



Comparative Analysis of Air Emission from Cement Plant Using TDF as Partial Substitute for Coal

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Tyre derived fuel (TDF) has a potential advantage for its use as a co-fuel with coal in rotary kilns of cement plants. In this study we analyzed the emissions of selected criteria pollutants; CO₂, CO, NO_x, SO_x and PM to suggest suitable proportion of TDF to replace coal when different proportions of TDF and coal were used as cement kiln feed. Emissions of CO₂, CO, NO, NO₂, SO₂ and total PM from stack and concentrations of CO, NO_x, SO_x and PM₁₀ in ambient air were assessed using USEPA recommended methods. Substitution such as 85% coal and 15% TDF, 80% coal and 20% TDF, 75% coal and 25% TDF and 70% coal and 30% TDF were applied in this study. Results of the current study revealed a rise in CO and TPM stack emissions with an increase in proportions of TDF and crossed standards at 80% coal and 20% TDF. Decline in NO_x emissions up-till 15% proportion of TDF was observed and remained lower than the emission of NO_x at 100% coal use in spite of an increase in its concentration at each increased proportion of TDF above 15% TDF use. No significant difference ($p > 0.05$) in ambient air pollutant concentration with and without TDF use was observed. The study suggested 15% TDF substitution as a preferable proportion for the selected cement plant.

Keywords: *air emission, cement industry, tyre derived fuel, coal, fuel substitution*

1. Introduction

It is of significance to ponder upon environmental implications of cement production because of its abundance in world market. On average approximately 1 ton of concrete is produced each year for every human being in the world (Lippiatt and Ahmad, 2005). Asia is the biggest producer among the world cement producing regions, as it contributed 67.60% of the total cement produced worldwide (Ali et al., 2011). Cement manufacturing process is a multistage process. Each step of cement production is associated with certain emissions (Chen et al., 2010). To meet energy demand of this industry, petroleum, oil, natural gas and coal are mostly used (Eckert et al., 1997). Among these, coal is at priority to generate energy for cement manufacturing (CEC 2003), more prominently in developing countries. Burning of fuel especially coal releases considerable amount of carbon dioxide in the atmosphere (Photovoltaics Bulletin, 2002; O'Keefe et al., 2010; Ali et al., 2011).

The processes involving combustion of fuel are mostly responsible for emissions of NO_x and SO_x, CO and O₃ along with PM (particulate matter), while crushing, grinding and mixing are major contributors to particulate matter (Sabah and Whab, 2006). High temperature in cement plant provides very favorable conditions for NO_x formation (USEPA, 1994). Burning of high sulfur coal results in higher SO₂ emission than burning of lower sulfur coal (0.4-1%) (Krylove, 2002). Pakistani coal has high sulfur content (Sharif et al., 2005; Ali et al., 2008). Sulfur content in Pakistani coal ranges from as much low as 0.20% to very high proportion i.e. 15% (Private Power and Infrastructure Board, 2004). There are 29 cement producing companies in Pakistan. Total production of cement increased from 8 million tons in 1996 to 22.50 million tons in 2007 (State Bank of Pakistan, 2007). Increase in production cannot be achieved independent of the energy requirements. It

has been estimated that cement industry consumes about 12-15 percent of the total energy used in industry (Madlool et al., 2011). It reveals that the cement manufacturing is an energy intensive process (Deborah et al., 2009). Coal is a cheaper energy resource therefore is widely used to derive energy in energy intensive processes (Smith, 2003; Gineys et al., 2011). To meet the elevating energy demand in industrial sector there is a need to adopt rational measures to avoid tradeoff between the environment and economic prosperity. Since last decade efforts have been made to find out a better option for fossil fuel substitution with waste materials in cement manufacturing (Gineys et al., 2011). Substitution of cement plant fuel with tyres and sewage sludge, refused derived fuel, waste carpets and resultant change in emissions has been studied worldwide (Carrasco et al., 2002; Lemieux, 2003; Abad et al., 2004; Pipilikaki et al., 2005; Conesa et al., 2008). Using waste tyres as fuel is a more economical option. It not only reduces the demand for coal acquisition, but it is also less expensive (Pipilikaki et al., 2005), and a good practice for reuse of waste tyres instead of incinerating or dumping in landfill (Conesa et al., 2008). Fuel type is one of the most significant factor influencing air emissions (Giannopoulos et al., 2007; Boughton, 2007; Zabaniotou and Theofilou, 2008; Conesa et al., 2008). This study aims to find the suitable proportion of tyre derived fuel (TDF), which could be used as co-fuel in combination of coal in cement kiln. 1 ton of TDF can replace 1.2 tons of coal as analyzed for kiln of the selected cement plant. It has higher calorific value i.e. 31400kJ/kg (Pipilikaki et al., 2005). If coal were fully replaced by TDF, the average requirement of TDF would be as 259 (250-267tons/day) tons per day. The present study compares the air emissions by use of coal as the only heat source and at different proportions of coal and TDF in kiln. Comparisons were made for both stack emissions and ambient air. Stack emissions analysis included CO₂, CO, NO, NO₂, SO₂ and total PM. While ambient air analysis included only CO, NO_x, SO_x and PM₁₀.

2. Methods

2.1. Study area

Study was conducted in Pioneers Cement Plant situated in Jauharabad (32°N 72°E) district Khushab in central Punjab Pakistan. Climate of Khushab is governed by very diverse components of land forms; plain area, hills, desert and agriculture area. The study was conducted in the month of March 2012. Data about wind direction, wind speed, and daily average temperature and humidity of Jauharabad (32°N 72°E) were obtained from the Pakistan Meteorology Department, as these are necessary to select sampling points and understanding the variations in pollutants concentrations (Kordova et al., 2011).

2.2. Cement production plant

The plant comprises two production lines (line1 and line 2), having daily production of clinker as 2000t/day-6000t/day. Both lines of production are well equipped with electrostatic precipitators and cyclones for dust collection. Each production setup has two stacks i.e., one stack for exhaust of combustion kiln and second is the stack for coal mill. The plant consists of raw material crusher, coal crusher, gypsum crusher, stackers, grinders, raw mixers, kiln, three cement mills and three silos. The height of the selected combustion kiln stack of line one is 200ft. The temperature in the cement kiln remains above 1550C⁰ at the time of clinker formation. The plant uses both Pakistani and Indonesian coal as its heat source. An average daily energy requirement is up to 5000MJ/ton. The amount of coal required in combustion kiln of production line 1 is 300 to 320 tons coal/day. American imported shredded TDF was used as a substitute in this study. TDF and coal ratio was adjusted as not to disturb the energy demand and temperature in the kiln during combustion. TDF was used in the proportion, which could not only replace more amount of coal with less amount of TDF, but also maintained the temperature in the kiln before and after substitutions. Five substitutions were given to reach the suitable coal and TDF ratio. Table 1 mentioned the percentage ratio adjustment scheme of two fuels in each of five proportional substitutions. Gaseous emissions and PM concentrations in the exhaust from stack at each substitution were determined.

Table 1. Proportions of coal and TDF in each substitution and combustion conditions

Substitution No	Proportion of coal and TDF, %	Amount of coal and TDF, t	Kiln temperature, C ^o	Oxygen in combustion exhaust, %
S0	100:0	310 : 00	1600	10.39
S1	85:15	264 : 39	1560	11.24
S2	80:20	248 : 52	1580	9.97
S3	75:25	233 : 65	1575	9.75
S4	70:30	217 : 77	1550	10.60

2.3. Ambient air quality monitoring

Selection of sampling points was based on identification of source activity and place for its accomplishment and wind direction. Fuel burning during clinker formation is the main source of NO_x, SO_x, CO₂, PM and CO from a cement plant (Walters et al., 1999; Baris et al., 2000; Deja et al., 2010; Lei et al., 2011). Steps involving physical processing of raw material such as crushing, pilling, and grinding are of significance as being the source of particulate matter (Sabah and Wahab, 2006; Branquinho et al., 2008). Ambient air gaseous parameters and PM₁₀ in nearby fields were analyzed at 100% coal and substitution 4. The sample of ambient air gaseous parameters and PM₁₀ in nearby fields was collected at a point of stack deposition. Plume of selected stack was falling on ground approximately at the distance of 2km from the source in the direction of wind. The sampling point was observable due to falling of hazy colored plume contributed by PM, and other gaseous emissions. In both cases that were before and after fuel substitution this point was selected for measurement of ambient NO_x, SO_x, CO, and PM₁₀ levels contributed by cement kiln. For ambient air, CO was measured using HORIBA APMA 360 analyzer (equivalent method RFCA-0895-106, USEPA, 2011), NO_x with help of HORIBA APNA 360 NO_x analyzer (RFNA-0196-111, USEPA 2011), SO₂ using HORIBA APSA 360 SO₂ analyzer (EQSA0197-114, USEPA, 2011),

particulate matter was collected using integrated air sampler (IO 2.3, USEPA, 2011) and the amount of PM₁₀ was calculated by a gravimetric method (USEPA, 1999).

2.4. Measurement of stack emissions

For stack emissions parameters; CO₂, CO, NO_x, SO₂, and total PM were measured using Testo 350 XI Germany as recommended (Baghdar et al. 2010, Udeozor and Nzeako 2012). The temperature in kiln was similar while measuring contaminants on 100% coal use and after substitutions with TDF. The amount of oxygen was also measured in combustion exhaust stack using Testo 350XI.

3. Results and discussion

The meteorological data re the day of monitoring were represented in Table 2. General meteorological parameters exhibit a key role in site selection. Analysis of gaseous emissions and total PM at 100% coal use with respect to variation in meteorological parameters revealed that with 11% rise in the average 24 hour temperature and 1.3% decrease in relative humidity, CO₂ emission from stack elevated up to 3%, CO up to 20%, NO_x up to 5%, no change in SO₂ concentration, and 8% decrease in TPM was assessed.

Table 2. Meteorological conditions at each day of experiment

Substitution	Average temperature, C°	Wind speed, m/s	Relative humidity, %	Precipitation, mm
S0	19.00	1.028	77.00	0.00
S1	17.50	0.00	81.00	0.00
S2	19.50	1.028	77.00	0.00
S3	18.50	1.028	82.00	0.00
S4	19.75	1.028	83.00	0.00

The highest concentration in stack exhaust was found for CO a well known hazardous gas (494.39 mg/Nm³). Stack emissions analysis did not show any SO₂ contamination (Table 3). All the concentrations obtained after substitutions were compared to the concentrations at 100% coal use. At all the substitutions average temperature and humidity did not vary more than 11% and 7%, respectively, and precipitation was 0 at all days of experiment. The

temperature in kiln did not vary significantly before and after different substitutions. It varied between 1550C° -1600C°. So the mentioned variations in emissions may be solely regarded to fuel substitution. At S1 decrease of 11% (p>0.05) in CO₂ and 51% in NO_x was assessed, While elevations in release of CO and TPM (5.4% and 1.13%, respectively) were negligible (P>0.05) as compared to 100% coal use. No change in SO₂ was observed.

Table 3. Concentration of gaseous pollutants and PM from the stack of cement plant at different proportions of coal and TDF

Parameters (gaseous pollutants and PM)	24 hour average concentration of pollutants for 100% coal and different substitution of TDF				
	S0	S1	S2	S3	S4
CO ₂ , %	9.45 ± 0.13	8.55 ± 1.00	9.40 ± 0.40	9.39 ± 0.02	9.25 ± 0.20
CO, mg/Nm ³	614.99 ± 28.00	650.39 ± 80.00	951.20 ± 272.00	949.10 ± 38.20	1,228.90 ± 17.3
NO _x , mg/Nm ³	309.39 ± 4.0	153.10 ± 17.00	216.29 ± 23.00	206.29 ± 24.00	220.29 ± 41.10
SO ₂ , mg/Nm ³	0.00 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.90 ± 0.51	1.49 ± 1.00
Total PM, mg/Nm ³	408.59 ± 33.20	413.29 ± 0.00	444.49 ± 26.00	541.00 ± 77.00	2,831.90 ± 247.40

Concentrations of all selected parameters were within the limits defined by Pak. EPA (2011) except total PM, which crossed the standard ($300\text{mg}/\text{Nm}^3$) not only at S1 but at 100% coal as well. At S2 there was no considerable ($P>0.05$) change in CO_2 concentration (2%), but remarkable decline ($p<0.05$) in NO_x i.e. 30% was observed. Results revealed that CO , SO_2 and TPM concentration increased in stack emissions with increase in proportion of TDF used. Increase in emissions of CO (35.3%) and TPM (8.1%) was observed exceeding the concentrations measured at S1. CO concentration as $951.20\text{mg}/\text{Nm}^3$ crossed the NEQS i.e. $800\text{mg}/\text{Nm}^3$. The trend was disturbed at S3. The parameters which were on decrease at S1 (CO_2 and NO_x) showed further decline in concentrations at S3. Slight decrease in CO_2 i.e. 2.1% and NO_x (33.3%) was assessed. There was incline in CO (35%), SO_2 (100%) and TPM (25%). This increase in CO led to violation of the national air quality standard for CO . At S4 an increase in all the considered pollutants has been observed except CO_2 . CO_2 decreased up to 3.6% and was also lower than the concentration at S3. Although the kiln temperature did not vary significantly, but increase in oxygen and decrease in CO_2 at S1 and S4 might be due to decrease in the temperature. 29% decrease in NO_x was assessed as compared to 100% coal use, while the concentrations higher than the concentrations at S3. CO increased 50%. It crossed the concentration emitted at S3 and national standard as well. SO_2 reached at $1.49\text{ mg}/\text{Nm}^3$ from 0 which was the highest among all substitutions, and is within the standard limit ($1700\text{mg}/\text{Nm}^3$). TPM was considerably elevated, i.e. 86% and also crossed the values obtained at S3. It reached up to $2831.90\text{ mg}/\text{Nm}^3$ and crossed the standard ($300\text{mg}/\text{Nm}^3$). Use of waste material and used tyres was adapted as a tool to reduce CO_2 and NO_x emissions reduction from the cement kiln to comply with regulatory requirements. Studies reported the achievement of lower CO_2 , NO_x emissions as a result of fuel substitution (Conesa et al. 2008; Deja et al., 2010). CO_2 showed a continuous declining trend with increase in the amount of TDF and decrease in the amount of coal. Literature supports the view that there is no significant change in

stack emissions during the use of TDF in comparison with coal (Pipilikaki et al., 2005; Middleton et al., 2008). The pilot scale study conducted by Carrasco et al. supports the results of this study. It states 37% increase in CO emission, 24 % increase in SO_2 and 11% decrease in NO_x emissions, when TDF is fed to kiln along with coal (Carrasco et al., 2002). Carbon and nitrogen content of waste material, combustion properties, feeding place, and retention time are often controlling factors for the CO and NO_x emission (Lemieux et al., 2004). Coal is responsible for greater NO_x emissions as compared to TDF. In this study substitution of coal with TDF leads to lesser NO_x emissions (including NO and NO_2). The trend in NO_x emission in this analysis has not been linear, as NO_x emissions from cement manufacturing are always excessively variable (Walters et al., 1999). SO_2 elevated non-significantly ($p>0.05$) only at higher proportion of TDF i.e. 25% and 30%. In clinker formation about 75-80% of SO_2 is absorbed by lime stone and some of proportion remains at the bottom of the combustion kiln. So SO_2 pollution from kiln remains negligible (Yang et al., 2003). A large increase in PM in S3 and S4 is unusual in this experiment; increase in the amount of TDF can be a good justification for this. Raw material input and operational conditions could also be reasons for increase in TPM emissions from stack.

The level of pollutants in nearby fields of a point source is often the point of concern for regulatory authorities and health scientists (Hajra et al. 2011). Ambient air analysis at a considerable distance from the point source (Table 4) that is stack of the cement plant implores that there was no significant variation ($p>0.05$) in the concentrations of these pollutants as compared to the values obtained at 100% coal use. Comparison between the ambient air concentrations in nearby field at the point of stack deposition for 100% coal and S4 indicated no correlation between stack emissions and ambient air concentration. Ambient air concentrations at the point of stack deposition did not exceed the USEPA (2011), WHO (1998), PAK EPA (2012), and WB (2012) ambient air quality standards.

Table 4. Ambient air concentration outside the vicinity of cement plant at 100% coal use and maximum proportion of TDF

Sr. No	Parameters	24 hour average concentration at S0	24 hour average concentration at S4
1	CO , mg/m^3	0.36 ± 0.007	0.65 ± 0.006
2	NO_2 , $\mu\text{g}/\text{m}^3$	20.30 ± 0.900	19.50 ± 0.006
3	SO_2 , $\mu\text{g}/\text{m}^3$	14.00 ± 0.041	12.30 ± 0.006
4	PM_{10} , $\mu\text{g}/\text{m}^3$	140 ± 0.041	138.00 ± 0.041

Micrometeorological parameters play a critical role in dilution, dispersion, and sampling (Goyal et al., 1996). The sampling point in nearby field was the same at 100% coal use and at the maximum level of substitution i.e. S4, because of the fact that wind was calm and wind speed (1.028 m/s) was the same on the day of substitution, which led to the deposition of exhaust at the same place as it was on the day of 100% coal use. Although in the present study,

temperature and precipitation did not play a governing role in variations in the ambient air concentration in nearby field, because there was no significant change in the average temperatures of the two respective days and precipitation was also 0, therefore the change in relative humidity could be the contributing factor. Decrease in NO_2 , SO_2 and PM_{10} at substitution 4 can be attributed to increase of 6% in relative humidity. There was no huge difference in ambient air

pollutants concentration at 100% coal use and substitution 4. No change in wind speed and direction on the two different days of measurement provides a good reason for it, as both are primary factors in dilution of pollution (Berkowicz et al. 1996). The study was limited due to unavailability of ambient air quality data of pollutants before deposition due to continuity of the manufacturing process. The obtained concentration of pollutants cannot be wholly regarded to stack emissions, as natural sources of the area always contribute to the ambient pollution level. PM level also depends upon land form of the area (Middleton et al., 2008; Ward and Lange, 2010), and the distance from stack or stack height (Godoy et al., 2009). The cement plant under consideration contains and is surrounded by the unpaved barren landscape leading to the greater air PM level. Ambient air analysis at minimum and medium substitution of coal for TDF other than these proportions is also lacking, which could provide a clearer picture for selection of most suitable coal TDF proportion. A considerable decrease in NO_x emissions and decline in CO₂ with increase in each proportion of TDF renders it preferable fuel. Results of the study suggest only proportional substitution of coal with TDF due to a considerable rise in CO and PM concentration from stack emissions with an increase in the amount of TDF substituted as fuel.

4. Conclusions

This industrial scale fuel substitution experiment demonstrated the potential for waste tyres to be used as a fuel in cement kiln taking the environment as priority. Comparison of results obtained from 100% coal use and 15% TDF use indicated only negligible increase in CO and TPM from stack emissions. While all other pollutants decreased considerably from their concentration level at 100% coal use. At 20% and 25% TDF substitution there was a decline in CO₂ and NO_x, but a significant increase in CO and TPM. Similarly at the maximum 30% substitution, although decline in NO_x emissions was considerable, CO and TPM immensely increased. The ambient air concentrations of parameters CO, NO_x, Sox, and PM₁₀ did not show significant difference as compared to the ambient air levels of gaseous emissions and PM₁₀ at 100% coal use. The study suggests 85% coal and 15% TDF as the most suitable proportion of the TDF among all proportions used in current studies for the selected cement plant, which could not be generalized for other cement plants of different properties.

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Emisijų į aplinkos orą iš cemento gamyklos, naudojant PAK kaip dalinį anglių pakaitalą, palyginamoji analizė

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Iš padangų atgautas kuras (PAK) turi potencialų pranašumą, nes jis panaudojamas kaip priedas anglies kurui sukamosiose krosnyse cemento gamyklose. Atlikus tyrimą išnagrinėti išmetamų teršalų kriterijai: CO₂, CO, NO_x, SO_x ir KD. Jie parinkti tokie norint parinkti tinkamą PAK, galintį pakeisti anglį, kiekį, nes iki šiol įvairios PAK ir anglies proporcijos buvo naudojamos kaip cemento džiovinimo krosnies kuras. CO₂, CO, NO, NO₂, SO₂ ir bendros KD (BKD) emisijos iš kamino bei CO, NO_x, SO_x ir KD₁₀ koncentracijos aplinkos ore buvo vertinamos taikant JAV AAA rekomenduojamus metodus. Tyrimui buvo naudojamos tokios medžiagų proporcijos: 85 % anglies ir 15 % PAK, 80 % anglies ir 20 % PAK, 75 % anglies ir 25 % PAK ir 70 % anglies ir 30 % PAK. Tyrimo rezultatai parodė, kad didesnis CO ir BKD emisijų kiekis iš kamino dėl didesnės PAK dalies ir buvo didesnis nei standartas – 80 % anglies ir 20 % PAK. NO_x išmetimų kiekis sumažėjo, naudojant iki 15 % PAK dalies, sudarančios kurą, ir išliko mažesnis nei NO_x emisijos kiekis, naudojant 100 % anglies kurą, nepaisant to, kad jo koncentracijos didėjo, didėjant PAK daugiau nei 15 %. Nustatyta, kad aplinkos oro teršalų koncentracijų pokytis nebuvo reikšmingas ($p > 0,05$) nei esant PAK, nei be šio kuro. Tyrimas parodė, kad 15 % PAK yra geriausias tirtos cemento gamyklos pasirinkimas.