Cradle-to-gate environmental life cycle assessment of limestone calcined clay cement (LC3)

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The manufacturing of cement contributes to approximately 5-7% of global anthropogenic carbon dioxide emissions, necessitating the need for reducing the environmental impact. Limestone Calcined Clay Cement (LC^3) has emerged as a promising alternative to ordinary Portland cement (OPC), leveraging widely available resources like clay, limestone and gypsum to partially replace the carbon intensive Portland clinker. One ton of Portland cement is associated with about one (1) ton of CO_2. This study aimed to assess and compare the CO_2 emissions of theoretical binary and LC^3 cement types against 100% Ordinary Portland Cement (OPC). Considered were: OPC with 30% calcined clay replacement, and LC^3, composed of 50% clinker, 30% calcined clay, 15% limestone, and 5% gypsum.

The study, limited to a cradle-to-gate analysis, utilised the life cycle assessment software tool SimaPro 8.1 with the Ecoinvent Database version 3. The life cycle inventory dataset for each material was compiled, and the ReCiPe midpoint (H) method was employed to generate and report the results in CO_2 equivalents.

The results indicated that LC^3 exhibited significantly lower CO_2 emissions compared to both OPC and binary OPC with 30% calcined clay replacement. This research demonstrates LC^3's potential as a highly impactful alternative supplementary cementitious material (SCM), particularly in reducing CO_2 emissions in the cement industry and acquiring significant carbon credits and/or reducing carbon tax where applicable.

Keywords: Calcined clay, CO2 emissions, life cycle assessment, limestone calcined clay cement, ordinary Portland cement, supplementary cementitious materials

1. Introduction

Globally, the cement industry faces significant pressure to mitigate and minimize carbon dioxide (CO_2) emissions. Cement manufacturing is responsible for emitting 780 – 1000 kg of CO_2 for every ton of cement produced, contributing to approximately 5-7% of the world's anthropogenic CO_2 emissions (Suryawanshi et al., 2015; Krajči et al., 2015). In response to growing global climate change concerns, the cement industry is actively exploring strategies to minimize its environmental impact and align with international goals outlined in the Paris Agreement (UNFCC, 2023). This agreement, which has been signed by 194 countries emphasizes the urgent need to strengthen measures aimed at limiting the global temperature rise to below 2°C above pre-industrial levels and to pursue further efforts to keep the increase below 1.5°C (UNFCC, 2023). For the cement industry, available mitigation strategies include increasing energy efficiency, utilising of alternative fuels, partially replacing clinker with supplementary cementitious materials (SCMs) to reduce the carbon footprint of the cement, and implementing carbon capture and storage solutions (UNEP, 2019).

The use of SCMs in cements is already a global practice, standardised, for instance, by prEN 197-1:2018 in Europe, and ASTM C 595:2019 in the United States (Rodrigues et al., 2022). The most used SCMs are fly ash (FA), ground granulated blast furnace slag (GGBFS) and, to a lesser extent, silica fume (SF). However, geographical constraints and limited supplies of these SCMs (as depicted in Figure 1.1) pose challenges due to the growing demand for cement.

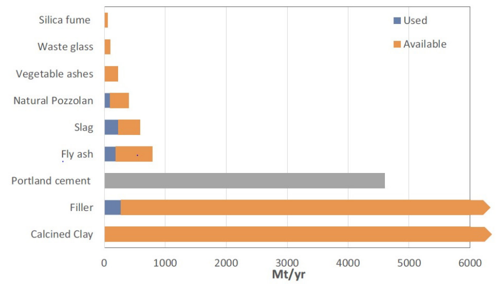

Figure 1.1: Use and estimated availability of possible supplementary cementitious materials and fillers globally in comparison to the amount of cement produced (UN Environment, 2018)

Currently, the South African cement sector produces approximately 13 million tons of cement annually, with an installed capacity to manufacture over 20 million tons per year. The market is projected to grow at a Compound Annual Growth Rate (CAGR) of 2.5% between 2023 and 2028, reaching a value of around 15.5 million tons, driven by increasing urbanisation and infrastructural development (Expert Market Research, 2023). However, in South Africa, fly ash is localised in Mpumalanga, while GGBFS and silica fume are localised to smelters in Gauteng and the North West provinces. Thus, there is a need to find alternative SCMs from local sources that are abundantly and ubiquitously available (Antoni et al., 2012).

Calcined clays have been used as an alternative SCM (Wild et al., 1996; Si-Ahmed et al., 2012). Kaolinitic clays, used to produce calcined clays, are abundantly and ubiquitously available. The deposits of kaolinitic clays are dispersed across various regions in South Africa, including Makana in Eastern Cape, Hammanskraal outside Pretoria, Zebediela in Limpopo, Potchefstroom, Ndwendwe, Kwazulu Natal, Cullinan, and Bronkhorstspruit areas in Mpumalanga and Western Cape and across the rest of Africa, as illustrated in Figure 1.2. This widespread availability of kaolinitic clays makes calcined clays an accessible and viable option for green cement production. In their natural form, kaolinitic clays are valuable materials used for ceramics, and as fillers for paper, paint, polymers and related materials (Rashad, 2013; Shan et al., 2016). However, when calcined under the right conditions, kaolinitic clays convert to calcined clays (Sabir et al., 2001; Rashad, 2013; Krajči et al., 2015; Shan et al., 2016).

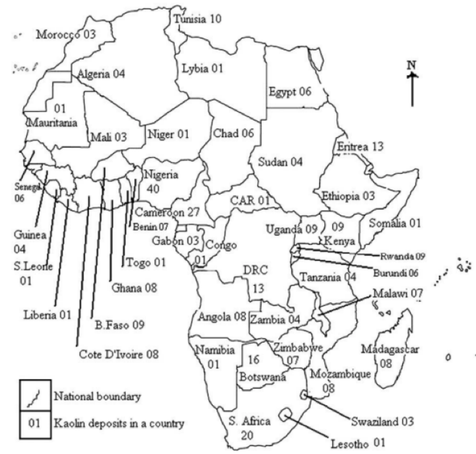

Figure 1.2: Kaolinitic clay deposits in Africa (Ekosse, 2010)

Calcined clays can be used to produce a broad range of products including cement blends, geopolymer binders, shotcrete, pre-cast products etc. The calcined clay-based cement blends have significantly lower carbon footprints, increased durability and higher strength compared to most other commercially available cements (Sabir et al., 2001; Krajči et al., 2015). The calcined clay-based cement blends can be applied in the same manner as ordinary Portland cement.

Despite its immense potential, using calcined clay as a cement supplementary cementitious material has failed to receive industry-wide use due to its high

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market price. Traditional production methods involving rotary kilns, flash calciners and multiple hearth furnaces are capital-intensive and operationally complex. However, researchers have developed a cost-effective process for beneficiating South Africa's huge reserves of kaolinitic clays to produce metakaolin (calcined clay) using a patented coal-fired vertical shaft kiln (VSK). The technology has been demonstrated at a semi-industrial scale (Dumani and Mapiravana, 2017).

In recent years, there has been a growing interest in Limestone Calcined Clay Cement (LC^3) as a viable solution to mitigating carbon dioxide (CO_2) emissions in the cement industry (Scrivener et al., 2018; Sharma et al, 2021). LC^3 represents a new category of ternary blended cements, combining Ordinary Portland Cement (OPC) with calcined clay and limestone (Antoni et al., 2012; Joseph et al., 2023). This approach allows for substantial reductions in CO_2 emissions, making LC^3 a promising alternative to ordinary Portland cement (Malacarne et al., 2021; Rodrigues et al., 2022; Huang, et al., 2023; Barbhuiya et al., 2023; Basavaraj et al., 2023). LC^3 cements permit high levels of clinker substitution, above 50%, with a common composition comprising 50% Portland clinker, 30% calcined clay, 15% limestone, and 5% gypsum (Antoni et al., 2012; Bishnoi et al., 2014; Jaskulski et al., 2020). Commonly referred to as LC^3-50 in literature, this blend has been extensively studied, demonstrating mechanical parameters comparable to OPC just after seven days of hydration, provided the clay contains at least 40% kaolin (Alujas Díaz et al., 2015; Avet et al., 2016; Scrivener et al., 2018; Jaskulski et al., 2020; Sharma et al, 2021, Qian et al., 2023). However, LC^3 encompasses various formulations tailored for specific applications and regulatory requirements (Blouch et al., 2023).

The abundance of raw materials needed for LC^3, namely kaolinitic clays and limestone, which are widely available worldwide, coupled with its superior mechanical and durability properties, positions LC^3 as a sustainable alternative to ordinary Portland cement (Scrivener et al., 2018; Jaskulski et al., 2020; Sharma et al., 2021; Musbau, 2021; Zhou et al., 2022). Calcined clay replaces clinker, significantly reducing emissions, while limestone acts as a filler material. Additionally, LC^3's manufacturing process aligns with existing cement industry practices, requiring no specialized equipment or skills (Bishnoi et al., 2014; Emmanuel et al., 2016; Scrivener et al., 2018).

Industrial trials conducted in Cuba and India have successfully demonstrated that LC^3, with only 50% clinker content, performs similarly to Portland cement, which typically contains over 90% clinker (Bishnoi et al., 2014; Vizcaìino-Andreìs et al., 2015; Emmanuel et al., 2016.) LC^3's environmental friendliness, significantly reduces CO_2 emissions compared to OPC, makes it an attractive choice, particularly in regions where other supplementary cementitious materials are not readily available (Malacarne et al., 2021).

Several life cycle assessment (LCA) studies have verified LC^3's positive environmental impact, confirming reductions of up to and above 30% in CO_2 emissions compared to other commercially available cements (Sánchez Berriel et al., 2016; Cancio Díaz et al., 2017; Scrivener et al., 2018; Gettu et al., 2018; Malacarne et al., 2021; Martinez, Junior et al., 2023; Huang, et al., 2023). Junior et al. (2023) conducted a study to assess the environmental impact of six LC^3 blends prepared from metakaolin and limestone filler in ratios of 2:1, 1.5:1, and 1:1, with 45% and 60% replacement of OPC. OPC and Portland composite cement (PCC) were used as reference binders. The results revealed that LC^3 cements exhibited a reduction in energy consumption of up to 28% and total CO_2 emissions of up to 38% compared to commercial OPC-based cements.

Sánchez Berriel et al. (2016) evaluated and compared the economic and environmental impact of producing three types of cement: traditional Portland cement, commercial blended cement with 15% zeolite content (PPC), and LC^3-50. The results showed that using LC^3 led to a reduction in production costs of around 30% and CO_2 emissions by 40%. The feasibility, environmental benefits, and global scalability of LC^3 position it as a promising supplementary cementitious material (SCM) for partial replacement of traditional cement (Zhang et al., 2020; Malacarne et al., 2021; Rodrigues et al., 2022).

The aim of this chapter was to assess and compare the CO_2 emissions of OPC, binary OPC blended with 30% calcined clay and LC^3-50 utilising the calcined clay produced using the VSK technology. The unique aspect of this research lies in the utilisation of calcined clay produced through a vertical shaft kiln technology, contributing novel insights to the existing body of knowledge.

2. Methodology

A life cycle assessment (LCA) study was conducted to investigate the CO_2 emissions associated with the

production of three types of cement; namely ordinary Portland cement, Portland calcined clay and limestone calcined clay cement (LC^3). Life Cycle Assessment (LCA) is a comprehensive and systematic methodology based on the international standards ISO 14040-44, used to evaluate the environmental impact of a product or process throughout its entire life cycle, from cradle to grave. The primary objective of LCA is to quantify and assess the resources consumed and emissions released at various stages of the product's life cycle, including raw material extraction and processing, manufacturing, transportation, use, maintenance, reuse, recycling, and final disposal. Figure 2.1 illustrates the generic life cycle stages of a construction product for LCA.

In an LCA study, environmental impacts are assessed by considering factors such as energy, land, water, materials, and other resources, as well as various types of emissions to the air, water, and soil. LCA methodology involves a detailed analysis of inputs and outputs, accounting for all relevant environmental factors. This systematic analysis helps identify potential environmental "hotspots" along the life cycle of the product or process. Moreover, LCA is an iterative process, allowing for continuous refinement and improvement.

LCA studies are structured into four mandatory phases (refer to Figure 2.2):

- Goal and scope definition: This phase involves stating the reasons and intended application of the study, defining system boundaries, specifying the functional unit to be used in the investigation, and clearly listing assumptions and limitations.
- Inventory analysis: This phase encompasses data collection and modelling of the product system under study.
- Impact assessment: In this phase, potential impacts associated with the investigated impact categories are calculated. Optional steps include normalization and weighting of results.
- Interpretation: This phase involves presenting and interpreting the results of the study, considering the initial intended goal and scope. The aim is to draw conclusions and make recommendations based on the findings.

Goal

The purpose of the study was to evaluate and compare the CO_2 emissions of producing three types of cement; namely ordinary Portland cement, calcined clay as a

Figure 2.1: Life cycle stages of construction product (Saint-Gobain, 2017)

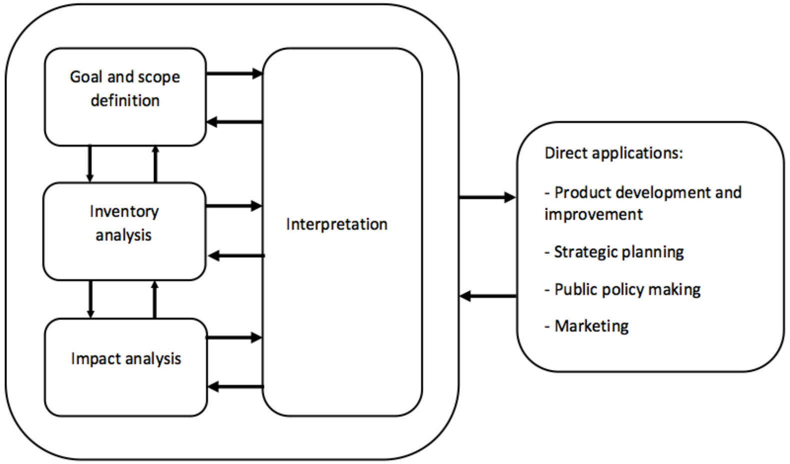


Figure 2.2: Life cycle assessment framework (ISO/SANS:14040,2006)

partial replacement for OPC, and limestone calcined clay cement (LC³). These cements are denoted as OPC, PCC, and LC³, respectively. OPC was also used for the composition of the LC³ cement. The following cements are analysed in this study:

- 100% ordinary Portland cement
- OPC with 30% replacement with calcined clay
- LC³ with a composition of 50% clinker, 30% calcined clay, 15% limestone, and 5% gypsum

Scope

The functional unit of analysis was selected as 1 kg of cement produced. The system boundary defines the scope of the analysis, and in this study, a cradle-to-gate system has been considered, as illustrated in Figure 2.3. This system includes raw material extraction and processing, as well as the transportation of raw materials to the cement production plant, and the actual cement production process, all of which are depicted in Figure 2.4.

Life cycle inventory analysis

The life cycle inventory (LCI) analysis phase involves data collection and modelling. LCI gathers relevant inputs (such as energy and materials) and outputs (such as emissions and wastes) of the product system being studied, which are then scaled to relate to the functional unit. The inventory for all processes in the cradle-to-gate life cycle of the cements was prepared qualitatively first and then quantitatively.

Data sources

The LCA software tool SimaPro 8.1, along with Ecoinvent Database version 3, was utilised to compile the Life Cycle Inventory (LCI) dataset for each material in this study. Table 3.2 provides an overview of the materials and transportation inventory considered for each material. South African datasets were used whenever they were available. In instances where no South African dataset was available for a specific material, the Rest of the World (RoW) dataset was chosen as a proxy and modified to align with the local context.

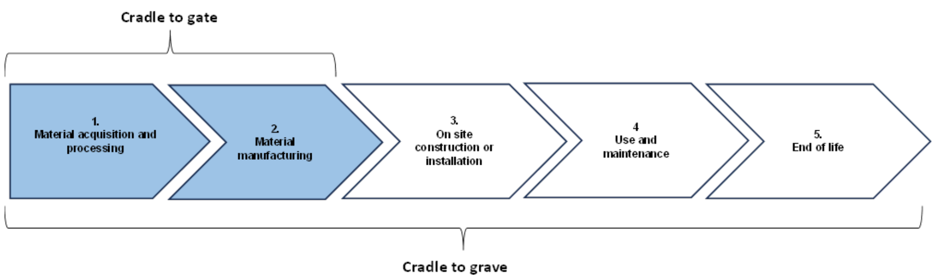
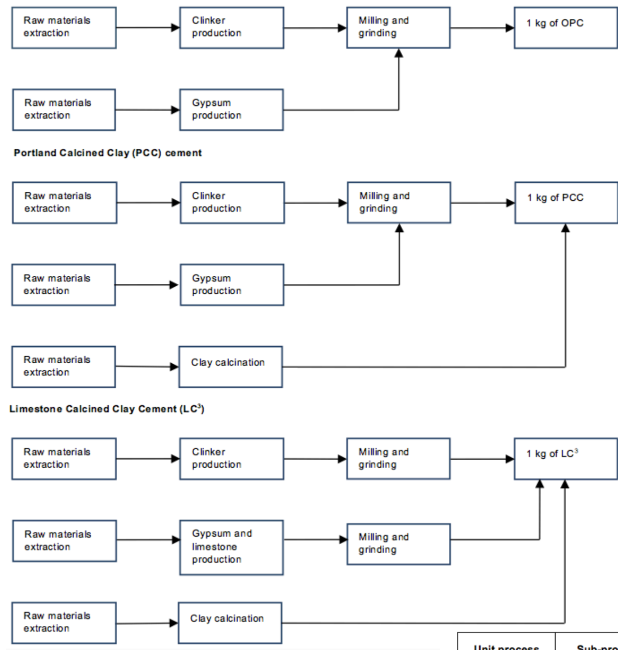


Figure 2.3: Generic life cycle stages of construction product



Caption: Figure 2.4: System boundaries of this study

Ordinary Portland cement 52.5 N and limestone were sourced from local suppliers and transported to the site. The kaolinitic clay was obtained from a local mining company and transported to a supplier for crushing. The crushed clay was then calcined using a vertical shaft kiln on a semi-industrial scale. Subsequently, a company milled the calcined clay using a ball mill. The milled calcined clay, along with OPC and limestone, was utilized to produce the PCC and LC³ cements on-site.

The energy needed for the complete calcination of the kaolinitic clay was determined through Thermogravimetric Analysis (TGA) and Differential Scanning Calorimetry (DSC). The process took into account a vertical shaft kiln with a 23% fuel efficiency, and the total mass loss during calcination, obtained as 13% based on TGA results. This method aligns with the approach utilised by Pillai et al. 2019 and Malacarne et al. 2021 to calculate energy for complete calcination of clay.

Life cycle impact assessment

In this phase, the potential environmental impacts are calculated based on the inventory. This study considered only climate change, that is, global warming potential. The ReCiPe midpoint (H) methodology, included with the LCA software, was utilized to generate and report the results in CO_2 equivalents.

Unit process	Sub-process	Assumptions	Source
Ordinary Portland cement	Manufacturing	Cement, Portland {RoW) production| Alloc Def, U, used as proxy	Ecoinvent 3 database
	Transporting	Road distance = 74 km Transport = lorry, 16-32t	Ecoinvent 3 database
Calcined Clay	Mining and extraction	Kaolin {RoW} | production | Alloc Def, U used as a proxy dataset kaolin mining and extraction	Ecoinvent 3 database
	Crushing	Limestone, crushed, for mill {RoW} | production | Alloc Def, U used as proxy	Ecoinvent 3 database
	Transporting	Road distance = 62 km Transport = lorry, 16-32t	Ecoinvent 3 database
	Calcining	0.0475 KWh/kg clay Fuel efficiency of the vertical shaft kiln is 23%	TGA and DSC
	Transporting	Road distance = 94 km Transport = lorry, 16-32t	Ecoinvent 3 database
	Milling	Quicklime, milled, loose {RoW} | production | Alloc Def, U used as proxy	Ecoinvent 3 database
	Transporting	Road distance = 79 km Transport = lorry, 16-32t	Ecoinvent 3 database
Limestone	Manufacturing	Limestone, crushed, for mill {RoW} | production | Alloc Def, U used as proxy	Ecoinvent 3 database
	Transporting	Road distance = 50 km Transport = lorry, 16-32t	Ecoinvent 3 database

Table 2.1: Life cycle inventory data sources and assumptions used in the study for the 3 different types of cement.

Results and Discussion

The percentage contributions of total CO_2 emissions arising from different processes and constituents associated with the three types of cement; namely, ordinary Portland cement (OPC), OPC with 30% replacement with calcined clay (PCC), and limestone calcined clay cement (LC^3), are depicted in Figure 3.1. As expected, the results indicate that the clinker production stage emerges as the environmental 'hotspot' in all the cements. OPC has the highest impact, while LC^3 has the least impact due to their highest and lowest clinker contents, respectively. Ordinary Portland cement (CEM I) contains 95-100% clinker and 0-5% minor additional constituents (SANS 50197-1) while LC^3, in this study, contains 50% clinker. Several studies, including those by Huntzinger and Eatmon (2009), Chen et al. (2010), Pillai et al. (2019), and Ige and Olanrewaju (2023), have reported the same findings, highlighting that clinker production is the main contributor to CO_2 emissions in cement.

The analysis also indicates that calcined clay production process significantly contributes to PCC and LC^3 cements. However, the calcination process of clay requires a lower temperature of 600–800 °C (Sabir et al., 2001; Krajči et al., 2015; Shan et al., 2016) compared to 1450 °C for OPC production (Ige and Olanrewaju, 2023), resulting in lower environmental impacts.

The contribution to climate change, expressed in CO_2 equivalents for each of the cements being studied, is represented in Table 3.1. The results indicate that LC^3 has a significantly lower CO_2 impact compared to OPC and PPC, with CO2 emissions from LC^3 measured at 0.668 kg CO_2eq/kg of cement. Similar values have been reported in studies by Cancio Díaz (2017), Gettu et al. (2019), Malacarne et al. (2021), and Junior et al. (2023).

Cement	kg CO_2eq/kg of cement
OPC	1.01
PCC	0.835
LC^3	0.668

Table 3.1: CO2 emissions for each of the cement evaluated in the study.

The comparison of CO_2 emissions for OPC, blended cement PCC, and LC^3 is illustrated in Figure 3.2. OPC exhibits the highest CO_2 emissions, approximately 34% higher than LC^3 cement. These findings align with those reported by Malacarne et al. (2021), who observed reductions of up to 38% in CO_2 emissions

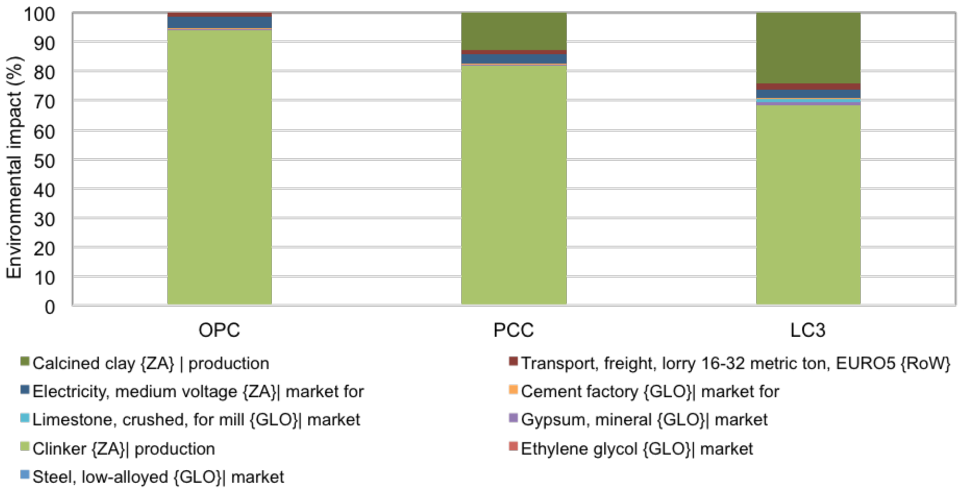


Figure 3.1: Climate change contribution analysis arising from different processes associated with the three types of cement in the study.

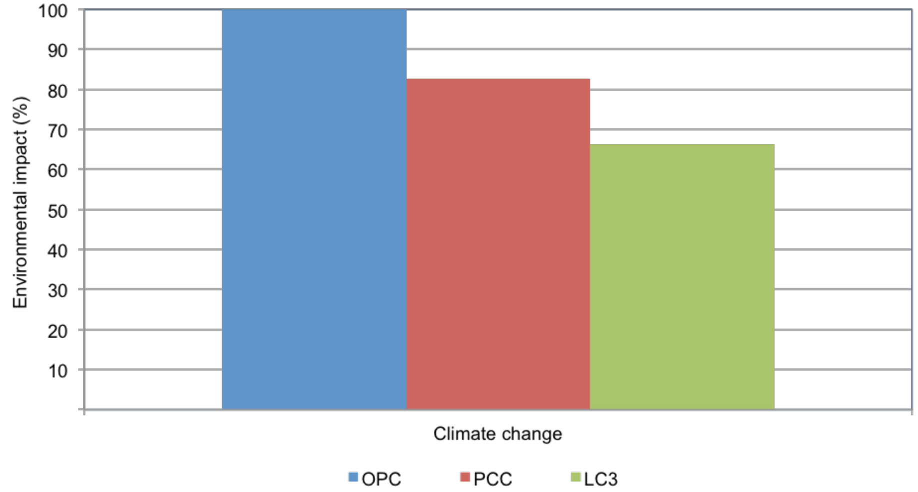


Figure 3.2: Relative CO2 emissions of cement production.

compared to OPC, and Sanchez Berriel, who reported up to a 30% reduction in CO_2 emissions.

Conclusions

This study quantified and compared the CO_2 emissions associated with the production of ordinary Portland cement (OPC), OPC with 30% calcined replacement, and LC^3, comprising 50% clinker, 30% calcined clay, 15% limestone, and 5% gypsum. The results demonstrated that LC^3 exhibited significantly lower CO_2 emissions in comparison to both OPC and binary OPC with 30% calcined clay replacement. LC^3 displayed a substantial reduction in CO_2 emissions, up to 34% less than OPC and OPC with 30% calcined replacement. These findings emphasize the significant potential of LC^3 as an alternative ordinary Portland cement to meet the increasing demand for cement with a low carbon footprint in the cement industry.

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