Inventory of kiln stacks emissions and health risk assessment: Case of a cement industry in Southwest Nigeria

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Abstract

Cement production is a significant source of air pollution as both gaseous and particulate materials released are detrimental to the ecosystem. This work was carried out in a cement industry located in Southwest Nigeria. The emission rates of carbon monoxide (CO), nitrogen oxides (NO_x), carbon dioxide (CO_2) and sulphur oxides (SO_x) released from the cement kilns using fuel oil, natural gas (NG) and coal were garnered for a year. Thereafter, the estimated emission quantities of the pollutants were employed to obtain the emission inventory of the cement plant. Uncertainty analysis associated with the emissions was evaluated using Analytica® (4.6). Total amounts of pollutants emitted from the plant were 4.86 tonne (t)(NO_x), 18.2 t (SO_x), 2.270 Kt (CO_2) and 1.17 t (CO). Uncertainty range of –149.38% to 149.38% was connected to all the pollutants. Results showed that the quantities of pollutants discharged from the cement industry were considerably higher than recommended. The evaluated air quality indices for CO, NO_x, and SO_x implied that the health risk on exposure to these gases was hazardous. This study revealed that NG and wastes are the best fuel for kiln firing to reduce the amounts of pollutants emitted into the microenvironment of the plant.

Keywords: fuels, cement industry, pollutants, emission rates, kilns

Introduction

Fossil fuels of coal, natural gas (NG), petroleum coke, and fuel oil (FO) are the main global energy sources used in the industrial sector, especially cement manufacturing industries (Uson et al. 2013; Chatziaras, Psomopoulos, and Themelis 2016). A cement kiln consumes around 30–40% of the total energy utilized for cement production. The choice of a fuel largely depends on the country, environmental effect, storage, processing, handling, fuel type, cost, and availability. Cement plants typically are renowned for their intensive raw material, fuel (fossil), carbon, energy consumption, and emission (Pudasainee et al. 2009; Uson et al. 2013; Barcelo et al. 2014). It has been reported that the global cement sub-sector accounts for about 10–15% of the energy used in the industrial sector globally, reaching a peak of 120 kWh/t of cement (Uson et al. 2013). Subject to the fuel type, 60 kg–150 kg of the fuel is consumed to produce a tonne of cement (Cembu and theoretically requiring 1750 MJ to produce a tonne of clinker (Hendriks et al. 2002). Over 150 countries of the world manufacture cement and/or clinker, with China ranked first (661 MMt), followed by India (100 MMt) and the United States (90 MMt) in 2001 (Shen et al. 2014).

Emissions of gases (carbon dioxide (CO_2), carbon monoxide (CO), sulphur oxides (SO_x), nitrogen oxides (NO_x), methane (CH_4), hydrogen sulphide (H_2S) etc.), particulates (black carbon, organic carbon, benzene, particulate matters etc.), dusts and heavy metals (mercury, arsenic, chromium, lead etc.; when wastes are used) from cement industries are of serious global concern as these pollutants are the major cause of environmental pollution. Sources of emissions in the industry include: excavation works, dumping and tipping activities, conveyor belts, kilns, raw material and cement mills, of which kiln emissions (process-related and fuel combustion) is mainly responsible for the emission of pollutants. Globally, cement production is one of the highest emitters of CO_2 (Benhelal et al. 2013). The production of cement accounted for the discharge of 2.37 Gt of pollutants into the atmosphere in the year 2000. About 1.8 Gt of CO_2 (6%) out of around 28.3 Gt of global CO_2 estimate was emitted in 2015 (Benhelal et al. 2013; Uson et al. 2013). However, this value was reduced to 5% in the last few years due to the utilization of alternative materials and fuels, and energy efficiency improvements (Chatziaras, Psomopoulos, and Themelis 2016).

Numerous studies have been conducted on cement production relating to energy efficiency and savings, reduction of pollutants' emission, inventory and assessment of emissions, usage of alternative materials and fuels, modelling of energy recovery and emission forecasting, environmental impact assessment and life cycle of greenhouse gas (GHG) emission (Pudasainee et al. 2009; Chen et al. 2010; Atabi, Ahadi, and Bahramian 2011; Lei et al. 2011; Ilalokhoin et al. 2013; Uson et al. 2013; García-Segura, Yepes, and Alcalá 2014; Shen et al. 2014; Abdul-Wahab et al. 2016; Chatziaras, Psomopoulos, and Themelis 2016). Atabi, Ahadi, and Bahramian (2011) employed scenario analysis to examine the effect of different policies in reducing CO_2 released from the cement industry in Iran. They found that an integrated scenario resulted in the highest CO_2 reduction (13%) as against the business as usual scenario compared to those of fuel switching (4.9% reduction) and energy efficiency (9.8%) for a period of 15 years (2005–2020). A study on the emission of harmful air pollutants (volatile organic compounds and heavy metals) from three cement kilns fuelled by co-burning waste showed that the pollutants were within the recommended emission limits with efforts towards their reduction being made (Pudasainee et al. 2009).

In addition, the release of CO_2 from a cement plant was modelled and used to assess its effect on the

workplace and microenvironment (Abdul-Wahab et al. 2016). It was observed that the maximum level of CO_2 predicted by the model for a period of 1 hr was higher than the acceptable levels for the selected days (winter and summer) while the CO_2 emissions within and outside the plant were found to be higher for the line sources than the point sources during winter days. Shen et al. (2014) employed a factory-level sampling technique (Tier three) for the first time to obtain the CO_2 emission inventory of China using a bottom-up approach for various types of cement and clinker production. They reported that earlier studies overestimated carbon emissions from cement production in China because technology transition (wet process to dry process), substitution of raw materials and fuels, lime content disparity, blend additive utilization, and clinker-to-cement ratios were neglected in their works.

Studies on characterization, modelling and inventory of emissions from cement plants in Nigeria are very scarce in the public domain despite the global concern raised over the enormous and adverse effect of pollutants related to this source as mostly reported in the literature (Pudasainee et al. 2009; Chen et al. 2010; Lei et al. 2011; Chatziaras, Psomopoulos, and Themelis 2016). Amos et al. (2015) and Oyinloye (2015) both examined the effects of cement dust and emissions on the soil properties (physicochemical) and inhabitants within 10 km radius of Ashaka Cement Company Plc., located at Ashaka, Gombe State (Nigeria) and Lafarge West African Portland Cement Company (WAPCO) (Ewekoro plant) in Ogun State, Nigeria, respectively. Both studies reported the serious impact of the cement particulate on the soil, environment and health risk of inhabitants within a 5 km radius of both plants. Ideriah and Stanley (2008) experimentally evaluated the concentrations of suspended particulate matter (SPM) and NO_2 in the air around Atlas and Eagles cement industries located at Port Harcourt, Nigeria. Their results revealed that the concentrations of SPM and NO_2 at Atlas cement were considerably lower than those at Eagle cement, with SPM concentrations in both industries and NO_2 at Eagle cement higher than the recommended limits. A similar study was conducted by Bada, Olatunde, and Oluwajana (2013) to assess the air quality around Ewekoro cement plant of WAPCO based in Ogun State, Nigeria at various distances (0–1500 m) from the plant location. They observed significant levels of SPM, particulates ($PM_{2.5}$ and PM_{10}), SO_x, NO_x, CO, and H_2S, which decreased with an increase in distance from the plant.

Previous studies have shown that the release of pollutants generated due to activities of the cement industries locally and globally are sources of serious environmental pollution (Lei et al. 2011; Benhelal et al. 2013; Shen et al. 2014; Abdul-Wahab et al. 2016). Air pollution from cement industries has been a prominent source of global warming and climate change (Uson et al. 2013). Globally, efforts have been geared towards the reduction of the quantities of pollutants discharged into the atmosphere via cement industries. Notable emission mitigation strategies include improvements of energy efficiency, switching from conventional fuels to alternative fuel (waste), emission capture and storage, raw material and cement blending with other materials (Pudasainee et al. 2009; Lei et al. 2011; Chatziaras, Psomopoulos, and Themelis 2016;). These schemes are primarily accompanied with cost, energy and environmental pollution reduction (Chatziaras, Psomopoulos, and Themelis 2016). Of these mitigation strategies, the substitution of traditional fuels with alternative fuels has been noted to be a key emission reduction technique. The use of wastes as fuel in cement kilns in the USA, Australia, Canada, Korea, Japan, and European countries (Ali, Saidur R, and Hossain 2011) is an established technology which is yet to be replicated in Nigeria and other African countries. The various wastes generated in Nigeria because of the country's large human population and the lack of facilities to recycle these huge wastes call for urgency in employing this potential and untapped energy source as a fuel for cement production in the country.

However, there is a scarcity of documentation on the epidemiological study of the effect of emissions on public health of residents near cement plants in the country. The data from the national emission inventory for the year 2000 revealed that cement industries are the major contributors of CO_2 emissions in the industrial processes sub-sector (64.52%), which signalled the urgent need to curb emissions from this source (Nigeria's Second National Communication 2014). Due to the alarming growth and tonnage of cement industries in Nigeria, and the utilization of traditional fuels which further portends an increase in environmental pollution and degradation subject to the discharge of obnoxious gases and particulates into the microenvironment, there arises the necessity to undertake an emission inventory, and to assess stack emissions and health risks due to cement production activities. In this present study, emission inventory, cement stack emission characterization, and associated health risk were undertaken for a major cement plant in Southwest Nigeria using three fuels for kiln firing.

Background of cement production in Nigeria

The production of cement in Nigeria dates back to 1957 with three plants being commissioned at different times by the Mid-Western (WAPCO), Eastern (Benue Cement Company (BCC)), and Northern (Cement Company of Northern Nigeria (CCNN)) regional governments. Cement production in Nigeria commenced as a way of substituting cement importation because of the huge domestic demand for cement. Presently, there is a progressive increase in the number of cement industries in Nigeria, which are distributed along the limestone deposit belts of the country. Key players in the industry include; Lafarge WAPCO Nigeria Plc, Ashaka Cement Company Plc, Edo Cement Company Limited, BCC, CCNN, Calabar Cement Company Limited, Nigerian Cement Company Limited, and Dangote Cement Limited. Cement demand in Nigeria has been reported to be the largest in sub-Saharan Africa with nearly 95% of the raw materials utilized to produce cement available locally (Industry Report 2011).

The energy (electricity) problem bedeviling the country is taking a critical toll on the cement industries in the country. This has led these industries like others to generate their own power as this sub-sector of the economy is energy intensive. Coal, NG and FO are being utilized as alternative energy sources to the public electricity for cement production. The amounts and types of pollutants released into the environment of cement plants are strongly linked to the composition (carbon and elemental components) of the fuel and raw materials, combustion conditions and variables, environmental and meteorological factors, and other relevant parameters. The challenges experienced by the cement manufacturers are an upsurge in the prices of fuel and scarcities in supply. This has resulted in the dwindling of both profits and production outputs of most of the plants. The cement companies reacted to these challenges differently. For example, AshakaCem switched to the use of coal by engaging in a coal-mining venture due to the high cost and unstable supply of low pour fuel oil (LPFO). Doing this, the overall energy cost was reduced, and the fuel supply was guaranteed to a large extent. The refurbishment of the roller press was also carried out to enhance the cement mill efficiency and reduce power consumption during cement grinding. In a similar reaction, Lafarge WAPCO switched to the use of NG to fire their kilns and generate power in order to guarantee the supply of power and hence, cut energy costs.

Materials and methods

Area of study and plant operation

This work was conducted at a cement industry sited in Southwest Nigeria, which is one of the major players and brands in the country. The sole purpose of non-declaration of the cement industry's name is to protect it against any form of issue, especially environmental matters that may arise from the outcome of this work. This present study was carried out between January and October 2016, which translated to a year as the two remaining months were used for annual general maintenance of the cement plants. The operating hours for the year under consideration was 7320 h (305 days). The cement factory has two plants (three production lines) with a combined capacity of over 2 million tonnes of cement annually. The kiln in each production line of the cement industry is fired using coal, FO, and NG as fuels.

Data source and collection

Data on cement kiln stack emissions spanning a decade were sought from many of the cement industries in the country. All the contacted industries turned down our requests citing company's policies and implications of such data as reasons. However, our untiring efforts yielded the data utilized in this study with the condition of anonymity regarding the specific source of data. Daily records (emission rates) of the point source emissions of a GHG (CO_2) and precursor gases (CO, SO_x, and NO_x) emanating from the kiln end of each production line of the cement industry through the stack were collected. The obtained emission rates were for a period of 7320 h of running of the plants. The kiln stack emissions were measured at a time-weighted average concentration using the various gas analyzer sensors embedded into the cement kiln system. The stack emission rates were computed automatically with the use of other parameters such as fuel consumption, emission factor, clinker tonnage, emission concentrations, etc. as described in the literature (Abdul-Wahab et al. 2016). In addition, the data on total cement produced per annum spanning 26 years (1986 to 2011) and CO_2 emitted (1958 to 2014) from cement production for the country were sourced from the literature (Mojekwu, Idowu, and Sode 2013; US NOAA). The quantity and concentration (Srujan 2014) of each pollutant released into the atmosphere due to the fuels used in the kilns for clinker production were calculated as expressed in Eqs. (1) and (2), respectively, from the obtained emission rates. The total amount of all the pollutants released from the kiln stacks was also estimated using Eq. (3).

$$E_{Q_{i,j}} = E_{R_{i,j}} \times t \tag{1}$$

$$P_{C_{i,j}} = 1.81 E_{R_{i,j}} \tag{2}$$

$$E_T = \sum_{i=1,j=1}^{3} E_{Q_{i,j}} \tag{3}$$

where:

i, j = fuel (NG, FO, and coal) and pollutant (NO_x, CO, SO_x, and CO_2), respectively;
$E_{R_{i,j}}$ Emission rate for individual fuel and pollutant (kg/day or kg/month;
$E_{Q_{i,j}}$ Emission quantity for individual fuel and pollutant (kg);
$P_{C_{i,j}}$ Concentration of individual pollutant and fuel (mg/m^3);
E_T Total emission quantity from the cement plant (kg or tonne);
t = time (h).

Data analysis

The daily and monthly averages and the total of the emission rates, quantities and concentrations for the pollutants (CO_2, CO, SO_x, and NO_x) associated with the three fuels used in kiln firing were analyzed. These were carried out for both the fuels and pollutants. Statistical analysis involving analysis of variance (ANOVA) and correlations were performed on the emission rates, quantities and concentration data obtained for all the pollutants from the fuels utilized in the cement kilns and other data (CO_2 emissions and cement production) used in this study.

Procedure for emission uncertainty evaluation

The uncertainty associated with the release of the kiln stacks emissions (CO_2, CO, SO_x, and NO_x) in the studied cement industry was estimated using the emission (quantity) data as the input variables. The emission data for each individual pollutant is the sum of the emissions from all the cement kilns (fuelled using NG, FO, and coal) for the period of operation of the kilns (7320 h).

The fitting of the emission data (input variables) into the suitable probability distribution function and the modelling of the emission uncertainty estimates were carried out using EasyFit® 5.6 (evaluation version) and Analytica® (4.6) software, respectively. Emission data of each pollutant were fed into EasyFit® 5.6 to perform the probability distribution fitting. The best fitted distribution as ranked by the software and the corresponding values were thereafter input into the Analytica® for uncertainty estimation modelling. The use of Analytica® consisted of the input, uncertainty propagation (input variables) and output stages. The input model was developed using the fitted probability distribution of the input variables. Tier 2 method was employed as it has been endorsed for national GHG inventories (IPCC 2006) reporting and computation. Hence, the choice of Analytica® software and the use of Latin hypercube sampling (LHS) for simulation of the input model. In this study, LHS was selected over Monte Carlo Simulation (MCS) as a numerical simulation technique because it offers a better approximation with large number of samples. The propagation of the input model consisting of the probability distributions of the input variables was performed using LHS. Median Latin hypercube and Minimal Standard were set as the default for sampling and random number generation, respectively. Iterations of the simulation was carried out until there was no further change in the value of the standard deviation. Thereafter, the mean value of each output variable was obtained from the simulation. Further information for estimating emission uncertainty can be read in previous studies published by Giwa, Nwaokocha, and Layeni (2016; Forthcoming). The uncertainty related to the release of the pollutants was estimated using Eq. (4). A flow chart of this study including the modelling procedure is provided in Figure 1.

$$R\ (\%) = \left(\frac{X - S}{S}\right) \times 100 \ \textit{and} \ \left(\frac{Y - S}{S}\right) \times 100 \tag{4}$$

where:

X = 2.5th percentile of mean (simulated)
Y = 97.5th percentile of mean (simulated)
S = Mean (simulated)
R = Relative uncertainty

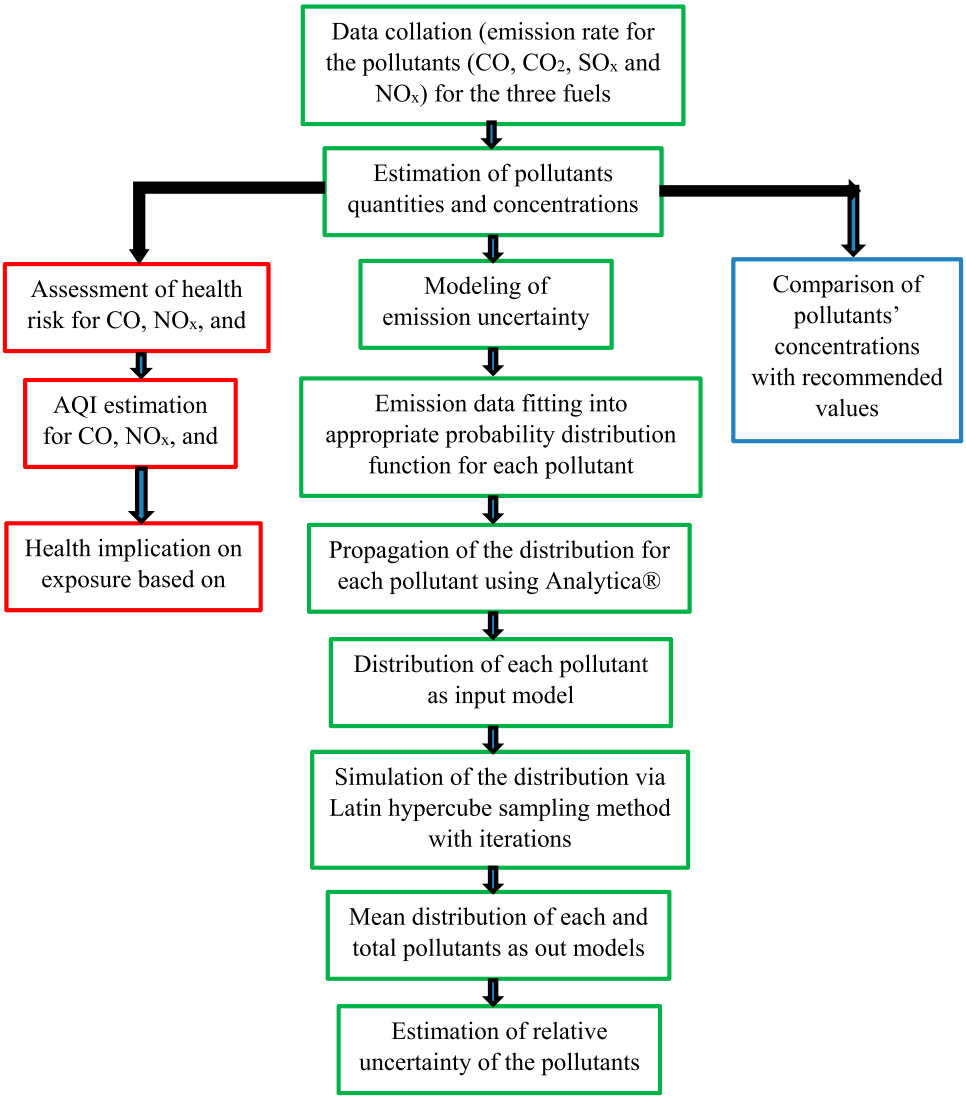

Figure. 1: Study flow chart.

Table 1: Air quality index descriptor.

Breakpoints					
$PM_{2.5}$ (μg/m^3)	CO (ppm)	SO_2 (ppm)	NO_2 (ppm)	AQI	Category
0.0–15.4	0.0–4.4	0.000–0.034	–	0–50	Good
15.5–40.4	4.5–9.4	0.035–0.144	–	51–100	Moderate
40.5–65.4	9.5–12.4	0.145–0.224	–	101–150	Unhealthy
65.5–150.4	12.5–15.4	0.225–0.304	–	151–200	Unhealthy
150.5–250.4	15.5–30.4	0.305–0.604	0.65–1.24	201–300	Very unhealthy
250.5–350.4	30.5–40.4	0.605–0.804	1.25–1.64	301–400	Hazardous
350.5–500.4	40.5–50.4	0.805–1.004	1.65–2.01	401–500	Hazardous

Source: Rim-Rukeh, 2015

Air quality index estimation

A comparison of the obtained results for this study with outdoor air quality standards (American Society of Heating, Refrigerating, and Air-Conditioning Engineers (ASHRAE) and World Health Organization (WHO)) was carried out. This was to examine their compliance with the stipulated values prescribed by these standards. In addition, an assessment of the health risk connected to NO_x, SO_x, and CO was carried out with the use of Eq. (5) as given in the literature for estimating air quality index (AQI) (Rim-Rukeh, 2015). Table 1 presents the AQI signifier.

$$I_p = (C_p - BP_L)\left(\frac{I_H - I_L}{BP_H - BP_L}\right) + I_L \quad (5)$$

where:

I_P = pollutant index;
C_P = rounded pollutant concentration;
BP_H = breakpoint equal or greater than C_P;
BP_L = breakpoint equal or less than C_P;
I_H = AQI value matching BP_H;
I_L = AQI value matching BP_L;

Results and discussion

Kiln stack emissions from clinker production

The environmental issue is one of the major problems facing cement production globally. Several pollutants are released through cement kiln stacks into the atmosphere which impacts the environment adversely and causes global warming, climate change, acid rain, and respiratory and heart-related health diseases, deterioration of air quality, etc. Data on emission rates of CO_2, CO, SO_x, and NO_x released from the cement plants under consideration were obtained via clinker formation in the kilns operated using three fuels (coal, FO (low pour) and NG). It is worth noting that CO and NO_x as reported in this work, were direct products of fuel combustion while the CO_2 and SO_x were sourced from both combustion of the fuels, and calcination of CO_2 and SO_x reaction with limestone during clinker formation in the cement kilns.

NO_x emission rates

The sum of the daily emission quantity of NO_x released via kiln stack due to clinker production for each month of the 7320 h operation of the cement kilns is presented in Figure 2. The daily mean emission of NO_x released from the kilns was estimated to be 1.62 kg (2.94 mg/m^3) (NG), 7.08 kg (12.82 mg/m^3) (FO) and 7.23 kg (13.08 mg/m^3) (coal) for the period of operation of the plants. In this study, 4.86×10^3 kg of NO_x (consisting of 10.18% NG, 44.45% FO, and 45.36% coal) was released from the plants for 7320 h of operation. The highest amounts of NO_x for coal, FO and NG were recorded in the months of October (232.7 kg), August (227.8 kg), and January (57.3 kg), respectively. Similarly, the lowest quantities of NO_x emitted from the plants were in the months of February, January, and April for coal, FO, and NG, respectively (see Figure 2). The total NO_x released in relation to the fuels was 495.3 kg, 2160.77 kg, and 2205.2 kg for NG, FO, and coal, respectively. It can be seen from Figure 2 that coal and FO emitted significantly higher amounts of NO_x compared to NG as fuels used for firing the cement kilns. This can be connected to the gaseous nature and carbon content of NG. Due to stable ventilation and the high operating temperature of rotary kilns, a high quantity of NO_x has been reported regarding this kiln type (Lei et al. 2011). In addition, the obtained values of NO_x in this work are moderately lower than those of cement plants with similar output capacities reported in the literature, though the fuel type used, and other features of these plants were not given (Shen et al., 2005).

The concentrations of NO_x for all the fuels reported in this work are considerably higher than the daily maximum limit (50 μg/m^3; daily) recommended by WHO (WHO 2000). Also, the IAQ index of the emission of NO_x from the cement industry to the atmosphere gave a value higher than 500, implying that the health risk linked to this pollutant on exposure is hazardous. NO_x is mainly formed by the oxidation of atmospheric nitrogen during fuel combustion and the chemical reaction of raw materials, mostly at a very high temperature in the presence of oxygen. Consequently, acid rain and smog, and the formation of secondary pollutants such as black carbon and ozone in the atmosphere around the cement industry, due to the excessive and long-time discharge of NO_x, are highly anticipated. This will negatively impact the residents, microenvironment and ecosystem causing chronic respiratory infection and pollution of air, soil, water, and vegetation in the area where the cement industry is located.

SO_x emission rates

The monthly emission amounts of SO_x from the fuels used in the production of clinker in the studied plants is

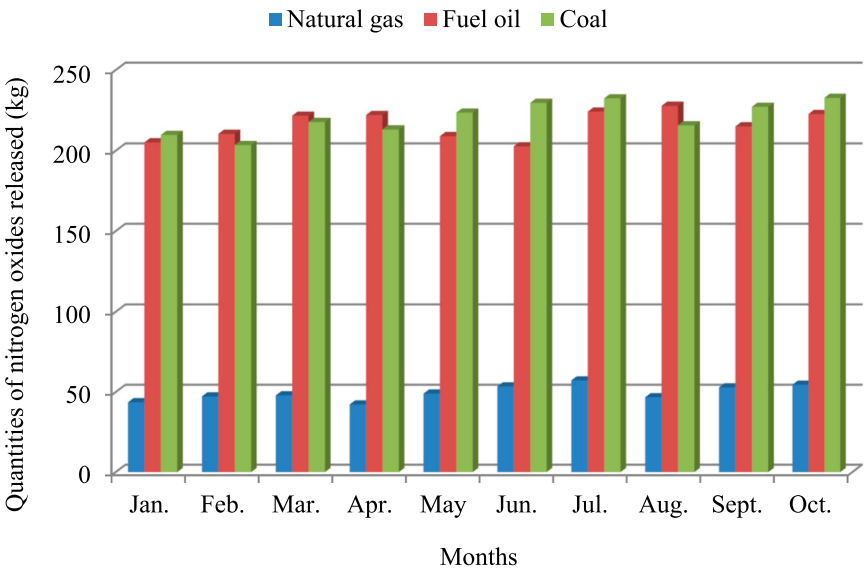

Figure 2: NO_X released into the atmosphere (kg/month).

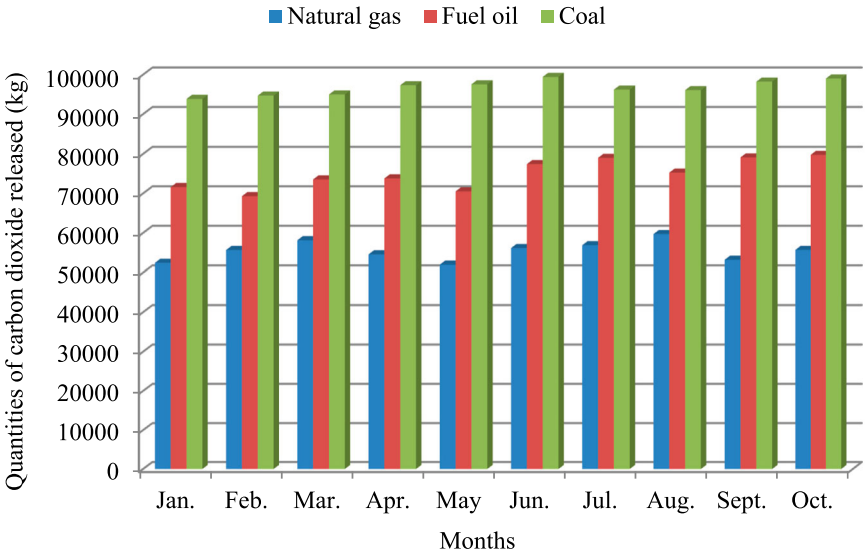

Figure 3: CO_2 released into the atmosphere (kg/month).

illustrated in Figure 3. The total and daily averages of SO_x released for the operation of the kilns was 5.82 kg and 0.019 kg (0.035 mg/m^3) for NG, 5685.27 kg and 18.64 kg (33.73 mg/m^3) for FO, and 12549.33 kg and 41.15 (74.45 mg/m^3) for coal, respectively. Estimated total emission of SO_x from the plants was 1.82×10^4 kg, consisting of 0.03% from NG, 31.17% from FO, and 68.80% from coal. For NG, FO and coal, maximum and minimum amounts of SO_x released monthly were 1.02 kg (September) and 0.07 kg (July), 643.6 kg (September) and 469.3 kg, and 1381.4 kg (April) and 1161.9 kg (June), respectively. SO_x emitted from the use of NG as fuel is very low and insignificant compared to those of coal and FO, with the quantities of SO_x released from coal more than twice that of FO for most of the months (Figure 3). This can be connected to the very low sulphur content of NG in comparison to moderate and high sulphur content of FO and coal, respectively. The obtained emission quantities for the period considered in this study are moderately less than those reported in previous works with similar plants' tonnage (Shen et al., 2005). It is pertinent to know that the characteristics of these plants were not given to enhance better comparison.

With the estimated daily concentrations of SO_x for the fuels, it is observed that only the kiln fired using NG has SO_x values moderately less than the maximum (50 µg/m^3; daily) prescribed by WHO (WHO 2000). This shows that the quantities of SO_x discharged via the kiln stack into the atmosphere using FO and coal as fuels are detrimental to the environment. The sources of SO_x are from the burning of fuels and chemical reactions of raw materials in the kiln system. The emission of SO_x causes acid rain and secondary pollutants' formation, which adversely affects physical structures (building walls and roofs), ecosystems, the environment and human health (acute respiratory infection), is probably highly pronounced in the microenvironment where the industry is located. The estimation of the AQI, which

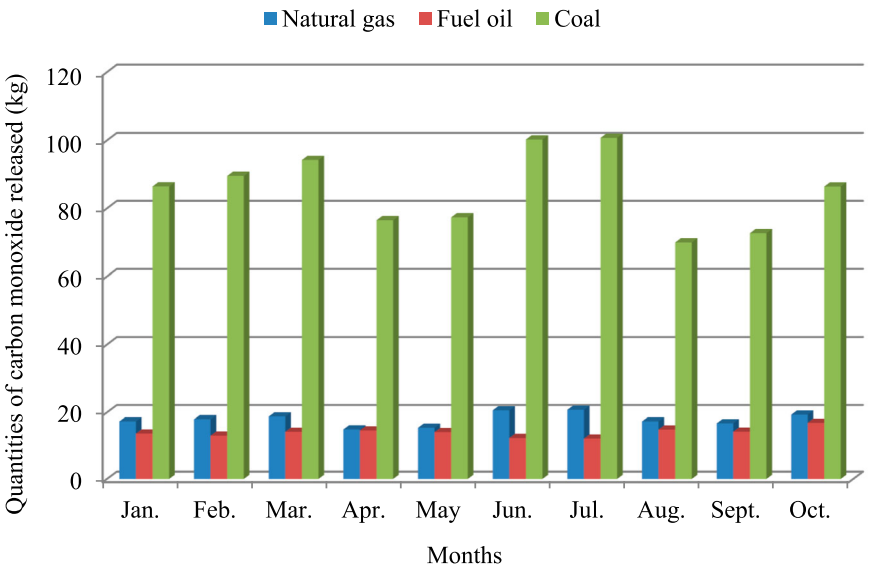

Figure 4: CO released into the atmosphere (kg/month).

indicates the health risk connected to this pollutant, was higher than 500, meaning, exposure to SO_x released from the cement industry is hazardous.

CO_2 emission rates

The calcination process and fossil fuel combustion are responsible for close to 50% and 40%, respectively, of the total CO_2 emissions during the process of cement manufacturing (Benhelal et al. 2013; Barcelo et al. 2014; Abdul-Wahab et al. 2016). Figure 4 presents the emission quantities of CO_2 released monthly by producing cement clinkers using coal, NG, and FO as fuels. When coal was used as fuel, an estimated total amount of CO_2 discharged from the kiln stack was 5.55×10^5 kg, with daily and monthly averages of 1.82×10^3 kg (3.30×10^3 mg/m^3) and 5.55×10^4 kg, respectively. For FO and NG, however, the total, daily and monthly averages of CO_2 emissions were 7.50×10^5 kg, 2.46×10^3 kg (4.45×10^3 mg/m^3) and 7.50×10^4 kg, and 9.67×10^5, 3.17×10^3 kg (5.74×10^3 mg/m^3) and 9.67×10^4 kg, respectively. The estimated total CO_2 discharged from the plants was 2.27×10^6 kg (24.4%; NG, 33.0%; FO, 42.9%; coal). As can be noticed in Figure 4, CO_2 emitted from all the fuels are more (in magnitude) compared to those of SO_x and NO_x as presented in Figures. 2 and 3. The amounts of CO_2 discharged from the production of tonnes of clinker were highest when coal was used as fuel, followed by FO and then NG. This is directly linked to the carbon contents of the fuels (coal > FO > NG) as the same quality of limestone and other raw materials were used in clinker production.

With a daily outdoor CO_2 concentration of 1.35×10^4 mg/m^3, which is more than the 100 mg/m^3 (100,000 ppm) considered to be very poor air quality, it is therefore apparent that the quantities of CO_2 released via cement kiln production from the plants are enormous. This contributes considerably to the local, national and regional source of CO_2, which causes global warming leading to climate change. Furthermore, cement production has been recognized as one of the foremost sources of CO_2 emission which leads to both global warming and climate change (Uson et al. 2013).

CO emission rates

All forms of fuel combustion have to do with the emission of CO, whether complete or incomplete. The burning of NG, FO, and coal as fuels in the cement kilns is always associated with CO emissions. The emissions of CO on monthly basis from these fuels are illustrated in Figure 5. The estimated daily means of CO released from the plants were 0.588 kg (1.06 mg/m^3), 0.460 kg (0.83 mg/m^3), and 2.80 kg (5.07 mg/m^3) for the use of NG, FO, and coal, respectively. The highest amounts of CO emitted were recorded in the months of July (20.79 kg) for NG, October (16.92 kg) for FO and July (100.76 kg) for coal whereas the lowest quantities were observed in the months of May (14.12 kg) for NG, July (12.16 kg) for FO and August (69.98 kg) for coal. In the context of this work, the amounts of CO released from the kilns were 179.22 kg (NG), 140.4 kg (FO) and 854.26 kg (coal) for the duration considered in operating the plants. Also, the total CO released from the plants was 1.17×10^3 kg (15.27%; NG, 11.95%; FO, and 72.78%; coal). Coal is observed to emit the highest amounts of CO in the order of four to five and four to eight compared with NG and FO, respectively (Figure 5). This is largely due to the high carbon content and solid nature of coal in comparison to those of NG and FO. The slight increase in the amount of CO released from the use of NG as fuel in clinker production relative to that of FO (see Figure 5) can be linked to the pre-treatment given to FO prior to its use for firing cement kiln. Compared to the other pollutants investigated in the work, CO is noticed to have the lowest magnitude of emission. This agrees with the literature stating that rotary kilns release lower quantities of CO compared to shaft kilns due to steady air inflow and increased operating temperatures (Lei et al. 2011).

The obtained daily concentration values of CO released from the plants are far higher than the

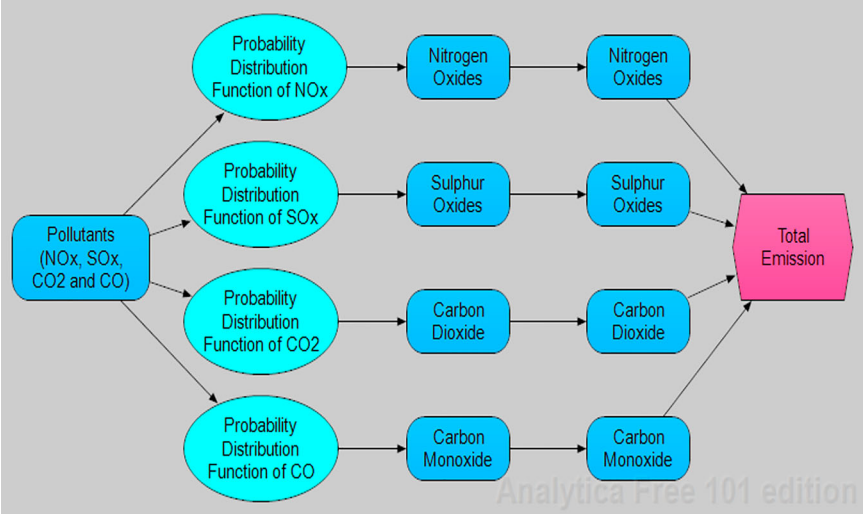

Figure 5: Influence diagram of developed model for uncertainty estimation.

Table 2: Correlation of pollutants from NG.

Pollutants	NO_x	SO_x	CO_2	CO
NO_x	1			
SO_x	0.0885	1		
CO_2	0.1539	−0.0500	1	
CO	0.5186	−0.4466	0.5804	1

recommendation (60 μg/m^3; daily maximum) of WHO (WHO, 2010). Therefore, the CO released from the plants into the environment portends danger to the micro-environment and public health (causing congestion of brain and lungs, tuberculosis, pneumonia and heart disease) on exposure. In addition, the health risk assessment of this pollutant as implied by the IAQ index (> 500) shows that exposure to this emission is hazardous.

Statistical analysis of emission rates data

NG emission rates data

The correlation coefficients between the pollutants released via the NG-fired kiln stack are presented in Table 2. Positive and moderate relationships are noticed to exist between CO and NO_x (0.5186), and CO and CO_2 (0.5804). Other pollutants have correlation coefficients indicating positive and weak relationships except for CO_2 and SO_x, and CO and SO_x, which have negative and weak correlations. The ANOVA test performed on the emission rates data of the pollutants from NG indicates that the data were statistically independent because $F_{observed}$ (5016.80) > $F_{critical}$ (2.8663). Also, at the 95% confidence interval, these data were found to be significant with a *p*-value of < 0.00001 for the ANOVA.

FO emission rates data

Correlation coefficients between the pollutants discharged from the use of FO as fuel for clinker production are provided in Table 3. It is observed that the correlation coefficients between all the pollutants showed positive-weak relationships between them. The ANOVA test conducted on the FO emission rates data indicates that the data were statistically not the same as $F_{critical}$ (2.8663) < $F_{observed}$ (3893.22). At the 95% confidence interval, the emission rates data were noticed to be significant as the *p*-value is < 0.00001.

3.2.3. Coal emission rates data

Table 4 gives the correlation coefficients between the pollutants emitted from using coal as fuel in the cement kiln. It is observed that only the relationship between CO_2 and NO_x is positive and relatively strong with a coefficient of 0.7476. Other correlations between the pollutants are noticed to be weak (positive and negative). ANOVA test conducted on the coal emission rates data revealed that the data were statistically not similar with $F_{critical}$ (2.8663) < $F_{observed}$ (26123.8). With a *p*-value < 0.00001 (at 95% confidence interval), it can be deduced that these data were significant to this study.

Table 3: Correlation of pollutants from FO.

Pollutants	NO_x	SO_x	CO_2	CO
NO_x	1			
SO_x	0.3420	1		
CO_2	0.3841	0.4809	1	
CO	0.2969	0.2030	0.2267	1

Table 4: Correlation of pollutants from coal.

Pollutants	NO_x	SO_x	CO_2	CO
NO_x	1			
SO_x	0.0098	1		
CO_2	0.7476	0.0303	1	
CO	0.2557	−0.1717	−0.0707	1

Uncertainty estimation

To estimate the uncertainties related to the emissions of CO_2, CO, SO_x, and NO_x, it was necessary to obtain suitable statistical (probability) distributions for the emission data, which were thereafter utilized to model the emissions of CO_2, CO, SO_x, and NO_x using Analytica®. EasyFit® was employed to fit the distribution for each individual pollutant using the emission data of that pollutant. The SO_x and NO_x emission data demonstrated uniform distribution while beta distribution was displayed by CO_2 and CO emission data. The values aligned to these distributions for all the pollutants were used as input variables in the input model to run the simulation by employing Analytica®. Figure 6 shows the influence diagram used in modelling the uncertainty linked to the emissions released from the cement plants. It is pertinent to know that the minimum, maximum and mean values (simulated) of the emissions obtained after running the simulation were at the 95% confidence interval. Also, the simulations were run using 500, 1000, 2000, 3000 and 4000 iterations, with the 3000th iteration giving the best result as there was no obvious change in standard deviation when compared to the 4000th iteration.

The upper and lower confidence levels of the simulated means, relative uncertainties, and means (simulated and estimated) for the release of CO_2, CO, SO_x, NO_x and total emission from the studied cement industry are provided in Table 5. In this study, the estimated means were obtained using the probability distributions subject to the probability range. In Table 5, the estimated means for all the pollutants are very close in numerical values to the simulated means. The data collection points (three kiln stacks), nature of data and their statistical distributions may be responsible for the slight differences in the mean

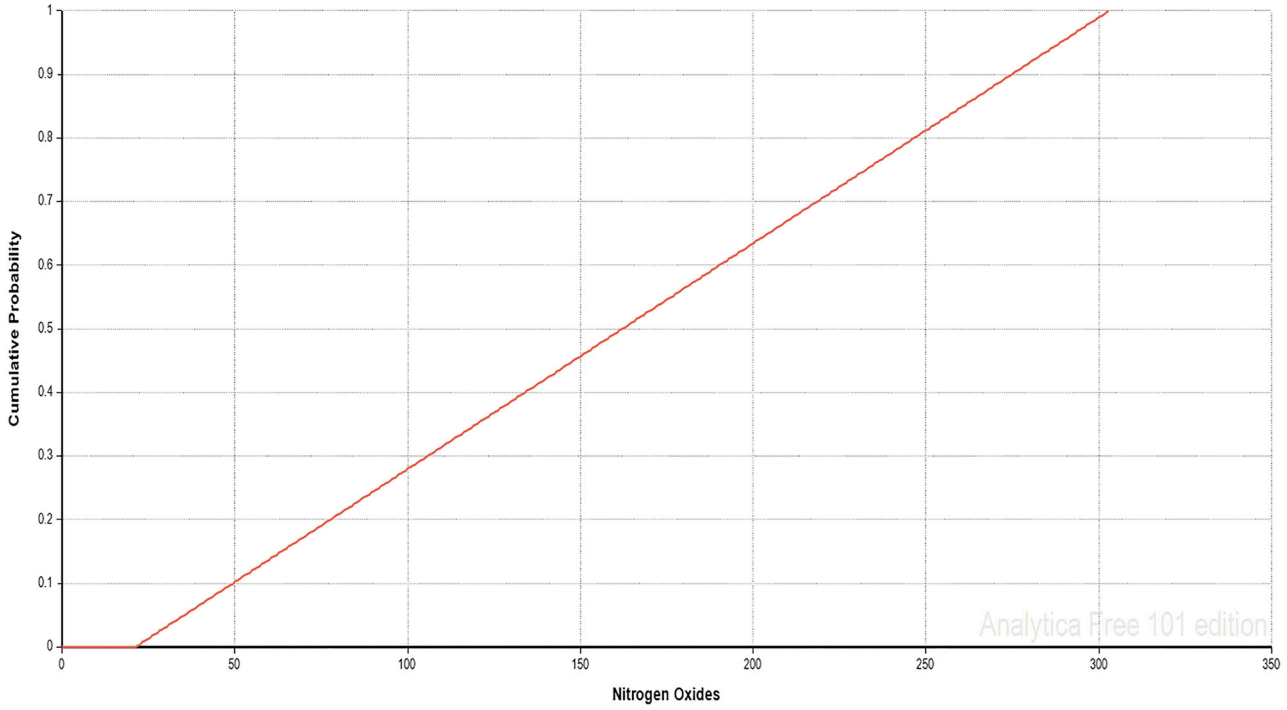

Figure 6: Cumulative distribution of nitrogen oxides emitted.

Table 5: Uncertainties associated with emissions (number of trails = 3000).

Pollutants	Min. (2.5th CL)	Simulated mean	Max. (97.5th CL)	Relative uncertainty	Estimated mean
NO_x	21.21	162	302.9	−86.91%–86.98%	162.04
SO_x	−300.2	608	1516	−149.38%–149.34%	608.01
CO_2	50.99 K	74.94 K	99.43 K	−31.96%–32.56%	75.72 K
CO	12.16	42.66	100.80	−71.50%–136.29%	39.13
Total emission	5.86 K	75.75 K	101.30 K	−32.86–33.73%	77.39 K

Note: CL = Confidence level: Negative random error = (2.5th percentile-mean)/mean; positive random error = (97.5th percentile-mean)/mean.

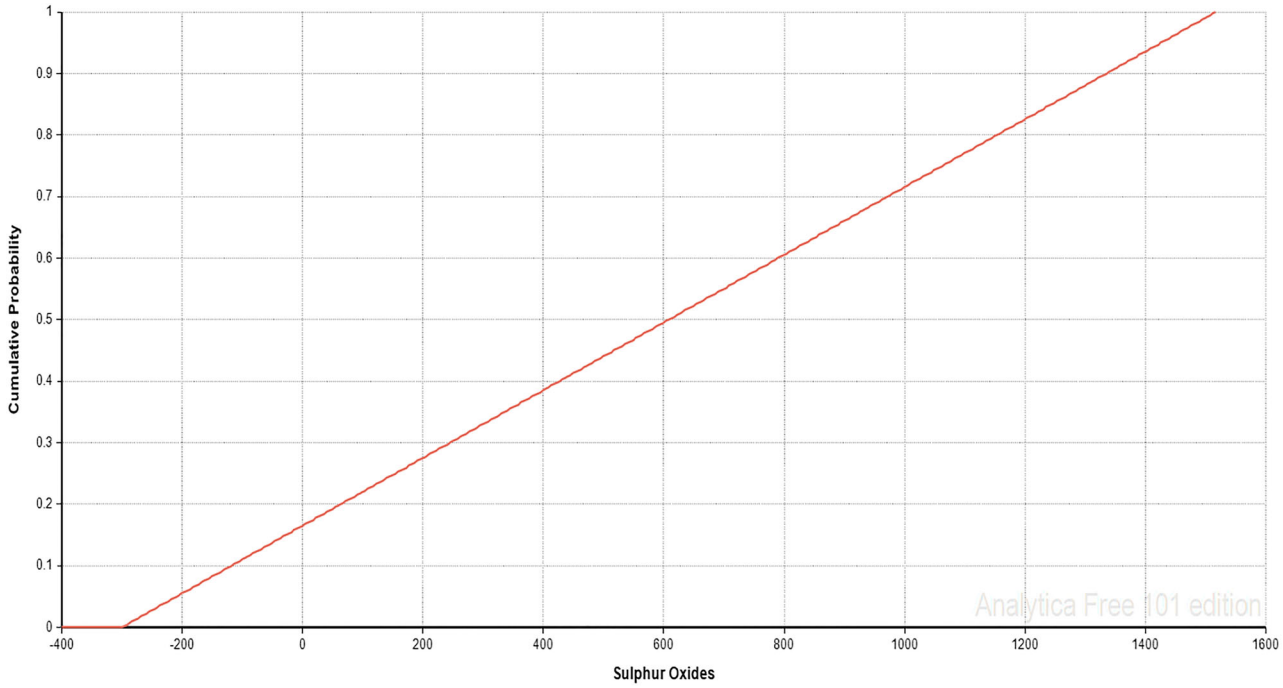

Figure 7: Cumulative distribution of sulphur oxides emitted.

values. The magnitudes of the values of the pollutants as given in Table 5 are reflections of the quantities of the pollutants discharged from the cement plants as earlier mentioned. The propagation of the uncertainties inherent in the emission data (inputs) collected for this study was used in estimating the uncertainties associated with the pollutants. As can be observed both in Table 5 and Figures 7–10, the ranges of uncertainties related to CO_2, CO, SO_x, NO_x and total emissions from the investigated cement industry are provided. For the total emission, the estimated range of relative uncertainty was –32.86% (lower limit) to 33.73% (upper limit), corresponding to monthly simulated and estimated means of 7.58×10^4 kg and 7.74×10^4 kg, and the lower and upper limits of 5.60×10^3 kg and 1.01×10^5 kg, respectively. In addition, the relative uncertainties associated with all the pollutants (CO_2: –31.96% to 32.56%; CO: –71.50% to 136.29%; SO_x: –149.38% to 149.34%; NO_x: –86.91% to 86.98%)

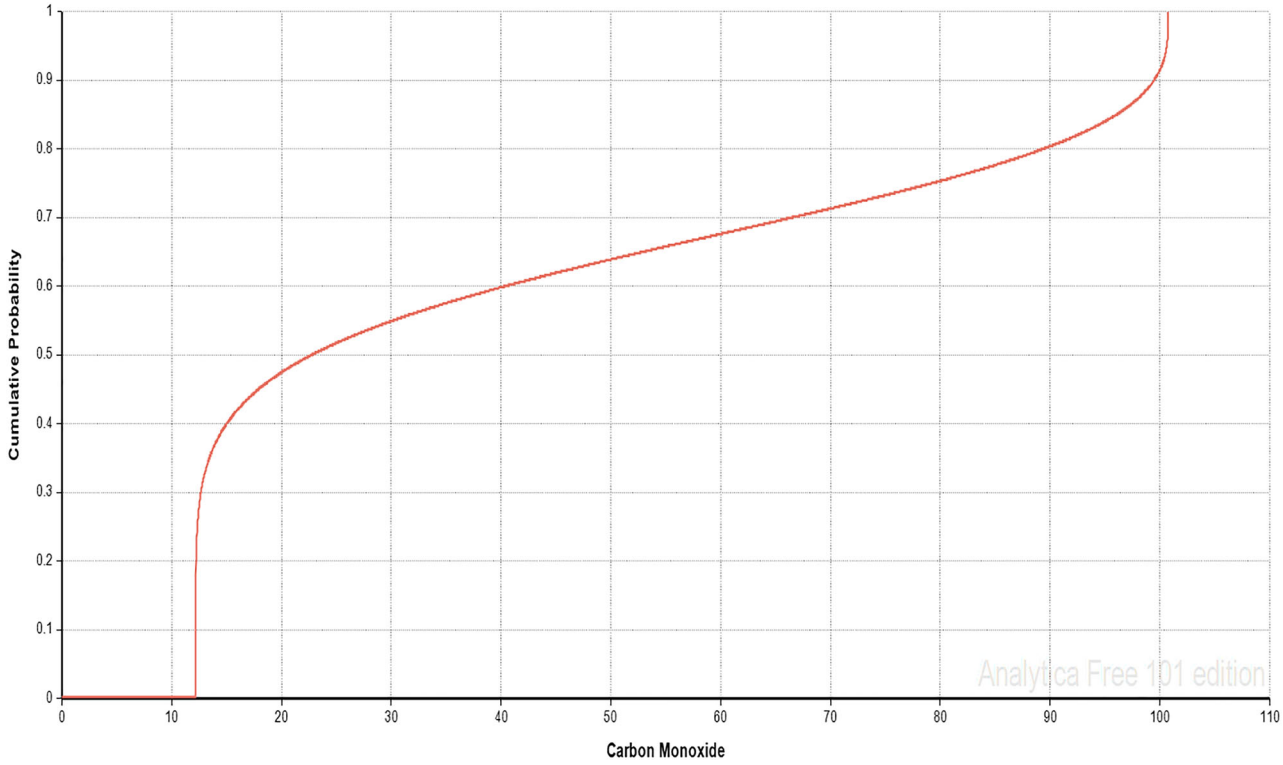

Figure 8: Cumulative distribution of carbon monoxide emitted.

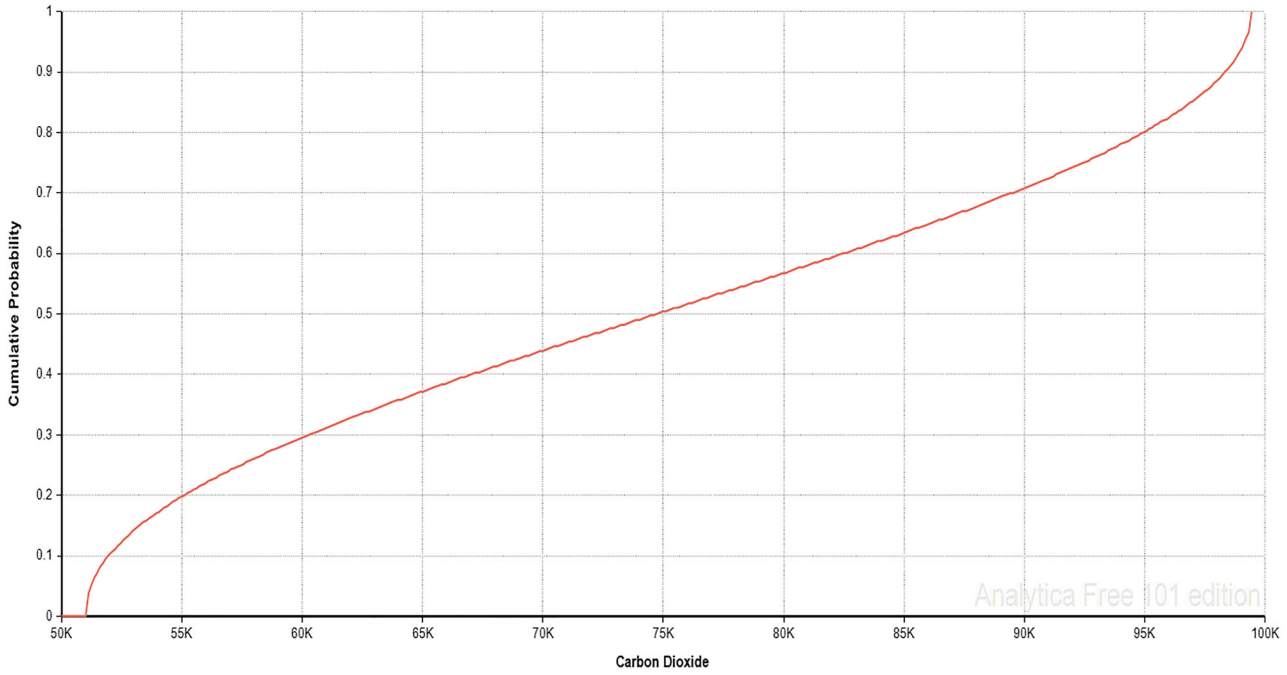

Figure 9: Cumulative distribution of carbon dioxide emitted.

were evaluated. The relative uncertainty range of 66.59% obtained for the emission of CO_2 from the cement industry is moderately higher that of 15–20% (China), 20.13–20.85% (previous study for China) and 3–5% (United States of America) reported in the literature for CO_2 uncertainty estimates of cement industries (Shen et al. 2014). The discrepancies in the uncertainty values have been primarily linked to the variations in the emission factors for the cement kilns (process-related), materials' compositions, fuels and modes of emission data collection. It is noticed that the ranges of the relative uncertainties were in the increasing order of CO_2 < NO_x < CO < SO_x. Uncertainty ranges of −10% to 1000% and ± 75% have been specified for N_2O and CO_2 emissions from flared gas in developing countries, but such recommendations are not available for cement industries (Giwa et al. 2017b). The model outputs showing the cumulative distributions of CO_2, CO, SO_x, NO_x and total emissions, and indicating the minimum and maximum confidence limits are presented in Figures. 7–10.

CO_2 emissions and cement production in Nigeria

Figure 11 illustrates the quantities of cement produced and the amounts of CO_2 emission associated with them. Data

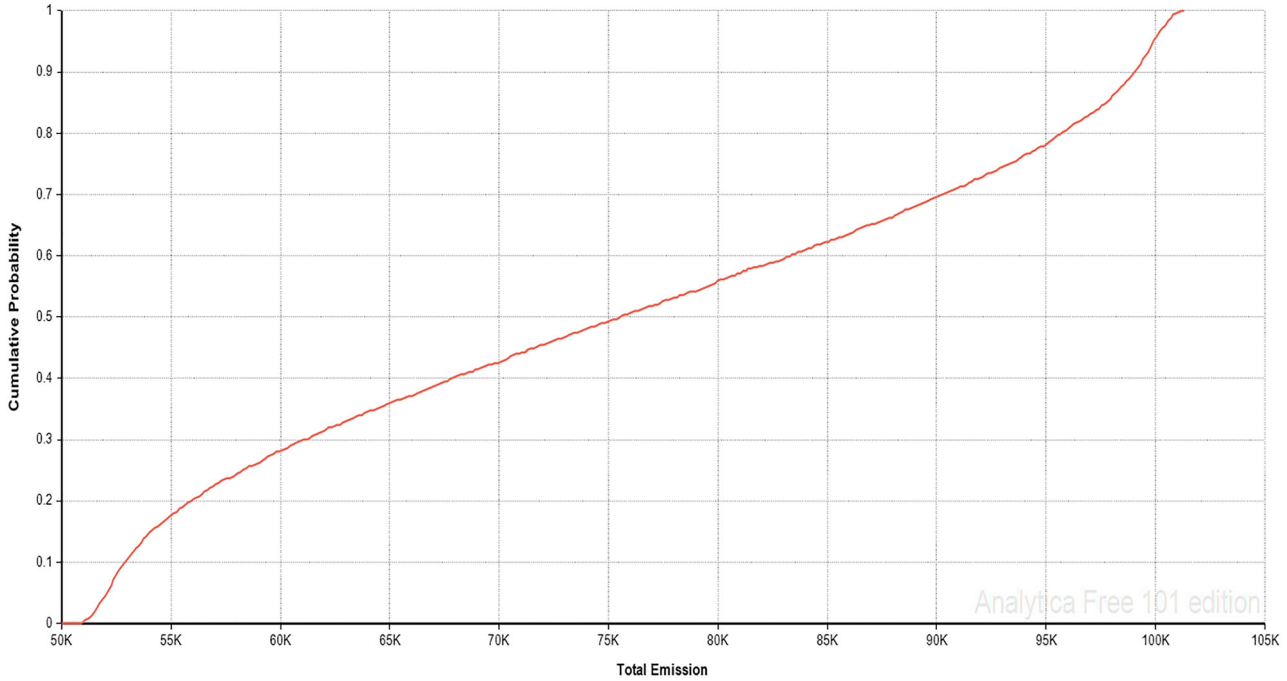

Figure 10: Cumulative distribution of total emissions released.

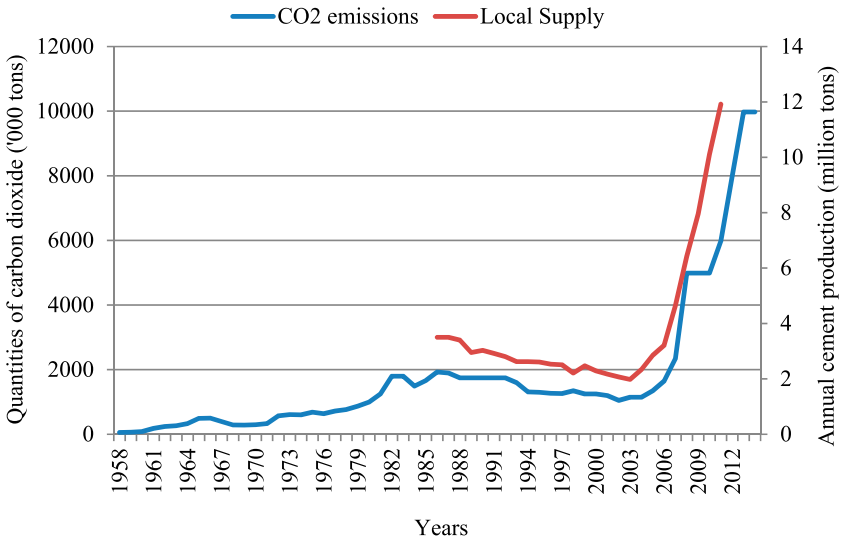

Figure 11: Carbon dioxide emission against cement production in Nigeria (Mojekwu et al., 2013; US NOAA).

on cement production and CO_2 released from cement industries in Nigeria are very scarce in the public domain, especially on the websites of national agencies saddled with this responsibility as is often the case for third world countries. As seen in Figure 11, the data on local supply (local production) of cement are only available from 1986 to 2011 (sourced from local literature) while the quantities of CO_2 released due to cement production activities in the country exist from 1959 to 2014 (United States National Oceanic Atmospheric Administration (US NOAA)). It is worth stating that the CO_2 emission data reported by NOCC for cement industries in Nigeria were collected using satellite, which is obviously a different technique compared with the method employed in this study to gather the emission rate data for the studied cement industry. There seems to be a nearly equal gap between the two lines representing the amounts of cement produced locally and quantities of CO_2 released from 1986 to 2011, except for the year 2008 (see Figure 11). This demonstrates a good relationship between the two parameters (though from different sources) as evidenced by a high coefficient of correlation (0.965). Furthermore, the data from the two sources were insignificantly the same ($F_{critical}$ (1.9375) > $F_{obsrved}$ (0.3248) and p-value = 0.9968).

In addition, CO_2 emissions from cement production in Nigeria were compared to those of South Africa, North Africa, sub-Sahara Africa and Africa as presented in Figure 12. The CO_2 datasets used in this study were available from 1960 to 2011 and the same values were observed with those of US NOAA for Nigeria. We chose the US NOAA CO_2 data for Nigeria because it has more data (1959 to 2014) which shows the trend of CO_2 emissions for cement production in Nigeria (US NOAA 2011; DataMarket 2018). Generally, the emissions of CO_2 from cement production appear to increase considerably over 50 years for Africa and North Africa, moderately for sub-Sahara Africa, fairly for South Africa and slightly for Nigeria. A surge in CO_2 emissions for North Africa was observed to have taken place in 1976, having been

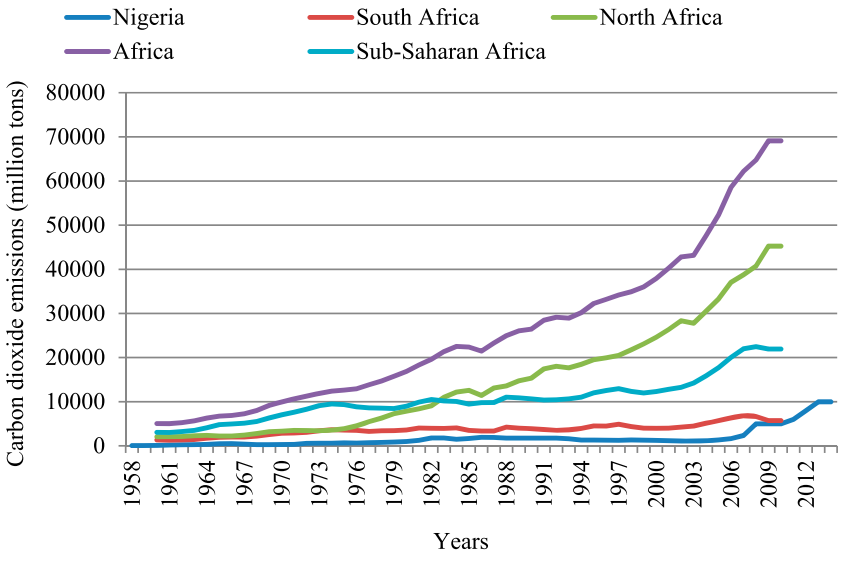

Figure 12: Trends of carbon dioxide emission from cement production (Source: US NOAA 2011; DataMarket 2018).

slightly above that of South Africa from 1960 to 1975. The amounts of CO_2 released from the cement industries in North Africa overtook that of sub-Sahara Africa from 1982 and thereafter increased progressively (see Figure 12). Comparison between Nigeria and South Africa in terms of cement production-based CO_2 emissions shows that Nigeria is fast overtaking South Africa. The trend as depicted in Figure 12 shows that the quantities of CO_2 released from Nigerian cement industries were more than those from South Africa, gradually increasing since 2007 with a possible turning point after 2011. This could be due to the establishment of additional cement industries in Nigeria and the modification of existing plants, which has substantially increased domestic cement production.

Much attention needs to be paid to the issue of emissions discharged into the environment by cement industries in Nigeria, and the attendant effects. Conducting an extensive emission inventory is paramount, as this would assist players, stakeholders and policymakers to thoroughly understand the trend and extent of emissions from this source, and the possible threat to both humans and environment, to facilitate decision making. To curb emissions from the cement industries, emissions must be monitored and reported, and mitigation strategies deployed declared, all in compliance with national emission guidelines.

Conclusion

The production of cement is mainly accompanied by the release of pollutants into the environment which is detrimental to both the microenvironment and the local environment. The emission rates of four pollutants were obtained from the utilization of three fuels in cement kilns for 7320 h of operation. This study revealed that the concentrations of the pollutants emitted into the environment were considerably higher than the recommended values. This showed that the air around the cement industry and its environment was highly polluted and could be a threat to both ecosystems and human beings. This is corroborated by the result of the health risk assessment of CO, NO_x, and SO_x, which revealed a hazardous status on exposure to the degraded air. Coal was found to emit the highest quantities of pollutants with CO_2 being the most discharged pollutants. The emission rates data for all the pollutants were found to be significant at 95% confidence interval with $F_{critical} < F_{observed}$ and p-values <0.0000001. Correlation coefficients between the pollutants for the fuels showed that only CO_2 and NO_x gave a positive and relatively strong correlation (0.7476) with others revealing weak (positive and negative) relationships between them. This study showed that the use of NG and wastes as fuel substitutes in cement kilns seems the best option to reduce the amounts of pollutants emitted, which are a continuous threat to the public health and microenvironment around the cement industry.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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