



Environmental analyze of cement production with application of wastes

Maria Luiza Grillo Renó¹

Larissa Ferrini Ferrari Alves²

José Carlos Escobar Palacio³

Lidiane La Picirelli de Souza⁴

Felipe Orlando Centeno González⁵

Pedro Jessid Pacheco Torres⁶

Abstract: The cement production is benefiting worldwide by use of waste fuel since 30 – 40% of total costs are attributed to energy cost. The application of mineralizer has also economic advantages due to reduction of clinkering temperatures. In this context the present work proposes analyzing different option of waste fuels through case studies (tires, plastics, woodchips and switchgrass) and the use of mineralizer in clinker production. The results showed that the waste fuel plastic emitted 8% less CO₂ than woodchips, and all case studies emitted SO₂ below of limit established by Directive PE-CONS 31/10. The mineralizer applied reduced the specific heat consumption in 12% and the CO₂ emissions were decreased in 4%.

Keywords: Cement production; mineralizer; waste fuel; environmental analysis.

¹ UNIFEI – Universidade Federal de Itajubá

² UNIFEI – Universidade Federal de Itajubá

³ UNIFEI – Universidade Federal de Itajubá

⁴ UNIFEI – Universidade Federal de Itajubá

⁵ CUC – Universidad de la Costa

⁶ CUC – Universidad de la Costa

1. Introduction

The cement production has undergone development from 2000 year. The annual global cement production has reached 2.8 billion tons and is expected to increase to some 4 billion tons per year (Schneider et al., 2011). Cement is one of the most important building materials worldwide, it is used mainly for the production of concrete, then the cement consumption and production is closely related to construction activity and, therefore to economic activity.

Cement sub-sector represent an energy intensive industry, approximately 12 – 15% of total industrial energy use. The typical primary energy consumption for cement production is 75% fuel and 25% electrical energy (Madloul et al., 2011). Forward this high fuel consumption the cement companies are introducing the industrial waste co-processing from a perspective that combines environmental policies with the interests of companies. The industrial wastes can provide part of the energy required to make cement.

The cement industry emits approximately 5% of the man-made and 3% of total global carbon dioxide (CO₂) emissions (EPA, 2013). CO₂ is emitted from the calcination process of limestone, from combustion of fuels in the kiln, as well as from power generation. The Figure (1) presents the global CO₂ production in 2005 year, and the participation of each sector.

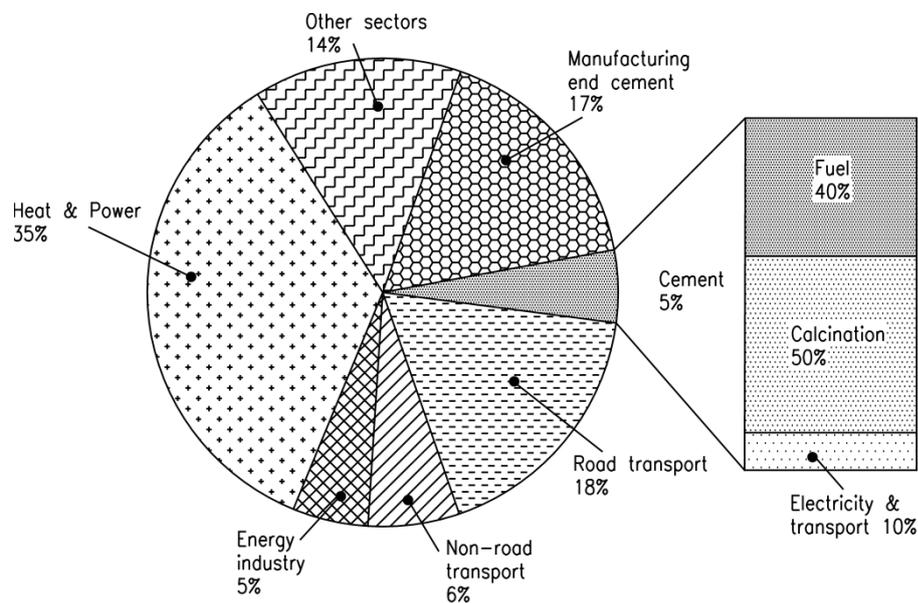


Figure 1: Global CO₂ production (WBCSD, 2005).

On this problem the Cement Sustainability Initiative (CSI) of the World Business Council for Sustainable Development (WBCSD) had encouraged the cement companies to address issues related to global sustainability. One of these issues is the industry's emissions of carbon dioxide (WBCSD, 2005). In Europe, in order to reduce energy consumption and CO₂

emissions, the industry is focusing on increasing the use of clinker substitutes in cement, the use of alternative fuels and energy efficiency measures, including modifications to the reaction chamber (kiln), recycling waste heat and carbon capture and storage of CO₂ from the waste gases.

In this context the present work have the aim applying different waste fuels, computing the emissions of SO₂ and CO₂ and mass consumption of each waste fuel. With the results is possible appointing which waste fuel was more favorable for the cement industry from environmental view point.

1.1 Portland cement production

In cement manufacturing, the main raw materials contain calcareous (high CaCO₃ content) and argillaceous (high silica and alumina content) materials. The raw materials must be crushed and proportioned so that the appropriate chemical composition of the raw material feed is reached. The chemical composition and the size distribution are crucial to both the quality of the product and the operation of the process (Kosmatka et al., 2002).

After crushing the raw material is sent to pyro processing stage, beginning with the raw materials are subjected to enough heat to allow the clinkering reactions to proceed. The main chemical reaction produces the calcium silicates. There is a combination of endothermic and exothermic reactions occurring in an extremely complicated chemical reaction sequence. In essence the following material transformation occurs in pyro processing stage (Table 1).

Table 1: Material transformation in pyro processing stage (Bhatta et al., 2004).

Process	Temperature
1. Evaporating free water, at temperature up to	100°C
2. Removal of adsorbed water in clay materials	100°C – 300°C
3. Removal of chemically bound water	450°C – 900°C
4. Calcination of carbonate materials	700°C – 850°C
5. Formation of C ₂ S, aluminates and ferrites	800°C– 1250°C
6. Formation of liquid phase melt	>1250°C
7. Formation of C ₃ S	1330°C – 1450°C
8. Cooling of clinker to solidify liquid phase	1300°C – 1240°C
9. Final clinker microstructure frozen in clinker	< 1200°C
10. Clinker cooled in cooler	1250°C – 100°C

From Table 1 is described the raw material transformation, one important phase of the production of the clinker is the calcination process. In this chemical reaction calcium carbonate decomposes in calcium oxide and carbon dioxide as Eq. (1) (Worrell et al., 2001):



Calcium oxide (CaO) is the main compound of cement clinker and inside the kiln it is reacted with other oxides such silica, alumina, iron oxide for the formation of $3\text{CaO} \cdot \text{SiO}_2$ (C3S), $2\text{CaO} \cdot \text{SiO}_2$ (C2S), $3\text{CaO} \cdot \text{Al}_2\text{O}_3$ (C3A) and $4\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot \text{Fe}_2\text{O}_3$ (C4AF). The main chemical reactions are presented below (Çamdali et al., 2004):



The C3S and C3A are responsible for the strength of the cement. The high percentages of C3S result in high early strength but also high heat generation as the concrete sets. C3A causes undesirable heat and rapid reaction properties, which can be prevented by adding CaSO_4 to the final product (Paula, 2009).

The kiln charge must spend sufficient time at high enough temperature to reach near complete reaction to form clinker minerals. The material bed must tumble adequately to ensure uniform heating through the mass. A summary of a typical kiln heat balance during clinkering reactions is presented in Table 2.

Table 2: Summarized data on a typical kiln heat balance for dry process (Bhatty et al., 2004).

Endothermic reactions	kJ/kg
Dehydration of clays	170
Calcination	1990
Heat of melting	105
Heating raw mats (1450°C)	2050
Sub total	4315
Exothermic reactions	
Crystallization dehydrated clay	40
Heat of formation, clinker minerals	420
Crystallization of melt	105
Cooling of clinker	1400
Cooling of CO_2	500
Cooling of water	85
Sub total	2550
Net heat for clinker formation (endothermic heat – exothermic heat)	1765
Heat losses related to inefficiencies during the kiln operation	
Evaporation of water	20
Heat losses, gas, clinker, dust	840
Radiation, convection loss	650
Total heat consumption = (endothermic heat – exothermic heat) + heat losses due to inefficiencies	3275

The final step in Portland cement manufacturing is grinding the blend of clinker and gypsum. Up to 5% (by weight) of gypsum is added to the clinker after it is cooled. The amount of gypsum is adjusted to regulate cement properties such as setting time (Kosmatka et al., 2002)

Depending on how the raw material is handled before being fed to the kiln, basically three different types of process can be distinguished: the dry, semi-dry/semi-wet and wet process. In dry process the feed material enters the kiln in a dry, powdered form. The dry feed is preheated in a tower of heat exchange cyclones in which the dry feed is preheated by hot exit gases from rotary kiln prior to entering the kiln. The calcination process can almost be completed before the raw material enters the kiln if part of the fuel is added in a special combustion chamber (pre-calciner kiln) as it showed in Fig. (2).

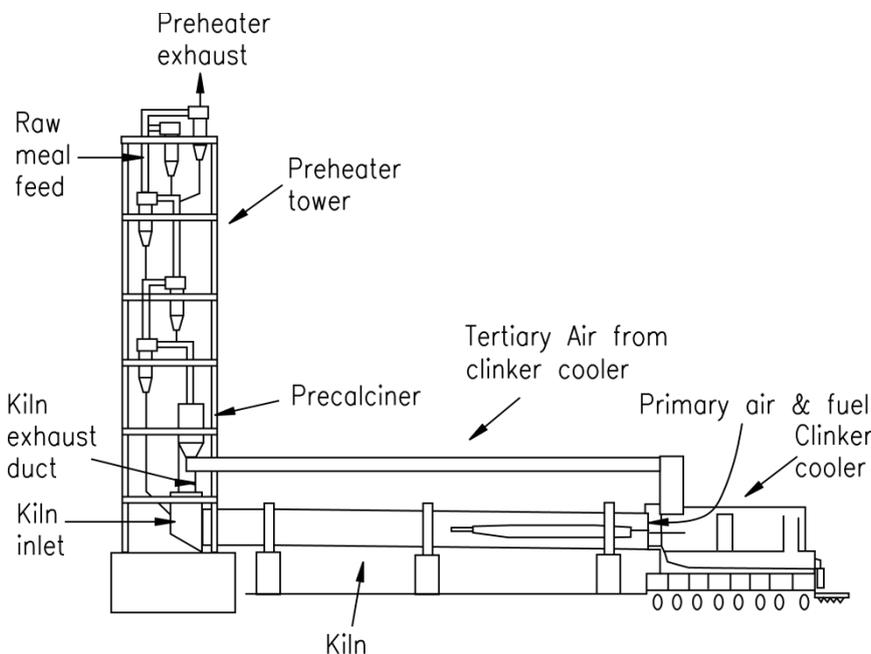


Figure 2: Precalciner kiln of cement industry

In the wet process the feed material goes to wet grinding and the resulting slurry, which contains typically 30 – 40% water. In the semi-dry or semi-wet process 10 – 20% water is either added to the ground dry feed material resulting in a feed material containing about 15 -20% moisture. Over past few decades the worldwide cement industry has invested in moving from wet process to the more energy-efficient dry process.

1.2 Waste fuel in cement industry

Nowadays the high degree of the industrial activity as well as the increasing standards of living has been accompanied by a growing waste generation which represents a serious

environmental problem. The possibility to use some industrial wastes in the cement production as source of raw materials or alternative fuel have been viable to cement industries production cost.

Cement kilns and calciner have technical conditions very favorable for use of alternative fuels, as example, the temperatures in the kilns of approximately 1500°C are higher than the threshold ignition temperature, as established by environmental regulations, long retention time of products under high temperature combustion, the high alkalinity atmosphere readily absorbs most acidic gases released by the oxidation of Sulphur and chlorides (Greco et al., 2004).

The waste fuel utilization in cement kiln is still processing, the advantage of cement kiln is the high temperatures reached inside (about 1450°C) combined with the highly oxidizing environment and the large residence time of material exposed to these conditions, represent an alternative recognized form of thermal destruction of industrial wastes.

While in some kilns up to 100% replaced rates have been achieved, in others process places, local waste markets and permitting conditions do not allow for higher rates of waste fuel. The waste fuel utilization requires the adaptation of the combustion process. Modern multi-channel burners designed for the use of alternative fuels and thermograph systems allow control of the flame shape to optimize the burning behavior of the fuels and the burning conditions for the clinker (Wirthwein & Emberger, 2010).

In a conventional preheater kiln (without precalciner), it is only possible to burn fuels in the kiln inlet with substitution rates of up to 25 to 30% (70 to 75% has to be fired in the main firing). On the other hand, in precalciner kilns usually up to 65% of the total fuel energy input is burned into the calciner. As consequence, in precalciner kilns, fluctuations in fuel quality have less impact on the kiln performance, and clinker quality (Schneider et al., 2011).

In relation to clinker quality, the burning behavior of most waste fuels differs significantly from the behavior of fossil fuels due to higher particle sizes, material densities and transport characteristics. Consequently it can change the temperature profile of the kiln including the clinkerization temperature, the length of the sintering zone and the cooling conditions. As result these changes can affect the quality of the clinker, as example, burning grade of the clinker, the porosity of the granules, the crystal size of the clinker phases or their reactivity (Schneider et al., 2011).

Other problem from the use of different fuels in the clinker burning is the formation of rings in the kiln caused by salts – such as KCl, K₂SO₄, Na₂SO₄, Ca₂SO₄ that are formed by the volatiles. This is also known as the cyclic phenomena, and the amounts of volatiles and especially sulfur, contained in the fuels have a direct effect on these phenomena (Pipilikaki et al., 2005).

The presence of chlorine in waste fuels has implications on cement kiln emissions and performance. Trace levels of chlorine in feed materials can lead to the formation of acidic gases

such as hydrogen chlorine and hydrogen fluoride. As well as, dioxins have potential to form if chlorine is present in the input fuel or raw materials. Formation can be repressed, however, by the high temperatures and long residence times that are standard in cement kilns (Karstensen, 2008). Take to account the wastes usually co-processed in cement industry are (Lamas et al., 2013):

- Used oil, including lubricant
- Soil contaminated with oil (fuel or lubricant), oily waste (rags, wood chips, sawdust, vegetation impregnated with oil and water, paper towel with oil)
- Solid waste compounds of non-toxic metal
- Slag molten of aluminum, iron, steel, and zinc
- Waste of non-metallic minerals
- Waste of paper, cardboard, polymerized plastic and textile materials
- Waste of acid washing of benzene, originating from distillation of coal tar coke
- Wastes from incineration or thermal treatment of contaminated soil
- Dirt solvents, asphalt emulsion (in solvent), mix resin with solvent.

One waste fuel used for many cement industries is the used tire, especially since the accumulation of used tires is an environmental liability, since the improper disposal of used tires can turn them into sources of environmental and public health problems (Caponero et al., 2003). In Brazil, the National Environment Council – CONAMA approved a resolution, with the force of law, the resolution n° 258 of August 26, 1999, to regulate the final destination of waste tires. The resolution established a timetable for the destruction of these tires since 2002. Therefore the use of scrap tires as waste fuel is considered, among the ways to recycle tires, as the most viable means for its disposal. Actually in Brazil 57% of scrap tires that are discarded each year were intended to cement kilns (Lamas et al., 2013).

But the use of tire as a fuel is limited to a maximum of 30% for both domestic and international cement industry due to the presence of heavy metals in their composition, mainly zinc. This has the effect of lowering the initial resistance, but ensures higher final strength of the cement. In study realized by (Pipilikaki et al., 2005) the total tire used as fuel was 6%, it was concluded that no apparent problems occurred from the use of this waste fuel in the clinker burning.

The waste solvent and spent oil from different industries generally have high calorific value and those can be used in cement kiln. The work Vos et al. (2007) concluded that the maximum and minimum calorific values of solvent and spent oil were 29 MJ/kg and 36 MJ/kg respectively. Solvent and waste oil contain less minerals compared to petroleum cokes and coal hence a little additional raw meal is need to ensure the quality of the cement. On the other side

Vos et al. (2007) reported that CO₂ emission was reduced while solvents and paint sludge were applied as waste fuel. The work Seyler et al. (2005) showed a reduction in heavy metal emissions when waste solvents were mixed with fossil fuel.

The plastic solid waste is the global environmental problem, the study Siddique et al. (2008) reported that approximately 8% of plastic solid waste is incinerated and 7% is recycled leaving the 85% remainder to be landfilled. The application of plastic solid waste as fuel was studied by Swart (2007) through two scenarios. The baseline burn consisted of only coal, whereas the trial burn consisted of a blend of coal, plastics and waste tires. The results of the trial were that concrete compressive strengths increased slightly, CO emissions were reduced, and NO_x, SO₂ and VOC were increased compared to the coal only burn.

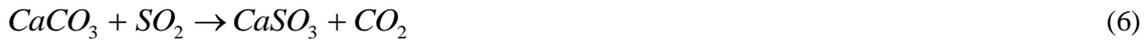
The application of biomass fuels in cement industries had increased, especially since biomass fuels have lower concentrations of nitrogen and sulfur, oxides produced during the combustion process are significantly less than emissions produced by fossil fuels. The use of biomass in cement manufacturing is less common in industrialized countries and appears to be concentrated in more rural developing regions such as India, Thailand, and Malaysia.

1.3 Emissions of cement industry

As mentioned the carbon dioxide emission from cement production is a by-product of a chemical conversion process (calcination) that is used in the production of clinker. During the cement production process, calcium carbonate (CaCO₃) is heated in a cement kiln at a temperature of about 1300°C to form lime (i.e., calcium oxide or CaO) and CO₂ (EPA, 2013).

According to the CSI's getting the Numbers Right database (GNR), the average specific CO₂ emissions amounted to 652 kg/ton cement product in 2010. These emissions have been progressively reduced due to the strong economic incentive for the cement industry to minimize fuel energy consumption. CO₂ reduction of some 30% in the last 25 years is expected by the adoption of more fuel efficient kiln processes, increased utilization of renewable alternative fuels or the other waste derived fuels and the production of blended cements with mineral additions substituting clinker (WBCSD, 2006).

Sulfur compounds enter the kiln system either with the fuels or with the raw materials. In raw material the sulfur compounds are present mainly as sulphates (calcium sulphate CaSO₄) or as sulphides (i.e. pyrite or marcasite FeS₂). The sulphates are thermally stable up to temperatures of 1200°C, and they are decomposed to produce SO₂. Part of the SO₂ combines with alkalis and is incorporated into the clinker structure; the remaining part of SO₂ is carried back to the cooler zones of the kiln system where it reacts with calcined calcium oxide or with calcium carbonate thus being reintroduced to the sintering zone again. The main chemical reactions of SO₂ absorption is presented below (Santos, 2007):



According Santos (2007) the absorption of sulfur by Equations (6) – (10) corresponds to 93% of total sulfur compounds from raw material and fuel. Other emission from cement manufacturing is the nitrogen oxides (NO_x). Its formation is favored by high process temperatures inside the kiln. Essentially all NO_x emissions are formed during fuel combustion by two primary mechanisms:

- Oxidation of molecular nitrogen present in the combustion air, named thermal NO
- Oxidation of nitrogen compounds present in the fuel

The thermal NO is generated in the burning zone through reaction between oxygen and nitrogen as follows:



The fuel nitrogen is only partially converted into NO during combustion and this reaction occurs through the temperature range of the combustion process. NO_x formation from nitrogen contained in fuels occurs through a complex series of reactions. N₂ is oxidized into NO and is reduced to N₂ by reacting with intermediate combustion products (Rother, 1997).

The NO_x formations mechanisms take to account the energy efficiency of cement production and the heat input need to produce a unit quantity of cement. A high thermal efficiency would lead to less consumption of heat and fuel and consequently less NO_x emissions. Then, the NO_x control approaches applicable to the cement industry may be grouped in two categories:

- Combustion control approaches where the emphasis is on reducing NO_x formation;
- Post combustion control approaches which control the NO_x formed in the combustion process.

With relation of emissions of ‘particulate matter’ or dust from cement kiln, these emissions have been reduced over the last two to three decades due to regular improvements in design and operation, including increase use of modern de-dusting equipment. The emission of volatile organic compounds (VOCs) is not a significant in cement production. Under normal circumstances the VOC content of the exhaust gas from cement kilns ranges between 1 and 60 mg/Nm³. Other pollutants such as acid gas can be considered as minor and not material considering the trace amounts of Chlorine and Fluor present in raw materials and fuels (WBCSD, 2006).

2. Cases study

The present work considered the application of different waste fuels in clinker production (tires, plastics, woodchips and switchgrass) together with traditional fuels. The amount of each fuel was based in BEN (2011), corresponding 75% petroleum coke, 1.2% mineral coal, 1.5% charcoal, 22.3% waste fuel.

Thus four case studies was stabilized, the case study 1 use the waste fuel tires, case study 2 (plastics), case study 3 (woodchips), case study 4 (switchgrass). The selection of these waste fuels was due the application of tires for more than 20 years by Brazilian cement industry, the increase of using renewable energy sources (woodchips and switchgrass). With relation to the plastics they are environmental problems by its large production, and then burned as fuel is a viable alternative for it. The composition of waste fuels applied is presented in Table 3.

Table 3: Waste fuel composition (Akkapeddi, 2008).

Component (wt. %)	Tires	Plastics	Woodchips	Switchgrass
Carbon	75.94	8.06	52.64	50.25
Hydrogen	6.53	64.23	5.83	5.70
Nitrogen	0.52	1.27	0.15	1.22
Oxygen	0.46	0.22	40.53	37.37
Sulfur	2.00	17.46	0.02	0.19
Al ₂ O ₃	1.15	0.48	0.93	1.57
CaO	1.68	92	54.61	13.99
Fe ₂ O ₃	84.72	0.54	1.79	1.06
K ₂ O	0.17	0.13	17.28	24.72
MgO	0.33	1.75	9.83	9.02
Na ₂ O	0.19	0.17	0.38	0.96
P ₂ O ₅	0.12	0.14	2.80	8.49
SiO ₂	4.91	2.12	3.27	34.86
SO ₃	0.51	0.41	3.33	4.53
Lower Heating Value (kJ/kg)	38969	29665	19510	18985

The composition of traditional fuels (petroleum coke, charcoal and mineral coal) are showed in Table 4:

Table 4: Traditional fuel composition (Berguerand & Lyngfelt (2008), Machado & Andrade (2004), Paula (2009))

Component (wt. %)	Petroleum coke	Charcoal	Mineral coal
Carbon	81.32	71.91	70.12
Hydrogen	2.87	4.20	4.51
Sulfur	6.02	-	1.28
Oxygen	0.45	11.36	1.13
Nitrogen	0.88	1.11	2.25
Water	8.00	7.80	0.85
Ash	0.46	3.62	19.86
Lower Heating Value (kJ/kg)	31750	27900	26928

With relation to raw material, it was analyzed a production of clinker of 34.72 kg/s, and the production of 1 kg clinker requires 1.56 kg of raw material for a cement plant operating as a dry process (based on real data on the Brazilian cement industry). Therefore, the cement plant consumes approximately 54.16 kg raw material/s. The composition of the raw material considered for this study is shown in Table 5.

Table 5: Composition of raw material.

Component (wt. %)	Value
CaCO ₃	75.21
MgCO ₃	3.57
P ₂ O ₅	0.13
SiO ₂	12.79
Al ₂ O ₃	3.44
Na ₂ O	0.03
Fe ₂ O ₃	1.86
K ₂ O	0.80
Cl	0.014

Estimating the burning in the kiln occurs with 1.7% of free oxygen, and the calciner it follows with 2.4% of free oxygen (medium value of the cement industry), the specific heat consumption is according with Table 2 (3275 kJ/kg). Thus it is possible computed the composition of clinker produced (Equations (1) – (5)) and emissions of SO₂ and CO₂ through combustion stoichiometric calculations and Eq. (6) – (10).

3. Results

Computing the clinker composition of each case study and the emissions of CO₂ and SO₂ reaches the following results in Table 6 and Table 7.

Table 6: Clinker composition.

Component (wt. %)	Case study 1	Case study 2	Case study 3	Case study 4
	Tires	Plastics	Woodchips	Switchgrass
CaCl ₂	0.04	0.04	0.04	0.04
CaSO ₄	1.62	3.01	0.72	0.32
K ₂ SO ₄	1.69	1.67	2.51	2.97
Na ₂ SO ₄	0.24	0.23	0.28	0.39
MgO	8.42	8.31	8.26	8.43
C2S	42.44	41.79	41.67	45.11
Cal free	2.26	2.23	2.21	2.26
C4AF	3.56	2.28	2.31	2.34
C3A	3.15	4.33	4.28	4.43
C3S	36.58	36.10	37.73	33.72

Table 7: Emissions of SO₂ and CO₂

Gas (kg/ton clinker)	Case study 1	Case study 2	Case study 3	Case study 4
	Tires	Plastics	Woodchips	Switchgrass
SO ₂	1.35	1.90	1.36	1.40
CO ₂	844	799	864	863

From Table 6 and 7 is observed that the different waste fuels did not change significantly the clinker composition. On the other hand the emissions showed differences. The plastics had a great advantage in relation to CO₂ emissions, it emits 8% less than the case study 3 that applied woodchips, but emits 39% more SO₂ than the case study 3.

However the Directive PE-CONS 31/10 (2013) established the limit of SO₂ emission of 200 mg/Nm³ and the case study 2 emits only 1.32 mg/Nm³. Thus the SO₂ emissions are not a great problem for cement industry.

Now, computing the mass consumption of each waste fuel, the results gotten are showed in Table 8:

Table 8: Consumption of waste fuel

Waste fuel	Amount (kg/ton clinker)
Case study 1 (tires)	18.74
Case study 2 (plastics)	24.62
Case study 3 (woodchips)	37.43
Case study 4 (switchgrass)	38.47

From Table 8 the case study 1 was more advantageous than case study 2, the main cause is the higher lower heating value of tires (31750 kJ/kg). This result is important to transport system necessary to carry the waste fuels to cement industry, with lower demand of tires in relation to plastics result in costs reduction with transport system.

4. Conclusions

From Table 8 the case study 1 was more advantageous than case study 2, the main cause is the higher lower heating value of tires (31750 kJ/kg). This result is important to transport system necessary to carry the waste fuels to cement industry, with lower demand of tires in relation to plastics result in costs reduction with transport system.

The present study aimed to do an evaluation of different waste fuels in clinker production from environmental view point, especially the CO₂ and SO₂ emissions. The waste fuels woodchips and switchgrass were the largest emitters of CO₂, however, it is important to take into account that these biomass absorbed CO₂ by photosynthesis process during growth stage, thus minimize the impact of CO₂ emission.

The waste fuel plastics had lower CO₂ emissions, the great advantage to clinker production, as well as the waste management since the illegal disposal contribute to an array of environmental problems.

The tires were the more advantageous with respect to waste fuel consumption. Its high lower heating value caused the demand of a lower mass consumption. Consequently reduce the costs with transport system that carry the waste fuels. Besides the cogeneration of tires help to solve the problem of thousands of waste tires disposed in landfills.

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