air pollutants generated from indoor charcoal combustion on indoor air quality, 10 kinds of commercial common charcoal in Taiwan were chosen to perform combustion experiments (Table 1). Charcoals were burned in a tube furnace under different temperature and airflow conditions to simulate charcoal combustion at high temperature during barbecue. Charcoals used in this research were all cut into smaller than 40 mm for the longest side and greater than 20 mm for shortest side. Charcoal was conditioned at room temperature and relative humidity below 50% for 24 h in a drying cabinet before combustion. The conditioned charcoal of 30 ± 0.5 g for each batch combustion experiment was burned at setting temperature and airflow rate. The total sampling time of combustion airborne pollutants was 60 min. The charcoal was flaming at first 20 min and smoldering at the following 40 min. After 1 h, the charcoal fire was put out. The residual unburned charcoal was conditioned again and weighted. Each combustion experiment was performed in triplicate. The emitted air pollutants from charcoal combustion including $PM_{2.5}$, PM_{10} , HC, CO, CO_2 , NO_x , formaldehyde and acetaldehyde were examined in this study. Finally, emission rates and emission factors of these combustion pollutants were calculated based on the monitoring results.

2.2. Experimental system

Fig. 1 shows the experimental system of charcoal combustion. At first, a pump sucked outdoor air into a mist eliminator and diffusion dryer to remove moisture. The dry air then flowed into a high efficiency particulate air (HEPA) filter to filter particulate matters. Finally, zero air was generated by using an activated carbon tube to further remove organic compounds from the outlet air of HEPA. The generated clean air was performed blank analysis before each experiment to ensure its cleanliness. Zero air was continuously sent into the combustion device (Model D-55, Deng Yng instrument Co., Ltd., Taiwan) to perform charcoal combustion test. The designed air flow rates were 10L/min and 20L/min corresponding to the

Table 2
Specifications of portable direct-reading devices for flue gas monitoring.

Pollutant	HC (as n-hexane)	CO	CO_2	NO_x (as NO)	$PM_{2.5}/PM_{10}$
Model	HM5000	HM5000	HM5000	HM5000	DustTrak 8520
Manufacture	Infrared Industries	Infrared Industries	Infrared Industries	Infrared Industries	TSI
Examine principle	Non-dispersive infrared (NDIR)	Non-dispersive infrared (NDIR)	Non-dispersive infrared (NDIR)	Electrochemical	Light scattering, laser diode
Measuring range	0–100000 ppm	0–10%	0–20%	0–5000 ppm	0.001–100 mg/m ³ –10 μm (size)
Resolution	1 ppm	0.01%	0.01%	1 ppm	0.001 mg/m ³
Accuracy	±2%	±2%	±2%	±2%	±1% to calibration aerosol
Response time	2 s	2 s	2 s	2 s	Adjustable from 1 to 60 s

flow velocities of 8.5 cm/s and 17 cm/s, respectively. Both the air velocities were satisfied the indoor air flow standard in excellent grade (<20 cm/s) recommended in the indoor air quality guideline of Hong Kong.

The combustion device (250 × 350 × 400 mm) was consisted of tube furnace, quartz tube, front and back stainless steel doors of furnace, and quartz carrier. The diameter and effective heating length of the tube furnace were 55 mm and 300 mm. The internal diameter, external diameter, and length of quartz tube were 50 mm, 53 mm, and 1000 mm, respectively. The quartz carrier was a half-cylinder with 35 mm in external diameter and length 154 mm. The conditioned charcoal was loaded in quartz carrier before combustion. In general, the ignition and burning temperature of charcoal was about 350 °C and 400–600 °C. The actual ignition and burning temperature will differ with carbon content of charcoal. Therefore, combustion temperatures were set at 450–550 °C to explore the differences of emitted air pollutants during charcoal combustion in this study. The flue gas was exhausted from the combustion device and cooled by flowing cooling water. The gaseous and particulate air pollutants in flue gas were measured continuously by two Dust-Trak aerosol monitors and one multi-gas analyzer, respectively. In order to protect the multi-gas analyzer, the moisture and particles must be removed by a diffusion dryer and a HEPA before measurement. Carbonyl compounds were collected in two impingers containing 2,4-dinitrophenylhydrazine (DNPH) and perchloric acid solution. The collected solution was filtrated and then analyzed by a high performance liquid chromatography (HPLC).

2.3. Sampling and analysis

Due to portability and affordability, a multi-gas analyzer (HM5000, Infrared Industries, USA) was used to measure gaseous air pollutants emitted from charcoal combustion including CO₂, CO, HC, NO_x. The measuring principle of CO₂, CO, and HC are non-dispersive infrared method; however, NO_x is electrochemical method. The suitable operation temperature of the multi-gas analyzer was in the range of 2–43 °C. The measuring range, resolution, accuracy and response time of this analyzer are shown in Table 2. Two DustTrak aerosol monitors (Model 8520, TSI Inc., USA) were used to measure PM_{2.5} and PM₁₀ during charcoal combustion. The DustTrak performs a real-time measurement for particulate matters based on 90° light scattering. The suitable operation temperature and relative humidity of the DustTrak were 0–50 °C and 0–95%, respectively. The other operation characteristics were also as shown in Table 2. McNamara et al. [27] developed a correction factor of 1.65 for DustTrak to measure wood smoke. Because the charcoal combustion was similar to wood burning, the DustTrak also adopted 1.65 as correction factor to examine PM_{2.5} and PM₁₀ in this study. The multi-gas analyzer was calibrated against the zero and span calibration gases each month. The DustTrak aerosol monitors were zero calibrated using a HEPA filter before sampling. In addition, these instruments were sending back to manufacturers for regular maintenance and calibration once per year.

The gaseous carbonyl compounds generated from charcoal combustion were collected on two series absorption bottles containing 20 mL solution of DNPH and perchloric acid. The sampled solution was further filtered with a 0.45 μm membrane filter. Aldehydes were analyzed by injecting 10 μL of filtrate to a 1200 series HPLC coupled with diode array detector (DAD) (Agilent, USA). The HPLC reversed phase column for DNPH-carbonyl derivatives separation was Agilent C18 (250 × 4.6 mm, 5 μm) (Agilent, USA). The mobile phase consisted of two solvents: mixture A, 40:60 (v/v) of pure water/acetonitrile; solvent B, 100% acetonitrile. The gradient program was mixture A for 7 min, followed by a linear gradient from mixture A to solvent B in 13 min. The flow rate was maintained at 1.0 mL/min until the end of the analysis. The

DNPH-carbonyl derivatives were quantified by the absorbance at 360 nm. Calibration curves ($R^2 \geq 0.995$) for quantifying formaldehyde and acetaldehyde were prepared by diluting a standard of aldehyde/ketone DNPH mix (Supelco, USA) with pure acetonitrile solvent into different concentrations. The recoveries of formaldehyde and acetaldehyde were evaluated using 50–1000 μg/L of standard solutions to ensure the accuracies of the method. The recoveries of formaldehyde and acetaldehyde from seven repeated experiments were 99.6–106.3% and 99.6–105.6%, respectively. The precisions of this method for analyzing 50 μg/L of formaldehyde and acetaldehyde were 0.5% and 1.2%, respectively. The limit of detection (LOD) was defined as 3-fold of standard deviation determined based on seven repeated measurements using 50 μg/L of standard solution. The LODs of this method for analyzing formaldehyde and acetaldehyde were 3.1 ppb and 2.6 ppb, respectively.

To understand the characteristics of 10 kinds of charcoal, heating value analysis, proximate analysis, and ultimate analysis were also conducted in this study. The higher heating value (HHV) or calorific value was measured by the test method of Taiwan Environmental Protection Administration (TEPA) NIEA R214.01C [28]. The moisture was determined using test methods of NIEA R213.21C [29] and Taiwan Bureau of Standards, Metrology and Inspection (TBSMI) CNS 10821 [30]. The ash was detected via test methods of NIEA R205.01C [31] and CNS 10822 [32]. The volatile matter and fixed carbon were analyzed following test methods of CNS 10823 [33] and CNS 10824 [34], respectively. The elemental analysis was performed with the element analyzer (Elementar Vario EL cube, Hanau, Germany) following NIEA R403.00C [35].

2.4. Data analysis

The emission factor (EF, mg/kg of fuel) and emission rate (ER, mg/h) of generated air pollutant from charcoal combustion were calculated by Eqs. (1) and (2), respectively.

$$EF = \frac{\sum_{i=1}^n C_i Q_i t_i}{m_o - m_f} \times 10^3 \quad (1)$$

$$ER = \sum_{i=1}^n C_i Q_i \times 60 \quad (2)$$

where C_i (mg/Nm) is air pollutant concentration; Q_i (Nm³/min) is sampling flow rate; t_i (min) is sampling time; m_o (g) is charcoal weight before combustion; m_f (g) is charcoal weight after combustion; i (min) is the i th sampling time; n is the final sampling time. Statistical software of IMB SPSS 21.0 was used to analyze the effect of experimental variables on the emitted air pollutants from charcoal combustion.

3. Results and discussion

3.1. Basic properties of the charcoals

Fig. 2 shows the heating values of 10 types of charcoal used in the current study. The highest gross heating value (wet-based) was found in Binchōtan (B1), while the lowest was found in eco-friendly charcoal (C1S). Generally speaking, traditional charcoal made from firewood has higher heating value than green synthetic charcoal. For example, the heating values of T2, T3, and B1 are higher than those of C1S, C2S, C3S, and I3S. In this study, sawdust briquette charcoal (I1S) was found to have the highest heating value among the green charcoals, and the value is even higher than those of traditional charcoal like longan charcoal (T3), mangrove charcoal (I2). This may be related to its high-temperature manufacturing process.

The proximate analysis results are shown in Table 3. It was shown that the charcoals used in the current study have very low

Table 3
Characteristics of commercial charcoals.

Charcoal ID	Proximate analysis (%)					Ultimate analysis (%)				
	Moisture	Volatile matter	Fixed carbon	Combustible matter	Ash	C	H	N	O	Unknown
C1S	3.3	23.2	47.6	70.8	25.9	52.7	2.4	2.0	16.8	26.2
C2S	3.1	26.4	48.1	74.4	22.5	62.4	4.3	0.9	11.9	20.6
C3S	4.0	18.1	56.3	74.4	21.6	66.3	2.6	0.7	15.9	14.6
I1S	7.4	5.9	80.6	86.5	6.1	80.8	2.1	0.5	10.5	6.2
I2	7.3	37.4	53.0	90.4	2.2	62.9	4.6	0.3	29.3	3.0
I3S	7.3	27.2	48.1	75.3	17.4	55.4	2.9	0.7	23.9	17.0
T1S	9.8	17.3	51.5	68.8	21.4	57.0	2.5	0.5	17.2	22.7
T2	3.8	28.9	65.3	94.2	1.9	73.8	4.1	0.9	19.1	2.2
T3	4.2	21.6	70.0	91.6	4.2	74.9	3.3	0.5	18.1	3.3
B1	9.9	2.7	85.2	87.9	2.3	84.6	1.2	0.4	10.1	3.7

Table 4
Emission factors and emission rates of air pollutants at different charcoal combustion temperatures.

Emission factor (mean ± standard deviation)									
Temperature (°C)	HC (mg/kg)	CO (g/kg)	CO ₂ (g/kg)	NO _x (mg/kg)	PM _{2.5} (mg/kg)	PM ₁₀ (mg/kg)	HCHO (mg/kg)	C ₂ H ₄ O (mg/kg)	
450	1842.3 ± 462.3	142.5 ± 3.5	970.3 ± 30.5	316.0 ± 170.9	19.8 ± 6.6	31.1 ± 7.8	5.3 ± 0.2	2.4 ± 0.9	
500	1157.6 ± 659.4	168.0 ± 25.4	945.6 ± 131.4	320.8 ± 56.6	165.9 ± 34.1	199.4 ± 35.3	6.7 ± 1.9	1.3 ± 0.7	
550	1594.6 ± 95.0	175.7 ± 8.0	920.2 ± 32.2	251.3 ± 13.2	241.3 ± 103.6	292.8 ± 112.0	7.8 ± 1.2	1.6 ± 0.2	
<i>p</i> -value	0.264	0.097	0.368	0.368	0.097	0.097	0.264	0.264	
Emission rate (mean ± standard deviation)									
	Temperature (°C)	HC (mg/h)	CO (g/h)	CO ₂ (g/h)	NO _x (mg/h)	PM _{2.5} (mg/h)	PM ₁₀ (mg/h)	HCHO (μg/h)	C ₂ H ₄ O (μg/h)
Whole stage (0–60 min)	450	40.1 ± 9.0	3.1 ± 0.4	21.4 ± 2.5	7.0 ± 4.1	0.4 ± 0.2	0.7 ± 0.2	117.2 ± 10.2	51.6 ± 17.8
	500	26.0 ± 14.6	3.8 ± 0.6	21.4 ± 3.2	7.2 ± 1.2	3.7 ± 0.7	4.5 ± 0.7	150.7 ± 44.0	30.3 ± 16.8
	550	37.5 ± 2.3	4.1 ± 0.2	21.7 ± 0.7	5.9 ± 0.3	5.7 ± 2.5	6.9 ± 2.6	183.3 ± 29.5	36.8 ± 5.7
<i>p</i> -value		0.264	0.097	0.717	0.529	0.097	0.008	0.264	0.264
Flaming stage (0–20 min)	450	42.1 ± 8.9	2.0 ± 0.3	21.4 ± 2.5	11.8 ± 4.4	1.1 ± 0.5	1.7 ± 0.7	122.0 ± 11.0	154.9 ± 53.3
	500	31.2 ± 12.6	3.8 ± 0.7	24.3 ± 3.3	10.6 ± 1.6	6.7 ± 1.9	7.9 ± 1.7	274.9 ± 92.1	90.9 ± 50.4
	550	33.8 ± 12.1	3.6 ± 0.7	24.6 ± 1.5	11.3 ± 4.0	8.3 ± 3.9	9.3 ± 3.8	352.0 ± 53.6	110.5 ± 17.1
<i>p</i> -value		0.264	0.097	0.097	1.000	0.097	0.097	0.097	0.264
Smoldering stage(20–60 min)	450	39.2 ± 9.0	3.7 ± 0.4	21.4 ± 2.7	4.6 ± 4.7	0.1 ± 0.1	0.2 ± 0.1	114.8 ± 16.3	N.D.
	500	23.4 ± 17.0	3.8 ± 1.0	19.9 ± 4.2	5.5 ± 2.3	2.2 ± 0.3	2.8 ± 0.4	88.6 ± 20.0	N.D.
	550	39.4 ± 2.6	4.4 ± 0.2	20.2 ± 1.1	3.2 ± 2.0	4.4 ± 1.7	5.7 ± 2.1	98.9 ± 18.4	N.D.
<i>p</i> -value		0.097	0.264	0.717	0.368	0.007	0.005	0.264	1.000

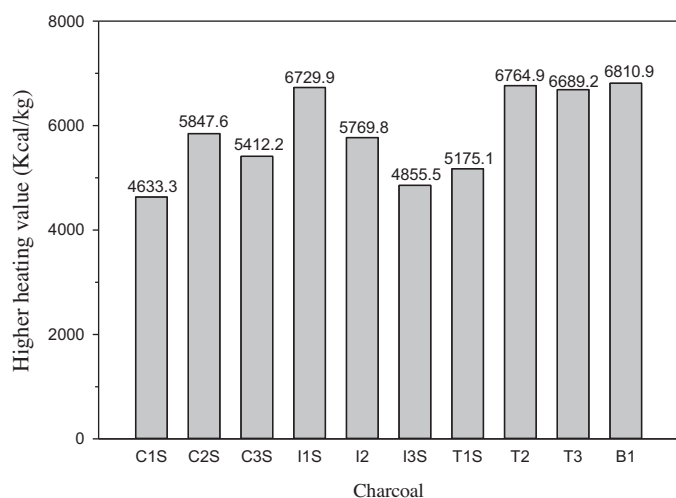


Fig. 2. Higher heating values of ten kinds of charcoal.

moisture contents, all below 10%, with the lowest being 2.24%. In terms of the combustible matter, those of the traditional charcoal and the sawdust briquette charcoal (T2, T3, I1S, I2, and B1) all reach as high as 90%, while those of the green charcoal (C1S, C2S, C3S, I3S,

and T1S) only reach 70%. The percentage of volatile matter is around 20–30%, except for those of sawdust briquette charcoal (I1S) and Binchōtan (B1). This is probably related to the high-temperature manufacturing process. Both types of charcoal are made at a temperature higher than 1000 °C, and most of the volatile matter would have been removed during the process. Therefore, the volatile matter only takes up a very small portion, and the percentage of fixed carbon is very high (>80%). The resulting Spearman's statistic confirms the highly significant positive correlation between fixed carbon percentage and heating value ($r=0.88$), showing that the heating value of charcoal is related to the extent of carbonization. The ash content exhibits a similar trend; while normal charcoal and sawdust briquette charcoal have ash contents below 10%; the ash content in green charcoals can reach as high as 20%. One exception is the mangrove charcoal (I2), which has a high percentage of combustible matter (90.42%) and a low percentage of ash (2.24%), while the volatile matter takes up 37.41%. Therefore, the heating value is only 5769.8 kcal/kg. In general, the more combustible matter, the higher the heating value. Meanwhile, the heating value generated by fixed carbon is higher than that by volatile matter.

The elemental analysis results (Table 3) show that both sawdust briquette charcoal (I1S) and Binchōtan (B1) have very high carbon contents (>80%), and the other samples also exceeded 50%. The correlation coefficient between carbon content and heating value reached 0.94 (Fig. 3). This proves again that there exists a

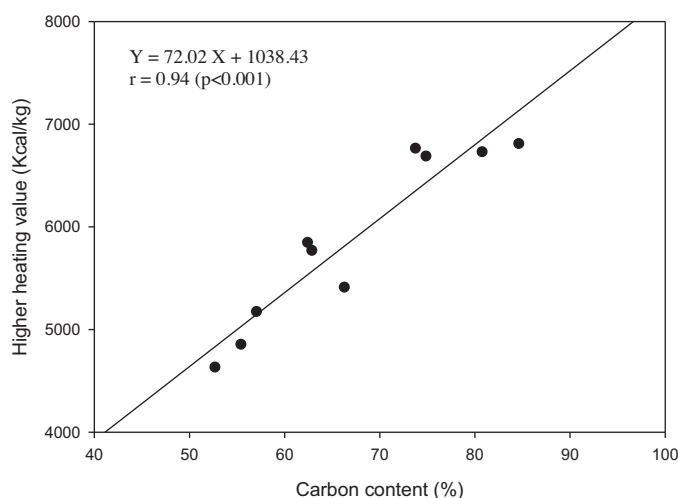


Fig. 3. Correlation between carbon content and heating value.

significant correlation between the heating value and the extent of carbonization. The nitrogen content was low in general, all below 1% except for that of C1S, which was 1.96%. The nitrogen content of C1S was twice as high as those of other charcoals, which is related to the fact that hay was used as the adhesive agent for this charcoal. The non-carbonized hay had relatively high nitrogen content. Moreover, from the elemental analysis it was shown that mangrove charcoal (I2) had higher hydrogen and oxygen contents, which were the main elements in volatile matter. This type of charcoal retained the reddish color from visual observation, and the high percentages of hydrogen, oxygen, and volatile matter were relevant to the extent of carbonization of mangrove charcoal. In addition to the contents of carbon, hydrogen, and oxygen, it was shown that the green charcoals had higher percentages of unknown elements, and the content of unknown elements in Binchōtan (B1) was higher than those in traditional charcoals (T2, T3, I2, and B1). The content of unknown elements, as a whole, had a significantly high correlation with the ash content ($r = 0.94$), and therefore, it was considered that the unknown elements belong to the incombustible ash.

3.2. The influence of combustion temperature on charcoal combustion pollutant emissions

Three different temperatures (450, 500, and 550 °C) were tested at an airflow rate of 10 LPM for the combustion of sawdust briquette charcoal (I1S), which is one of the most commonly used charcoals in barbecue restaurants. Friedman's statistical analyses of the air pollutants' EF at different temperatures (Table 4) show that the EFs did not have significant differences ($p > 0.05$) at 450–550 °C during whole stage (60 min) combustion. Furthermore, when EF results were divided into flaming stage (first 20 min of the combustion) and smoldering stage (last 40 min of the combustion) before the Friedman test was conducted (Table 4), a significant difference was observed between the emission rates of PM_{2.5} and PM₁₀ at the smoldering stage around 450–550 °C ($p < 0.05$). Similarly, temperature is highly correlated to the ER of PM_{2.5} and PM₁₀ at the smoldering stage ($p < 0.05$); the higher the temperature, the higher the ER at the smoldering stage. On the other hand, temperature does not have a significant impact on other emission factors of air pollutants.

3.3. The influence of charcoal types on emissions of pollutants

Tables 5 and 6 show the EF and ER of air pollutants generated from 1 h of combustion of ten types of charcoal at 500 °C and

Table 5
Emission factors of air pollutants for combustion of different types of charcoal.

Charcoal ID	Emission factor (mean ± standard deviation)									
	HC (mg/kg)	CO (g/kg)	CO ₂ (g/kg)	NO _x (mg/kg)	PM _{2.5} (mg/kg)	PM ₁₀ (mg/kg)	HCHO (mg/kg)	C ₂ H ₄ O (mg/kg)	Combustion rate (%)	PM _{2.5} /PM ₁₀
C1S	3048.0 ± 768.4	128.2 ± 22.1	776.0 ± 45.8	834.1 ± 491.0	235.3 ± 27.5	286.8 ± 43.7	8.0 ± 6.1	45.0 ± 12.7	93.8 ± 2.2	0.82 ± 0.04
C2S	11,209.3 ± 1275.8	168.8 ± 20.5	835.8 ± 21.4	674.6 ± 93.6	9905.4 ± 1221.6	12041.3 ± 1377.8	519.9 ± 200.8	769.5 ± 31.6	70.7 ± 6.3	0.82 ± 0.02
C3S	5276.8 ± 394.4	300.3 ± 31.3	986.8 ± 60.0	161.1 ± 98.6	368.9 ± 47.3	471.8 ± 78.4	9.8 ± 2.5	43.0 ± 5.1	77.5 ± 3.2	0.79 ± 0.03
I1S	1965.3 ± 216.1	214.7 ± 16.6	878.7 ± 96.3	462.1 ± 68.4	153.9 ± 11.4	201.2 ± 19.8	6.1 ± 0.4	1.3 ± 0.3	88.0 ± 1.1	0.77 ± 0.03
I2	9313.5 ± 2997.9	108.5 ± 19.3	855.1 ± 92.1	277.0 ± 44.2	955.7 ± 112.6	1189.8 ± 138.3	26.7 ± 11.4	135.9 ± 52.1	81.3 ± 4.1	0.82 ± 0.02
I3S	4057.5 ± 560.0	67.7 ± 56.4	723.7 ± 43.2	413.6 ± 149.3	282.6 ± 91.6	329.9 ± 101.8	57.8 ± 10.4	102.6 ± 4.8	81.6 ± 1.8	0.85 ± 0.02
T1S	3989.9 ± 1176.7	179.5 ± 25.1	644.4 ± 53.1	158.5 ± 22.9	707.2 ± 46.7	854.5 ± 71.9	38.6 ± 14.8	93.5 ± 41.5	96.1 ± 5.1	0.83 ± 0.02
T2	8062.8 ± 753.6	84.3 ± 1.0	1113.6 ± 83.9	823.9 ± 357.3	325.1 ± 67.7	440.7 ± 96.4	19.8 ± 2.9	36.9 ± 5.2	84.6 ± 2.0	0.74 ± 0.02
T3	4013.4 ± 75.7	107.6 ± 9.8	1096.6 ± 42.4	621.1 ± 209.4	67.1 ± 15.0	87.4 ± 15.0	15.1 ± 3.5	10.6 ± 3.1	81.6 ± 5.4	0.76 ± 0.04
B1	3923.9 ± 1191.0	76.6 ± 14.4	1225.2 ± 39.0	831.9 ± 295.9	12.0 ± 2.4	21.6 ± 1.2	16.2 ± 4.6	4.3 ± 0.8	66.5 ± 7.7	0.56 ± 0.09
p-value	0.005	0.003	0.003	0.011	0.002	0.002	0.003	0.002	0.002	

Table 6
Emission rates of air pollutants for combustion of different types of charcoal (a) (1/3), (b) 2/3 (c) 3/3.

(a)									
Emission rate (mean ± standard deviation)									
	Charcoal ID	HC (mg/h)	CO (g/h)	CO ₂ (g/h)	NO _x (mg/h)	PM _{2.5} (mg/h)	PM ₁₀ (mg/h)	HCHO (μg/h)	C ₂ H ₄ O (μg/h)
Whole stage(0–60 min)	C1S	85.6 ± 20.5	3.6 ± 0.6	21.8 ± 1.2	23.5 ± 14	6.6 ± 0.8	8.1 ± 1.2	222.7 ± 169.0	1262.2 ± 340.8
	C2S	238.5 ± 9.5	3.6 ± 0.8	17.9 ± 2.0	14.4 ± 1.6	210.6 ± 9.2	256.2 ± 11.3	10,881.1 ± 3538.8	16,506.7 ± 2033.1
	C3S	123.8 ± 4.3	7.0 ± 0.6	23.2 ± 2.3	3.8 ± 2.5	8.7 ± 1.5	11.1 ± 2.3	229.9 ± 58.4	1007.0 ± 76.5
	I1S	51.7 ± 5.7	5.7 ± 0.4	23.3 ± 2.5	12.1 ± 2.2	4.1 ± 0.3	5.3 ± 0.5	162.7 ± 9.9	35.5 ± 8.6
	I2	227.4 ± 64.1	2.7 ± 0.4	21.1 ± 3.1	6.8 ± 1.1	23.5 ± 2.6	28.5 ± 2.9	650.3 ± 252.9	3307.2 ± 1106.8
	I3S	98.3 ± 15.0	1.6 ± 1.3	17.5 ± 1.4	10.1 ± 3.8	6.8 ± 2.1	8.0 ± 2.3	1395.6 ± 220.7	2490.2 ± 155.8
	T1S	114.7 ± 29.6	5.2 ± 0.9	18.6 ± 1.6	4.6 ± 0.5	20.4 ± 0.6	24.9 ± 0.9	1121.1 ± 450.1	2720.0 ± 1268.9
	T2	205.2 ± 15.5	2.1 ± 0.1	28.4 ± 1.8	20.9 ± 8.9	8.3 ± 1.7	11.3 ± 2.5	504.2 ± 82.8	938.7 ± 117.2
	T3	98.8 ± 6.5	2.6 ± 0.2	27.0 ± 2.4	15.4 ± 6.3	1.7 ± 0.4	2.2 ± 0.5	368.7 ± 74.0	257.5 ± 65.8
	B1	79.7 ± 32.3	1.5 ± 0.4	24.3 ± 2.3	16.5 ± 6.3	0.2 ± 0.1	0.4 ± 0.1	315.1 ± 49.5	85.5 ± 12.4
p-value		0.005	0.003	0.007	0.011	0.002	0.002	0.002	0.002
(b)									
Emission rate (mean ± standard deviation)									
	Charcoal ID	HC (mg/h)	CO (g/h)	CO ₂ (g/h)	NO _x (mg/h)	PM _{2.5} (mg/h)	PM ₁₀ (mg/h)	HCHO (μg/h)	C ₂ H ₄ O (μg/h)
Flaming stage(0–20 min)	C1S	126.6 ± 37.6	4.2 ± 0.8	34.2 ± 3.7	66.3 ± 46.2	19.4 ± 2.2	23.7 ± 3.5	401.3 ± 351.7	3724.8 ± 996.5
	C2S	474.6 ± 19.4	3.6 ± 1.0	17.6 ± 4.1	8.2 ± 2.2	570.7 ± 19.6	697.1 ± 30.8	31,302.3 ± 10,291.8	49,282.6 ± 6133.1
	C3S	229.4 ± 30.8	8.6 ± 1.4	32.1 ± 4.8	4.7 ± 4.3	25.2 ± 4.2	32.3 ± 6.6	492.2 ± 186.9	3021.0 ± 229.6
	I1S	56.5 ± 11.8	7.8 ± 0.2	29.5 ± 2.2	16.8 ± 6.1	8.7 ± 0.5	11.0 ± 1.0	341.1 ± 10.0	106.4 ± 25.8
	I2	492.1 ± 175.4	4.5 ± 0.6	29.7 ± 7.3	10.5 ± 7.5	70.0 ± 8.4	84.8 ± 9.8	1646.7 ± 607.5	9905.9 ± 3339.1
	I3S	151.1 ± 37.8	2.6 ± 0.7	25.6 ± 0.4	17.2 ± 3.3	17.6 ± 7.3	20.5 ± 8.4	2237.2 ± 503.8	7347.0 ± 526.1
	T1S	181.9 ± 23.6	6.3 ± 1.9	27.7 ± 3.9	7.5 ± 1.3	55.4 ± 4.4	66.8 ± 6.1	3037.7 ± 1404.0	7755.8 ± 3799.9
	T2	477.0 ± 23.0	4.7 ± 0.2	36.7 ± 4.0	27.6 ± 8.1	24.1 ± 4.8	32.3 ± 6.8	1226.9 ± 144.1	2816.1 ± 351.6
	T3	143.0 ± 12.5	3.9 ± 0.1	38.4 ± 4.8	22.3 ± 10.9	4.9 ± 1.2	6.2 ± 1.2	835.8 ± 140.8	772.6 ± 197.3
	B1	70.4 ± 18.5	2.4 ± 0.8	24.5 ± 3.7	19.6 ± 6.8	0.7 ± 0.2	1.0 ± 0.3	717 ± 145	256 ± 37
p-value		0.003	0.010	0.013	0.019	0.002	0.002	0.002	0.002
(c)									
Emission rate (mean ± standard deviation)									
	Charcoal ID	HC (mg/h)	CO (g/h)	CO ₂ (g/h)	NO _x (mg/h)	PM _{2.5} (mg/h)	PM ₁₀ (mg/h)	HCHO (μg/h)	C ₂ H ₄ O (μg/h)
Smoldering stage(20–60 min)	C1S	65.1 ± 12.3	3.3 ± 0.5	15.6 ± 0.7	2.1 ± 3.7	0.2 ± 0.1	0.3 ± 0.1	133.4 ± 77.6	30.9 ± 30.8
	C2S	120.4 ± 20.8	3.7 ± 0.6	18.1 ± 1.0	17.5 ± 3.0	30.5 ± 5.0	35.7 ± 5.6	670.5 ± 165.9	118.7 ± 31.9
	C3S	71.0 ± 13.0	6.3 ± 0.5	18.8 ± 1.6	3.4 ± 1.7	0.4 ± 0.1	0.5 ± 0.2	98.7 ± 8.1	N.D.
	I1S	49.4 ± 2.7	4.6 ± 0.7	20.2 ± 2.7	9.7 ± 5.2	1.8 ± 0.3	2.5 ± 0.5	73.5 ± 19.5	N.D.
	I2	95.1 ± 14.8	1.7 ± 0.3	16.8 ± 1.8	5.0 ± 2.5	0.2 ± 0.4	0.4 ± 0.5	152.2 ± 75.9	7.8 ± 13.5
	I3S	71.9 ± 20.8	1.2 ± 2.0	13.5 ± 2.0	6.5 ± 4.5	1.4 ± 0.7	1.7 ± 0.9	974.8 ± 147.1	61.8 ± 40.3
	T1S	81.0 ± 40.5	4.7 ± 0.5	14.1 ± 1.4	3.1 ± 0.3	3.0 ± 1.4	3.6 ± 1.7	162.8 ± 41.8	202.1 ± 11.0
	T2	69.4 ± 11.7	0.9 ± 0.0	24.2 ± 0.7	17.6 ± 12.2	0.4 ± 0.2	0.7 ± 0.3	142.9 ± 84.8	N.D.
	T3	76.7 ± 12.6	2.0 ± 0.3	21.3 ± 2.1	12.0 ± 6.4	0.1 ± 0.1	0.1 ± 0.1	135.1 ± 48.4	N.D.
	B1	84.4 ± 39.3	1.1 ± 0.3	24.2 ± 1.7	15.0 ± 11.7	0.1 ± 0.0	0.2 ± 0.1	114.0 ± 27.7	N.D.
p-value		0.136	0.005	0.003	0.033	0.005	0.004	0.059	0.003

20 LPM. Table 5 shows that after 1 h of combustion, most charcoal achieved more than 80% of the combustion rate except for C2S, C3S, and B1. Binchōtan (B1) had the lowest combustion rate of all, only achieving 66%. Among the charcoals, charcoal briquettes (C2S) emitted the largest amount of pollutants; the emission factors of HC, PM_{2.5}, PM₁₀, CH₂O, and C₂H₄O all ranked first among the charcoals. The lowest EF for PM_{2.5} and PM₁₀ were found in Binchōtan (B1). In addition, sawdust briquette charcoal (I1S) emitted the lowest amount of carbonyl compounds (CH₂O and C₂H₄O).

3.3.1. Hydrocarbons

The 3 types of charcoal that had the highest EF of HC compounds were charcoal briquettes (C2S), mangrove charcoal (I2), and acacia charcoal (T2), in order from highest to lowest. The average EF of HC compounds of all ten types of charcoal was in the range of 1953–11209 mg/kg, which is consistent with other studies on the emissions of HC compounds from burning wood [22,23,26]. During the flaming stage, significant differences between the EF and ER of

HC were found in sawdust briquette charcoal (I1S) and charcoal briquettes (C2S) ($p=0.02$). During the smoldering stage, no significant difference was found between the ER of HC ($p=0.136$), which means that there is no significant difference between the HC compound emission at the smoldering stage for any kind of charcoal. At the flaming stage, the ER of HC had a highly positive correlation with the hydrogen content of the charcoal ($r=0.839$), and a moderately positive correlation with the content of volatile matter ($r=0.734$). On the other hand, at the smoldering stage, HC only exhibited a low positive correlation with the hydrogen content ($r=0.463$) and volatile matter content ($r=0.285$). This is because during the burning stage, the charcoal temperature increases when heated, the water gradually evaporates and large amounts of combustible HC compounds are released. The HC compounds released burn at a high temperature, and therefore, HC and the hydrogen content of the charcoal itself has a close relationship. As the amount of the combustible HC gases released from the surface of the charcoal decreases, the burning process enters the smoldering stage

[22]. Therefore, the HC of different types of charcoal exhibit no significant differences.

3.3.2. Carbon dioxide and carbon monoxide

The average EF of CO and CO₂ generated from the combustion of ten types of charcoal were 68–300 and 724–1225 g/kg, respectively. The EF of CO₂ obtained in this study were all lower than those obtained in other studies on different kinds of biomass burning, while the EF of CO were similar to those obtained in other studies [22,23,25,26]. At the flaming stage, CO₂, CO, and the basic composition of charcoal all exhibited low correlations. At the smoldering stage, CO₂ exhibited moderate or high correlation with the heating values ($r = -0.530$), fixed carbon ($r = 0.789$), combustible matter ($r = 0.659$), carbon content ($r = 0.832$), and ash content ($r = -0.530$). CO exhibited a moderate negative correlation with combustible matter ($r = -0.647$), and a moderate positive correlation with ash ($r = 0.609$). Due to the complexity of the combustion reaction during the flaming stage, which would not be dominated by any one of the single components of the charcoal, there was little correlation between basic charcoal components during the flaming stage. In the smoldering stage, since the amount of volatile gases decreased significantly and the release rate stabilized, the amount of CO₂ released was mainly determined by fixed carbon. When there were no big differences between the surface areas of charcoal, the higher the density of the surface carbon, the more CO₂ was released. During the smoldering stage, the amount of CO released was influenced by the ash content. When there is more ash contained in the charcoal, there would be more ash covering the surface of the charcoal during the smoldering stage that would hinder the reaction between oxygen gas and the carbon on the surface of the charcoal, generating more CO.

3.3.3. Nitrogen oxides

In this work, the 3 types of charcoal that released the largest amount of nitrogen oxides (NO_x) were the eco-friendly charcoal (C1S), Binchōtan (B1), and acacia charcoal (T2). The average NO_x EF of those 10 types of charcoal was in the range of 159–834 mg/kg, which was higher than that of the wood and charcoal combustion [23], but lower than those of charcoal burning by Andreae and Merlet [22] and wood burning reported by Zhang et al. [21]. At the flaming stage, NO_x had a low correlation with the basic properties of the charcoal, while at the smoldering stage, it exhibited a moderately positive correlation with the heating value ($r = 0.627$) and the combustible matter ($r = 0.508$). Nitrogen oxides generated during combustion can be categorized into 3 types, the thermal NO_x, fuel NO_x, and prompt NO_x [36]. The generation of thermal NO_x requires temperature over 1000 °C. Since the burning temperature was in the range of 450–550 °C in this study, charcoal burning would not generate thermal NO_x. The production of fuel NO_x depends on the nitrogen content, and prompt NO_x is generated from the prompt collision between the hydrocarbons around the flame area and the nitrogen gas in the air [37]. The ultimate analysis results in Table 3 show that, except for the eco-friendly charcoal (C1S), the nitrogen content of all the other charcoal is lower than 1%, and no significant differences were found. Fig. 4a shows that the NO_x peak concentration of C1S combustion occurs before 2 min, and large amounts of hydrocarbons are released from 3 min, which does not meet the conditions for the generation of prompt NO_x. Therefore, this peak value is related to fuel NO_x. On the other hand, except for the eco-friendly charcoal (C1S), the peak concentration of NO_x generated during the combustion of other charcoals all occurred after that of the hydrocarbon peak concentration (Fig. 4b). Therefore, it was concluded that prompt NO_x was the main determinant for the amount of nitrogen oxides generated.

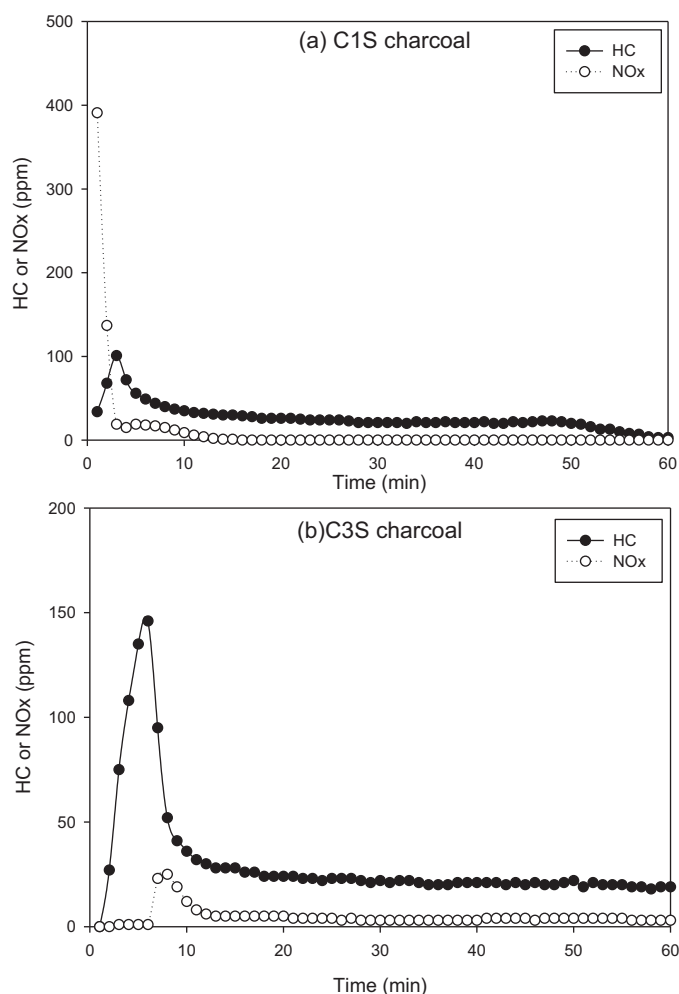


Fig. 4. Concentrations of hydrocarbons and nitroxide during (a) C1S charcoal combustion and (b) C3S charcoal combustion.

3.3.4. Particulate matter

Tables 5 and 6 show that the 3 types of charcoal with the highest EF and ER of PM₁₀ and PM_{2.5} are charcoal briquettes (C2S), mangrove charcoal (I2), and eco-friendly charcoal (T1S). The PM_{2.5} EF of charcoal briquettes (C2S) was more than ten times higher than that of other types of charcoal, and it was 800 times that of the Binchōtan (B1). Furthermore, it was higher than those of the residential wood [25] and Queensland trees [24]. The ER of PM_{2.5} and PM₁₀ exhibited a low correlation with the basic charcoal compositions at the smoldering stage, while at the flaming stage, they exhibited moderately positive correlations with volatile matter (PM_{2.5}: $r = 0.550$; PM₁₀: $r = 0.530$) and hydrogen content (PM_{2.5}: $r = 0.650$; PM₁₀: $r = 0.640$), moderately negative correlations with fixed carbon (PM_{2.5}: $r = -0.579$; PM₁₀: $r = -0.548$) and carbon content (PM_{2.5}: $r = -0.554$; PM₁₀: $r = -0.524$). This indicates that the higher the ratio of volatile matter and hydrogen content, the larger the amount of particulates released. On the other hand, the higher the ratio of fixed carbon and carbon content, the smaller the amount of particulates released. As previously mentioned, the hydrogen content had a high correlation with hydrocarbons ($r = 0.839$). It was therefore speculated that the particulates were mostly hydrocarbons that decomposed on the surface of charcoal as a result of cooling at the site of incomplete combustion, and were not relevant to the ash content of the charcoal itself (PM_{2.5}: $r = 0.257$; PM₁₀: $r = 0.250$).

The ratio of $PM_{2.5}/PM_{10}$ from charcoal combustion was mostly within the range of 0.5–0.9 (Table 5), the lowest ratio of 0.56 was found in Binchōtan (B1). The average atmospheric particulate matter in the Taiwan area was about 0.61 for $PM_{2.5}/PM_{10}$ [38], and the value of $PM_{2.5}/PM_{10}$ obtained from charcoal combustion in this study was significantly higher than that. Cheng et al. [39] reported that in the Taiwan Taichung area, the value of $PM_{2.5}/PM_{10}$ for agricultural waste burning and non-agricultural waste burning was 0.79 and 0.60, respectively. This suggests that the particulates generated during burning contain more fine particles $PM_{2.5}$, which are in agreement with the results obtained in the current study.

3.3.5. Carbonyl compound

Tables 5 and 6 show that during the flaming stage, large amounts of formaldehyde and acetaldehyde are produced from the combustion of different types of charcoal. However, during the smoldering stage, formaldehyde is the dominant compound emitted. Ho et al. [40] studied the carbonyl compositions generated in Hong Kong commercial kitchens using natural gases, and reported that formaldehyde, acetaldehyde, and acrolein accounted for the major carbonyl emissions. It was therefore demonstrated that formaldehyde and acetaldehyde were the major carbonyl compounds emitted no matter what cooking style was used. The highest carbonyl compound EF was found in charcoal briquettes (C2S). This charcoal type was mainly used for outdoor barbecues, and it is claimed that it can be ignited without fire. Table 5 shows that C2S combustion generates much higher EF of hydrocarbons, carbonyl carbons, and particulates than those produced by other charcoal combustion. It was speculated that during the manufacturing of this type of charcoal, more combustible substances are added, which leads to the emission of high concentrations of pollutants during the flaming stage. During the smoldering stage, the amount of pollutants emitted tends to decrease.

Lower carbonyl EF was found in sawdust briquette charcoal (I1S) and Binchōtan (B1), both of which are common types of charcoal used for indoor barbecues. The common feature of these two types of charcoal is that both are manufactured through carbonization at a high temperature (1000 °C), and therefore, have a relatively high degree of carbonization. The results of the proximate and ultimate analyses in Table 3 show that the fixed carbon content and carbon content of these two types of charcoal are over 80%, and therefore, the amount of carbonyl compounds emitted is extremely small. When the combustion reaches the smoldering stage, the amount of formaldehyde and acetaldehyde emitted decrease markedly. Therefore, it was speculated that carbonyl emissions are associated with the addition of adhesive agents during the manufacturing process of the charcoal.

4. Conclusions

There are many different types of charcoal used in barbecues that vary in quality. A list of 8 air pollutants from charcoal combustion was assembled that covers ten types of charcoal commonly used in Taiwan. The emission amounts of these air pollutants were associated with the basic properties of the charcoal. Among them, charcoal briquettes (C2S) were found to have the highest risk, with the highest EF of hydrocarbons, particulates, and carbonyl compounds. Nitrogen oxides generated from charcoal combustion were mostly fuel and prompt NO_x , which have a severe impact on human health. Since most of the pollutants have higher emission amounts during the flaming stage, it is recommended that barbecue restaurants first burn the charcoal to the smoldering stage in a well-ventilated environment before sending the charcoal to the customers at the dining area. Alternatively, the restaurants could install a local exhaust system at the barbecue area in order to

decrease public exposure to air pollutants. The information regarding the amount of air pollutants emitted from charcoal combustion in this study can be used as a reference for the public or barbecue industry, especially when selecting a suitable type of charcoal. The findings can also be used as the database for health risk assessments to further discuss the harm and risk of using charcoal indoors.

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