

# Particulate black carbon and gaseous emission from brick kilns in Greater Dhaka region, Bangladesh

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#### Abstract

Eighteen brick kilns of three brick-making technologies (Fixed Chimney Kiln (FCK), Zigzag, and Hoffmann) were selected to measure the concentrations of particulate matter ( $PM_{2,5}$ ) with Aerocet 531S (USA) sampler, black carbon (BC) with Magee Scientific, OT-21 Soot scan Transmissometer (USA), and gaseous pollutants (CO2, CO, SO2, NO3, and volatile organic carbon (VOC)) with Aeroqual 500 gas sampler (New Zealand) to understand the emission scenario from brick sector in Greater Dhaka region, Bangladesh. Emission factor (EF) of each pollutant was computed from their respective concentration for three brick kiln technologies. Ambient  $PM_{2.5}$  and  $PM_{10}$  were measured in brick kiln premises and 1 km far from the respective kilns to see the effect on the surrounding areas. The PM<sub>2.5</sub> concentration was found on an average of  $141 \pm 86$ ,  $128 \pm 72$ , and  $110 \pm 53$  mg/m<sup>3</sup> in FCK, Zigzag, and Hoffmann kilns, respectively. The average BC concentration was found 16.6 ± 7.1 (FCK), 11.8 ± 4.2 (Zigzag), and  $8.9 \pm 4.4$  (Hoffmann) mg/m<sup>3</sup>. FCK has a greater emission of CO, whereas Zigzag has a higher CO<sub>2</sub> emission. A comparatively higher value of CO<sub>2</sub> and lower value of CO indicates effective combustion of coal, and this is found to be more efficient for Zigzag and Hoffmann compared to traditional FCK. SO<sub>2</sub> and VOC emissions were depending not only on the kiln types but also on the fuel qualities. From EF calculation, approximately 4526 t of PM2.5, 340 t of BC, 209,776 t of CO2, 8700 t of CO, 19,441 t of SO<sub>2</sub>, and 835,450 t of VOC per year found to emit from 1000 brick kilns. The conversion of traditional FCK to improved one, i.e., Zigzag and/or Hoffman is not a straight forward solution, as CO<sub>2</sub> emission was higher in Zigzag whereas BC and PM<sub>2.5</sub> emissions were higher in FCK. Therefore, considering EF of various pollutants from these three types of kilns, conversion of FCK to Zigzag or Hoffmann could be a better choice.

Keywords Brick kilns · Black carbon · Gaseous pollutants · Emission factor

## Introduction

Brick-producing industry is a vital sector for the severe air pollution problem in a country like Bangladesh. More than 6000 brick kilns are preset and contributed significantly to the worst air quality in Bangladesh (UNDP, 2015; Motalib et al. 2015). Most of the kilns produce fired clay red brick which is the major building material in this country and also widely used throughout the South Asia (World Bank 2011).

Abdus Salam asalam@gmail.com Approximately 87% of the 1.5 trillion clay bricks are produced annually in Asia and 18% of those are made in South Asia (India, Bangladesh, Nepal, and Pakistan) (Weyant et al. 2014). Greater Dhaka region of Bangladesh, which is a densely populated area, produced 3.5 billion bricks per year nearly from 1000 brick kilns (Guttikunda and Goel, 2013). Most of these bricks are manufactured traditionally from clay by burning of coal and biomass without proper pollution control.

Emissions of aerosols and gaseous pollutants from brick kilns have considerable respiratory health effect of on-site employers (Pariyer et al. 2013; Kaushik et al. 2012; Zuskin et al. 1998) and subsequently deteriorate the ambient air quality (Guttikunda and Khaliquzzaman, 2014; Joshi and Dudani, 2008). Around 4.5 million people die each year from the effects of outdoor air pollution reported last year by Lancet Commission on Pollution and Health and Bangladesh ranked the top most position for human deaths (Landrigan et al. 2017). Another study revealed that India's air quality has worsened fastest in past decade

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(Lelieveld and Poschl, 2017). Some major cities in South Asia like Dhaka (Begum et al. 2010), New Delhi (Guttikunda et al. 2013), and Kathmandu (Tuladhar and Raut, 2002) are facing severe air quality problems due to the clusters of brick kilns situated around the cities. For example, brick kilns have been responsible for 38% of PM<sub>2.5</sub> pollution in Dhaka Metropolitan Area (Begum et al. 2006, 2008, 2011). The earlier study suggested that estimated emissions from 1000 brick kilns in Greater Dhaka region were 23,300 t of PM<sub>2.5</sub>, 15,500 tof SO<sub>2</sub>, 302,000 t of CO, 6000 t of BC, and 1.8 million tons of CO<sub>2</sub> per year (Guttikunda et al. 2013). However, much lower emissions have found in the current study which will be discussed later.

Among various pollutants emitted from brick kilns, black carbon (BC) emission due to incomplete combustion is a primary concern as it has both health and climate impact and it is known to have second highest radiative forcing after CO<sub>2</sub> (Bond et al., 2013). In India (Reddy and Venkataraman, 2002) and China (Wang et al. 2012), brick kilns are estimated to produce 7% of BC emissions on a national scale. SO<sub>2</sub> emission is also considered as a main emission from brick kilns, and it has found that most of the coals used in Bangladeshi kilns are imported from India and Indonesia, which have a higher level  $(\sim 5\%)$  of sulfur (Skinder et al. 2014; Bhanarkar et al. 2002). Currently, in developing countries, SO<sub>2</sub> is the main issue regarding air pollution problems as it contributes both to urban pollution and to regional acid depositions (Cofala et al. 2004). Volatile organic carbon (VOC) emission mainly depends on fuel quality and burning efficiency of the kilns. A higher amount of VOC and SO<sub>2</sub> can emit if low-grade coal is used.

In this study, three types of kilns, namely Fixed Chimney Kiln (FCK), Zigzag, and Hoffmann, were selected for the experimental purpose. Among the total kilns in Bangladesh, 92% of them are traditional FCK, around 5% are Zigzag, and 1% is Hoffmann (World Bank 2011). FCK is a continuous type of kilns where firing zone is moving through the stationary green bricks and firing pattern is straight for green bricks. The height of the chimney is 120 ft, and the kiln can be made circular or elliptical in shape (Heierli and Maithel, 2008). Three types of zones (brick cooling, preheating, and combustion zone) usually exist in FCK, where the air of cooling zone picks up heat from the fired bricks whereas in preheating zone, unfired bricks are heated by the warm combustion gases (Heierli and Maithel, 2008). Though FCK is ideally suited for the present level workers, it is an outdated technology and thermally lowefficient kilns. Most of the Zigzag kilns in Bangladesh are transformed from FCK by reducing the chimney height to 60 ft and increasing the length of the kiln gallery by zigzagging the firing chambers. Here, the fire follows a zigzag path throw 44 to 52 chambers separated by a batch of green bricks, which ensures longer combustion time. It has a laden connected to the chimney with induced draught fan. It requires electricity and generator which are used for that induced draught fan. Due to lesser fuel usage and better brick stacking and flue gas scrubbing in a water-filled duct which presents to the outlet chimney. Zigzag kiln becomes more sophisticated and effective technology (Maithel, 2012). Hoffmann is not popular in Bangladesh as it is very expensive to build and requires regular maintenance. The chimney is built outside the kiln structure and is approximately 50-70 ft. In Hoffmann, the firing path is elliptical and about 16-22 barrel arched chamber (like a railway tunnel) is used in the kiln gallery. In this kiln, the firing zone is stationary, but the bricks are pulled horizontally using carts on rails (Weyant et al. 2014). Evenly firing is controllable by the use of the dampers and careful stoking. In addition, strong fan draught system assists a high rate of fire traveling to ensure proper combustion. Hoffmann kilns can be operated throughout the year regardless of adverse weather condition such as in monsoon. Although their primary investment is high enough, they can return it in a profitable way within few years (World Bank 2011). Hoffmann is similar to tunnel kilns where natural gas is used instead of coal and due to banned of supplying natural gas in brick sector; tunnel kilns in Bangladesh are also using coal as fuel. Hoffmann and tunnel kilns are quite popular in Vietnam and China (Maithel, 2012; Heierli and Maithel, 2008), and very few are present in Bangladesh (World Bank 2011).

It is very important to modernizing brick kilns to reduce the short-lived climate pollutants, such as BC and alternate fuels are necessary to minimize the dependency on coal (United Nations Environment Program and World Meteorological Organization 2011; US Environmental Protection Agency, 2012). There is still a large uncertainty exist in emission factors due to regional variations in fuel use and technologies (Zhao et al. 2011; Bond et al., 2013). Therefore, it is necessary to quantify direct measurement of particulate emission (BC, PM2.5) and gaseous pollutants (SO<sub>2</sub>, CO, CO<sub>2</sub>, NO<sub>2</sub>, VOC) from most common fuel and kiln types. Emission factor (EF) estimation of these pollutants from those relevant kilns will help policy makers to take appropriate policy intervention to mitigate the air pollution from brick kilns. To corroborate this idea, this study reports emissions from 18 selected kilns of seven different clusters (Ashulia, Dhamrai, Gajipur, Fotulla, Rupgonj, Narayanganj, and Savar) in and around Dhaka megacity. This study represents emission factors of BC, PM2.5, CO2, CO, SO2, and VOC for FCK, Zigzag, and Hoffmann types of kilns. Moreover, ambient PM25 and PM10 were also measured in brick kiln campuses and 1 km far from the kilns to see the effect in the surrounding areas. However, findings of this study will help to reduce the emission uncertainties in future.

# Methodology

#### Brick kiln and cluster selection

In the Greater Dhaka region, nearly 1000 brick kilns were previously mapped in the vicinity of DMA (Guttikunda et al.



Fig. 1 Location of brick kilns in Greater Dhaka region, Bangladesh, with dots and also indicating seven clusters with star in red color. Eighteen brick kilns were chosen for sampling of these seven clusters

2013). At present, a total of 925 brick kilns are found registered and among them, 870 kilns are actively running. Based on

direct field observation and global positioning system (GPS), the locations of these kilns have been identified in this study. Later, using geographical remote sensing data, a latest GISbased mapping of these kilns has been done (Fig. 1). Ten FCK, six Zigzag kilns, and two Hoffmann kilns were selected from clusters of kiln. For the selection of 18 sampled kilns, random selection was not possible as kiln owner's permission was required for sampling in kilns. Among the selected clusters in Dhaka Metropolitan Area (DMA), it has been found that Savar (25%), Gazipur (19%), and Narayanganj (18%) have the major proportion of brick kilns. Field survey suggested that most of the kilns (51%) are situated nearby rivers and almost 5% brick kilns are placed nearby communities. They also have taken place over the middle of open and cultivable lands (18%). These nearby rivers and canals also serve as the major routes for transporting coal, soil, and also fired brick from and to their destinations. Most of the kilns are operated usually through 5-6-month period from mid-November to mid-April before monsoon period started. Modern brick kilns as Hoffmann can produce bricks throughout the year as it does not depend on weather. Average production rate of bricks, major types of fuel used, and their imported places were figured out by field survey at individual kiln.

# Sampling

#### Black carbon (BC) and PM<sub>2.5</sub>

Black carbon (BC) and PM2.5 concentrations were measured from the flue gas which was directly collected through the hole created in the chimney. A standard 20-25-ft height from the top layer of bricks was maintained for sampling in most of these kilns, and a minimum of three repetitive runs was carried out in each test to maintain the data quality. Also, a blank sample was carried out in every kiln for measurement of BC. From each brick kiln, multiple sampling was carried out at different conditions like before, after, and time of fuel loading for the high quality of dataset. In some Zigzag kilns, where the creation of a hole in the chimney was not permitted, we opened the lid which concealed the pathway of fume and then collected the sample of gaseous measurement for a short period. Isokinetic source sampler (Apex instrument) was used for BC sampling, and this sampler is used according to method 201A (EPA 2010). Fine particulate control device extracted BC samples from stack emission and BC loaded on 47-mm quartz fiber filters (Gelman, membrane filters, type tissue quartz 2500 gat-up) placed at 47-mm stainless steel filter holder at the end of 1.5-m heated probe.

The pitot tube was used to measure stack velocity, and flue gas temperature was measured by thermocouples at the nozzle location. Most of the cases, stack velocity was lower than 3/ ms and it was difficult to collect BC by this lower velocity in an isokinetic manner. Hence, BC sampling was carried out at selected appropriate target flow rate, and as BC is generally accepted to be ultrafine particulate, sampling bias on this particles are expected to be minor. A diluent sample was collected simultaneously with colocated to the BC sample, and flow rate was controlled by the maintaining the static pressure at the sample location.

Tedlar bags were used to collect the integrated dry sample for the measurement of CO<sub>2</sub>%, and bag samples were analyzed with a portable flue gas analyzer (Viasensor model G-100). Loaded filters were analyzed directly by Magee Scientific OT-21 Soot scan Transmissometer. All filters were treated in the furnace at 800 °C for about 2 h prior to sampling to remove any traces of residual carbon. Filters were stored in individual Petri dishes after first oven treatment. After sampling, the loaded filter was measured against a blank filter at 880 nm wavelength in the OT-21. After this initial measurement, a run filter was treated again at 800 °C in the furnace for 15 min to burn off any organic carbon or mineral matter (e.g., fly ash) and leave inorganic residues. A subsequent 10-min gap needed to cool the burned filter before operating the second measurement against the same blank filter. Thus, BC emissions were correlated with the differences in the two sample measurements. Duration of sampling, i.e., mass loaded on the filter, is done based on the attenuation factor instrument response between 20 and 120, as too dark sample prevents the light transmission through the sample.

 $PM_{2.5}$  concentration was determined gravimetrically from the weight difference of (collected on quartz filters with a isokinetic particulate matter sampler) between loaded and unloaded filters, which was used for BC analysis subsequently.

#### **Gaseous pollutants**

Gaseous pollutants (CO<sub>2</sub>, CO, SO<sub>2</sub>, VOC, NO<sub>x</sub>, and O<sub>3</sub>) were also directly measured from flue gas by low volume (0.5 LPM) Aeroqual 500 real-time sampler through an electrochemical gas sensor. Ambient particulate matter (PM) were measured by another sensor-based device AEROCET 531S (flow rate 2.83 LPM). All the samples were collected before, during, and after fuel loading. Sulfur content from different coal sampled used in kilns were measured as per standard measurement techniques ASTM D3177-89 (1997). Realtime and integrated sample measurements were averaged for the presentation (Table 1).

### **Emission measurements**

#### Stack emission

Stack emissions of pollutants greatly varied due to kiln technology, different grades of coal used, and also their feeding practices. Specific fuel consumption (SFC) is an important parameter for measurement of emission factors. Therefore, key monitoring-based datasets are necessary to estimate the

Table 1Measurements andanalysis techniques in the brickkilns—Dhaka, Bangladesh

Parameter measured	Sample location	Analysis method	Types of presentation
Black carbon (BC)	Measured from collected flue gas	Optical attenuation method	Average of single point observations
Gaseous pollutants (CO <sub>2</sub> , CO, SO <sub>2</sub> , NO <sub>2</sub> , VOC)	Measured from collected flue gas	Electro-chemical gas sensor technique	Real-time measurements
Particulate matter (PM <sub>2.5</sub> , PM <sub>10</sub> )	Collected from flue gas and ambient air	Gravimetric (stack) and electro-chemical sensor technique (ambient)	Integrated sample taken over hours (ambient)

SFC (Suresh et al. 2016). This can be derived from the data collected during field measurement (Table 2). In this study, SFC varied between 42–69, 54–68, and 46–54 g/kg of fuel for FCK, Zigzag, and Hoffmann kilns, respectively. The brick production rates varied per day from 10,000 to 45,000 per kiln, and it depends on manufacturing capacity of the kiln with the availability of labor and raw materials.

# Black carbon emission measurement

BC emission varies according to fuel used, type of kiln, and their operating conditions. BC emission is determined in terms of energy consumed (mg/MJ) and in terms of fuel consumed (mg/kg). The average concentration of BC can be calculated through attenuation factor (ATN) measurement for a blank and a sample filter through Magee Scientific OT-21 Sootscan transmissiometer by the following equation:

$$ATN = 100 \times ln$$
 (blank transmission/sample transmission) (1)

BC density is calculated by using ATN value at 880 nm with the following equation:

$$\delta_{BC} = (ATN_1 - ATN_2) / \sigma_{ATN} \tag{2}$$

where,

$\delta_{BC}$	carbon density ( $\mu$ g/cm <sup>2</sup> )
$\sigma_{ATN}$	specific attenuation coefficient (cm $^{2}/\mu g$ ) (16.6 cm $^{2}/\mu$
	μg)
ATN <sub>1</sub>	attenuation of sample before heat treatment
$ATN_2$	attenuation of sample after heat treatment

BC density is converted to stack concentration by using the area of the filter and volume of gas sampled:

$$C_d = (\delta \times A \times 35.31) / V_{ms} \tag{3}$$

where,

- $C_d$  measured carbon concentration (µg/dscm) (dscm denotes dry gas standard cubic meter)
- A collection area of the filter,  $14.5 \text{ cm}^2$  for 47 mm filter

 $V_{ms}$  volume sampled through dry gas meter, dry gas standard cubic feet (dscf)

# Determination of energy-based emission of BC (mg/MJ)

BC concentrations were corrected for dilutions to result in emissions. Pollutant concentrations can be transformed into emissions using procedures in Method 19 (EPA 2007) where fuel is the primary source of carbon and air is the primary source of oxidant and diluents. Derived Eq. (3) can be further used for measurement of BC emission (E) by the following equation:

$$E = (F_c \times 10^6 \times C_d \times 100\%) / (\% CO_2)$$
(4)

where,

Ε	black carbon emission (mg/MJ)
$F_c$	carbon based F factor $(4.84 \times 10^{-7} \text{ dscm/J})$
$C_d$	measured BC concentration (mg/dscm)
%CO <sub>2</sub>	carbon dioxide concentration - dry basis, %

 Table 2
 Types of brick kilns and key features investigated in this study

Type of kilns	Number of kilns ( <i>n</i> )	Major fuel used	Coal used per 100,000 bricks	Averaged fuel consumption (kg/day)	Average production of bricks/day	Specific fuel consumption (g/kg of fired brick)
FCK	10	Coal with occasionally biomass	20–22 t	4700	25,000	67
Zigzag	6	Coal	18–19 t	4181	22,000	61
Hoffmann	2	Coal	14–15 t	2388	16,000	49

#### Emission factor (EF) determination of BC

By using BC emission from Eq. (4), the emission factor of BC can be calculated as follows.

$$EF = (E \times GCV) /_{1000}$$

$$\tag{5}$$

where,

EF	black carbon emission factor (mg/kg)
E	black carbon emission (mg/MJ)
GCV	gross calorific value of fuel (kJ/kg)

By multiplying Eq. (5) with specific fuel consumption (kg/ kg of brick) gives another emission factor in terms of mg/kg of fired brick used.

# Emission factor estimation of PM<sub>2.5</sub> and gaseous pollutants

Data for brick production per day, the weight of bricks, types, and amounts of fuel used was collected from each sampling brick fields. Emission factor based on fuel used and energy content derived from pollutant concentrations are discussed as follows (Rajarathnam et al. 2014):

Emission rate ER  $\left(\frac{mg}{hr}\right)$ 

= concentration of pollutant  $(mg/m^3)$ 

× volumetric flow rate of flue gas  $(m^3/hr)$  (6)

where volumetric flow rate can be determined by multiplying velocity of flue gas (m/s) with the area of the stack  $(m^2)$ .

Mass production rate of bricks 
$$\left(\frac{kg}{h}\right)$$
  
= Production rate of bricks  $\left(\frac{bricks}{h}\right)$   
× brick weight(kg) (7)

Equation (7) can be further divided by Eq. (6) to give emission factor based on brick mass (g kg of brick).

Mass-based emission factor (EFm) can be calculated as follows:

 $EFm (g/kg of fuel used) = {^{ER (g/hr)}}\!/_{FC(kg/hr)}$ (8)

where FC is the fuel consumption rate (kg/h).

If Eq. (8) is divided by gross calorific value (GCV), then energy input-based emission factor (EFe) can be determined as follows:

$$EFe (g/MJ) = {}^{EFm (g/kg)} /_{GCV(MJ/kg)}$$
(9)

where GCV is the gross calorific value of the fuel (MJ/kg). Mass production rate of bricks can be found as follows:

Mass production rate of bricks 
$$\left(\frac{kg}{h}\right)$$

>

= production rate of bricks (bricks/h)

$$\langle \text{ brick weight } (\text{kg})$$
 (10)

Equation (6) divided by Eq. (10) can give emission factor based on mass of fired brick as follows:

Emission factor based on brick mass  $\left(\frac{g}{kg} \text{ of brick}\right)$ 

$$= \text{ER}\left(\frac{g}{h}\right)/\text{mass production rate of bricks}\left(\frac{\text{kg}}{h}\right) \quad (11)$$

All the results are represented here are the average population of 92% of FCK, 5% of Zigzag, and 1% of Hoffmann which are based on Bangladesh scenario (World Bank 2011).

Overall, Eqs. (1-5) are used to derive different types of emission factors for BC and Eqs. (6-11) are used for PM<sub>2.5</sub> and various gaseous pollutants.

# **Results and discussion**

Concentrations of all pollutants are presented in Table 3 and their corresponding emission factors are represented in Table 4. In addition, Table 5 has given for the comparison with other literature values. The following subsections discuss all the results in details for all types of kilns.

#### PM<sub>2.5</sub> concentration

 $PM_{2.5}$  concentration was found on an average of  $141 \pm 86$ ,  $128 \pm 72$ , and  $110 \pm 53 \text{ mg/m}^3$  in FCK, Zigzag, and Hoffmann, respectively (Table 3). FCK was the most  $PM_{2.5}$  emitter while Hoffmann was lowest. Repetitive direction

Pollutants	FCK	Zigzag	Hoffmann
BC	16.6 ± 7.1	11.8 ± 4.2	$8.9 \pm 4.4$
PM <sub>2.5</sub>	$141 \pm 86$	$128\pm72$	$109 \pm 53$
CO <sub>2</sub>	$5254\pm2021$	$6995\pm2667$	$2350\pm758$
СО	$264\pm75$	$177 \pm 81$	$74 \pm 21$
$SO_2$	$578\pm354$	$332\pm196$	$316\pm219$
VOC	$23,204 \pm 2560$	$25,266 \pm 3563$	$22,939 \pm 2760$
$NO_x$	$0.74\pm0.63$	$1.6\pm0.75$	$1.2\pm0.58$

All samples are presented in their averaged values from specific kilns

changes of flue gas and impinging on the walls happened in Zigzag, lower coal consumption, and better technology used in Hoffmann could be the reason that flue gas had the less particulate load. It is also understandable that due to the presence of more soot particle (BC) and incomplete combustion in FCK, black fumes were more enhanced in FCK compared to Zigzag and Hoffmann. As BC is considered fine particles, the fraction of BC was more in FCK. This can be justified from BC/PM<sub>2.5</sub> ratio from Table 3. BC fractions among PM<sub>2.5</sub> were 11.8% (FCK), 9.2% (Zigzag), and 8.1% (Hoffmann), respectively. Prescribed suspended particulate matter (SPM) level for Bangladeshi brick kilns is 1000 mg/m<sup>3</sup> by the Ministry of Environment and Forests (MoEF), Government of Bangladesh. In India, 750 mg/m<sup>3</sup> is considered for PM emission. Some of the PM levels suggested by earlier studies were 143–766 mg/m<sup>3</sup> (SSEF, 2012), 113–514 mg/m<sup>3</sup> (TERI, 2015), and 172 mg/m<sup>3</sup> (Suresh et al. 2016).

#### Black carbon concentration

BC concentrations varied between 0.02 to 1.3 mg/m<sup>3</sup> in three types of kilns before coal feeding with the corresponding average of  $0.95 \pm 0.45$  (FCK),  $0.22 \pm 0.09$ (Zigzag), and  $0.07 \pm 0.03$  (Hoffmann) mg/m<sup>3</sup>. After coal was loaded, BC concentrations escalated to  $16.6 \pm 7.1$ ,  $11.8 \pm 4.2$ , and  $8.9 \pm 4.4$  mg/m<sup>3</sup> (Fig. 2). BC concentration discharged from FCK was almost twice than Hoffmann kiln after fuel feeding (Table 3). More amount of coal is usually used in FCK compared to Zigzag and Hoffmann. Therefore, due to inefficient burning, more soot particles like BC emission attributed in FCK. In addition for Zigzag and Hoffmann, the induced draught fan, arduous path flow, continuous feeding in small quantities, and better mixing of fuel reflected better performance compared to FCK (Heierli and Maithel, 2008).

#### **Gaseous pollutant concentration**

Concentrations of gaseous pollutants in the flue gas that are measured from 18 selected kilns are presented in Table 3. First, the CO<sub>2</sub> emission is higher and CO emission is lower in Zigzag compared to traditional FCK. It might be possible due to the more efficient burning of fuel or complete combustion, more conversion of CO to CO<sub>2</sub> occurred in Zigzag than FCK. But it should also be noted that both CO<sub>2</sub> and CO is lowest in the case for more effective Hoffmann kilns where modern technology is used and also less amount of fuel used (SFC 0.49). Correlation ( $R^2$ ) between CO and CO<sub>2</sub> was found 0.35 in this study, whereas Weyant et al. (2014) suggested 0.4 for Indian brick kilns. Concentrations of CO across the 18monitored kilns ranged from 30 to 322 mg/m<sup>3</sup> with an average level of FCK, Zigzag and Hoffmann were 264 ± 75,  $177 \pm 81$ , and  $74 \pm 21$  mg/m<sup>3</sup>, respectively. During the time of

coal feeding, high levels of CO were observed and their maximum concentration declined gradually within few minutes after the fuel feeding operation.

Second, concentrations of SO<sub>2</sub> varied extensively between 44.5 and 740 mg/m<sup>3</sup> in various kilns with corresponding average values of  $578 \pm 354$  (FCK),  $332 \pm 196$ (Zigzag), and  $316 \pm 219$  (Hoffmann) mg/m<sup>3</sup>. SO<sub>2</sub> concentrations manifold based on sulfur content present in fuel and do not fully depend on kiln technology. It was found during the sampling period that most of the imported coal had a higher content of sulfur and varied between 0.35 and 5.58%. On average,  $116 \pm 47$  mg/m<sup>3</sup> was reported in Suresh et al. (2016) which was considerably lower than this study, but another earlier study in India where Shakti Sustainable Energy Foundation (SSEF) revealed that SO<sub>2</sub> levels were in the range of 29–611 mg/m<sup>3</sup>, which lies in vicinity of our observed values (SSEF, 2012).

Third, NO<sub>x</sub> concentration was found much lower and found to be insignificant emission. Although a better oxidation trend can be observed in improved Zigzag and Hoffmann kiln than traditional FCK in terms of NO<sub>x</sub> formation. Oxidation of nitrogen in the atmosphere created NO<sub>x</sub> emissions in the brick making by burning, and this NO<sub>x</sub> can play a major role in ozone formation (Amison 1992). Chemical reactions are mainly responsible for the NO<sub>2</sub> formation, and it does not depend on direct emission (Mayer et al. 1999). Several studies have also shown that significant emission of NO<sub>x</sub> in the brick making has not been found (Kolkmeier et al. 1991).

Fourth, VOC concentration was very high compared to other gaseous pollutants. VOC level also depends on coal grade, and if poor quality coal is used, then their emission might be higher. Their concentrations fluctuated around 23,000 mg/m<sup>3</sup> for all types of brick kilns.

#### Emission factor (EF) of the pollutants

#### EF of PM<sub>2.5</sub>

Mass (per kg of fuel and brick) and energy (per MJ of fuel) based emission factors in three types of kilns are shown in Table 4. All types of EF are higher in FCK compared to others kilns. Hoffmann kiln produced lowest EF of  $PM_{2.5}$ . There was no previous specific literature value for EF of  $PM_{2.5}$  in Bangladeshi brick kilns. An earlier study in India suggested that EF of  $PM_{2.5}$  in FCK was 0.18 and in Zigzag was 0.13 g/ kg of fired brick (Maithel et al. 2012). EF of PM (both fine and coarse) was found much higher in FCK (0.89) than Zigzag (0.24) reported by Rajarathnam et al. (2014). Table 5 shows a comparison of  $PM_{2.5}$  emission factors with other literature values available in South Asia and are in close agreement with these.

Bangradesh								
Pollutants		BC	PM <sub>2.5</sub>	CO <sub>2</sub>	СО	SO <sub>2</sub>	VOC	
Emission factors	g/kg of fuel (Coal)	FCK	$0.43\pm0.18$	6.12 ± 4.3	242 ± 92	7.5 ± 6.8	26.7 ± 17.6	1085 ± 947
		Zigzag	$0.31\pm0.09$	$5.88\pm3.2$	$379 \pm 144$	$6.8\pm5.9$	$18.5\pm13.4$	$1371\pm526$
		Hofmann	$0.25\pm0.07$	$4.73\pm2.4$	$237 \pm 162$	$5.6\pm3.1$	$33.7\pm15.3$	$1953\pm972$
	g/kg of fired brick	FCK	$0.03\pm.02$	$0.44\pm0.31$	$14.9\pm5.8$	$0.44\pm0.41$	$1.8\pm1.6$	$76.7 \pm 47.2$
		Zigzag	$0.02\pm0.01$	$0.42\pm0.29$	$21.4\pm8.6$	$0.41\pm0.38$	$1.1\pm0.87$	77.6 ± 35.9
		Hofmann	$0.01\pm0.01$	$0.34\pm0.18$	$12.6\pm5.9$	$0.31\pm0.18$	$1.8\pm0.87$	$98 \pm 48.3$
	g/MJ of fuel	FCK	$0.02\pm0.01$	$0.27\pm0.21$	$10.7\pm4.1$	$0.33 \pm 0.30$	$1.2 \pm 0.8$	$48.4 \pm 41.6$
		Zigzag	$0.014\pm0.004$	$0.25\pm0.18$	$16.9\pm6.4$	$0.31\pm0.26$	$0.8\pm0.6$	$61.2 \pm 23.4$
		Hofmann	$0.011 \pm 0.003$	$0.21 \pm 0.11$	$10.9 \pm 4.9$	$0.24 \pm 0.16$	$1.5 \pm 0.78$	$87.4 \pm 41.8$

**Table 4**Emission factors in terms of fuel, brick, and energy for all pollutants in three types of kilns (FCK, Zigzag, and Hofmann) in Dhaka,Bangladesh

#### EF of black carbon

BC concentrations were used to measure energy-based emission factor (mg/MJ of fuel) (Eq. (4)), which are further used to derive mass-based emission factor of BC (mg/kg of fuel) (Eq. (5)). Average gross calorific value of coal is considered to 22.4 MJ/kg (Guttikunda et al. 2013). Energy input based emission factor (mg/MJ) of BC varied between 2.03 and 45.2 with highest levels for FCK (averaged 19.6  $\pm$ 8.3) and lowest for Hoffmann (averaged  $11.4 \pm 3.2$  mg/MJ). Mass-based emission factor (g/kg of fuel) varied from 0.04 to 0.99 for all types of kilns during the sampling period, and FCK was the most BC emitter while Hoffmann was the lowest (Fig. 2). Average EF of BC for FCK, Zigzag, and Hoffmann are  $0.43 \pm 0.18$ ,  $0.31 \pm 0.09$ , and  $0.25 \pm 0.07$  g/ kg of fuel used, respectively (Table 4). The average weight of a fired brick is considered to be about 3.1 kg, and this weight of brick aid to measure emission factor of BC in terms of g/kg of fired brick. The literature on emission factors of BC for brick kilns is very limited, especially with developing countries perspective (Weyant et al. 2014). Weyant et al. (2014) measured EF of BC by carbon balance method for some types of kilns operated in India, and our measured EF (0.002 to 0.05) of BC in terms of g/kg of fired brick is comparable to that study (Table 5).

#### **EF of gaseous pollutants**

#### CO<sub>2</sub> and CO

EF of gaseous pollutants are derived from their concentrations, flow rate from stack emission and specific fuel consumption as discussed before. EF (g/kg brick) of CO<sub>2</sub> for three types of kilns are  $14.9 \pm 5.8$  (FCK),  $21.4 \pm 8.6$  (Zigzag), and  $12.6 \pm 5.9$  (Hoffmann), whereas for CO the averaged values are  $0.44 \pm 0.41$ ,  $0.41 \pm 0.38$ , and  $0.31 \pm 0.18$  (Table 4). Table 5 demonstrates that our measured emission factors of CO<sub>2</sub> are 7-10 times lower than other reported South Asian kilns (Rajarathnam et al. 2014; Suresh et al. 2016; Guttikunda et al. 2013). However, CO emission factors are someway similar or comparable to other literature values (Table 5). CO emission factor (7.4 in terms of g/kg of fuel used) lies between 6.6 and 53.5 which is suggested by Weyant et al. (2014) and are in close agreement in the case for Zigzag. In terms of g/kg of fired brick, EF (0.41) lies in the lower range (0.35) studied by Rajarathnam et al. (2014).

Pollutants	Emission factors	Emission factors				
	g/kg of fuel	g/kg of brick	g/MJ of fuel			
BC PM <sub>2.5</sub>	$0.01-3^{\rm c}; 0.5^{\rm d}$ $0.6-4.4^{\rm c}; 6.1^{\rm d}$	$\begin{array}{c} 0.0010.27^{\rm c}; 0.02^{\rm d} \\ 0.030.5^{\rm c}; 0.43^{\rm d} \end{array}$				
CO <sub>2</sub> CO SO <sub>2</sub>	40–56 <sup>a</sup> ; 6.6–53.5 <sup>c</sup> ; 7.4 <sup>d</sup> 9–15 <sup>a</sup> ; 24.7 <sup>d</sup>	115 <sup>a</sup> ; 79–526 <sup>b</sup> ; 15.4 <sup>d</sup> 2.07–2.80 <sup>a</sup> ; 0.35–15.01 <sup>b</sup> ;0.41 <sup>d</sup> 0.57–0.71 <sup>a</sup> ; 1.5 <sup>d</sup>	92–181 <sup>b</sup> ; 10 <sup>d</sup> 0.32–5.17 <sup>b</sup> ; 0.32 <sup>d</sup> 0.1–1.78 <sup>b</sup> ; 1.1 <sup>d</sup>			

<sup>a</sup> Suresh et al. 2016

<sup>b</sup> Rajarathnam et al. (2014)

<sup>c</sup> Weyant et al. (2014)

<sup>d</sup> Present study

**Table 5**Comparison of emissionfactors of pollutants fromdifferent parts of Asia

**Fig. 2** Averaged data of BC concentrations and emission factor in terms of g/kg of fired brick for three types of kilns



# SO<sub>2</sub> and VOC

Although  $CO_2$  and CO emission factors are observed much lower in this study  $SO_2$  concentration, and their emission factors are slightly higher than other literature values. Emission factors of VOC are highest among other pollutants. High sulfur grade of coal used reflected higher emission factors of  $SO_2$ . In terms of g/kg of fuel and fired brick, EF is almost two times higher than Suresh et al. (2016) while in terms of g/MJ of fuel, it lies in the proximity presented in Rajarathnam et al. (2014). Another study in China reported that average emission factors per 1000 bricks were 6.35–12.3 kg of CO, 0.52–5.9 kg of  $SO_2$ , and 0.64– 1.4 kg of particulate matter (PM) (Le and Oanh, 2010).

Overall, emission factors of various pollutants fluctuated due to sufficient air for combustion or velocity of stack emission, kiln types, fuel choices, and also their feeding practices.

#### Total emission of pollutants estimated

In this study, estimated total emissions for 1000 brick kilns in Greater Dhaka region are found to be  $4526 \text{ t of PM}_{2.5}$ , 340 t of BC, 209,776 t of CO<sub>2</sub>, 8700 t of CO, 19,441 t of SO<sub>2</sub>, and 835,450 t of VOC per year. Except for SO<sub>2</sub> and VOC emission, all other values are 10–20 times underestimated than earlier studies suggested.

#### Impact of kiln emission on ambient air

Significant quantities of particulates and gaseous pollutants emitted from brick production and results in environmental degradation. Gaseous pollutants and PM were measured in few residential areas along with brick kiln premises. Several rooftops of six to eight storied buildings were visited according to the wind direction and the pollutants were measured by a portable device. Average data are presented for real-time

measurements taken in few localities particularly in Dhamrai, Ashulia, and Gazipur cities. Table 6 illustrates the comparison of these pollutants and it can be clearly perceived that all of these concentrations except CO were slightly higher or similar in residential areas than brick kiln premises. PM<sub>2.5</sub> concentrations were 2-3 times higher than WHO (2016) guideline value (65  $\mu$ g/m<sup>3</sup>) for Bangladesh. Similarly, PM<sub>10</sub> concentrations were also 3 times higher than WHO (2016) guideline value (150  $\mu$ g/m<sup>3</sup>). PM<sub>10</sub> was only carried out in the kiln premises with portable sensor and not used for flue gas measurement. In earlier studies, Begum et al. (2011) suggested that brick kilns contributes 38% of total PM levels in DMA. However, ambient PM values also depend on longrange transport as well as wind direction. Lower PM levels were observed particularly in some rainy days during sampling time and were increased during dry and hazy weather condition. CO<sub>2</sub> levels were slightly higher (not shown), but CO levels were much lower found in rooftop buildings compared to brick kiln premises. However, higher values greater than 10 ppm (11,400  $\mu$ g/m<sup>3</sup>) were observed in some Zigzag kiln premises where chimney height was half than FCK and it

**Table 6**Process variable in the ambient environment of the brick kilncampus in Greater Dhaka, Bangladesh. All units are in  $\mu g/m^3$ 

Types of pollutants	Ambient environment			
	Brick kiln premises	Residential area (nearly 1 km around kiln)		
PM <sub>2.5</sub>	$136\pm22$	$171 \pm 33$		
PM <sub>10</sub>	$458\pm160$	$480\pm56$		
СО	$6413\pm4466$	$343\pm229$		
$SO_2$	$549\pm0.15$	$454\pm249$		
O <sub>3</sub>	$19.6\pm13.9$	$26\pm17.8$		
NO <sub>x</sub>	$86\pm 36$	$147\pm98$		

could be possible to spread down some CO from stack emission to kiln premises (Guttikunda et al. 2013).

Average concentrations of SO<sub>2</sub> were found in the residential area during kiln active period was  $454 \pm 249 \ \mu g/m^3$ , and this value was higher than a study done in an Indian village suggested by Skinder et al. (2014). Coal containing higher proportion of sulfur might play a significant role in elevating the SO<sub>2</sub> concentration in the ambient air (Maity, 2011; Dwivedi and Tripathi, 2007). O<sub>3</sub> values fluctuated around 20  $\mu g/m^3$  in both kiln premises and surrounding areas. NO<sub>x</sub> values observed in residential areas and kiln premises were 86  $\pm 36$  and 147  $\pm 98 \ \mu g/m^3$ , respectively, and these values are comparable with Skinder et al. (2014) found in their study during peak season of brick production (Fatima, 2011; Skinder et al. 2014). Clearly, fume emitted from brick kilns affected the ambient air quality.

## Conclusions

The current study provides an assessment of air pollutant emission from three commonly used brick-making technologies in Bangladesh. Among three technologies, i.e., FCK, Hoffmann, and Zigzag kilns, the latter two shows less amount of coal consumption, and thus better performance in terms of lower black carbon emission. Therefore, conversion of FCK to the latter two will be at least a partial solution for reducing the emission from brick kilns. Average EF for FCK, Zigzag, and Hoffmann are  $0.43 \pm 0.18$ ,  $0.31 \pm 0.09$ , and  $0.25 \pm 0.07$  g/kg of fuel used, respectively, and suggest that Hoffmann was the lowest BC emitter whereas FCK was the highest.

Among the small-scale production, FCK accounted substandard environmental performance with high CO emission levels. Whereas, Zigzag shows lower emission rate of CO with a higher emission rate of  $CO_2$  and indicates efficient combustion. Hoffmann performed better regarding lower emission of both CO and  $CO_2$  and suggested effective consumption of fuel. A higher level of sulfur content found in coal used for brick production and thus reflected their elevated emission factor compared to earlier studies. Emission of VOC and  $SO_2$  mainly depends on fuel type, and if lowgrade coal is used, their emission is higher.

Total estimated emissions for 1000 brick kilns in Greater Dhaka region are found to be 4526 t of  $PM_{2.5}$ , 340 t of BC, 209,776 t of CO<sub>2</sub>, 8700 t of CO, 19,441 t of SO<sub>2</sub>, and 835,450 t of VOC per year. Concentrations of different gaseous pollutants, as well as particulate matter in brick kiln premises and the surrounding locations, suggest that air quality is deteriorated due to emission from kilns. Emission measurement from stack monitoring was not steady and fluctuated greatly during the operational time, and there is a need to progressively reconsider the stack emission standards for the brick industry in Bangladesh. Combined account of fuel efficiency, cost, and health benefits are also necessary for this rapidly growing sector to standardize the kiln efficiency and technology. Considering lower emission and better combustion techniques, Hoffmann and Zigzag have been found performing better compared to traditional FCK. Using highgrade coal with lower sulfur content including conversion of FCK to these modern kilns could be an effective solution for reducing the emission from brick kilns.

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