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Life cycle assessment of synthetic natural gas production from captured cement's CO_2 and green H_2

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ABSTRACT

Research on the environmental benefits of carbon capture and utilization (CCU) in cement production so far, has predominantly emphasized energy efficiency enhancements and CO_2 emission reductions at a CCU product level, neglecting broader environmental consequences for the sector. This research broadens this perspective by providing an extensive life cycle assessment (LCA) of a circular Portland cement (CPC) model. Synthesized methane is used as input fuel through green hydrogen and calcium-looping (CaL) post-combustion captured CO_2 from cement flue gas. Comparative analysis with ordinary Portland cement (OPC) reveals significant reductions in climate change and fossil resource use environmental impact categories. However, trade-offs are evident in acidification, water use, and minerals and metals resource consumption. The electrolysis system is a critical contributor due to the high electricity demand for hydrogen production, and its environmental impact depends largely on the renewable electricity source. The wind-based electrolysis model yields the most favourable results, followed by mixed (50% solar – 50% wind) and solar scenarios. These findings offer valuable insights for the cement industry, supporting stakeholders decision making on the adoption of sustainable circular production methods.

1. Introduction

The cement manufacturing industry is acknowledged as a significant contributor to carbon dioxide emissions, exacerbating climate change. As nations set their carbon neutrality targets for 2050, as requested in the EU Green Deal, it becomes imperative to investigate sustainable measures for mitigating the environmental impact of energy-intensive industries, including the cement sector. Identifying innovative solutions to minimise this impact is both a priority and a challenge. Carbon capture, utilization and storage (CCUS) has emerged as a promising solution to reduce CO_2 emissions from cement production [1].

Among various CCUS technologies, post-combustion calcium-looping (CaL) CO_2 capture has garnered considerable attention due to its suitability in the cement sector. Cement plants benefit from well-established limestone handling infrastructure, simplifying CaL integration into existing operations. Moreover, using resulting CaO-based sorbents in cement production holds potential for enhancing circularity and resource efficiency [2,3]. A substantial drawback emerges with the conventional use of coal as a fuel source in CaL systems, which significantly exacerbates CO_2 emissions, counteracting its intended environmental benefits [2].

Captured CO_2 can be used as a feedstock for fuel synthesis when combined with hydrogen. This process is central to the "Power-to-Gas" (P2G) pathway, which specifically focuses on utilizing electrolysis for

Abbreviations: ATIC, Technical Association for the Portuguese Cement Industry; BAT, Best Available Technique; BAU, Business-As-Usual; CaL, Calcium-looping; CCS, Carbon Capture and Storage; CCU, Carbon Capture and Utilization; CCUS, Carbon Capture, Utilization and Storage; CPC, Circular Portland Cement; EPD, Environmental Product Declaration; GLO[1], Global; LCA, Life Cycle Assessment; LCI, Life Cycle Inventory; LCIA, Life Cycle Impact Assessment; OPC, Ordinary Portland Cement; P2G, Power-To-Gas; PPA, Power-Purchase Agreement; PV, Photovoltaic; RER[1], Europe; RES, Renewable Energy Source; SNG, Synthetic Natural Gas; SOEC, Solid Oxide Electrolysis Cells; TRL, Technology Readiness Level, [1] SimaPro software region codes.

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hydrogen production [4]. Water electrolysis is strategically chosen due to its potential to harness renewable energy sources, such as wind and solar power, facilitating the production of green hydrogen. This approach differs from traditional hydrogen production methods that mainly depend on fossil fuels, which contribute to carbon emissions [5]. A noteworthy strategy involves the application of power-purchase agreements (PPAs), which enable the use of renewable electricity without physical proximity constraints between the electrolysis plant and renewable energy sources (RES) [6].

Green hydrogen production, however, poses some challenges. As the demand for hydrogen rises, the inefficiencies in its production from RES become more pronounced, especially on an industrial scale where electricity consumption increases accordingly [6,7]. Furthermore, renewable energy technologies, despite their benefits, carry their own environmental impacts, encompassing raw materials extraction, land use and greenhouse gas (GHG) emissions throughout their life cycles. Consequently, a life cycle assessment (LCA) is imperative in sustainability research, for a quantitative evaluation of the environmental footprint of diverse technologies [8].

Traditionally, cement industry research on CCUS has prioritized economic analysis, a critical factor influencing the adoption of carbon capture technologies. While environmental benefits are the motivation for decision-makers to evaluate these technologies, previous investigations have primarily emphasised improvements in energy efficiency and CO_2 emission reductions, neglecting a comprehensive assessment of the wider environmental impacts [9–13]. Furthermore, many research efforts concentrate solely on the capture phase, frequently employing amine absorption, without conducting a holistic evaluation of subsequent CO_2 pathways: carbon capture and utilization (CCU) or carbon capture and storage (CCS). Notably, studies exploring these aspects have predominantly favoured CCS, with limited attention to CCU. Those examining CO_2 utilization have typically compared the environmental impact of CCU products, like synthetic natural gas (SNG), against conventional counterparts (e.g., natural gas), primarily at the product level, sidestepping an evaluation of the direct technological integration's industry-wide implications [9,14,15].

This paper aims to address these research gaps through a comprehensive comparative LCA study of cement production. Its primary focus lies in contrasting the environmental impact of traditional cement production methods against a novel circular CCU model, which integrates P2G technologies. Additionally, a thorough analysis of the use of alternative RES, namely wind and PV energy, along with their combined application to enhance energy security, is conducted.

2. Methodological framework

The methodological framework adhered to specific LCA guidelines tailored for CO_2 utilisation, which are more restrictive than the general ISO requirements. This approach aimed to enhance transparency and foster comparability among LCA studies [16,17]. An attributional modelling approach was employed, encompassing four intertwined phases: goal and scope definition, life cycle inventory (LCI), life cycle impact assessment (LCIA) and interpretation.

During the goal and scope definition phase, the research's foundation, comprising its objectives, scenarios, boundaries, and declared unit of analysis, underwent comprehensive evaluation. In the LCI phase, both primary and secondary data were used as inputs to establish mass and energy balances, ensuring data accuracy and completeness. Moving into the LCIA phase, the material flows from the inventory were translated into impact categories using the specialised software Simapro 9.5, enabling the assessment of potential environmental burdens associated with each modelled scenario. Finally, in the interpretation phase, the extent to which the circular model could mitigate the environmental impacts tied to conventional cement production was quantified.

However, all phases of the LCA are intrinsically interconnected. Modifications made in one phase affect subsequent phases; for instance, initial data availability was not entirely known at the outset of the LCA. Therefore, an iterative approach was systematically applied and implemented.

2.1. Goal and scope

The primary objective of this LCA is to conduct a rigorous quantitative analysis of the environmental impact associated with the production of a Circular Portland Cement (CPC) proposed model, contrasting it with Ordinary Portland Cement (OPC). Moreover, within the CPC model, a comparative assessment of the use of alternative RES, specifically wind and solar PV, along with their combined application is performed. In parallel, a hot-spot analysis is executed to pinpoint the systems displaying the most pronounced environmental impacts. Through this comprehensive evaluation, the research aims to provide insights into the environmental performance of the proposed CPC production model and its renewable energy source, while concurrently identifying critical contributors to the overall environmental footprint compared to the current OPC model.

Cement serves as an intermediary product with an undetermined end-use. Consequently, a mass-based declared unit of 1 metric tonne was chosen. Fig. 1 illustrates the elements and cradle-to-gate system boundaries for both OPC and CPC simplified production models.

The OPC serves as the reference for the business-as-usual (BAU) cement production model. In the circular cement model, CO_2 emissions from the cement production unit are captured using CaL post-combustion technology. This captured CO_2 is then combined with H_2 produced through water electrolysis driven by RES, to obtain SNG rich in methane. This SNG acts as a fuel source for both cement production and the CaL plant, replacing conventional fuels.

Circularity is further enhanced by using the oxygen by-product from the electrolysis process to supply the CaL unit. This substitution removes the need for the conventional energy-intensive air separation unit (ASU) in the CaL unit. A critical assumption underlying this model is the complete transfer of all emissions, typical of industrial-scale cement plants, to the CO_2 capture unit. It is noteworthy that this assumption significantly influences mass and energy balances and subsequent LCIA.

2.2. Life cycle inventory

LCI involves a systematic inventory of the input and output of energy and material flows during the life cycle. It was constructed by integrating primary data with secondary data drawn from stoichiometric calculations and literature sources. These sources encompass process simulations involving mass and energy balance analyses and the utilization of datasets aligned with the closest resembling technology/process/material available in the ecoinvent database version 3.9.1 [18]. The models analysed were built as a function of the relevant systems.

The LCI was framed in the Portuguese cement sector context, which displays a notable degree of homogeneity, even though it consists of multiple plants from two different companies. This uniformity arises from the widespread adoption of a consistent production method across all units. Although there are variations in the specificities of production, such as capacity, equipment, quarry mineral attributes, and auxiliary material consumption, the sector shares fundamental technological characteristics. Primary data applied in this research were derived from all six cement plants operating in mainland Portugal.

Given the confidential nature of individual cement plant data, a meticulous anonymization process was imperative. To facilitate this, automated spreadsheets were customized for each company, enabling them to securely share their data with a neutral third-party entity, specifically the technical association for the Portuguese cement industry (ATIC). Upon receiving the data from all six factories, ATIC merged the information into a consolidation worksheet. Subsequently, the anonymized dataset was generated, combining inputs and outputs obtained from the six plants to form a comprehensive and representative model

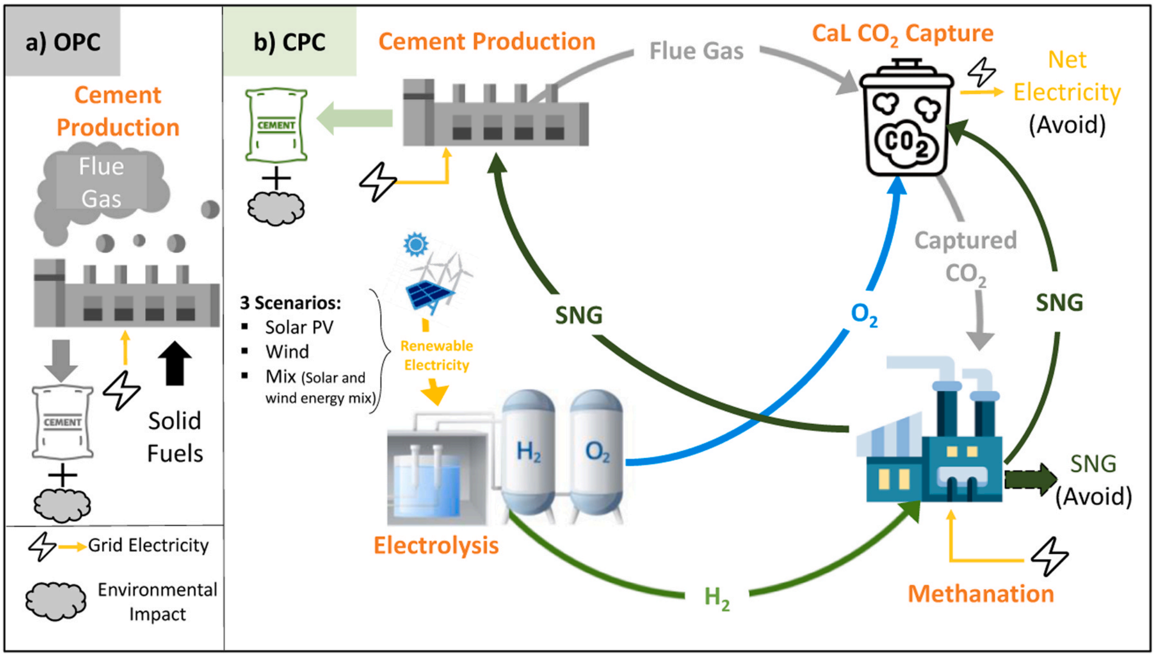

Fig. 1. System elements and boundaries for cement models: (a) OPC; (b) CPC.

for the sector.

In line with strict confidentiality regulations, only data that is at least two years old from the current analysis year can be legally used. Given that the core project commenced in 2020, the most recent legally usable dataset available pertains to the year 2018. Expert evaluation from the industrial partners indicates that the sector has undergone no perceptible changes during this interval, with no notable technological innovations introduced within the past four years. Consequently, the

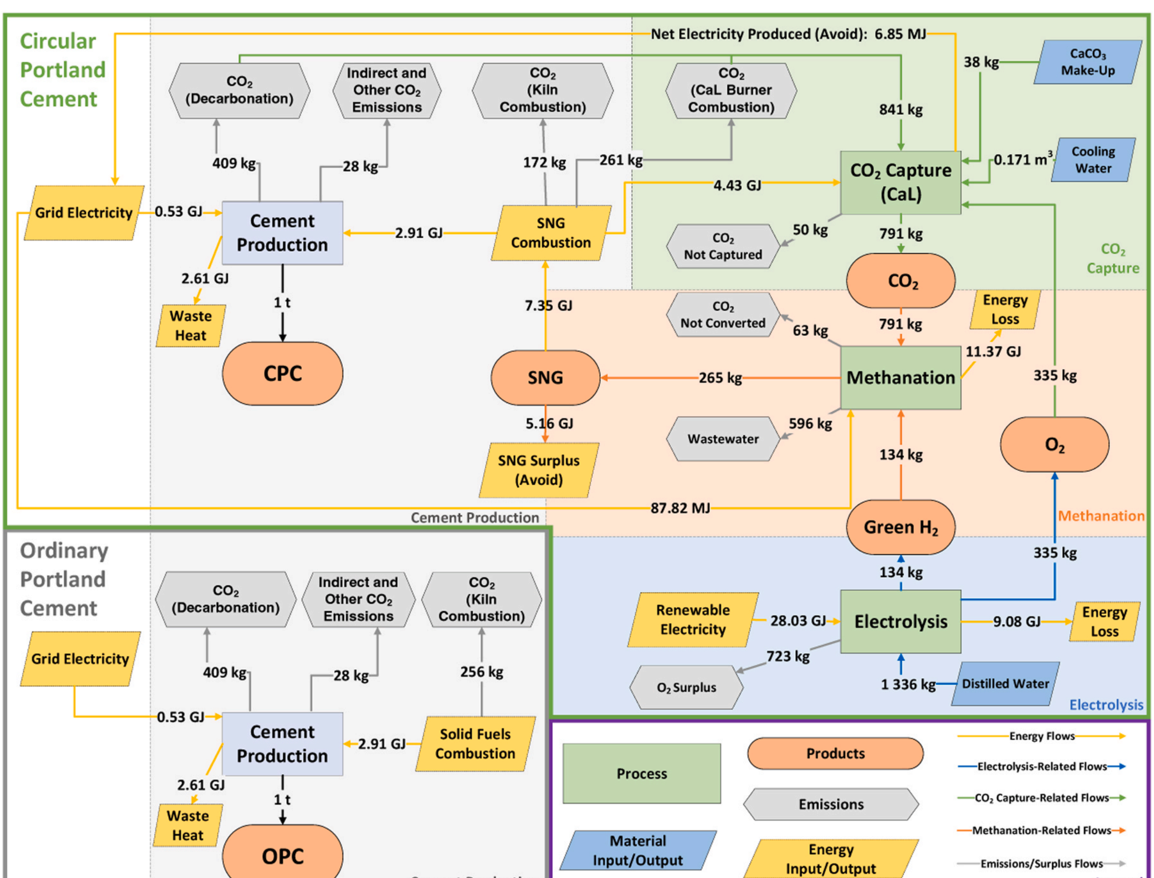

Fig. 2. Mass and energy balances for each model.

datasets specific to the year 2018 are deemed suitable for modelling the BAU Portland cement manufacturing process.

In situations where data specific to Portugal was unavailable, European average data (e.g., “RER” or “Europe Without Switzerland” in Ecoinvent) was employed. When European average data was also inaccessible, a proxy region would be used, such as “Switzerland” was utilized. If all else failed, global average (GLO) data was the fallback option, but the situations where these applied were marginal and do not significantly affect the results.

The grid electricity generation mix in Portugal was applied to both models, following data from [19]. Comprehensive inventory data, including inputs, outputs, and specific assumptions for each system, can be found in the Supplementary Information. Fig. 2 provides a streamlined overview of the mass and energy balances for each model.

2.2.1. Ordinary Portland cement

The OPC production, represented in the “Cement Production” block of Fig. 1, comprises a three-stage process: raw materials preparation, clinker production and grinding with additional components to produce cement. Initially, primary and secondary raw materials are blended to create a homogeneous powder. Naturally occurring calcareous deposits, such as limestone and marl, supply the calcium carbonate required for cement production. Supplementary materials, such as sand, shale and clay are also extracted to supplement silica, alumina and iron oxide to the raw mix. Quarrying heavy-duty machines extract these materials, which are subsequently crushed and transported to the cement plant [4,20,21]. As a foreground process, the internal quarries activities were modelled in higher detail to include the extraction procedure. Processes powered by grid electricity do not generate on-site (direct) emissions, however, they are considered as sources of indirect emissions.

Subsequently, the material undergoes grinding and enters a rotary kiln, passing through a pre-heater and pre-calciner. Fuels are employed to gradually heat the material until calcination occurs at approximately 900 ºC, releasing CO_2 from calcium carbonate ($CaCO_3$). At temperatures reaching up to 1450 ºC, clinker forms as CaO reacts and agglomerates with silica, alumina, and iron oxide [4,20,21]. Using data directly sourced from producers, a clinker-to-cement ratio of 0.779 was assumed, resulting in the calcination reaction contributing to 61 % of the total direct CO_2 emissions in cement manufacturing, releasing approximately 409 $kgCO_2/t_{cement}$. These emissions are then combined with those arising from fuel combustion, responsible for 256 $kgCO_2/t_{cement}$, constituting 39 % of the total direct emissions. Given the essential role of calcination in clinker production, a significant portion of these emissions is unavoidable.

A mix of different fuels was considered, guided by authentic data supplied by manufacturers. Petcoke is the most common fuel used in Portugal, although alternative fuels, including used tires and wood waste, are extensively employed. On average, the combination of residue-derived fuels (RDF) and biomass contribute to approximately 41 % of the calorific input.

Ultimately, the hot clinker is cooled rapidly using a grate cooler and is temporarily stored between production and grinding stages. Clinker is subsequently mixed with mineral additives, such as gypsum or fly ash, to produce blended cement. The final product is homogenized, stored in silos, and later dispatched to customers for transportation [4].

The Portuguese cement industry employs three transportation methods: road, maritime and rail. The freight transport of fuels and both externally-sourced natural and secondary raw materials to the plant gates to produce one declared unit was included in the model.

2.2.2. Circular Cement

In the cement circular model, there are two by-products produced besides cement: the SNG and electricity. Allocation is a method used to determine how the environmental burden of multiple products should be allocated. However, the ISO 14044 guidelines recommend minimizing the use of allocation by either dividing the unit process or expand the product system. In SimaPro, the "avoided products" method was employed to expand the system. This involves subtracting the impacts of the products avoided (as they are provided in the system considered) from the total impacts. System expansion operates on the principle that if the primary system generates, for example, surplus electricity that is fed back to the grid, it reduces the need to produce electricity at an alternative power plant [22].

In the current research, the environmental burden is entirely attributed to the cement product, while the electricity and SNG surplus produced are considered avoided products. Consequently, the cement product receives an environmental credit for avoiding this impact, while also bearing the additional emissions associated with the CO_2 capture, electrolyser and methanation units.

The CPC model encompasses four interconnected systems forming a closed loop: "Cement Production" and three P2G systems, namely “CaL CO_2 Capture”, “Electrolysis” and “Methanation”. This “Cement Production” system has the same energy requirements of the OPC production, including electricity for grinding and loading equipment, as well as fuels for heating in the pre-calciner and rotary kiln. The system was therefore similarly simulated in SimaPro. Notably, while the OPC model considered the utilization of the solid fuel mix employed in the Portuguese cement industry, the CPC model integrates the SNG generated from the "Methanation" unit.

To capture the CO_2 emitted from the flue gas during clinker production at an industrial scale, post-combustion CaL technology is applied. It relies on the reversible carbonation reaction, employing two interconnected circulating fluidized bed reactors, namely the carbonator and the calciner. Initially designed in the software “Aspen Plus” for a hypothetical cement plant conforming to the European Best Available Technique (BAT) standard for cement manufacturing [23], this CaL configuration was subsequently tailored to suit a specific Portuguese cement plant [24] and modelled in SimaPro accordingly in the present paper. The flue gases are sent to a carbonator, where CO_2 reacts with the CaO-based sorbent and forms $CaCO_3$ at around 650ºC. The calcium carbonate is transferred to the calciner, where oxy-combustion of SNG is carried out to reach a temperature above the calcination equilibrium temperature (950ºC). Note that the combustion process uses a mixture of flue gas (mainly CO_2 and H_2O) and pure oxygen, instead of air, to reduce nitrogen content. This saves energy for the calcination process, either lowering flue gas volume or increasing kiln capacity. Additionally, the temperature is controlled by mixing the oxidant with the combustion gases, resulting mainly in CO_2 and H_2O. After water condensation, a highly concentrated CO_2 stream is extracted [25].

These processes are aggregated in the “CaL CO_2 Capture” system, which generates a significant amount of thermal power by calciner combustion that is recovered as high-temperature waste heat for electricity production [23,24,26,27]. Note that the SNG fuel used is produced in the “Methanation” unit while the required oxygen is obtained directly through the water electrolysis instead of the ASU.

The potential application of the spent sorbent purge from the CaL system as a substitute for limestone in clinker production has been investigated. The feasibility of this use largely depends on the purge composition, particularly its sulphur ($CaSO_4$) content as excessive sulphur levels are known to compromise clinker quality. Consequently, the scope for utilizing a purge with high sulphur content is constrained [2]. Notably, the present study deliberately excluded the potential advantages of substituting limestone with the purge in the cement production process, thereby ensuring a conservative estimation.

Alkaline water electrolysis (AEL) technology was selected due to it suitability for large- applications [12,28–30]. The mass and energy balances were based on a model created in the HOMER open-source software by [12]. The “Electrolysis” system utilizes two electrodes immersed in an alkaline solution, separated by a diaphragm, to facilitate water splitting into H_2 and O_2 through the application of an electric current. The system therefore relies on distilled water and renewable energy as the main inputs to produce green hydrogen for SNG production and oxygen for CaL CO_2 capture [28,31,32]. Three scenarios for renewable energy production were considered: solar, wind and a combination of both solar and wind energy.

The "Methanation" system uses captured CO_2 from the "CaL CO_2 Capture" system, green H_2 from the "Electrolysis" system and electricity to produce the SNG main output through CO_2 hydrogenation [12,33]. The SNG produced is recirculated for clinker production and to the "CaL CO_2 Capture". Mass and energy balances relied on the catalytic process for SNG production through CO_2 hydrogenation developed in Aspen Plus by [12].

The "CaL CO_2 capture" and "Methanation" infrastructures were incorporated in the SimaPro model through proxy processes. The "Electrolysis" system infrastructure included both the AEL electrolyser infrastructure inventory from [34,35], as detailed in the Supplementary Information, and the associated renewable energy technology infrastructure, utilizing generic SimaPro processes modelled for Portugal.

The circular model faces limitations due to varying technology readiness levels (TRL). The methanation plant operates at TRL 6 [36,37], the CaL CO_2 Capture technology at TRL 7 [38], and the AEL electrolyser at TRL 8 [39,40]. Key assumptions are outlined in Table 1, and comprehensive inventory data, emissions, detailed assumptions, and calculations can be found in the Supplementary Information.

3. LCIA: Results and discussion

The impact assessment was performed in Simapro software employing the "EN15804 + A2 Method", an adaptation of the European Commission's "Environmental Footprint 3" method customized for Environmental Product Declarations (EPD) of construction products. Core environmental impact indicators were selected in accordance with EN 15804:A2 requirements [46]. Detailed scenario results can be found in Table 2.

The comparison of environmental impacts between the OPC and CPC models reveals notable disparities across various categories. In alignment with the cement industry's expectation to have this technology contributing to mitigate its climate change impact, the circular model exhibits promising outcomes, achieving a reduction of up to 513 kg CO_2-eq in the most favourable scenario (wind-based electrolysis). These reductions range from 17 % to 68 % across different scenarios, as shown in Fig. 3.

The analysis of the climate change components show an overwhelming dominant contribution from fossil CO_2, exceeding 98 % across all scenarios. Notably, it is important to clarify that the carbon originating from calcination reactions, although of mineral origin, is classified as "fossil" for LCIA purposes. Contributions from biogenic sources and land use are found negligible in all models. In the CPC models, the decrease in biogenic contribution is attributed to the exclusion of biomass fuels, whereas the increase in land use impact is linked to the substantial land requirements of RES, including both PV and wind energy. Nonetheless, these variations within the circular models are insignificant compared to the substantial reduction observed in the fossil category.

The reduction in climate change impact was expected due to the implementation of tail-end CaL technology, which captures a significant proportion of the CO_2 emitted, and its subsequent use to replace the current fuel mix. This mix presently comprises a notable fraction of fossil fuels (59 %) and alternative fuels (41 %), attributing to the fossil and biogenic specific categories, respectively. This technology captures both CO_2 from the flue gas of clinker production and all CO_2 formed in the calciner by fuel combustion, as shown in Fig. 4.

Consequently, there is a significant disparity in direct net CO_2 emissions (excluding indirect emissions from electricity) between the two systems, as depicted in Fig. 5.

The environmental impacts obtained from "EN 15804+A2 method" were subsequently normalised to the reference impact per person of EU-28, employing the "Environmental Footprint 3.1" normalisation set, as indicated in Fig. 6. Note that the ODP category is not included as it presented a normalised score close to zero across scenarios.

The comparative analysis shows that circular cement production from the proposed layout diminishes environmental impacts in the climate change and fossils resource use categories compared to the reference system. However, CPC exhibits unfavourable environmental outcomes particularly in minerals and metals resource use but also in terms of acidification and water use, in comparison to OPC. The remaining environmental categories are scenario-dependent.

The normalised environmental indicators show varying relative impact between the CPC and OPC models. The circular model notably manifests "minerals and metals resource use" and "eutrophication freshwater" as the most influential impact categories, whereas "climate change" and "fossil resource use" are more prominent in the OPC baseline scenario.

Eutrophication initiates with increases in nutrient loading into ecosystems, often constrained by nitrogen or phosphorus availability. The increased availability of these nutrients has stimulated primary production, resulting in adverse consequences, such as algal toxin accumulation and issues with taste and odour in drinking water [47]. The wind-based circular scenario exhibits a slightly more favourable marine eutrophication performance, while other circular scenarios perform worse than the BAU. Additionally, OPC exhibits a lower freshwater and terrestrial eutrophication than the circular model. This disparity can be attributed to the raw materials extraction process associated with renewable energy technologies. This offsets the benefits of CPC models, where captured CO_2 is converted into SNG for reintroduction as clinker kiln fuel, replacing solid fuels, including petcoke (a petroleum refining by-product). In contrast, the BAU scenario's reliance on petcoke production contributes significantly to various environmental impacts, such as marine eutrophication. However, it is crucial to note that this trade-off is advantageous only in the context of the marine eutrophication performance of the wind-based CPC scenario.

The results revealed statistically significant differences in environmental impacts among the circular models, emphasising the role of the

Table 1
LCI base assumptions.

System	Assumption	Value	Reference
General	Operational lifetime [years]	25	[3,41]
Cement Production	Operating hours [hours/year]	8 000	[13]
	Average annual clinker production capacity [t/year]	884,947	Primary data provided by ATIC
	Clinker/Cement Ratio [$t_{clinker}$/t_{cement}]	0.779	
	Direct CO_2 emissions [tCO_2/$t_{clinker}$]	0.823	
CaL CO_2 Capture	CaL CO_2 Capture Ratio	0.94	[13]
	Fuel Required [GJ/$t_{clinker}$]	5.690	[2]
	Net Surplus Electricity Production [MW]	0.270	
	Cooling Water Required [m^3/$kg_{CO2\ Captured}$]	2.16E-04	
	Oxygen Required [t/$t_{clinker}$]	0.430	
	Circulating CaO to CO_2 in the flue gas molar ratio (F_0/FCO_2)	0.02	
Electrolysis	AEL: H_2O to H_2 Mass Ratio	10.00	[42]
	Electricity for Electrolysis (AEL) [kWh/t_{H2}]	58,270	[43]
	Energy Efficiency	0.68	
	Wind-to-Solar Electricity Ratio (WtS)	0	Solar Scenario
		0.5	Mix Scenario
		1	Wind Scenario
Methanation	CO_2 to CH_4 Conversion Efficiency	0.92	[12]
	Electricity for Methanation [kWh/GJ_{SNG}]	1.95	[44]
	Energy Efficiency	0.53	[45]
	CO_2 Emissions/GJ_{SNG} [tCO_2/GJ]	0.0589	[18]

Table 2
Environmental impacts of each scenario.

Impact category			OPC	CPC		
Main	Specific	Unit	BAU	Solar	Mix	Wind
Climate Change	Total	kg $CO_{2\ eq}$	7.54E+02	6.26E+02	4.33E+02	2.41E+02
	Biogenic		1.23E+01	2.11E+00	1.34E+00	5.64E-01
	Fossil		7.41E+02	6.22E+02	4.30E+02	2.39E+02
	Land use and LU change		8.36E-01	2.17E+00	1.71E+00	1.26E+00
Eutrophication	Marine	kg N_{eq}	2.05E-02	3.13E-01	2.13E-01	1.14E-01
	Freshwater	kg P_{eq}	4.31E-01	6.57E-01	4.49E-01	2.41E-01
	Terrestrial	mol N_{eq}	1.84E+00	6.86E+00	4.58E+00	2.31E+00
Other	Acidification (AP)	mol H+ eq	7.07E-01	3.51E+00	2.24E+00	9.73E-01
	Ozone depletion (ODP)	kg $CFC_{11\ eq}$	5.04E-06	4.02E-05	2.34E-05	6.70E-06
	Photochemical ozone formation (POCP)	kg $NMVOC_{eq}$	1.59E+00	2.42E+00	1.63E+00	8.37E-01
	Water use (WDP)	m^3 depriv.	6.22E+01	6.15E+02	4.00E+02	1.86E+02
Resource use	Fossils	MJ	3.24E+03	2.05E+03	-2.84E+02	-2.62E+03
	Minerals and metals	kg Sb_{eq}	1.71E-04	2.09E-02	1.22E-02	3.44E-03

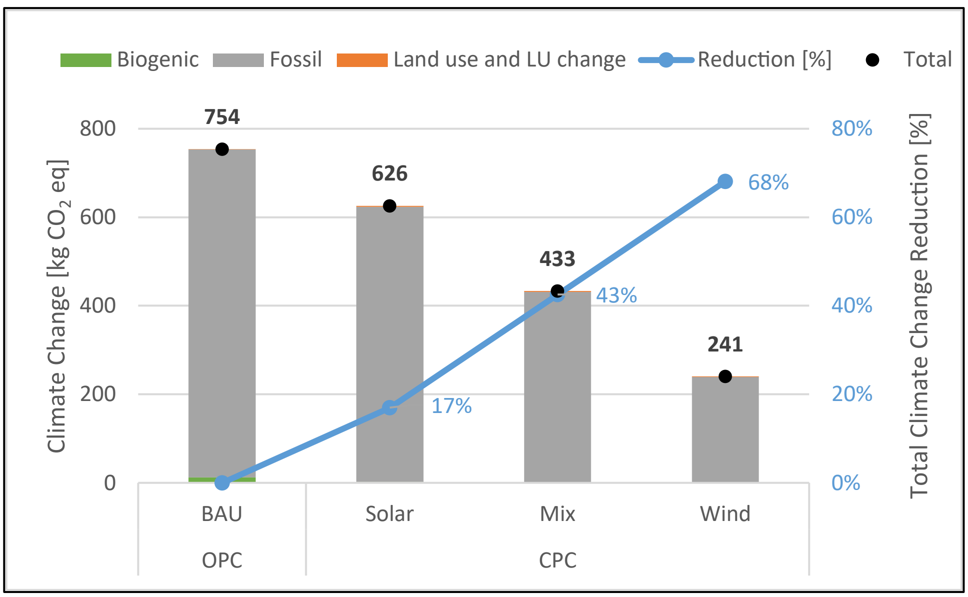

Fig. 3. Climate change indicators [kg CO_2 eq] and reduced impact in the circular models [%].

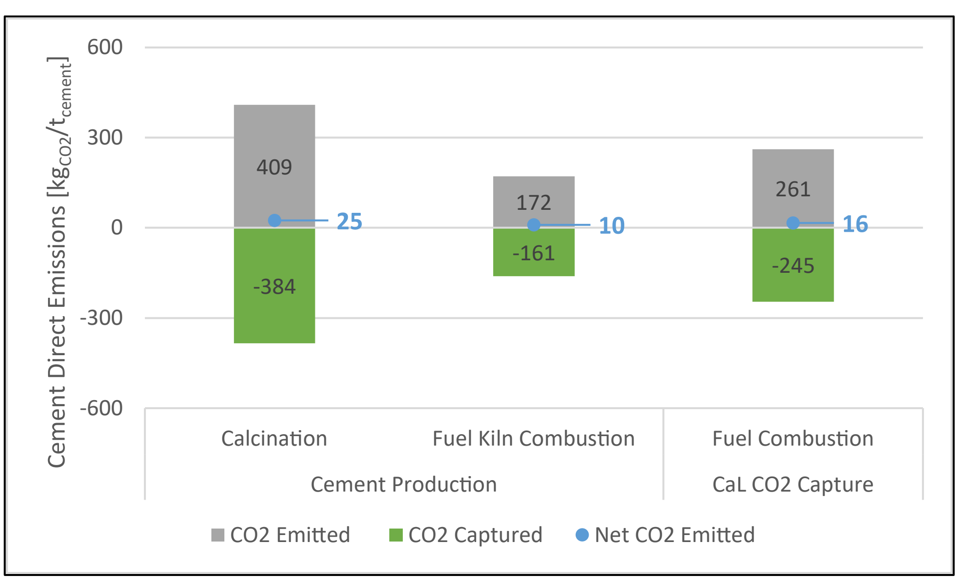

Fig. 4. CPC direct CO_2 emissions produced and captured [$kgCO_2/t_{cement}$].

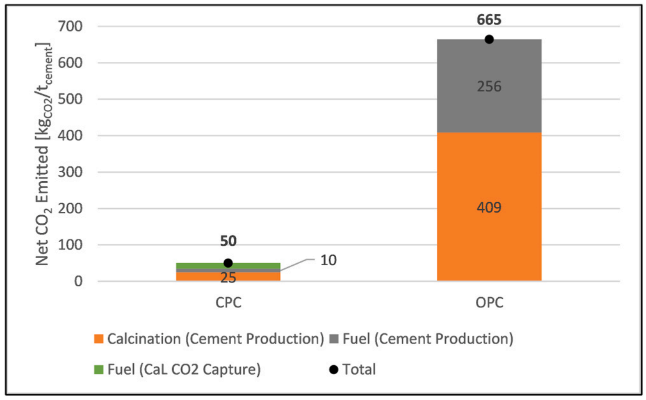

Fig. 5. Net CO_2 emitted in the CPC and OPC models.

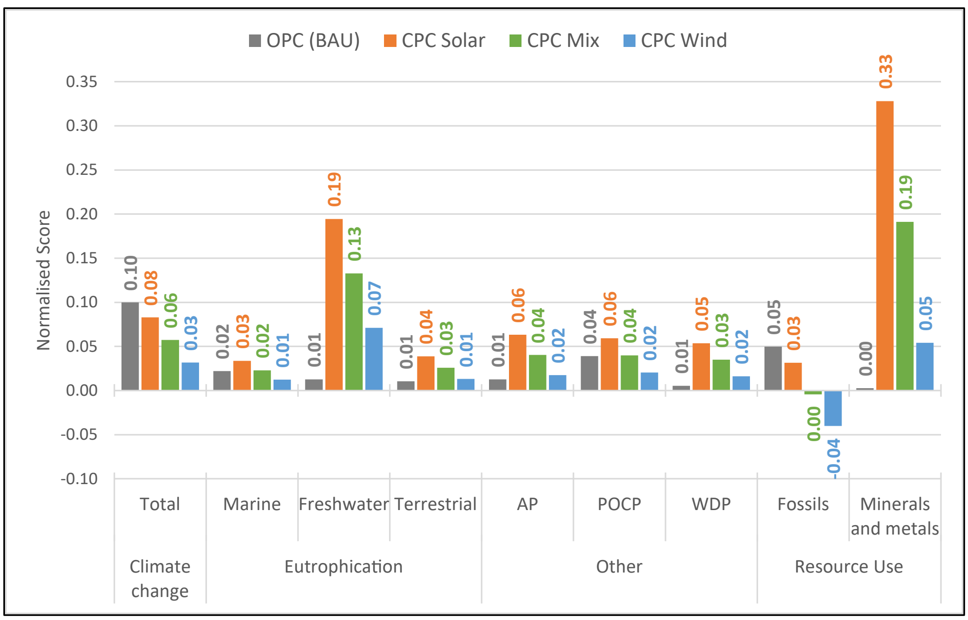

Fig. 6. EF 3.1 normalised results.

RES in the environmental performance. The wind energy based scenario exhibited the most favourable environmental outcome, followed by the mixed and finally the solar scenario. These results are supported by existing literature, including a recent cradle-to-grave LCA study affirming wind turbines' lower environmental impact compared to solar panels [48].

Fig. 7 illustrates a hot-spot analysis for the three CPC scenarios, highlighting environmental impacts mainly originate from electrolysis-related processes. Across categories, except for "climate change" and "fossil resource use" in the wind scenario, electrolysis-related processes consistently contributed from 63 % to 98 %. These findings align with a previous analysis of a similar design applied to a coal-fired power plant, which showed that the environmental footprint of electrolysis could reach 100 % [49].

The significant impact of electrolysis is attributed to its high electricity demand for producing the required amount of H_2 to convert CO_2 emitted at an industrial scale into SNG. Moreover, the adverse environmental impacts arising from oxygen production required for the CaL process have been allocated to the electrolysis category instead of the calcium looping, due to the absence of an energy-intensive ASU. Additionally, in contrast to the methanation, CaL CO_2 capture and electrolyser infrastructures, renewable energy infrastructure holds considerable influence on the environmental performance of the CPC models.

In particular, the solar PV infrastructure exhibits a relatively high climate change impact. In fact, in the solar-based CPC model, the PV farm infrastructure contributes around 79 % of the total climate change impact, which corresponds to an emission of 494 kg CO_2-eq per tonne of CPC produced. This result is in contrast to the public perception of solar energy as carbon neutral, as the infrastructure production results in substantial indirect carbon emissions.

The methanation system used in the circular models significantly reduces the use of fossil resources compared to the BAU. This is due to the fact that, it not only avoids the consumption of solid fossil fuels but also prevents the extraction of natural gas from nature, considering the excess production of SNG, resulting in a "negative" impact for this category.

Wind turbines require specific resources, although in lower impacts associated when compared to solar panels. Notably, wind turbines rely on materials such as steel, fiberglass, and iron and rare earth materials such as neodymium whereas solar panels require silicon, aluminium, and glass. The latter materials used in solar panels need more processing, which consequently demands higher energy input [50,51]. As such, the "minerals and metals resource use" environmental indicator in

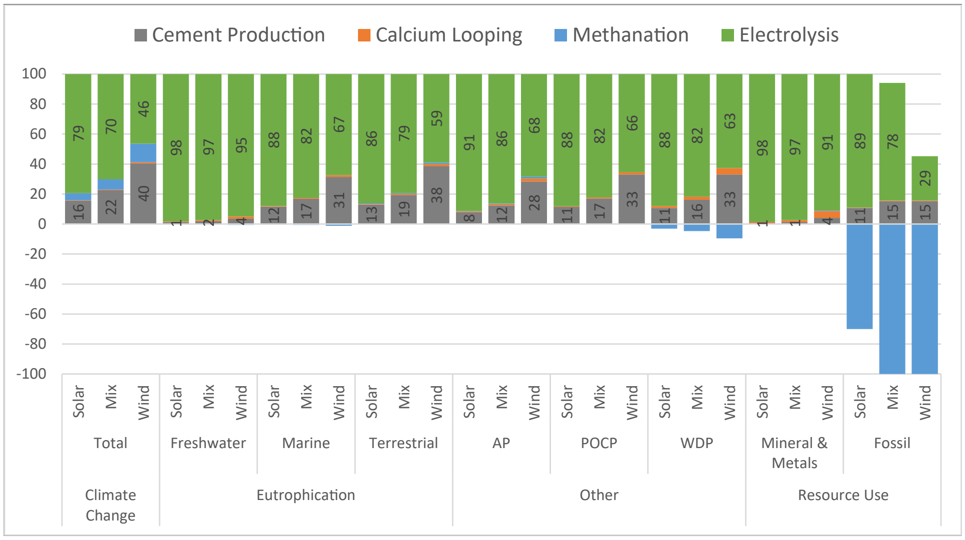

Fig. 7. Hot-spot analysis.

circular models, particularly in the solar scenario, is less favourable in comparison to the OPC model.

The increased water use in the circular models was expected since water is the main source of hydrogen for electrolysis. Furthermore, both solar and wind energy systems consume water during their manufacturing processes. However, operational water use for wind turbines is negligible as they require little maintenance and cleaning. Solar panels also have minimal operational water usage, primarily for periodic cleaning, but involve water-intensive procedures during production, such as cooling and rinsing, increasing the water demand compared to wind turbine production, as highlighted in a previous study [52].

Ozone depletion results consistently demonstrate a low impact across all models and scenarios. However, certain manufacturing stages for solar PV modules may contribute slightly more to ozone depletion compared to wind energy systems.

The raw materials used to produce electrolysers, solar panels and wind turbines have an impact on acidification and photochemical ozone formation (POCP) categories. As a result, the current cement model performs better. Nonetheless, the POCP environmental impact is lower in the wind CPC model compared to OPC. The reason for this is again the decommissioning of solid fuels, in particular petcoke, which has a significant impact on the above categories.

When assessing the environmental impact of CPC in comparison to the OPC production model, it is important to consider the wider environmental context. A narrow focus on climate change or fossil fuel consumption would favour CPC, with significant reductions in CO_2 emissions. However, models for CPC suggest increased water use and varying effects on eutrophication, acidification and minerals and metals resource use. These trade-offs must be weighted for a comprehensive evaluation. Additionally, renewable energy infrastructure has a crucial role in shaping environmental outcomes. Notably, electrolysis-related processes, particularly those tied to renewable energy infrastructure, significantly influence CPC's overall impact. It is imperative to comprehend these primary contributors for the environmental footprint of the circular cement production proposed. This study highlights the complexity involved in shifting towards circular cement production and the importance of considering diverse environmental aspects.

3.1. Uncertainty analysis

The LCA methodology has limitations, particularly regarding uncertainty. This is most apparent during the inventory data phase, where the absence of standardized and reliable data, as well as potential errors in data gathering, can result in an inventory that does not accurately represent reality. The values of certain inventory parameters can often vary significantly instead of remaining constant, which can significantly impact the accuracy and trustworthiness of LCA study outcomes. The Monte Carlo method is a numerical analysis technique based on probability and statistical theory. It uses random (or pseudo-random) numbers to solve various computational problems, providing a way to deal with the uncertainties present in LCA research [53,54].

The study employed a Monte Carlo simulation, with 20,000 iterations and a 95 % confidence level to assess the uncertainties in the LCA comparisons between the wind-based CPC and OPC models. Fig. 8 shows that in most environmental impact categories, one model outperforms the other, with over 90 % of the Monte Carlo runs significantly favoring one model. This significant percentage indicates a clear difference, suggesting minimal uncertainty in these comparisons. The notation "CPC Wind < OPC" indicates when the wind-based CPC model has a lower environmental impact than the OPC model. However, it is important to note that two categories, terrestrial eutrophication and water use in particular, have higher levels of uncertainty. The comparison indicates low uncertainty overall, but further investigation is needed to fully understand the environmental implications of choosing between the two models, particularly in the specific areas mentioned.

3.2. Sensitivity analysis

A sensitivity analysis was conducted to investigate how to minimize the environmental impact of the selected wind-based CPC model by optimizing key parameters within the electrolysis stage. This analysis builds upon the prior hot-spot analysis, which identified electrolysis as the primary contributor to environmental burdens, responsible for 63–98 % of the impact across various categories.

Due to its significant influence, the sensitivity analysis focuses on two critical LCI parameters that are specific to the electrolysis stage: water consumption and energy efficiency. Water consumption refers to the amount of water required by the electrolyser to produce hydrogen, measured in tonnes of water per tonne of hydrogen (t_{H2O}/t_{H2}). Energy efficiency represents the electricity demand of the electrolysis process, expressed as a percentage.

Three scenarios were modelled for each parameter: pessimistic (with increased water consumption/lower efficiency), medium (baseline), and optimistic (with decreased water consumption/higher efficiency). The specific values used for each scenario are presented in Table 3.

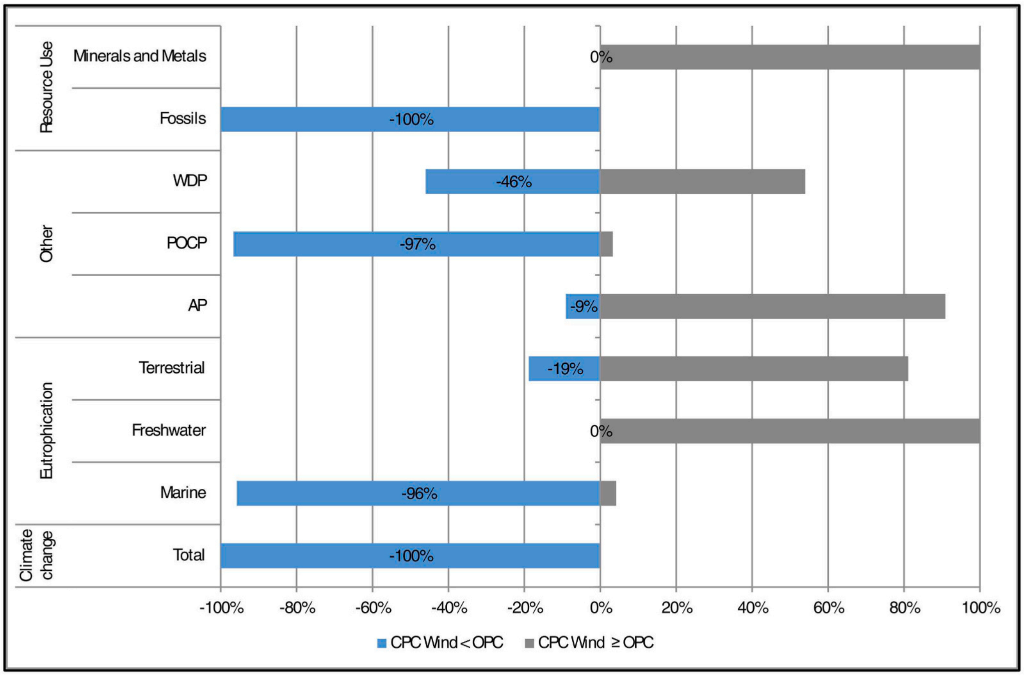

Fig. 8. Monte-Carlo simulation results of characterized LCIA comparison between wind-based CPC model and OPC.

The sensitivity analysis employed in this study is known as one-at-a-time (OAAT) or perturbation analysis. In OAAT, the sensitivity of the model output to a specific input parameter is determined by the ratio between the variation in the model results and of the studied parameter itself [55].

The results of the sensitivity analysis are presented in Fig. 9. Fig. 9a focuses on water consumption, while Fig. 9b explores the impact of energy efficiency. The *y*-axis quantifies the change in environmental impact, with negative values indicating a decrease in impact and positive values indicating an increase. The *x*-axis shows different environmental factors potentially affected by the LCI parameters.

The sensitivity analysis revealed that the environmental impact of the wind-based CPC model is highly sensitive to the electrolyser's energy efficiency across all impact categories. While variations in water consumption also affect the model's environmental impact, this effect is mainly observed within water-related categories. It is worth noting that reducing the electrolyser's water requirements can significantly decrease water usage and its associated burdens on freshwater and marine ecosystems. This analysis underscores the critical role of optimizing energy efficiency in electrolysers for minimizing the overall environmental footprint of the CPC model, with additional benefits attainable through reduced water consumption.

The analysis confirms that reduced water consumption generally results in a lower environmental impact across various categories, particularly within the "water use" impact category. This category specifically considers the volume of water used, taking into account regional water scarcity. Here, changing the water ratio by 7 % (red column) resulted in a 3.9 % increase in water use impact. Conversely, a 9 % decrease in the water ratio (green column) led to a 4.8 % decrease in the same impact category. The analysis also shows that water-related eutrophication impacts, affecting both freshwater and marine environments, are sensitive to changes in the water ratio to varying degrees. Freshwater eutrophication is more sensitive, which reflects the trend in water use. In contrast, marine eutrophication is less sensitive, with only a 1 % difference observed between the pessimistic and optimistic scenarios compared to the baseline.

The sensitivity analysis supports the hypothesis that energy efficiency has a significant impact on environmental performance in all categories. The model is highly sensitive to this critical parameter of the electrolysis process. The results show a clear trend, where improved energy efficiency leads to a lower environmental impact across various factors, highlighting the effect of electric energy consumption on the environmental profile of the CPC model. A decrease in electrolyser efficiency results in higher energy consumption to produce the same amount of hydrogen. This leads to an increased environmental impact across all impact categories. Specifically, a 2 % reduction in energy efficiency worsens the environmental impacts by 1–3 % depending on the category, compared to the baseline.

Conversely, improving the efficiency of electrolysers can significantly reduce their environmental impact. A 5 % increase in energy efficiency can lead to improvements ranging from 1 % to 7 % across various impact categories. This is particularly true for minerals and metals resource, where a 7 % improvement can be achieved due to lower reliance on renewable energy infrastructure, followed by acidification and photochemical ozone depletion, which can be improved by 5 %. For categories such as Climate Change or Eutrophication, a 5 % increase in energy efficiency results in an approximately 4 % reduction in environmental impact. However, the Water Use category shows the least sensitivity to energy efficiency improvements, with only a marginal 1 % enhancement noted. This is mainly due to the minimal impact of wind energy used in the selected CPC scenario on water consumption metrics.

Table 3
Analysed LCI parameters. NA=Not applicable.

Scenario LCI Parameter	Unit	Pessimistic	Medium	Optimistic
Water Ratio	[t_{H2O}/t_{H2}]	10.70	10.0	9.1
	[Variation %]	+7%	NA	-9%
Efficiency	[%]	66	68	73
	[Variation %]	-2%	NA	+5%

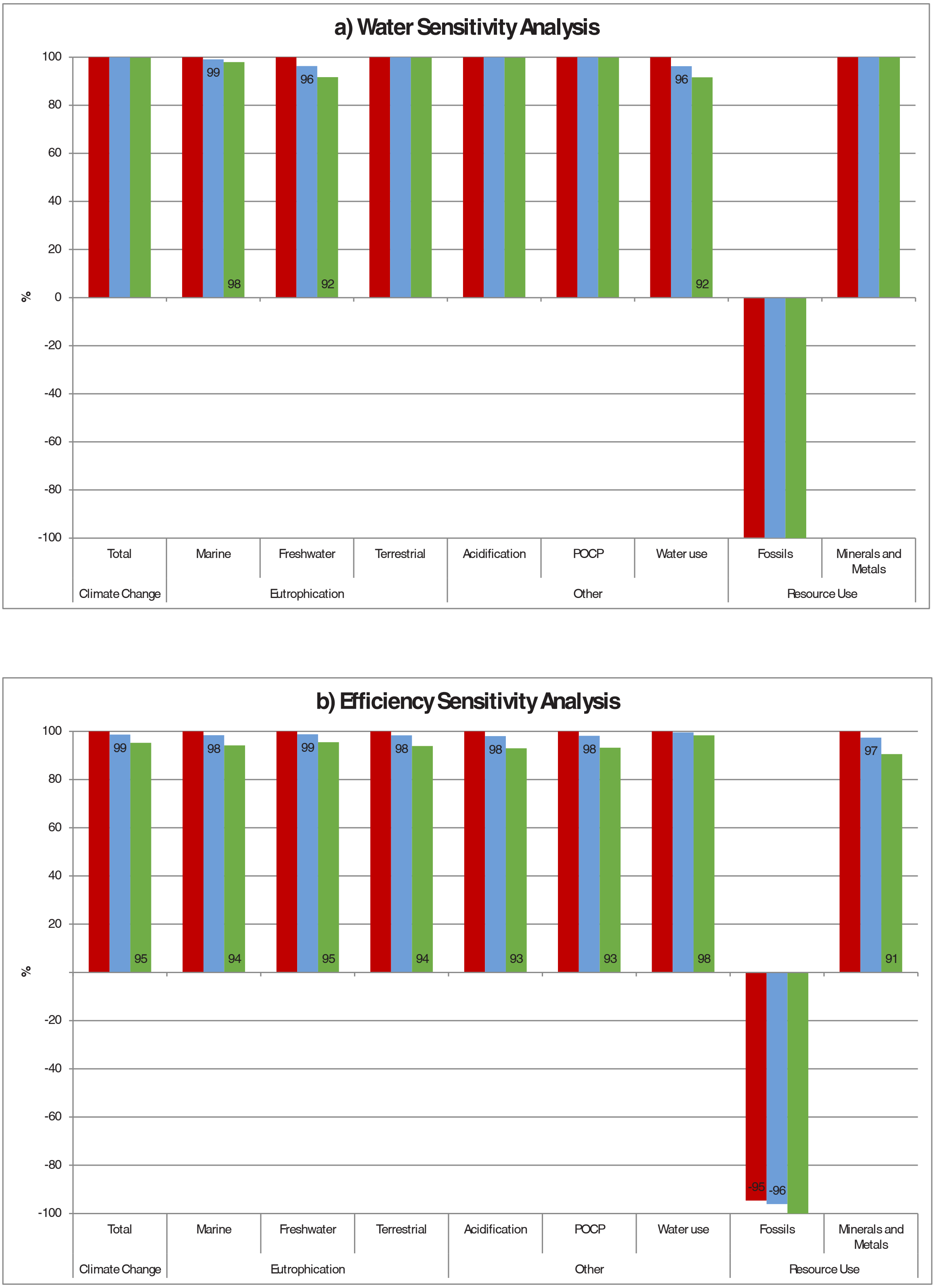

Fig. 9. Sensitivity analysis of water ratio (a) and energy efficiency (b) impact on environmental factors. Red denotes pessimistic, blue medium, and green optimistic scenarios. Only significant changes (different from ± 100 %) are shown.

4. Conclusion

The cement manufacturing sector's substantial carbon dioxide emissions have soured a critical need for mitigation strategies in the face of climate change. In response, a comprehensive life cycle assessment (LCA) was conducted to assess the environmental impact of a traditional ordinary Portland cement (OPC) model, when compared to a circular Portland cement (CPC) proposed model, incorporating CO_2 capture, green hydrogen production from various renewable energy sources (RES) and the production of synthetic natural gas (SNG).

This methodology provides significant advances on current carbon capture and utilization (CCU) research, which typically focuses on comparing the CO_2 emissions of CCU products like SNG to conventional counterparts such as natural gas. In contrast, this paper adopts a broader industry-wide perspective, assessing the environmental implications resulting from the direct technological integration of these innovative processes. It compares the environmental impact of producing one cement tonne across different models.

The assessment adhered to the "EN15804 + A2 Method", tailored for construction product environmental product declarations. The results revealed that when focusing solely on climate change and fossil resource use, CPC models demonstrate significant environmental advantages. The reductions in CO_2 emissions, driven by carbon capture and fuel substitution, present a promising pathway towards more sustainable cement production. This aligns with the primary objective of assessing CPC's viability in reducing its environmental footprint.

However, CPC models showed increased water use and diverse impacts in categories like eutrophication, minerals and metals resource use, and acidification. These trade-offs emphasize the need for a holistic approach when evaluating the environmental sustainability of CPC, considering various environmental dimensions.

Furthermore, this research highlighted the pivotal role played by the renewable energy infrastructure, often overlooked in RES's sustainability assessments. Electrolysis-related processes linked to renewable energy sources played a dominant role shaping CPC's overall environmental impact. Considering the 13 indicators used for the environmental impact assessment, wind-based CPC model displayed the most favourable outcomes, followed by the mixed and solar scenarios.

This research provides valuable insights for stakeholders within the cement industry as they navigate the challenges and opportunities in adopting the proposed CPC production model. However, it is vital to acknowledge that the proposed model considers the complete transfer of CO_2 emissions from cement plants, at an industrial-scale, to the CO_2 capture unit.

This research provides a critical tool for advancing the environmental sustainability of the cement industry, for full scale deployment. In fact, it conducts a thorough assessment of the circular cement production model proposed, its associated renewable energy sources, and offers a comparative analysis with the current OPC model. It also highlights key factors contributing to the overall environmental impact of the proposed model, supporting an informed decision-making process towards a more sustainable cement production future.

In essence, the transition to circular cement production is a complex endeavor with multifaceted environmental implications. While there are clear advantages in terms of reducing CO_2 emissions and fossil resource use, it is essential to weigh these benefits against potential trade-offs in other environmental categories. Recognizing electrolysis as the key system contributor is vital for understanding and optimizing the proposed CPC production's environmental footprint.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: M. Bacatelo reports a relationship with c^5Lab that includes: employment. F. Capucha reports a relationship with c^5Lab that includes: employment. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

The authors do not have permission to share data.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jcou.2024.102774.

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