



## Estimation and characterization of gaseous pollutant emissions from agricultural crop residue combustion in industrial and household sectors of Pakistan



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### H I G H L I G H T S

- Energy crisis has resulted in increased combustion of crop residues in Pakistan.
- Emission attributes of rice husk, rice straw, corncobs and bagasse were estimated.
- Rice straw had significantly higher gaseous pollutant emission factors.
- Bagasse had the highest value of total emission of gaseous pollutants.
- Rice straw and bagasse had >90% share in total gaseous pollutant emissions.

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### A B S T R A C T

A long-term energy crisis has resulted in increased combustion of biomass fuel in industrial and household sectors in Pakistan. We report results of a study on the emission characteristics of rice husk, rice straw, corncobs and bagasse since they are frequently used as biomass fuel and differed remarkably in physico-chemical and combustion characteristics. Emission concentrations and emission factors were determined experimentally by burning the biomass fuel using a burning tower. Modified combustion efficiency (MCE) of rice husk, rice straw, corncobs and bagasse was >0.97 indicating that combustion was dominated by flaming mode. Emission factors of gaseous pollutants CO, CO<sub>2</sub>, NO<sub>2</sub>, NO, NO<sub>x</sub> and SO<sub>2</sub> for rice straw were calculated to be 17.19 ± 0.28, 1090.07 ± 24.0, 0.89 ± 0.03, 1.48 ± 0.04, 3.16 ± 0.08 and 0.38 ± 0.03 g kg<sup>-1</sup> respectively which were significantly ( $p < 0.05$ ) higher compared to those from rice husk (14.05 ± 0.18, 880.48 ± 8.99, 0.19 ± 0.01, 1.38 ± 0.02, 2.31 ± 0.04 and 0.11 ± 0.03 g kg<sup>-1</sup>), corncobs (8.63 ± 0.12, 595.44 ± 10.38, 0.16 ± 0.01, 0.70 ± 0.01, 1.23 ± 0.02 and 0.02 ± 0.00 g kg<sup>-1</sup>) and bagasse (12.39 ± 0.08, 937.03 ± 9.07, 0.36 ± 0.03, 1.44 ± 0.02, 2.57 ± 0.04 and 0.18 ± 0.02 g kg<sup>-1</sup>). Total emissions of CO, CO<sub>2</sub>, NO<sub>2</sub>, NO, NO<sub>x</sub> and SO<sub>2</sub> were estimated to be 3.68, 230.51, 0.05, 0.36, 0.60 and 0.03 Gg for rice husk, 33.75, 2140.35, 1.75, 2.91, 6.20 and 0.75 Gg for rice straw, 1.11, 76.28, 0.02, 0.02 and 0.03 Gg for corncobs and 42.12, 3185.53, 1.22, 4.90, 8.74 and 0.61 Gg for bagasse respectively. Rice straw, however, had significantly ( $p < 0.05$ ) higher potential of gaseous pollutant emission factors. Bagasse had the highest values of total emissions followed by rice straw, rice husk and corncobs. Rice straw and bagasse, on cumulative basis, contributed more than 90% of total emissions of gaseous pollutants. Results reported in this study are important in formulating provincial and regional emission budgets of gaseous pollutants from burning of agricultural residues in Pakistan.

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## 1. Introduction

Pakistan, with an annual population growth rate of 2.4% and projected population of 18 million people, has been witnessing severe energy crisis over the last five years. At present, approximately 54% of energy requirement is met through fossil fuels such as oil and gas, and rest of the energy is obtained from biomass fuel such as wood and agricultural residues (Tahir et al., 2010). Crop residues are value added organic byproducts generated from harvesting and processing of agricultural crops.

Due to lack of knowledge regarding the significance of crop residues, they are often burned in the field (Samra et al., 2003). Agricultural open field burning is widely practiced in the rural areas and suburbs to dispose of biomass waste (Yevich and Logan, 2003). Several reasons favor burning of crop residue including cleaning and field preparation, meeting domestic energy requirements, fertilizing the field with ash and offering the pest control (Huang et al., 2012; Korontzi et al., 2006). However, the quantity of the crop residues burned and the fire intensity strongly influence the amount of carbon and nutrients released during the fire (Sharma and Mishra, 2001).

Crop residues and/or agricultural wastes are important domestic fuels since ancient times. Nearly half of the world population utilizes crop residues for domestic heating and cooking, especially in developing countries (Guoliang et al., 2008). According to estimates of Andreae and Merlet (2001) and Bond et al. (2004), burning of crop residues accounts for 540 and 475 Tg dry matter combustion per year respectively. Therefore, air quality deterioration, in cities located around major agricultural sectors, is perhaps not surprising (Cancado et al., 2006). There also have been extensive evidence of overlooking the emissions of trace gases from crop residue burning to a large extent, because these fires are often short-lived and do not offer significant time to be detected and quantified under natural conditions (Smith et al., 2007; Vander-Werf et al., 2010).

Field and domestic burning of crop residues consist of pyrolysis, smoldering and flaming processes, however, dominance of these processes and resultant gas emissions largely depend on the type of material being burnt (Andreae and Merlet, 2001). For example, agricultural residues usually follow flaming mode of burning that results in higher NO<sub>x</sub> concentrations, dung cakes are burnt through smoldering mode and burning fuel wood normally pass through all three stages of combustion (Saud et al., 2011).

Environmental problems associated with crop residue burning include smoke, trace gases and particulate matter (Bijay-Singh and Yadvinder-Singh, 2003). Concentrations of the greenhouse gases have increased over the past 50 years as a result of anthropogenic activities including agriculture, and have accelerated the rise in average global temperature (IPCC, 2001). In particular, uncontrolled and incomplete open-field burning results in emission of toxic air pollutants and greenhouse gases which affect the atmospheric chemistry (Andreae and Merlet, 2001; Kanabkaew and Oanh, 2011). Agricultural crop residue burning is also the prime source of the micron-sized aerosols which affect the composition of atmosphere (Awasthi et al., 2011; Saud et al., 2011). Trace gases emitted during burning, carbon monoxide and nitrogen oxide, are the main precursors of tropospheric ozone (O<sub>3</sub>), decreasing the concentrations of tropospheric hydroxyl radical (OH) (Mauzerall et al., 1998); the later holds potential threats to environment, ecosystem and human health (Cheng et al., 2000).

Emission factor is a crucial parameter used to estimate and quantify emission of trace gases and aerosols from biomass burning which describes compounds or substances emitted per amount of dry fuel burned (Andreae and Merlet, 2001; Yang et al., 2008). Emission factors of gaseous pollutants vary with time and space, and also depend on type, quality and composition of biomass fuel

(Shah et al., 1997). Emission factors, measured over longer time periods, are helpful in making emission inventories to control air pollution at local, national and regional levels. Emission factors, from different biomass burning, are integral components for making emission inventories and budgets.

Although studies on emissions from biomass burning are well documented across the globe (e.g. Delmas and Servant, 1982; Lacaux et al., 1993) including studies of Saud et al. (2011) in India and Zhang et al. (2008) in China, the research area is yet to be explored in Pakistan. It should be noted that there are limited emission factors available in developing countries, and those reported in the literature often varied dramatically due to difference in fuel properties and combustion conditions. In addition, emission factors measured in the laboratory may differ from those obtained in field measurements (Roden et al., 2006, 2009; Shen et al., 2010). Therefore, there is need to assess emission characteristics of biomass burning in Pakistan since severe energy crisis have forced large population to use firewood, crop residues and animal dung for meeting energy demands, especially in rural and peri-urban areas.

Keeping in context of the above discussion, a field scale study was performed to evaluate the emission characteristics of commonly burned agricultural biomass wastes in Pakistan i.e. rice straw, rice husk, corn cobs and bagasse. Furthermore, to our knowledge, this is the first study determining emission concentrations, emission factors and emission inventories of trace gases from burning of crop residues in Pakistan. The current study was designed to:

- investigate the emissions of different gaseous pollutants (CO, CO<sub>2</sub>, NO<sub>2</sub>, NO, NO<sub>x</sub>, SO<sub>2</sub>) from burning of rice straw, rice husk, corncobs and bagasse.
- characterize and compare the emission factors of rice straw, rice husk, corncobs and bagasse burning
- prepare emission inventories to estimate total emissions of trace gases

## 2. Materials and methods

### 2.1. Selection, sampling and preparation of crop residue samples

Rice straw, rice husk, corncobs and bagasse were used in this study because they are burnt in the agricultural fields as waste products and in homes and/or industries for energy in Pakistan. Samples of crop residues were collected in triplicate from farmers' fields and agricultural processing industry around Faisalabad and Kasur in Punjab, Pakistan (Fig. 1). Rice straw and bagasse were collected from Gatwala and corncobs were collected from Jarranwala, suburbs of Faisalabad. However, rice husk samples were obtained from Kasur. Samples were air dried under outdoor ambient conditions for several days before the start of experiment. When uniformly air-dried, samples were kept in sealed plastic bags.

### 2.2. Construction and design of burning tower

For this experiment, a metallic combustion tower was designed with an aim to facilitate the analysis by channelizing the smoke through one stack (Fig. 2). The tower consisted of an inverted funnel shaped cylindrical bottom having 1.2 m diameter and 1.0 m height. A stack with internal diameter of 0.2 m and length of 1.2 m was attached at the top end of the cylindrical bottom (Fig. 2a). The stack was at 1.2 m height from the ground. The cylindrical bottom was supported with iron rods to keep it at 0.2 m height from ground level (Fig. 2b). A metallic burning table of 0.4 m × 0.4 m dimension was also constructed using a coarse iron wire-gauze which has 0.2 m long legs at its four corners. The stack had an

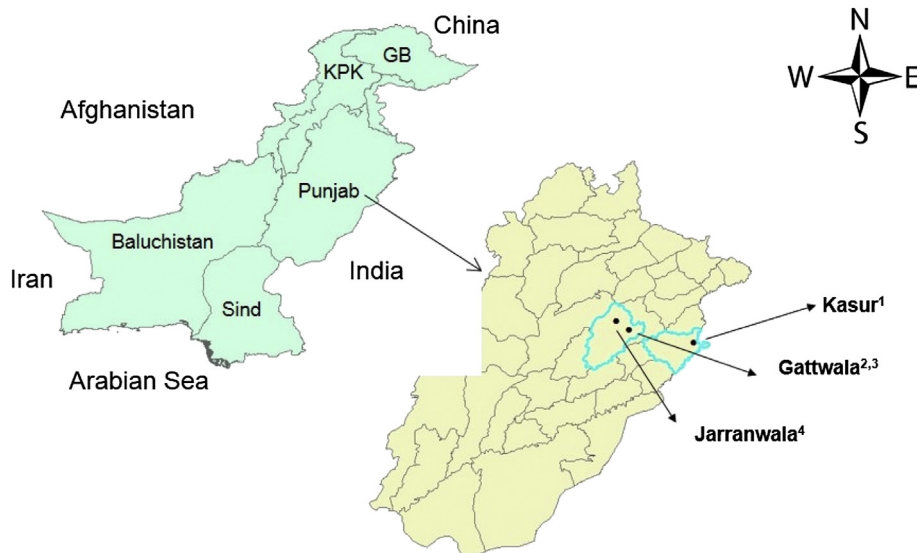


Fig. 1. Sampling locations of rice husk, rice straw, corncobs and bagasse in Punjab, Pakistan. <sup>1</sup>Rice husk, <sup>2</sup>Rice straw, <sup>3</sup>Bagasse, <sup>4</sup>Corncobs.

opening at 0.6 m height from its bottom for insertion of the instrumental probe and recording of different parameters. Keeping the burning material on the perforated metallic stand, at 0.2 m height, ensured uniform 3-D movement of gases and ample supply of oxygen to facilitate uniform burning under ambient conditions.

### 2.3. Emission analysis

#### 2.3.1. Principle of flu gas analyzer

Trace gas emissions were measured using a digital flu gas analyzer, testo 350-S (testo AG, Germany) by following a modified protocol of Li et al. (2009). The analyzer draws gases from the stack with the help of sampling probe. Gases pass through the sensors and sensors, based on the principle of selective ion potentiometry, measure the electrochemical potential differences. The range of the instrument for emission concentrations of CO<sub>2</sub> was 0–50 vol. % whereas range was 0–10000, 0–3000, 0–500 and 0–5000 ppm for

CO, NO, NO<sub>2</sub> and SO<sub>2</sub> respectively. The accuracy of the instrument for CO, NO, NO<sub>2</sub> and SO<sub>2</sub> emission concentrations was  $\pm 5\%$  of the measured value.

#### 2.3.2. Experimental process

Before each burning test, the selected crop residue was weighed and placed on metallic perforated burning table designed especially to facilitate residue burning (Fig. 3). After ignition, sampling probe of the analyzer was inserted into the stack to measure trace gas concentrations from the start to the end of each burning cycle of residue combustion at 10 s interval (Jenkins et al., 1996). Experimental conditions and design of the combustion tower allowed natural and uniform ventilation during each burning event. Time was noted for each burning event and when burning process was completed, ash was collected and weighed to calculate percent mass loss. Each crop residue burning test was repeated three times throughout the experiment; however, where

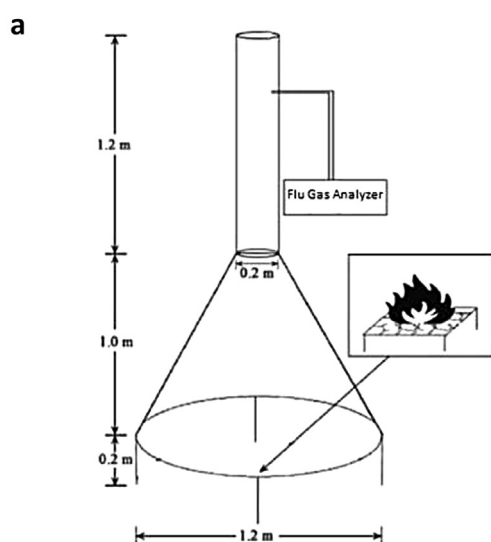


Fig. 2. Experimental set up: (a) schematic diagram, (b) actual burning tower.





**Fig. 3.** Air dried biomass on metallic stand before burring: (a) rice husk, (b) rice straw, (c) bagasse, (d) corn cobs.

appropriate, mass weighted means of the data are presented in tables and figures.

#### 2.4. Analytical protocols

##### 2.4.1. Moisture content and mass loss

Moisture content was measured gravimetrically by drying crop residue samples at 90 °C for 48 h in a pre-heated oven. Samples were cooled in a desiccator before they were reweighed and moisture content was calculated on percent dry mass basis. Mass loss was calculated by weighing the fuel samples before and after the combustion process was completed (ash content). Mass loss was also presented on percent mass basis.

##### 2.4.2. Total carbon (C), nitrogen (N) and sulfur (S) contents of biomass

Oven-dried crop residue samples from moisture content determination were further used for the measurements of total C, N and S contents. For C and N analysis, the oven-dried samples were ground using a ball mill (Retsch MM301) to homogenize the samples prior to analysis on a Carlo Erba Na 1500 CNS analyzer (Thermo Fisher Scientific, Waltham, MA). C and N content of the samples were calibrated using the standards atropine and acetanilide and an internal reference sample. For S analysis, 100 mg residue material was digested under high pressure with nitric acid and hydrogen peroxide in sealed Teflon vessels using a Milestone destruction microwave oven (MLS 1200 mega). After digestion, the samples were analyzed for S contents on an inductively-coupled plasma emission spectrophotometer (ICP, Spectroflame Flame VML2). Standard reference solutions for S were analyzed for calibration on ICP. C, N and S contents of residue samples were expressed on percent dry mass basis.

##### 2.4.3. Stack gas velocity, flu temperature, burning cycle and emission concentrations of gaseous pollutants

The stack gas velocity ( $\text{m s}^{-1}$ ), flu temperature ( $^{\circ}\text{C}$ ), burning cycle (s) and emission concentrations of CO, CO<sub>2</sub>, NO<sub>2</sub>, NO, NO<sub>x</sub> and SO<sub>2</sub>

(ppmv) were measured using the digital flu gas analyzer (testo 350-S). At the start of the burning cycle, the probe of the analyzer was inserted into the stack through the designed hole to record the said parameters for each fuel burning event every 10 s until the burning cycle was complete. Emission concentrations were used to calculate emission factors of the gaseous pollutants (Guoliang et al., 2008).

#### 2.5. Calculation of emission factors

Fuel based emission factors of gaseous pollutants represent mass of the specie released per unit fuel weight (Andreae and Merlet, 2001). Emission factors of gaseous pollutants were calculated using the mass balance equation described by Jenkins et al. (1996) and Guoliang et al. (2008) and were expressed on g per kg dry weight of the fuel:

$$E_i = \frac{10^{-3}}{m_{fd}} \int_{t_0}^{t_f} A_s u C_i \frac{w_i}{22.4} dt \quad (1)$$

Where

- $E_i$  = Emission factor for species  $i$
- $m_{fd}$  = Mass of crop residue used in the each burning test
- $t_0$  = Initial start time for each burning test
- $t_f$  = Finishing time for the test
- $A_s$  = Stack area ( $0.03 \text{ m}^2$ )
- $u$  = Average stack gas velocity
- $C_i$  = Sample concentration of species  $i$ , and
- $w_i$  = Molecular weight of species  $i$

#### 2.6. Quantification of crop residue production and burning in Pakistan

Crop residues production was estimated from crop production data (Government of Pakistan, 2011–12) and relevant residue

generation rate or ratio (Singh and Gu, 2010) using following relationship:

$$\text{Crop residues (Mt)} = \text{Crop production (Mt)} \times \text{crop to residue ratio} \quad (2)$$

Total amount of residue burnt, for each residue, was quantified as under:

$$\begin{aligned} \text{Total residue burnt (Mt)} &= \text{Total crop residue (Mt)} \\ &\times \text{Residue dry matter fraction} \\ &\times \text{Crop residue burnt (\%)} \end{aligned} \quad (3)$$

Dry matter fraction for each crop residue was obtained from Streets et al. (2003) and crop residue percent being burnt was estimated to be 25% for each crop residue (Iqbal and Goheer, 2008).

### 2.7. Estimation of total annual trace gases emissions

Total annual emission of each gaseous pollutant from burning of biomass fuel was calculated using following relationship described by Kanabkaew and Oanh (2011) and Yang et al. (2008):

$$\text{Total annual emissions} = M \times \text{EF} \quad (4)$$

Where,

$E$  = Total annual emission (Gg)

$M$  = Quantity of crop residues burnt in a year (Mt dry mass of residue)

$\text{EF}$  = Emission factors of gaseous species ( $\text{g kg}^{-1}$  fuel dry mass)

### 2.8. Modified combustion efficiency (MCE)

Ward et al. (1992) described combustion efficiency (CE) as the ratio of carbon released as  $\text{CO}_2$  to the total mass of carbon in the fuel biomass. CE may be considered helpful in determination of the completeness of the combustion as well as indication of process and/or processes dominant during the combustion. CE is usually measured as under:

$$\text{CE} = C_{\text{CO}_2} / C_{\text{Total}}$$

Where  $C_{\text{CO}_2}$  is the carbon emitted in  $\text{CO}_2$  form and  $C_{\text{Total}}$  is the total amount of carbon in gaseous and particulate emissions. In the current study,  $C_{\text{CO}_2}$  and  $C_{\text{CO}}$  were measured but particulate matter contents were not measured; hence, the modified combustion efficiency (MCE) was calculated following relationship proposed by Zhang et al. (2008):

$$\text{MCE} = C_{\text{CO}_2} / [C_{\text{CO}_2} + C_{\text{CO}}] \quad (5)$$

### 2.9. Statistical analysis

Data regarding moisture content (%), mass loss (%), C (%), N (%), S (%), flu temperature ( $^{\circ}\text{C}$ ), stack gas velocity ( $\text{m s}^{-1}$ ), burning cycle (s), gaseous pollutant emission concentration (ppmv) and gaseous emission pollutant factors ( $\text{g kg}^{-1}$ ), measured and/or calculated on replicate samples, were subjected to one way analysis of variance (ANOVA). Tukey's HSD postdoc test was used for multiple means comparisons technique only for those parameters where significant

treatment effects were found. However, where appropriate, figures and tables contain means of three replicates.

## 3. Results and discussion

### 3.1. Biomass characteristics

Physical and chemical characteristics of rice husk, rice straw, corncobs and bagasse are summarized in Table 1. Moisture content ranged from  $9.74 \pm 0.43\%$  for rice husk to  $12.06 \pm 0.18\%$  for bagasse. Mass loss percent values were  $85.66 \pm 0.17\%$  for rice husk,  $81.07 \pm 0.07\%$  for rice straw,  $97.06 \pm 0.04\%$  for corncobs and  $89.59 \pm 0.62\%$  for bagasse. Mass loss percent of corncobs was significantly ( $p < 0.05$ ) higher compared to that from rice husk, rice straw and bagasse.

C, N and S contents of crop residues are also shown in Table 1. C contents of corncobs and bagasse were  $44.70 \pm 0.04$  and  $43.87 \pm 0.10\%$  respectively which were significantly ( $p < 0.05$ ) higher than C content of rice husk ( $36.29 \pm 1.60\%$ ) and rice straw ( $39.16 \pm 0.05\%$ ). N contents were  $0.47 \pm 0.03\%$  for rice husk,  $0.59 \pm 0.04\%$  for rice straw,  $0.44 \pm 0.03\%$  for corncobs and  $0.62 \pm 0.02\%$  for bagasse. S contents of rice straw,  $0.17 \pm 0.01\%$ , were the highest among the crop residue used in this study and were significantly ( $p < 0.05$ ) different from rice husk, corncobs and bagasse.

The moisture content, mass loss, C, N and S contents of biomass fuel have a significant impact on the burning and emission characteristics of biomass. In our study, bagasse had the highest moisture contents compared to rice husk, rice straw and corncobs. The moisture content of rice straw ( $11.05\%$ ) was in accordance with the range of moisture content ( $10\text{--}12\%$ ) for rice straw previously reported by Buzarovska et al. (2008). However, the moisture content of the rice husk was higher compared to that  $7.20\%$  observed by Ileleji and Zhou (2008). This higher moisture content of rice husk could be attributed to regional climatic conditions. The results of mass oxidized (mass loss) for bagasse was similar to those reported by Sahai et al. (2011). However, mass loss values for rice straw, rice husk and corncobs was found to be  $81.07$ ,  $85.66$  and  $89.59\%$  which differed slightly from the reported  $90\%$  value of mass loss for these crop residues (Sahai et al., 2011). In this study, we have also reported C, N and S contents of crop residues since the chemical composition of the crop residue is an important factor in determining the emission factors of gaseous pollutants as argued by Zhang et al. (2008).

### 3.2. Burning characteristics

Flu temperature, stack gas velocity, burning cycle and modified combustion efficiency (MCE) for rice husk, rice straw, corncobs and bagasse are presented in Table 2. Flu temperatures of rice straw

**Table 1**

Moisture content, mass loss, total carbon, total nitrogen and total sulfur content of rice husk, rice straw, corncobs and bagasse.

Parameter	Rice husk	Rice straw	Corncobs	Bagasse
Moisture content (%)	9.74 (0.43) a	11.05 (0.27) ab	11.43 (0.28) a	12.06 (0.18) a
Mass loss (%)	85.66 (0.17) c	81.07 (0.07) d	97.06 (0.04) a	89.59 (0.62) b
Total carbon (%)	36.29 (1.60) b	39.16 (0.05) b	44.70 (0.04) a	43.87 (0.10) a
Total nitrogen (%)	0.47 (0.03) bc	0.59 (0.04) ab	0.44 (0.03) c	0.62 (0.02) a
Total sulfur (%)	0.06 (0.00) b	0.17 (0.01) a	0.03 (0.00) c	0.07 (0.00) b

Values are means of three replicates. Standard errors of means are enclosed in parenthesis. In a row, for specified parameter, means with different letters differ significantly from each other at  $p < 0.05$ .

(245.50 ± 6.16 °C) and bagasse (263.50 ± 5.01 °C) were significantly higher ( $p < 0.05$ ) compared to that for rice husk (115 ± 2.31 °C) and corncobs (197.57 ± 2.72 °C). Values of stack gas velocity were 12.33 ± 0.10, 14.34 ± 0.91, 14.17 ± 0.29 and 18.39 ± 0.30 m s<sup>-1</sup> for rice husk, rice straw, corncobs and bagasse respectively. Bagasse had significantly ( $p < 0.05$ ) higher stack gas velocity values compared to rice husk, rice straw and corncobs. There were significant ( $p < 0.05$ ) differences in length of burning cycle for rice husk, rice straw, corn cobs and bagasse. MCE ranged from 0.976 for rice husk and rice straw to 0.980 for bagasse. Bagasse had significantly ( $p < 0.05$ ) higher MCE compared to rice husk, rice straw and corncobs.

In this study, stack gas velocity was measured under ambient conditions since it determines speed of gaseous pollutant emissions from open burning of residue biomass and depends on the ambient environmental conditions like air flow to ensure optimum oxygen concentrations for complete and efficient burning (Wardoyo et al., 2006). Bagasse showed the highest value of stack gas velocity in this study. Burning cycle could also serve as important determinant of combustion efficiency of biomass and depends on physical and chemical characteristics of fuel biomass (Ward et al., 1992). MCE was measured to distinguish between flaming and smoldering mode of combustion during crop residue burning. MCE in our study was 0.976, 0.976, 0.978 and 0.980 for rice husk, rice straw, corncobs and bagasse respectively which falls in the range of 0.9–1.0 suggested by Reid et al. (2005) for fires following flaming as dominant mode of combustions. However, it is also an established fact that smoldering and flaming mode of combustions cannot be separated completely when biomass is burnt under field conditions. Nevertheless, MCE of crop residues in our study support the well-documented claim that agricultural crop residue burn under flaming mode under field and laboratory conditions (Saud et al., 2011; Zhang et al., 2008).

### 3.3. Emission factors of gaseous pollutants

Emission factors (EFs) of gaseous pollutants, calculated from emission concentrations, of rice husk, rice straw, corncobs and bagasse are shown in Fig. 4. The mean emission factors of CO<sub>2</sub> for rice husk, rice straw, corncobs and bagasse were 880.48 ± 8.99, 1090.1 ± 24.0, 595.44 ± 10.4 and 937.03 ± 9.07 g kg<sup>-1</sup> respectively (Fig. 4a). Emission factor of CO<sub>2</sub> of rice straw were significantly ( $p < 0.05$ ) higher compared to rice husk, corncobs and bagasse. Emission factors of CO from rice husk (14.04 ± 0.18 g kg<sup>-1</sup>), rice straw (17.19 ± 0.28 g kg<sup>-1</sup>), corncobs (14.04 ± 0.18 g kg<sup>-1</sup>) and bagasse (12.39 ± 0.08 g kg<sup>-1</sup>) followed order similar to that of CO<sub>2</sub> emission factors (Fig. 4b). Emissions factors of NO<sub>2</sub> were 0.19 ± 0.03, 0.89 ± 0.03, 0.16 ± 0.01 and 0.36 ± 0.03 g kg<sup>-1</sup> for rice husk, rice straw, corncobs and bagasse respectively (Fig. 4c). Emission factors of NO<sub>2</sub> from rice straw were significantly ( $p < 0.05$ ) higher compared to that from rice husk, rice straw, corncobs and bagasse. Emission factors of NO from rich husk, rice straw and bagasse were 1.38 ± 0.02, 1.48 ± 0.04 and

1.44 ± 0.01 g kg<sup>-1</sup> respectively (Fig. 4d); they were significantly ( $p < 0.05$ ) higher from NO emission factors of corncobs (0.70 ± 0.01 g kg<sup>-1</sup>). Emission factors of NO<sub>x</sub> ranged from 1.23 ± 0.01 g kg<sup>-1</sup> for corncobs to 3.16 ± 0.08 g kg<sup>-1</sup> for rice straw (Fig. 4e). Emission factors of SO<sub>2</sub> were 0.11 ± 0.03, 0.38 ± 0.03, 0.02 ± 0.00 and 0.18 ± 0.02 g kg<sup>-1</sup> from burning of rice husk, rice straw, corncobs and bagasse respectively (Fig. 4f). Emission factors of SO<sub>2</sub> from rice straw were found to be highest and significantly ( $p < 0.05$ ) different from rice husk, corncobs and bagasse.

Emission factor is an important tool to estimate total gaseous pollutant emissions to help making pollution inventories and policy decision to mitigate air pollution (van Leeuwen and Hermens, 1995; Andreae and Merlet, 2001; Yang et al., 2008). Emission factors for different crop residues have been widely reported in literature, especially of rice straw. The rice straw produced the highest emission factors of the trace gases in the current study. The emission factors of CO, CO<sub>2</sub>, NO<sub>2</sub>, NO, NO<sub>x</sub> and SO<sub>2</sub> from rice straw were calculated to be 17.19 ± 0.28, 1090.1 ± 24.0, 0.89 ± 0.03, 1.48 ± 0.04, 3.16 ± 0.08 and 0.38 ± 0.03 g kg<sup>-1</sup> respectively which showed considerable agreement with data of some previous studies on rice straw e.g. emission factors of CO<sub>2</sub> 101 g kg<sup>-1</sup> (Smith et al., 1993), NO<sub>x</sub> 3.43 g kg<sup>-1</sup> (Guoliang et al., 2008) and NO<sub>2</sub> 0.79 g kg<sup>-1</sup> (Zhang et al., 2008). However, emission factors of CO and SO<sub>2</sub> were found to be different than those reported in literature e.g. Jenkins et al. (1996) reported 31.41 and 0.62 g kg<sup>-1</sup> emission factors of CO and SO<sub>2</sub> respectively which were higher than those reported in our study. The emission factors of CO, CO<sub>2</sub>, NO<sub>2</sub>, NO, NO<sub>x</sub> and SO<sub>2</sub> from bagasse were 12.39 ± 0.08, 937.03 ± 9.07, 0.36 ± 0.03, 1.44 ± 0.02, 2.57 ± 0.04, and 0.18 ± 0.02 g kg<sup>-1</sup> respectively. Emission factors of NO<sub>x</sub> and NO for bagasse from this study were comparable to 2.6 g kg<sup>-1</sup> NO<sub>x</sub> (Dennis et al., 2002) and 1.7 g kg<sup>-1</sup> NO (Brocard et al., 1996). However, emission factors of CO, CO<sub>2</sub>, NO<sub>2</sub> and SO<sub>2</sub> from bagasse were lower from those previously reported e.g. CO 34.7 g kg<sup>-1</sup> and CO<sub>2</sub> 1130 g kg<sup>-1</sup> (Kanabkaew and Oanh, 2011), NO<sub>2</sub> 1.6 g kg<sup>-1</sup> (Brocard et al., 1996), SO<sub>2</sub> 0.23 g kg<sup>-1</sup> (Kato, 1996) and 0.50 g kg<sup>-1</sup> (Gadi et al., 2003). These differences in emission factors could be due to factors like moisture content and local climatic conditions (Goldammer et al., 2009), physical and chemical differences in the crop residue composition of different regions (Lobert and Warnatz, 1993) and, especially N contents for the variations in NO<sub>x</sub> emission factors (Zhang et al., 2008).

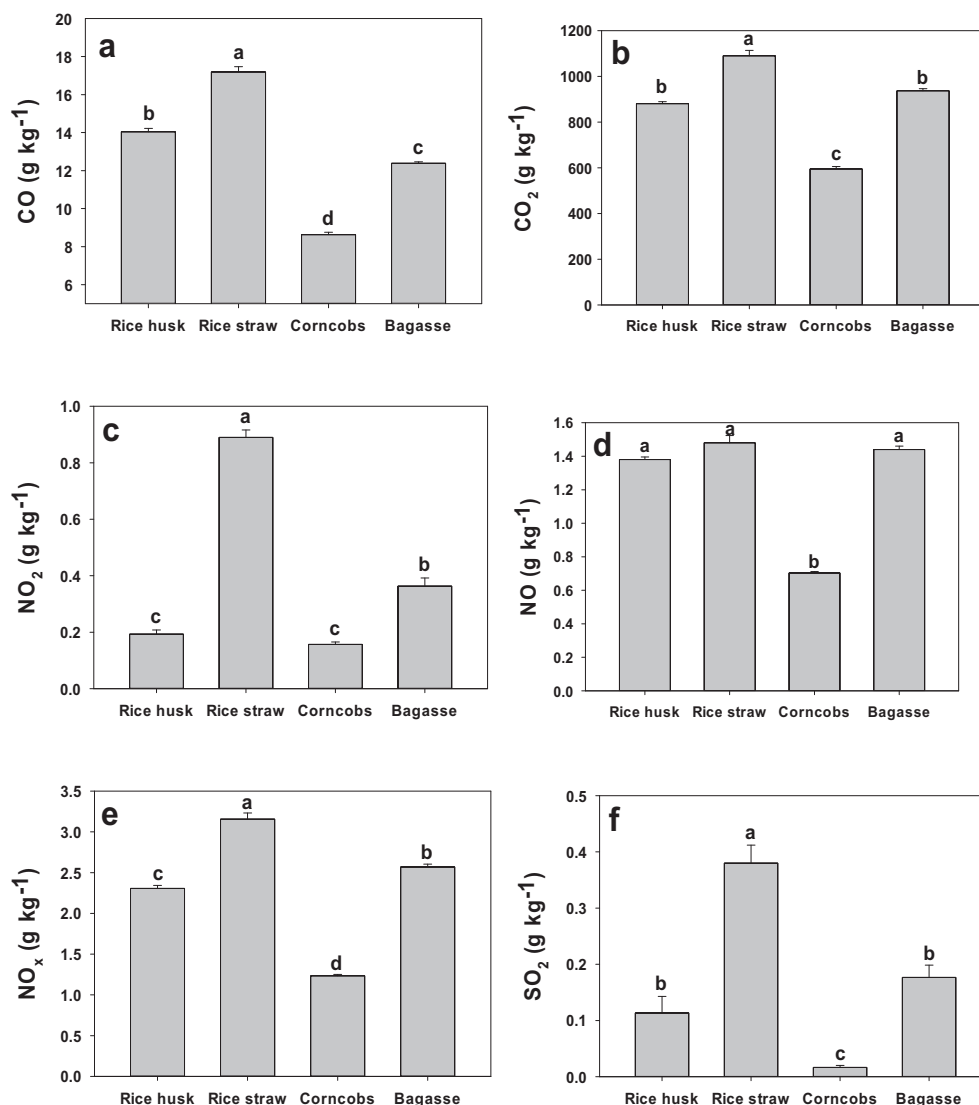
The emission factors from corncobs and rice husk have not widely been reported in the literature and this is perhaps the first attempt in this regard. The emission factors of CO, CO<sub>2</sub>, NO<sub>2</sub>, NO, NO<sub>x</sub> and SO<sub>2</sub> from burning of corncobs were observed to be 8.63 ± 0.12, 595.44 ± 10.38, 0.16 ± 0.01, 0.70 ± 0.01, 1.23 ± 0.02 and 0.02 ± 0.00 g kg<sup>-1</sup> respectively. The results of emission factors of SO<sub>2</sub> and NO<sub>x</sub> were in reasonable agreement with 0.04 g kg<sup>-1</sup> for SO<sub>2</sub> by Cao et al. (2008) and 1.27 g kg<sup>-1</sup> for NO<sub>x</sub> by Zhang et al. (2008) which were based on the burning of aggregated maize crop waste. However, the emission factors of CO and CO<sub>2</sub> from our study for corncobs differed from those reported by Andreae and Merlet (2001) for CO (53 g kg<sup>-1</sup>) and Zhang et al. (2008) for CO<sub>2</sub>

**Table 2**

Flu temperature, stack gas velocity, burning cycle and modified combustion efficiency (MCE) of rice husk, rice straw, corncobs and bagasse.

Parameter	Rice husk	Rice straw	Corncobs	Bagasse
Flu temperature (°C)	115 (2.31) c	245.50 (6.16) a	197.57 (2.72) b	263.50 (5.01) a
Stack gas velocity (m s <sup>-1</sup> )	12.33 (0.10) c	14.34 (0.91) b	14.17 (0.29) b	18.39 (0.30) a
Burning cycle (s)	953.33 (5.24) b	990 (4.04) a	618.33 (4.91) d	783.33 (8.25) c
Modified combustion efficiency (MCE)	0.976 (0.00) a	0.976 (0.00) a	0.978 (0.00) b	0.980 (0.00) c

Values are means of three replicates. Standard errors of means are enclosed in parenthesis. In a row, for specified parameter, means with different letters differ significantly from each other at  $p < 0.05$ .



**Fig. 4.** Gaseous pollutant emission factors from burning of rice husk, rice straw, corncobs and bagasse: (a) CO, (b) CO<sub>2</sub>, (c) SO<sub>2</sub>, (d) NO<sub>x</sub>, (e) NO<sub>2</sub>, (f) NO (units: g kg<sup>-1</sup>). Values are average of three replicates. Error bars are standard error of means ( $n = 3$ ). Bars with different letters differ significantly from each other at  $p < 0.05$ .

(1160 g kg<sup>-1</sup>). This difference could be due to the reason that they measured emission factors by burning aggregate maize crop residue; however, in contrast, we used corncobs which could result in different emission factors through changes in the composition of biomass (Lobert and Warnatz, 1993). The emission factors of CO, CO<sub>2</sub>, NO<sub>2</sub>, NO, NO<sub>x</sub> and SO<sub>2</sub> from burning of rice husk were  $14.04 \pm 0.18$ ,  $880.48 \pm 8.99$ ,  $0.19 \pm 0.01$ ,  $1.38 \pm 0.02$ ,  $2.31 \pm 0.04$  and  $0.11 \pm 0.03$  g kg<sup>-1</sup> respectively. The results suggested considerable differences in emission factors of rice straw and rice husk due to the very fact that they were sampled from different locations.

#### 3.4. Emission estimates, inventories and allocation of gaseous pollutants

Rice husk, rice straw, corncobs and bagasse are important residue producing crops being used as biomass fuel in Pakistan. In order to prepare emission estimates, inventories and allocations, quantity of crop residues was estimated to be 1232, 9240, 1281 and 19153 Mt for rice husk, rice straw, corncobs and bagasse respectively (Table 3). Based on the dry matter fraction (Streets et al., 2003) and percent of the crop residues being combusted (25%;

Iqbal and Goheer, 2008), total crop residue burned for rice husk, rice straw, corncobs and bagasse was found to be 262, 1964, 128 and 3400 Mt respectively (Table 4). Bagasse had the highest values for residue production and combustion followed by rice straw, rice husk and corncobs respectively.

Total emissions (Gg) from crop residues for CO, CO<sub>2</sub>, NO<sub>2</sub>, NO, NO<sub>x</sub> and SO<sub>2</sub> are presented in Table 5. Total emissions from bagasse were 42.12, 3185.53, 1.22, 4.90, 8.74 and 0.61 Gg for CO, CO<sub>2</sub>, NO<sub>2</sub>,

**Table 3**

Estimation of production of rice straw, rice husk, bagasse and corncobs in Pakistan in 2011–12.

Residue type	Crop production (Mt) <sup>a</sup>	Crop to residue ratio <sup>b</sup>	Total crop residue production (Mt) <sup>c</sup>
Rice husk	6160	0.20	1232
Rice straw	6160	1.50	9240
Corncoobs	4271	0.30	1281
Bagasse	58,038	0.33	19,153

<sup>a</sup> Government of Pakistan (2011–12).

<sup>b</sup> Singh and Gu (2010).

<sup>c</sup> Metric tons.



**Table 4**  
Estimation of residue burnt in Pakistan in 2011–2012.

Residue type	Total crop residue (Mt)	Dry matter fraction <sup>a</sup>	Crop residue/dry matter burnt (%) <sup>b</sup>	Total residue burnt (Mt) <sup>c</sup>
Rice husk	1232	0.85	25	262
Rice straw	9240	0.85	25	1964
Corncoobs	1281	0.40	25	128
Bagasse	19,153	0.71	25	3400

<sup>a</sup> Streets et al. (2003).

<sup>b</sup> Iqbal and Goheer (2008).

<sup>c</sup> Metric tonns.

NO, NO<sub>x</sub> and SO<sub>2</sub> respectively. Total emissions from bagasse for CO, CO<sub>2</sub>, NO and NO<sub>x</sub> were the highest compared to those from rice straw, rice husk and corncoobs. However, total emissions of NO<sub>2</sub> (1.75 Gg) and SO<sub>2</sub> (0.75 Gg) from the rice straw were found to be highest compare to the other crop residues (Table 5). Total emissions for each gaseous pollutant from burning of crop residue were 80.66 Gg for CO, 5632.67 Gg for CO<sub>2</sub>, 3.04 Gg for NO<sub>2</sub>, 8.19 Gg for NO, 15.70 Gg for NO<sub>x</sub> and 1.42 Gg for SO<sub>2</sub>. Calculated from Table 5, emission allocations for gaseous pollutants from rice husk and bagasse together accounted for 94.1, 94.6, 97.7, 95.4, 95.2 and 95.8% total emission of CO, CO<sub>2</sub>, NO<sub>2</sub>, NO, NO<sub>x</sub> and SO<sub>2</sub>. Our study also showed that the cumulative contribution of rice husk and corncoobs to the total emissions of gaseous pollutants was marginal.

Total emissions of gaseous pollutants from burning of rice husk, rice straw, corncoobs and bagasse were many fold lowered compared to those reported in studies from China (Zhang et al., 2008) and India (Saud et al., 2011). However, China and India are the largest countries of the world with remarkably higher agricultural crop production and crop residue generation. We estimated total emissions of gaseous pollutants considering burning of only 25% crop residue as is the case in China suggested by Gao et al. (2002) and Iqbal and Goheer (2008); however, recent energy crisis in Pakistan has led to far higher utilization of crop residues as biofuel which may mean that actual total emission could be higher. We have observed that bagasse contributed largely to the budgets of gaseous pollutants especially of CO, CO<sub>2</sub> and NO<sub>x</sub> in Pakistan. This could be related to the considerably higher emission factors and the most importantly, larger amounts of bagasse production as compared to rice straw, rice husk and corncoobs. The rice straw and the bagasse contributed more than 90% of total emission of gaseous pollutants. Field burning of rice husk, rice straw, corncoobs and bagasse is not commonly practiced in Pakistan; however, these crop residue are largely consumed in industrial and rural sectors. In addition, household income of large percent of farmers in Pakistan is low whereas energy supply and cost is becoming expensive so they usually opt to use crop residues to meet domestic energy requirements. The latter claim is supported by studies of Cao et al. (2008) and Chen (2001) who found that field burning of crop residues was related to income level of farmers.

**Table 5**  
Estimation of total gaseous pollutant emissions (Gg) from the crop residue burning in Pakistan in 2011–12.

Residue type	Total emissions (Gg) <sup>a</sup>					
	CO	CO <sub>2</sub>	NO <sub>2</sub>	NO	NO <sub>x</sub>	SO <sub>2</sub>
Rice husk	3.68	230.51	1.75	2.91	6.20	0.75
Rice straw	33.75	2140.35	0.05	0.36	0.60	0.03
Corncoobs	1.11	76.28	0.02	0.02	0.16	0.61
Bagasse	42.12	3185.53	1.22	4.90	8.74	0.03
<b>Total</b>	<b>80.66</b>	<b>5632.67</b>	<b>3.04</b>	<b>8.19</b>	<b>15.70</b>	<b>1.42</b>

<sup>a</sup> 1 Gg = 1 × 10<sup>9</sup> g.

We have reported emission factors and total emissions from combustion of crop residues which are commonly used in industrial and household sectors of Pakistan. According to best of our knowledge, this is the first study reporting emission inventories of gaseous pollutants from burning of agricultural residues. Results described in the study are assumed to be helpful in making national and provincial estimates of gaseous pollutants from frequently consumed agricultural residue biomass. However, it should be noted that variations in fuel properties and combustion conditions could lead to rather rough estimates of emission factors with high degree of uncertainty.

#### 4. Conclusions and future research

Recent energy crisis has led to increased dependency on agriculture-based biomass fuel combustion in agro-industrial and household sectors in Pakistan. Rice husk, rice straw, corncoobs and bagasse represent common biomass fuels in Pakistan. Biomass fuels differed markedly for physical, chemical and combustion characteristics. Modified combustion efficiency (MCE) ranged from 0.976 to 0.980 indicating flaming as the mode of combustion under ambient conditions. This study reports experimentally measured gaseous pollutant emission concentrations, emission factors and emission inventories of rice husk, rice straw, corncoobs and bagasse combusted under ambient outdoor conditions using specially designed burning tower. Emission factors of CO, CO<sub>2</sub>, NO<sub>2</sub>, NO, NO<sub>x</sub> and SO<sub>2</sub> were determined to be 14.05 ± 0.18, 880.48 ± 8.99, 0.19 ± 0.01, 1.38 ± 0.02, 2.31 ± 0.04 and 0.11 ± 0.03 g kg<sup>-1</sup> for rice husk, 17.19 ± 0.28, 1090.07 ± 24.0, 0.89 ± 0.03, 1.48 ± 0.04, 3.16 ± 0.08 and 0.38 ± 0.03 g kg<sup>-1</sup> for rice straw, 8.63 ± 0.12, 595.44 ± 10.38, 0.16 ± 0.01, 0.70 ± 0.01, 1.23 ± 0.02 and 0.02 ± 0.00 g kg<sup>-1</sup> for corncoobs and 12.39 ± 0.08, 937.03 ± 9.07, 0.36 ± 0.03, 1.44 ± 0.02, 2.57 ± 0.04 and 0.18 ± 0.02 g kg<sup>-1</sup> for bagasse. Results of emission factors of gaseous pollutants from burning of rice husk, rice straw, corncoobs and bagasse were in reasonable agreement with those reported elsewhere. Total emissions of CO, CO<sub>2</sub>, NO<sub>2</sub>, NO, NO<sub>x</sub> and SO<sub>2</sub> from burning of biomass fuels were estimated to be 80.66, 5632.67, 3.04, 8.19, 15.70 and 1.42 Gg respectively. On cumulative basis, rice straw and bagasse contributed more than 90% of total emissions of gaseous pollutants. Results of this study are important in formulating provincial and regional budgets of gaseous pollutants from burning of agricultural residues. However, biomass fuels like cotton sticks and dung cake needs to be assessed for their role in emission of gaseous pollutants in future since they also represent important biofuels in rural sectors of Pakistan.

#### Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.atmosenv.2013.11.046>

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