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Trace gas and particle emissions from open burning of three cereal crop residues: Increase in residue moistness enhances emissions of carbon monoxide, methane, and particulate organic carbon

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HIGHLIGHTS

- Emission factors for open burning of straw of rice, wheat, and barley were obtained.
- Emission factors for open burning of rice husks were also obtained.
- The effect of straw moisture content on the emission factors was evaluated.
- Increased moisture content enhanced emissions of CO, CH_4, and organic C particles.
- Moistness-adjusted emission factors are necessary for actual open burning.

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ABSTRACT

We determined emission factors for open burning of straw of rice, wheat, and barley, as well as rice husks, and we incorporated the effects of moisture content on the emission factors for the straw. A closed system that simulated on-site backfiring of residues on the soil surface under moderate wind conditions was used to measure the gas and particle emissions from open burning of the residues on an upland field. Two moisture content conditions were evaluated: a dry condition (air-dried residues, 11–13% by weight) and a moist condition (20%). When a linear regression model with the initial moisture content of the residue as the explanatory variable showed good correlation between the primary emission data of a substance and the moisture content, the regression model was adopted as a function to give the emission factors. Otherwise, the unmodified primary data were used as the emission factors. The magnitudes of the gas and particle emissions differed among the residue types. For example, carbon monoxide (CO) emissions from straw of rice, wheat, and barley and rice husks burned under the dry condition were 27.2 ± 1.7, 41.8 ± 24.2, 46.9 ± 2.1, and 66.1 g kg^{-1} dry matter, and emissions of methane (CH_4) were 0.75 ± 0.01, 2.01 ± 0.93, 1.47 ± 0.06, and 5.81 g kg^{-1} dry matter, respectively ($n = 2$ for straw with the standard deviation; $n = 1$ for husks). Emissions of carbon-containing gases and particles (e.g., CO, CH_4, and particulate organic carbon) were higher under the moist condition than under the dry condition, which suggests that emission factors for open burning should incorporate the effects of moisture content except open burning performed in the dry season or arid zones.

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1. Introduction

Open burning of crop residues is conducted worldwide, and large-scale open burning is particularly common in Asia (Yevich and Logan, 2003). The amount of crop residues burned in Asia in a typical year around 2000 was estimated to be 250 Tg (Streets

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et al., 2003). Open burning of rice residues (both straw and husks) is standard practice in Asia (Singh et al., 2008; Gadde et al., 2009).

Open burning of crop residues has several advantages. On-site burning dispenses with the need for removing and subsequently treating the residues. In addition, open burning allows for rapid and complete residue removal, which is required for double or triple cropping; for example, in flooded paddy fields, floating straw remaining from the previous cropping interferes with seeding and transplanting during the subsequent cropping (Singh et al., 2008). Residual ash remaining after burning acts as a fertilizer and is a particularly good source of potassium. Burning effectively eliminates pathogens on residues, although it has limited utility for soil disinfection (Kutcher and Malhi, 2010).

Open burning may have opposing effects on the carbon cycle as follows: on croplands where residues are not usually removed, open burning may reduce carbon pool in the soil (which is enhanced by plowing residues without burning them), as suggested by the fact that burning of sugarcane residues results in the loss of soil organic carbon (Galdos et al., 2009); whereas on croplands where residues are usually removed, on-site burning and plowing of the burned residues (char) may enhance long-term carbon sequestration. Open burning also affects emissions of methane (CH_4), as a potent greenhouse gas. Flooded paddy fields are an important source of atmospheric CH_4 (IPCC, 2013), and application of fresh organic matter such as straw to the fields increases CH_4 emissions (Yagi and Minami, 1990; Delwiche and Cicerone, 1993); therefore, burning rice residues and then plowing them into the soil can be expected to reduce CH_4 emissions (Haefele et al., 2011; Knoblauch et al., 2011), in addition to sequestering carbon in the soil.

However, open burning of crop residues has harmful environmental effects. For example, it produces air pollution both directly and indirectly and reduces visibility as a result of the gas and particle emissions (Streets et al., 2003). Open burning of crop residues is a source of various important atmospheric constituents as follows: greenhouse gases (carbon dioxide [CO_2], CH_4, and nitrous oxide [N_2O]; Ministry of the Environment, 2011), reactive gases such as carbon monoxide (CO; Galanter et al., 2000), and particles (including elemental carbon [EC] and organic carbon [OC]; Hays et al., 2005). The global emissions of CO_2, CH_4, N_2O, CO, EC, and OC resulting from agricultural residue burning in the late 1990s were estimated to be 818, 1.5, 0.04, 50, 0.37, and 1.8 Tg yr^{-1}, respectively (Andreae and Merlet, 2001).

Accurate emission factors (EFs) for atmospheric constituents released by crop residue burning are indispensable for developing better emission inventories for assessment of the environmental impacts of crop residue burning. The magnitudes of gas and particle emissions during open burning depend strongly on the type of crop and the burning method (U.S. EPA, 1995; Turn et al., 1997; Hays et al., 2005). Moisture levels in crop residues also affect EFs; for example, residue moistness has been shown to be positively correlated with particle emissions (Darley et al., 1974; Oanh et al., 2011), in which the positive and consistent correlation of particle emissions with moisture contents in a range of 5–35 % by weight was found (Darley et al., 1974). It is also hypothesized that high moisture content enhances emissions of gases originated from incomplete combustion, typically CO. However, data on the effects of moisture content are sparse. Although many studies have been conducted to obtain EFs for crop residue burning (e.g., U.S. EPA, 1995; Andreae and Merlet, 2001; Hays et al., 2005; Sahai et al., 2007; Zhang et al., 2008; Oanh et al., 2011), few of these studies considered the effects of residue moistness on EFs, and therefore current emission inventories do not explicitly incorporate the effects of residue moistness on gas and particle emissions.

Accordingly, in this study, our goals were to obtain EFs of trace gases and particles for open burning of three cereal crop residues (rice, wheat, and barley) by means of the same burning method and to evaluate the effects of residue moisture content on the emissions. We conducted open burning experiments with rice straw, wheat straw, barley straw, and rice husks. Even though incineration of rice husks is common practice in Japan and perhaps in other Asian countries, no reliable EFs for open burning of rice husks are available. Several laboratory experiments have been conducted with a fully closed apparatus so that combustion is complete and mass balance and EFs can be obtained (e.g., Lobert et al., 1991; Christian et al., 2003); however, these experiments required an extensive facility. Therefore, we used a hood system with forced ventilation to simulate on-site burning, which had the advantages of being portable and reproducing some of the conditions of real-world open burning; for example, the crop residues were burned on the soil surface, and some of the residues remained unburned. However, collection of the fine fractions of burned ash was difficult because the ash was in contact with soil, and the degree of combustion slightly fluctuated owing to the uncontrolled burning. The key features of the present study were as follows: (1) a hood system with forced ventilation was used to simulate on-site burning of crop residues on the soil surface with ignition from the leeward side (backfiring); (2) EFs for burning of straw from three major cereal crops were compared; (3) the effects of residue moisture content on EFs were evaluated; (4) EFs for rice husks were determined; and (5) emissions of greenhouse gases, other trace gases, and particles were measured simultaneously. The target species were the greenhouse gases CO_2, CH_4, and N_2O; the trace gases CO, nitric acid (HNO_3), nitrous acid (HNO_2), ammonia (NH_3), hydrochloric acid (HCl), and sulfur dioxide (SO_2); and total particulate matter (PM), OC, and EC, as well as particulate nitrate (pNO_3), ammonium (pNH_4), chloride (pCl), and sulfate (pSO_4).

2. Materials and methods

2.1. Crop residues

The crop residues used for the burning experiments were straw of paddy rice (*Oryza sativa* L.), wheat (*Triticum aestivum* L.), and barley (*Hordeum vulgare* L.) and husks of paddy rice. All crop residues were air-dried indoors prior to the burning experiments. Straw was cut to a length of approximately 20 cm. Rice husks were used in the form obtained after threshing and winnowing. The rice and barley residues originated from cropland in Okayama Prefecture in western Japan; the cropland was a reclaimed area with gray lowland soils including marine deposits as the parent material. Summer rice and winter barley are double-cropped at the site as follows: rice is seeded in early summer, and the paddy is submerged after germination; in late autumn, the paddy is drained, the rice is harvested, and the field is burned and tilled; barley is seeded at the end of autumn and harvested in early summer; the field is burned and tilled; and then rice is seeded again. In this region, open burning of crop residues is conducted by means of conventional methods. The wheat straw originated from cropland in Hokkaido in northern Japan. The cropland was an upland field with volcanic ash soils, and wheat is cropped as part of a 4-year rotation cycle, with a typical cycle consisting of potatoes, winter wheat, sugar beets, and legumes. In this area, wheat straw is generally not burned in the field except to disinfect residues.

Many early studies performed burning experiments using air-dried straw with a moisture content of approximately 10% by weight. However, straw is not well dried in a field except in the dry season or in arid zones. For example, the moisture contents of rice straw spread on paddy fields were 14.2% in Japan (Miura and

Kanno, 1997) and 26% on average in Thailand (Oanh et al., 2011). Thus, a moisture content of 20% was set as a moist condition in the present study. The initial moisture contents of the four air-dried crop residues were determined by drying them in an oven at 80 °C for 72 h. For the open burning experiments, two moisture content conditions were evaluated for each of the three types of straw: a dry condition and a moist condition. For the dry condition, a 200-g pack of air-dried straw was used for each burning experiment. For the moist condition, a weight of air-dried straw sufficient to yield 200 g when humidified to a moisture content of 20% was place in a large plastic bag and evenly misted with the necessary amount of deionized water from a spray bottle, and the bag was then allowed to stand overnight. Note that the resulting straw was not palpably moist. Only one moisture content condition, a dry condition, was evaluated for the rice husks because rice husks are nonflammable; in addition, only 100 g of rice husks was used for the burning experiment because the husks took a long time to burn out in a trial run.

2.2. Burning experiments in a forced-ventilation combustion hood

We used a portable combustion hood (Fig. 1) to simulate typical open burning of crop residues, backfiring in particular, albeit on a small scale. The stainless steel open-bottomed hood was operated under closed-system conditions. The back of the hood was equipped with an inlet for ambient air and two fan motors for stable forced ventilation. The exhaust was blown out to the atmosphere through an outlet on the roof. An exhaust duct with an inner diameter of 160 mm and a five-port sampling manifold was horizontally connected to the exhaust outlet via an elbow; the end of the duct was open to the atmosphere. The hood was placed on the soil surface in a field, and crop residues placed on the soil surface inside the hood were ignited. A portion of the exhaust air was continuously sampled starting after ignition and continuing to the end of combustion. In addition, burning experiment without soil was not conducted in this study because of the premise that open burning is usually conducted on soils.

Burning experiments were conducted at an upland field at the National Institute for Agro-Environmental Sciences, Japan, on November 24 and 25, 2011, from approximately 10:00 to 16:00 local time on both days. The average air temperature was 15.2 °C. For each type of straw, four burning experiments were conducted: two experiments under the dry condition and two under the moist condition. However, for each moisture condition, we measured PM, OC, and EC from a single sample collected continuously over the course of the two consecutive burning experiments to obtain sufficient sample quantities for analysis. Straw was evenly and loosely broadcast to a thickness of ~5 cm over 50 × 50 cm area of the soil surface inside the hood. For the rice husks, only one burning experiment was conducted under the dry condition. The soil surface inside the hood was hollowed to a depth of 10 cm, a handful of barley straw (a combustion enhancer) was placed in the hollow, and air-dried rice husks were heaped on the straw. The barley straw was then ignited, and sampling was started after smoke from the straw disappeared completely.

The hood was forcibly ventilated by means of the two fan motors, and the airflow rate could be varied continuously using a compact transformer. The cross-sectional distribution of the wind speed from the center of the duct to the edge of the duct was determined at the end of the duct by means of a hot-wire flowmeter (Climomaster 6542, Kanomax). The ventilation rate was calculated prior to the experiments. The ventilation rate was fixed at 4.25 m^3 min^{-1} (20 °C, 1013 hPa), and the wind speed just above the straw layer was approximately 0.6 m s^{-1}.

The steps of the combustion procedure were as follows: place the residue; start ventilation; ignite the residue from the leeward side with a gas torch; immediately close the front flapper of the combustion hood and seal the gaps between the flapper and the main body of the hood with tape (side gaps) and with soil (lower gap); start sampling of the exhaust air; and stop sampling at the end of the smoldering period. The residue remaining after combustion, which consisted of a mixture of unburned residue, char, and ash, was collected. As mentioned above, the fine fractions of char and ash were not collected, owing to contamination with soil particles; however, the fine (pulverized) char and ash are lightweight enough to be excluded from determining the weight of residue remained after combustion (Iwata, personal communication). Finally, the burned soil was removed and replaced with fresh soil for the next experiment.

The target gases and particles as inorganic salts were sampled by means of the following procedure. An L-tube made of stainless steel was attached to a sampling port of the exhaust duct, and a diaphragm air pump (APN-240, Iwaki) was used to suction a portion of the exhaust air from the upstream side parallel to the flow direction: the airflow rate was fixed at 3 L min^{-1} by a mass-flow controller (MQV0020, Azbil). Between the L-tube and the pump was placed a filter pack consisting of three filter holders (NL-I, NILU) in the following order from the upstream stage: (1) a glass-

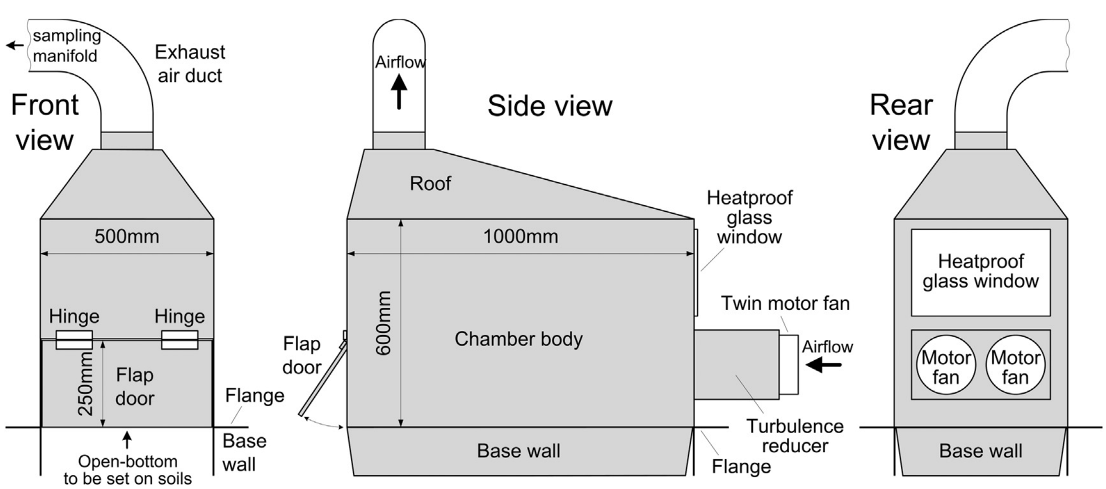

Fig. 1. Schematic of the hood system for open burning of the crop residues.

fiber filter (T60A20, Pall) to collect pNO_3, pNH_4, pCl, and pSO_4; (2) a cellulose filter (51A, Advantec) impregnated with potassium carbonate to collect HNO_3, HNO_2, HCl, and SO_2; and (3) a cellulose filter (51A, Advantec) impregnated with phosphoric acid to collect NH_3.

The exhaust from the air pump was divided into two streams. One stream was released to the atmosphere via a check valve, and the other stream was diverted to the inlet of another diaphragm air pump (APN-085, Iwaki) with an airflow rate fixed at 0.6 L min^{-1} by a mass-flow controller (MQV0020, Azbil). The exhaust from the second pump was forced into an aluminum foil gas sampling bag (volume, 10 L) for determination of the mixing ratios of CO_2, CH_4, N_2O, and CO. Because coexisting SO_2 and NO react to form N_2O during sample storage (Muzio and Kramlich, 1988), the potassium chloride-impregnated filter at the second stage of the filter pack served as a filter for removal of SO_2 from the downstream gas sampling for N_2O.

Another sampling line was used to sample particles via a cascade impactor (AN-200, Tokyo Dylec Corp) with a flow rate of 28.3 L min^{-1} for particle sizes of >7.0, 2.1–7.0, and <2.1 μm. Quartz-fiber filters (diameter, 80 mm; 2500QAT-UP, Pall) were used to collect the particles. An L-tube made of stainless steel (outer diameter, 15 mm) was placed inside the duct and connected to the sampler via a stainless steel tube (outer diameter, 15 mm; length, 0.5 m). Owing to high particulate concentrations at the initial stage of burning, the sample gases were diluted by a ratio of 9.0 ± 0.2 with filtered ambient air by means of a diluter (DI-1000, Dekati) for the straw burning experiments.

2.3. Weighing and chemical analysis

The unburned residue, char, and ash remaining after combustion were collected, combined, and dried in an oven at 80 °C for 72 h to determine the dry matter (DM) weight of the remaining residue. The combustion ratio (%) was calculated by the following equation: $(DM_{ini} - DM_{aft})DM_{ini}^{-1} \times 100$, where DM_{ini} and DM_{aft} are the DM weights (g DM) of the initial residue and the residue remaining after combustion. Portions of the air-dried crop residues not used for the burning experiments were analyzed to determine the initial carbon and nitrogen contents with an NC analyzer (Sumigraph NC-22F, Sumika Chemical Analysis Service). The carbon and nitrogen contents of the remaining residue after combustion were not analyzed, because of the difficulty in collecting all of the residue, including the fine fractions.

As soon as possible after each burning experiment, the sample filter of the filter pack was placed in a clean polypropylene test tube, which was sealed with a cap to avoid contamination. The target ion species collected on the filter were extracted ultrasonically with 10 mL of deionized water and were then analyzed by ion chromatography (ICS-1600, Thermo Scientific). Unused filters were also analyzed to correct for the background levels of the target ion species. The mean air concentration was calculated by dividing the collected amount of each target ion species by the total airflow through the filter pack. The quantities of emitted target ion species were obtained by multiplying the mean air concentration by the total airflow through the exhaust duct.

The exhaust gas collected in the sampling bag was analyzed to determine the mean mixing ratios of CO_2, CH_4, N_2O, and CO during the burning experiment. A gas chromatograph (GC-2014, Shimadzu) equipped with a thermal conductivity detector, a flame ionization detector, and an electron capture detector was used for CO_2, CH_4, and N_2O, respectively. For CO, a gas chromatograph (GC-9A, Shimadzu) equipped with a flame ionization detector via a methanizer (MTN-1; Shimadzu) to reduce CO into CH_4 was used. The emission quantities of these gases were calculated from the mixing ratio (corrected by the mixing ratio in the ambient air) and the total airflow through the exhaust duct.

The mass concentration of size-resolved particles, collected by a cascade impactor, was derived from the difference between the weight of the quartz-fiber filter before sampling and the weight after sampling. Each filter was weighed on a microbalance with a readability of 1 μg (M5P-F, Sartorius AG) in an air-conditioned chamber (CHAM-1000, Horiba) in which the temperature and relative humidity were controlled at 21.5 °C and 35%, respectively. Each filter was weighed twice, and the two masses were averaged. If the difference between the weights exceeded 10 μg, the old data were ignored and the sample was reweighed. The quartz-fiber filters were conditioned in the chamber for 48 h before being weighed. Total carbon, EC, and OC of the samples were determined with a thermal/optical carbon analyzer (DRI Model 2001 Carbon Analyzer, Desert Research Institute) (Chow et al., 1993) by means of the IMPROVE protocol (Chow et al., 2001). Punched quartz-fiber filters (diameter, 8 mm) were analyzed. Pyrolysis of OC during each analysis was corrected by the laser reflectance. Filter weighing and carbon analysis were performed by particle size, and the results aggregated for all the size ranges were then obtained.

2.4. Calculation of emission data and EFs

The emission quantities derived from the experiments were converted into quantities per unit weight of initial residue as DM, and the resulting quantities were used as the primary data for gas and particle emissions from open burning. The primary data were not corrected in terms of the combustion ratio. In the present study, we treated the initial moisture content of the residue as a candidate explanatory variable for the EF of each target species. We excluded the combustion ratio as an explanatory variable because the combustion ratios did not differ substantially among the burning experiments for each type of residue (see Table 1). In addition, the combustion method was also excluded as an explanatory variable because the combustion method was the same for all the straw types.

Linear regression analysis of the emission factor as the objective variable and the moisture content as the explanatory variable was conducted for the straw burning experiments for which two data points per moisture content condition were available (only one data point per condition was available for PM, OC, and EC). We accepted the resulting model as the EF when the adjusted coefficient of determination was higher than 0.4. Otherwise, the arithmetic mean of the primary data was chosen as the EF. The emission quantities of PM and OC for the straw burning experiments increased with increasing moisture content, as will be discussed later. Therefore, linear models were also derived for PM and OC, although one data point per moisture content condition did not allow to evaluate the model fitness. For EC emitted during the straw burning experiments, we used the arithmetic means of the emission quantities as the EFs. For rice husks, the results of one experiment were used as the EFs.

3. Results and discussion

3.1. Gas and particle emissions

The characteristics of the crop residues and the conditions for the burning experiments are summarized in Table 1. The carbon contents were less than 40% for rice straw and husks and more than 40% for wheat and barley straw. The nitrogen content of rice straw (0.547%) was higher than that of the other residues, and therefore, the rice straw had the lowest C/N ratio (71) among the residues. The combustion times for the rice and barley straw were similar

Table 1
Characteristics of crop residues and conditions for open burning experiments.

Residue		Rice straw		Wheat straw		Barley straw		Rice husk
Carbon content	(% DM)	38.9		44.2		42.9		38.2
Nitrogen content	(% DM)	0.547		0.315		0.387		0.312
C:N ratio	(–)	71		140		111		123
Moisture content	(%)	10.6	20.0	12.0	20.0	11.2	20.0	13.1
Initial residue	(g DM)	178.9	160.0	176.0	160.0	177.5	160.0	86.9
Replication[a]		2	2	2	2	2	2	1
Combustion time	(min)	3.4 ± 0.4	3.6 ± 0.6	14.7 ± 1.8	18.7 ± 3.3	3.3 ± 1.1	3.3 ± 0.0	86.1
Remained residue	(g DM)	54.8 ± 3.4	52.0 ± 3.2	49.4 ± 15.2	46.4 ± 10.4	30.2 ± 5.4	29.9 ± 1.0	12.3
Combustion ratio	(% DM)	69.3 ± 1.9	67.5 ± 2.0	71.9 ± 8.7	71.0 ± 6.5	83.0 ± 3.1	81.3 ± 0.6	85.8

DM, dry matter. Uncertainty values denote the standard deviation.
[a] For PM, OC, and EC, a single sample collected continuously over the course of the two consecutive burning experiments to obtain sufficient sample quantities for analysis.

(~3 min), whereas the combustion time for wheat was significantly longer (15–19 min) according to a *t*-test ($p < 0.05$). The wheat straw smoldered slowly with occasional flaming. By contrast, the rice and barley straw flamed vigorously after ignition. The rice husks were nonflammable and smoldered for approximately 90 min even though 100 g of husks was used for the experiment. Barley straw showed the highest combustion ratio (>80%), and the ratios for rice and wheat straw were similar to each other (~70%). Moisture content did not significantly affect combustion time or combustion ratio for any of the straw species ($p > 0.05$).

Primary data for gas and particle emissions are listed in Table 2, part of which is illustrated in Fig. 2. Carbon dioxide emissions were similar for all the residue types except the dry wheat straw and the rice husks, which emitted relatively low amounts of CO_2. The CO and CH_4 emissions from the rice straw were lower ($p < 0.05$) than those of the wheat straw, which took a long time to combust with smoldering, and those of the barley straw, which flamed vigorously and showed a high combustion ratio (Table 1). The nonflammable rice husks also showed high CO and CH_4 emissions. During combustion, oxidized substances such as CO_2, N_2O, and SO_2 are produced at the highest concentrations during the flaming stage, and incompletely oxidized and reduced substances such as CO, CH_4, and NH_3 are produced during the smoldering stage (Lobert et al., 1991). Therefore, we interpreted our results as indicating that the long smoldering of the wheat straw enhanced the CO and CH_4 emissions and that the rapid flaming of the barley straw resulted in partial oxygen deficits at the burning points, which resulted in higher CO and CH_4 emissions than the rice straw. For the rice and barley straw, HNO_2 was the most abundantly produced of the nitrogenous gases (N_2O, HNO_2, HNO_3, and NH_3; NO and NO_2 were not measured), whereas the wheat straw showed high emissions of both HNO_2 and NH_3. Nitrous oxide and NH_3 were the most abundant nitrogenous gases produced by rice husks. Note, however, that some of the nitrite ion that collected on the alkaline impregnated filters and was treated as HNO_2 in the present study might have originated from NO_2 (Noguchi et al., 2007). For the particles, PM and OC emissions from the wheat straw and rice husks, which smoldered for a long time, were larger than the emissions from the rice and barley straw. The EC emissions from the rice straw (particularly in the dry condition) and rice husks were smaller than the emission from the wheat and barley straw. The pNO_3 emissions were relatively small in amounts and similar among all the residue types. The pSO_4 emissions from the barley straw (particularly in the moist condition) were larger than the emissions from other types of residues.

A concentration ratio of CO_2 to CO (CO_2/CO ratio) is a useful index of combustion temperature (Khalil and Rasmussen, 2003), and therefore the degree of incomplete combustion. The CO_2/CO ratios of the rice and barley straw significantly decreased with the

Table 2
Gas and particle emissions from open burning of crop residues.

Residue	Rice straw		Wheat straw		Barley straw		Rice husk
Moisture content (%)	10.6	20.0	12.0	20.0	11.2	20.0	13.1
Replication	2	2	2	2	2	2	1
Gas (g kg^{-1} DM)							
CO_2	803 ± 65	946 ± 49	526 ± 9	952 ± 97	983 ± 62	1068 ± 25	652
CO	27.2 ± 1.7	59.4 ± 0.7	41.8 ± 24.2	77.3 ± 10.7	46.9 ± 2.1	93.3 ± 4.0	66.1
CH_4	0.75 ± 0.01	2.47 ± 0.16	2.01 ± 0.93	3.62 ± 0.42	1.47 ± 0.06	4.55 ± 0.40	5.81
N_2O	0.033 ± 0.003	0.057 ± 0.005	0.018 ± 0.000	0.013 ± 0.005	0.019 ± 0.001	0.027 ± 0.002	0.121
HNO_2	0.305 ± 0.046	0.493 ± 0.167	0.309 ± 0.251	0.244 ± 0.022	0.229 ± 0.034	0.469 ± 0.006	0.089
HNO_3	0.010 ± 0.000	0.010 ± 0.001	0.004 ± 0.001	0.006 ± 0.001	0.009 ± 0.000	0.009 ± 0.002	0.010
NH_3	0.059 ± 0.045	0.025 ± 0.020	0.125 ± 0.045	0.326 ± 0.025	0.024 ± 0.019	0.018 ± 0.008	0.356
HCl	0.062 ± 0.003	0.022 ± 0.006	0.027 ± 0.009	0.031 ± 0.023	0.069 ± 0.009	0.051 ± 0.008	0.018
SO_2	0.231 ± 0.127	0.029 ± 0.000	0.014 ± 0.005	0.015 ± 0.001	0.337 ± 0.066	0.048 ± 0.012	0.009
Particle (g kg^{-1} DM)							
PM[a]	2.2	9.1	15.0	19.5	5.8	8.0	9.6
OC[a]	1.0	4.5	9.3	13.5	1.8	3.0	6.0
EC[a]	0.207	0.501	0.706	0.943	0.807	0.698	0.269
pNO_3	0.006 ± 0.002	0.008 ± 0.000	0.006 ± 0.002	0.005 ± 0.001	0.005 ± 0.001	0.008 ± 0.001	0.007
pNH_4	0.083 ± 0.020	0.245 ± 0.092	0.034 ± 0.016	0.044 ± 0.015	0.081 ± 0.013	0.218 ± 0.011	0.043
pCl	0.30 ± 0.02	0.69 ± 0.14	0.12 ± 0.08	0.10 ± 0.06	1.53 ± 0.00	2.24 ± 0.16	0.13
pSO_4	0.027 ± 0.000	0.063 ± 0.003	0.031 ± 0.011	0.033 ± 0.010	0.121 ± 0.025	0.232 ± 0.023	0.020
Reference (ratio as carbon)							
CO_2/CO	18.8	10.1	8.0	7.8	13.3	7.3	6.3
(OC + EC)/PM	0.53	0.55	0.67	0.74	0.44	0.46	0.65

DM, dry matter. Uncertainty values denote the standard deviation.
[a] For PM, OC, and EC, a single sample collected continuously over the course of the two consecutive burning experiments to obtain sufficient sample quantities for analysis.

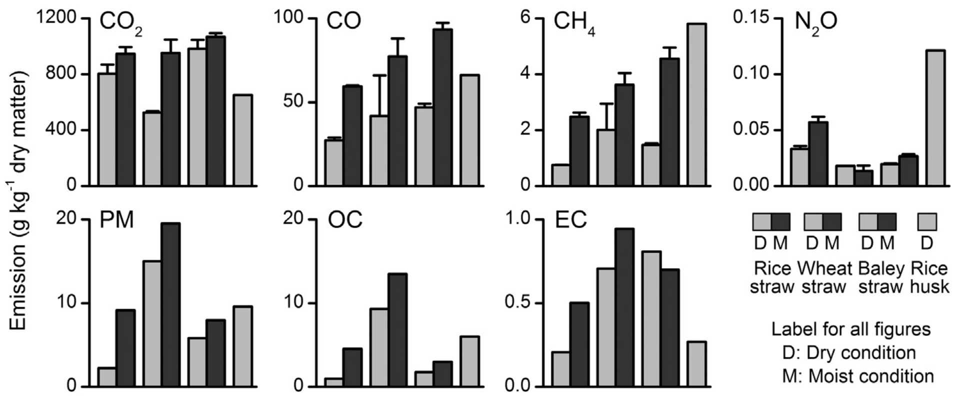

Fig. 2. Emissions of CO_2, CO, CH_4, N_2O, PM, OC, and EC during open burning of the crop residues. Error bars denote standard deviations ($n = 2$).

increase in the moisture content (Table 2) ($p < 0.05$), which indicated that the increase in moistness enhanced the degree of incomplete combustion. By contrast, the CO_2/CO ratios of the wheat straw were similar between the dry and moist conditions; however, those were smaller than the other straw even at the dry condition, which reflected the smoldering combustion with occasional flaming (Table 2). The CO_2/CO ratio of the rice husk was lowest among the residues, which supported the nonflammable property of rice husks.

Table 3 shows the correlation coefficients for the relationships between the moisture contents of the three types of straw and the emissions of gaseous and particulate species ($n = 4$ for each type of straw, 2 replicates per moisture content). The correlations observed for the wheat straw differed from those observed for the rice and barley straw, and this difference may have been due to the aforementioned difference in the combustion conditions (i.e., the wheat straw smoldered for a long time). Moisture content was positively correlated with emissions of carbonaceous gases, and the correlations were particularly strong for emissions of CO and CH_4 from rice and barley straw. Emissions of N_2O was positively correlated with the moisture content of rice and barley straw but was negatively correlated with the moisture content of wheat straw. The positive correlations support the hypothesis that emission of these gases during flaming combustion is enhanced by high moisture content, owing to incomplete combustion. For rice and barley straw, increased moisture content resulted in decreased emissions of HCl and SO_2 but increased emissions of pNH_4, pCl, and pSO_4. Burling et al. (2010) reported that HCl and SO_2 were exclusively emitted at flaming combustion. Therefore, it is expected that high moisture content inhibits flaming combustion and then decreases HCl and SO_2 emissions, which was confirmed in the present study. In addition, vaporization of the emitted pSO_4 to gaseous species is perhaps negligible according to the very low vapor pressure of sulfuric acid (e.g., Marti et al., 1997).

As supported by the findings reported by Darley et al. (1974), the PM and OC emissions were enhanced by high moisture content, although no replication for the moisture content was available in the burning experiments (Table 2 and Fig. 2). For all the straw types, the emissions of PM and OC under the moist condition were larger than the emissions under the dry condition; however, the magnitudes of the increases differed among the straw types. Emissions of PM and OC under the moist condition were 310% and 360% higher than the emissions under the dry condition for the rice straw, 30% and 44% higher for the wheat straw, and 37% and 70% higher for the barley straw, respectively. The effect of moisture content on the PM and OC emissions was particularly large for the rice straw. Although the magnitudes of the effects were similar for the wheat and barley straw, the amounts of PM and OC emitted from wheat straw were particularly large, perhaps as a result of incomplete combustion during the long smoldering period. The increases in the OC emissions due to increased moisture accounted for 51%, 91%, and 58% of the increase in PM emissions for the straw of rice, wheat, and barley, respectively. Thus, the increased PM emissions due to the increased moisture were ascribed mainly to the increased OC emissions, which accounted for 50–60% of the PM emission increase during the flaming combustion of rice and barley straw, and 90% during the smoldering combustion of wheat straw. The relationship between moisture content and EC emissions was unclear.

Table 3
Correlation coefficients for the relationship between moisture content and gas and particle emissions ($n = 4$; 2 replicates for each moisture content).

	Rice straw	Wheat straw	Barley straw
CO_2	0.870	0.975*	0.786
CO	0.998**	0.802	0.995**
CH_4	0.996**	0.844	0.991**
N_2O	0.975*	−0.701	0.970*
HNO_2	0.734	−0.248	0.990*
HNO_3	−0.255	0.863	−0.397
NH_3	−0.562	0.968*	−0.259
HCl	−0.987*	0.184	−0.830
SO_2	−0.846	0.164	−0.974*
pNO_3	0.716	−0.264	0.957*
pNH_4	0.866	0.413	0.993**
pCl	0.946	−0.197	0.976*
pSO_4	0.995**	0.136	0.955*

** and * denote the significant correlation with $p < 0.01$ and $p < 0.05$, respectively.

Table 4 lists a comparison of gas and particle emissions among relevant studies. The CO_2 emissions of wheat straw in the present study were smaller than the other reported values, which might be ascribed to the combustion condition (i.e., a long time smoldering). The CO and CH_4 emissions in the present study well agreed with the other reported values. Previous studies support our findings that high moisture content enhanced emissions of some gases and particles. Miura and Kanno (1997) reported that emissions of CO and CH_4 during open burning of rice straw at a moisture content of 14.2% (70 and 4.1 g kg^{-1} DM, respectively) were larger than those at a moisture content of 10.6% (44 and 2.1 g kg^{-1} DM, respectively). However, the CH_4 emissions in their study were larger than those in the present study, which might result from further incomplete combustion due to the thicker straw layer for burning in their study (15 cm) compared to the present study (5 cm). Oanh et al. (2011) reported that emission of CO (97 g kg^{-1} DM) was high during open burning of rice straw on paddy fields with a high moisture content (26% on average). These results, as well as ours, strongly

Table 4
Comparison of gas and particle emissions from previous studies and from the present study.

Residue	Moisture content (%)	Gas or particle emission (g kg^{-1} DM)						Note	Source
		CO_2	CO	CH_4	PM	OC	EC		
Rice straw	–	–	41	1.2	4.0	–	–		a
	9	–	–	–	–	0.9	0.50	Particles as PM10	b
	10.6	1111	44	2.1	–	–	–		c
	14.2	711	70	4.1	–	–	–		c
	–	791	64	–	–	–	–		d
	26	1147	97	–	9.4	3.1	0.53	Particles as PM10	e
	10.6	803	27	0.7	2.2	1.0	0.21	Table 2	This study
	20.0	946	59	2.5	9.1	4.5	0.50	Table 2	This study
Wheat straw	–	–	54	1.3	6.0	–	–		a
	7	–	–	–	–	2.2	0.80	Particles as PM10	b
	–	1558	141	–	–	–	–		d
	14	1787	28	3.6	–	0.3	0.16		f
	12.0	526	42	2.0	15.0	9.3	0.71	Table 2	This study
	20.0	952	77	3.6	19.5	13.5	0.94	Table 2	This study
Barley straw	–	–	78	2.2	11.0	–	–		a
	7	–	–	–	–	3.2	1.2	Particles as PM10	b
	11.2	983	47	1.5	5.8	1.8	0.81	Table 2	This study
	20.0	1068	93	4.6	8.0	3.0	0.70	Table 2	This study
Rice husk	13.1	652	66	5.8	9.6	6.0	0.27	Table 2	This study
Agricultural residues	–	1515	92	2.7	–	3.3	0.69	2006 IPCC Guidelines	g

–: Information is not available.
Sources: a, U.S. EPA (1995); b, Turn et al. (1997); c, Miura and Kanno (1997); d, Zhang et al. (2008); e, Oanh et al. (2011); f, Sahai et al. (2007); g, Andreae and Merlet (2001).

support the idea that high moisture content results in high emissions of CO and CH_4 during open burning, at least for rice straw. Particle emissions in the present study were similar to those of the other studies, except the high PM and OC emissions for the wheat straw in the present study; the OC emissions in the present study were approximately 3 times those reported for agricultural residues by Andreae and Merlet (2001) (Table 4). This discrepancy implies that how the residue burns (rapid flaming combustion versus slow smoldering combustion) also has an important effect on particle emissions.

3.2. EFs with the effect of moisture content

Table 5 shows the EFs obtained in the present study. Note that an applicable range of moisture contents for the regression models is specified to avoid extrapolations, and the upper or lower limit value derived from the regression model should be used instead of the extrapolations when the moisture content is beyond the range shown in the table. On the basis of the adoption criterion for the regression model (i.e., adjusted coefficient of determination > 0.4), the EFs of CO_2, CO, and CH_4 were functions of moisture content for all the straw types. For the other target species, the results differed between the two combustion conditions (i.e., rapid flaming combustion for the rice and barley straw and long smoldering combustion with occasional flaming for the wheat straw). The moisture content also affected the EFs of N_2O, HCl, SO_2, pNH_4, pCl, and pSO_4 for the rice and barley straw, those of HNO_2 and pNO_3 for the barley straw, and those of HNO_3 and NH_3 for the wheat straw. The EFs of PM and OC were functions of the moisture content in the present study, although only one data point per moisture condition was

Table 5
Emission factors for gas and particles during open burning of crop residues (See Table 2 for rice husks).

	Rice straw		Wheat straw		Barley straw	
Gas (g kg^{-1} DM)						
CO_2	$EF = 642 + 1520\ Moi$	(0.635)	$EF = -112 + 5320\ Moi$	(0.925)	$EF = 875 + 968\ Moi$	(0.427)
CO	$EF = -9.1 + 342\ Moi$	(0.999)	$EF = -11.5 + 444\ Moi$	(0.464)	$EF = -12.7 + 530\ Moi$	(0.986)
CH_4	$EF = -1.20 + 18.4\ Moi$	(0.988)	$EF = -0.40 + 20.1\ Moi$	(0.569)	$EF = -2.48 + 35.2\ Moi$	(0.974)
N_2O	$EF = 0.006 + 0.253\ Moi$	(0.926)	$EF = 0.016$		$EF = 0.010 + 0.083\ Moi$	(0.912)
HNO_2	$EF = 0.399$		$EF = 0.277$		$EF = -0.079 + 2.74\ Moi$	(0.969)
HNO_3	$EF = 0.010$		$EF = 0.001 + 0.024\ Moi$	(0.618)	$EF = 0.009$	
NH_3	$EF = 0.042$		$EF = -0.177 + 2.51\ Moi$	(0.907)	$EF = 0.021$	
HCl	$EF = 0.107 - 0.427\ Moi$	(0.961)	$EF = 0.029$		$EF = 0.092 - 0.206\ Moi$	(0.533)
SO_2	$EF = 0.458 - 2.15\ Moi$	(0.573)	$EF = 0.014$		$EF = 0.708 - 3.30\ Moi$	(0.923)
Particle (g kg^{-1} DM)						
PM[a]	$EF = -5.54 + 73.4\ Moi$	(–)	$EF = 8.14 + 57.0\ Moi$	(–)	$EF = 3.09 + 24.3\ Moi$	(–)
OC[a]	$EF = -3.01 + 37.7\ Moi$	(–)	$EF = 3.11 + 51.8\ Moi$	(–)	$EF = 0.17 + 14.1\ Moi$	(–)
EC[a]	$EF = 0.354$		$EF = 0.824$		$EF = 0.753$	
pNO_3	$EF = 0.007$		$EF = 0.006$		$EF = 0.001 + 0.035\ Moi$	(0.875)
pNH_4	$EF = -0.101 + 1.73\ Moi$	(0.625)	$EF = 0.039$		$EF = -0.096 + 1.57\ Moi$	(0.978)
pCl	$EF = -0.151 + 4.23\ Moi$	(0.842)	$EF = 0.107$		$EF = 0.624 + 8.09\ Moi$	(0.929)
pSO_4	$EF = -0.013 + 0.383\ Moi$	(0.986)	$EF = 0.032$		$EF = -0.021 + 1.27\ Moi$	(0.868)
Applicable range of moisture content (%) for regression model						
	Rice straw: 10.6–20.0		Wheat straw: 12.0–20.0		Barley straw: 11.2–20.0	

DM, dry matter; *EF*, emission factor; *Moi*, moisture content at the initial condition (%)/100.
Values in parentheses denote the adjusted coefficient of determination.
[a] For PM, OC, and EC, a single sample collected continuously over the course of the two consecutive burning experiments to obtain sufficient sample quantities for analysis.

used to determine the EFs. We expect that the current EFs will be confirmed in their accuracy with additional data from future studies. In addition, the soil at the study site was moderately dry during the experiments. Moisture exchanges between the soil and residues were negligible because the burning experiments were conducted immediately after the placement of residues.

Linearity of the effect of residue moisture content on gas and particle emissions is important to obtain EFs as a function of the residue moisture content, although the two moisture levels in the present study assumed a linear relationship. Fig. 3 shows the relationship between the residue moisture content and the emissions of CO_2, CO, CH_4, PM, OC, or EC obtained by the present and early studies (Table 4). Significant linearity regardless of straw types was found in CO and CH_4 ($p < 0.05$). Future research needs to evaluate the nonlinear responses of gas and particle emissions to the residue moisture content.

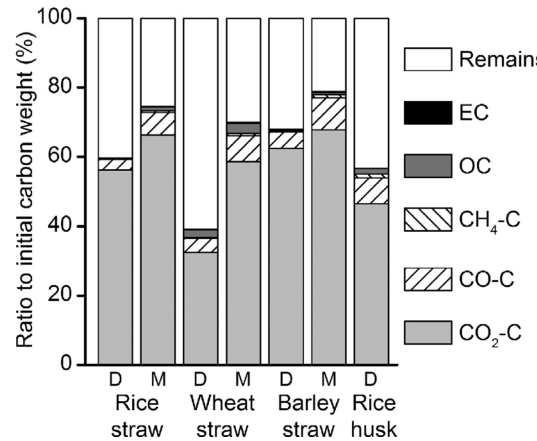

Fig. 4. Carbon balance for emitted gases and particles.

3.3. Carbon balance

It is also important to determine the proportion of the total carbon in the crop residues that is emitted to the atmosphere as gases and particles as a result of open burning. During open burning, the residues have various fates (partly unburned, charred, or cinereous), and the combustion ratio also fluctuates. Fig. 4 shows the carbon balance in the present study for each combination of residue type and moisture content. Theoretically, the difference between the amount of emitted carbon and the initial amount of carbon corresponds to the carbon remaining in the burned residue. We found that the emitted carbon accounted for 39–68% and 70–79% of the initial carbon under the dry and moist conditions, respectively. It is interesting that for wheat straw, a higher moisture content resulted in a remarkably larger proportion of emitted carbon. The combustion method we simulated (backfiring under moderate wind conditions) might be involved in this outcome. Carbon dioxide emission from the dry wheat straw was especially low, even though the combustion ratios for the dry wheat straw and dry rice straw were similar (Table 1). Therefore, it is possible that some of the wheat straw was charred during the smoldering combustion. Determining the fate of the residues (unburned, charred, or cinereous) is also important for evaluation of the carbon balance as well as the combustion ratio. However, because we simulated actual practice (i.e., burning on the soil surface), we were unable to collect all the char and ash after the combustion experiments because it might have been contaminated with soils particles.

For all the three types of straw, the moist condition increased the emissions of CO_2 as a typical oxidized substance, whereas the emissions of CO and CH_4 as typical substances of incomplete combustion also increased (Fig. 4). Relevant experimental results were as follows: (1) the emissions of $CO_2 + CO + CH_4$ as the weight ratio of carbon were higher in the moist condition than in the dry condition (Fig. 4), which suggests that a higher moisture content (within a range of 10%–20%) increases overall gaseous emissions of carbon; and (2) the emissions of CO and CH_4 increased and the ratio of CO_2/CO decreased in the moist condition (Table 2), which indicates that the moisture content affects compositions of emitted gases, and particularly enhances emissions of gases originated from incomplete combustion. Accordingly, a moist condition might increase overall gaseous emissions of carbon including CO_2, CO, and CH_4; however, the strengthened incomplete combustion due to the moist condition does not necessarily result in a decrease in CO_2 emission.

4. Conclusions

In this study, we obtained the EFs of trace gases and particles during open burning of straw of rice, wheat, and barley and husks

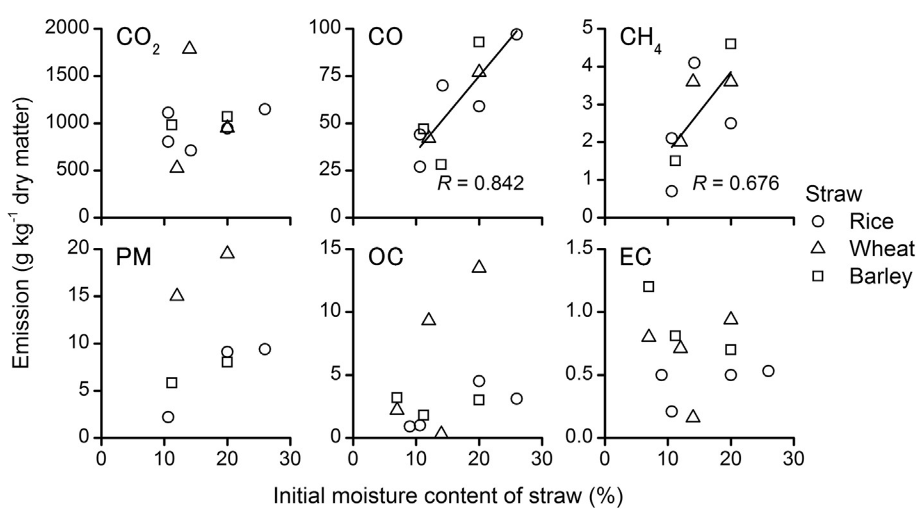

Fig. 3. Relationship between the residue moisture content and the emission of CO_2, CO, CH_4, PM, OC, or EC based on the present and early studies (see Table 4).

of rice, and we accounted for the effects of residue moisture content for the three straw types. The gas and particle emissions differed among the residue types. In particular, a large difference was observed between wheat straw and straw of rice and barley, and this difference was attributable to the difference in the combustion behavior of the straw types: the wheat straw underwent long smoldering combustion with occasional flaming, whereas the rice and barley straw underwent rapid flaming combustion. For the rice and barley straw (flaming combustion), increasing the moisture content enhanced the emissions of CO, CH_4, N_2O, and particles as inorganic salts and reduced the emissions of HCl and SO_2; whereas for the wheat straw (smoldering combustion), an increase in the moisture content was less correlated with the emissions of gases and particles as inorganic salts, except that increased emissions of CO_2 and NH_3 were observed. An increase in the moisture content enhanced the emissions of OC and therefore PM; however, the relationship between moisture content and particulate carbon emissions differed between the rice straw and the barley straw, even they showed similar combustion behavior. Furthermore, the slow smoldering combustion of the wheat straw resulted in high OC emissions.

Our results indicate that EFs that account for the effect of moisture content should be obtained for gases and particles, if the emissions of which are affected by moisture content, because crop residues burned in the field are not always dried well. It seems a normal situation that on-site burning is conducted for uncontrolled crop residues with high moisture content except cases in the dry season or arid zones. However, the existing EFs for open burning of crop residues are generally derived from experiments using air-dried residues (~10% in moisture content) and ignore the effects of moisture content on emissions.

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