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ABSTRACT

Cement production needs to reduce its contribution to climate change urgently. This industry is, however, difficult to electrify and decarbonize and is yet expected to increase its output to accommodate the need of an increasing population. CCS and, more recently, CCU have been presented as promising solutions to align with the Paris Agreement objectives. Given the lack of application of these technologies at scale in cement production, it remains dubious under which conditions they can deliver the expected reductions. In this study, we answer this question by developing a prospective Life Cycle Assesment model for an existing cement plant in Denmark. Results show that the extensive use of biomass in the fuel mix combined with decarbonized electricity are necessary conditions for CCS and CCUS. CCUS could allow the cement plant to mitigate up to 106% of its operational emissions and deliver up to 90′000 tons of synthetic kerosene annually to Denmark in 2050. Considering direct and indirect CO_2-eq. emissions, such savings would bring the cement to near climate-neutrality. A variation test was run to illustrate how the results change when assuming different values for relevant parameters.

1. Introduction

The Paris Agreement sets a framework for balancing anthropogenic emissions of greenhouse gases-GHG at the source and removing residual emissions to reach net zero by the second half of this century. This current ambition represents a significant challenge for carbon- and energy-intensive industries that are difficult to decarbonize and electrify, such as cement production. This process generates CO_2 emissions from both the avoidable combustion of fuel and the unavoidable calcination reaction (also referred to as "process CO_2"), which accounts for around 50% of the emissions (Vilella and Arribas, 2016). In this context, the cement industry is exploring solutions to reduce the sector's carbon emissions, such as energy savings, Carbon Capture and Storage-CCS, and alternative fuels and materials (Benhelal et al., 2013). Among these, CCS has an abatement potential of 36–42% (GCCA, 2021). CCS entails the capture and sequestration of gas in geological formations or depleted oil and gas fields. Carbon Capture and Utilization-CCU entails the transformation of captured CO_2 into a valuable product, and CCUS simultaneously stores and uses CO_2 (IEA, 2020a). In the case of cement production, several capture technologies are considered suitable today. Among them are chemical absorption, membranes, oxyfuel, and cryogenic separation. The most mature technology for CO_2 capture is the post-combustion chemical absorption process using Monoethanolamine-MEA as a solvent (Bhadola et al., 2020; Kepler Cheuvreux, 2021; UNECE, 2021). This method has a typical capture efficiency of 90% in commercial plants; it can produce 99% purity CO_2 and be retrofitted to the existing cement kilns (Hoenig et al., 2007). A drawback of this technology is that it needs large amounts of Low-Pressure Steam-LPS to separate the CO_2 from the amine, making it energy-intensive. In addition, the compression and liquefaction of CO_2 and other downstream activities, such as producing methanol from recycled CO_2, consume substantial amounts of electricity. This is referred to as the "energy penalty" associated with CCUS, and it is one of the biggest challenges for its implementation (García-Gusano et al., 2015; Giordano et al., 2018; Saunier et al., 2019). All CCUS technologies have an energy and environmental footprint that, if neglected, may significantly overestimate the emission reductions they expect to achieve. Therefore, life cycle GHG emissions associated with CCUS should be quantified before implementing these technologies.

Several studies perform the Life Cycle Assessments-LCA for CCUS

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systems, mainly considering applications in the power sector (Cruz et al., 2021), while only a few consider the cement industry: García-Gusano et al. (2015) assess only the capture process, while Meunier et al. (2020) evaluate CCU with conversion to methanol, Volkart et al. (2013) & An et al. (2019) address CCS with storage in saline aquifers and Cavalett et al. (2022) in depleted oil and gas fields. None of the studies assess the entire CCUS value chain, and addressing the implementation of CCS and CCU simultaneously strengthens the understanding of these options that are often presented as opposites but are both necessary for reducing carbon emissions (Cuéllar-Franca and Azapagic, 2015). Aside from the most recently published article by Cavalett et al. (2022), all previous studies analyze the impacts of a capture facility at newly-built cement plants or assume Best Available Technology-BAT hypothetical plants. However, the potential of CCUS to help reach climate-neutral cement production depends on the specific operational parameters under which the cement plants are operated. In addition, the post-capture downstream activities, such as transport, storage, and conversion, are geographically and context-dependent. Hence, understanding the effects of implementing CCUS in this industry requires plant-specific assessments of different value chains (IEA, 2018).

Moreover, these studies report a wide range of GHG reductions, namely, 15% (García-Gusano et al., 2015), 39–78% (Volkart et al., 2013), 57–62% (An et al., 2019), 25% (Meunier et al., 2020) and 74–91% (Cavalett et al., 2022). Such variation in results indicates that implementing these technologies does not consistently deliver the goal of CO_2-neutral operation for cement production. The uncertainty around CCUS performance is high, especially considering that it will be implemented in the incoming years as its application to cement production is still emerging. The insufficient understanding of the environmental impact across the complete CCS and CCUS value chains hinders informed decision-making toward progressing with the adoption of these technologies.

The decarbonization pathways proposed by the Paris Agreement should lead to profound changes in the power sector, thereby affecting the future carbon footprint of energy-intensive activities. Consequently, the potential carbon emission reductions achievable via CCUS differs depending on whether the system is modelled under current or future conditions regarding the energy mix carbon intensity. Prospective LCA has been proposed to address and incorporate such system changes to better assess emerging technologies' performances (Thonemann et al., 2020). Among various features, this approach entails including future technological and efficiency developments in the LCA background system. This type of LCA uses Integrated Assessment Modelling-IAM that produces a stylized representation of the climate, land, water, energy, and industrial systems under various scenarios to reflect worldwide changes in different economic sectors (Schwanitz, 2013). The prospective approach improves the temporal consistency when modelling emerging technologies that will be implemented in the future (Mendoza Beltran et al., 2020). In this regard, only two studies include elements of temporal consistency by including changes over time in the relevant energy mix in their model (Cavalett et al., 2022; Volkart et al., 2013). Since CCS and CCUS are emerging technologies, applying a prospective assessment approach to model these technologies under future conditions is relevant.

The current study adopts a more critical approach than existing ones, as it identifies the conditions under which carbon capture technology can help mitigate the climate impacts of cement production. Therefore, it challenges the assumption that CO_2 reductions are reached inherently by implementing CCS or CCUS. Also, unlike most published studies, a comprehensive consequential and prospective modelling framework is used to quantify the future environmental performance of CCUS technology across multiple climate scenarios. Its mitigation efficiency in the cement industry might depend on how fast other sectors decarbonize (i.e., transport, electricity).

This study presents the LCA of cement clinker production following the implementation of CCS or CCUS by 2050 in an actual cement plant in Denmark. The modelling includes all the activities of the value chain: capture, liquefaction, transport, permanent storage, and conversion of the biogenic fraction to methanol for e-kerosene production. A post-combustion system with amines is used as a capture technology. Also, a power-to-methanol plant followed by a conversion to e-kerosene is assumed for the conversion path. We address the energy penalty by (1) assuming the energy requirements and solvent make-up of Shell's Cansolv, a solvent with a lower energy requirement than the traditional MEA, (2) considering energy integration between the cement and the capture plant, and (3) exploring three different options for generating the needed LPS: electrical boiler, gas boiler, and heat pump. To improve the robustness of the results and account for uncertainty and variability, moving beyond the static approaches used in previous studies, a wide range of potential scenarios are calculated, and the pre-conditions in which the system reached climate neutrality are described. This allows us to identify which other measures need to be taken together with CCUS to deliver the required GHG emission savings. In this study, carbon neutrality is reached when the total GHG emissions of the process under assessment are equal to zero, and it does not consider compensation for the historical cement plant emissions.

2. Methods

2.1. Case study: cement production and flue gas characteristics

The study is based on the case of Aalborg Portland, a Danish cement plant that currently produces approximately 4500 tons of grey clinker per day. This production line is fed with liquid chalk slurry from a quarry and features a preheater system that enables a semi-dry process. As a result, a substantial amount of excess heat is generated, which is presently being recovered and supplied to the district heating network. This heat could potentially be utilized in a capture unit if implemented. The flue gas from the kilns has an average temperature, pressure, and flow rate of 122 °C, 1.004 atm, and 411 Nm^3/h, respectively; the average CO_2 concentration is around 21%Vol. While these characteristics may vary in a future scenario, they were used as base parameters for the capture plant concept study, cf. SI.1 for more details. The carbon emissions from this production line are around 1.3 $MtCO_2/y$, of which 10% is biogenic due to the utilization of combustible waste with a high caloric capacity, such as Refuse-Derived Fuel (RDF), meat and bone, sewage sludge, paper sludge, and wood fiber boards, hereafter referred to as alternative fuels-AF (GCCA, 2021). Among these, RDF and paper sludge contain a fossil fraction. This facility aims to increase the use of AF with a higher biogenic content to mitigate emissions of GHG. Table 1 shows the fuel mix in 2020 (taken here as a baseline) and by 2050 for a conservative and ambitious scenario regarding the biogenic content for AF. The ambitious scenario assumes that petroleum coke and coal will be phased out by 2050.

2.2. Value chain: capture plant, transportation, storage site, and utilization route

2.2.1. Capture plant

This study models a conceptual capture facility of approximately 1.3$MtCO_2/y$ of capacity. The capture plant consists of a post-combustion amine-based process installed in the tail-end of the grey rotatory kiln.

Table 1
Type of fuels for clinker production by type, year, and biogenic content, with respect to energy content.

Fuel	Baseline		Conservative		Ambitious	
	2020	Biogenic content	2050	Biogenic content	2050	Biogenic content
Fossil	40%	–	23%	–	0%	–
AF	60%	25%	77%	36%	100%	57%

Specific conditions and composition of the flue gas were considered to design the capture plant. The capture process is assumed to start with a flue gas condensation unit that aims to remove pollutants and cool down the gas stream to 30–40 °C. This is followed by the absorption-desorption loop in which the flue gas first enters contact with a CO_2-selective solvent in a packed bed absorption column. The CO_2-amine solution is then pumped into a desorber tower where the solvent is regenerated using LPS at 130–150 °C, releasing CO_2 at 99% purity. The regenerated amine is returned to the absorption tower to be used again. An ion exchange reclaimer unit is needed due to the degradation of the amine: the solvent is recovered, and some brine sludge is generated. Finally, the CO_2 is compressed to 15 bar and cooled to −28 °C in a liquefaction unit using ammonia. Temporary storage in tanks at the plant's harbor is included.

Since the cement plant cannot produce all the required heat to operate the capture plant, this study examines three distinct steam sources: a natural gas boiler, an electric boiler, and a Mechanical Vapor Recompression-MVR heat pump. The MVR heat pumps upgrade available low-quality heat (SINTEF, 2017). In this case, they convert water at 75 °C into LPS at 135 °C. A Coefficient of Performance-CoP of 2.85 is assumed for the MVR pump. The water the boilers use comes from the flue gas condenser and must be previously deionized. The flue gas generated by the combustion of natural gas is fed into the capture plant. In addition, the study considers energy integration, which entails using excess heat and water from the cement production in the capture plant (including the liquefaction unit) and the generation of district heating to improve overall energy efficiency. A cooling system is necessary during the summer when the demand for district heating is low. In addition, a maximum heat supply capacity of 100 MW is assumed during winter. Table S1 in SI.1 shows the summer and winter heat and cooling demand.

Furthermore, the model includes the proprietary solvent Shell's Cansolv 103–47% of cyclic amines blend, 48% water, and 5% propylene glycol (Criterion Catalyst and Technologies, 2018). This solvent presents superior kinetics compared to conventional amines, high loading capacity, and more resistance to degradation (Vega et al., 2020). The energy requirement for capture plants using this solvent has been reported to range between 2.33 and 2.89 MJ/kgCO_2 captured (Singh and Stéphenne, 2014; Wang et al., 2021; Young et al., 2019). This study assumes an energy requirement of 2.5MJ/kgCO_2 and a solvent makeup of 0.588 kg/kgCO_2 (cf. Details in SI.1).

2.2.2. *Transportation, storage, and conversion path*

Given that shipping costs for CO_2 per ton can be 62% less expensive compared to offshore pipelines for transport distances between 500 and 1500 km (Al Baroudi et al., 2021), the model incorporates CO_2 transportation via sea shipping. Furthermore, injection for permanent storage takes place in a depleted oil and gas field located in the North Sea, 480 km from the cement plant.

Lastly, while various products can be derived from captured CO_2, methanol produced through the hydrogenation of CO_2 exhibits the most promising potential for short-term implementation (Artz et al., 2018; Chauvy et al., 2019; Garcia-Garcia et al., 2021; Thonemann, 2020). Considering that the hydrogen generation process through water electrolysis is the primary contributor to GHG emissions in this specific utilization method, utilizing low carbon intensity power sources such as wind, solar, hydro, and nuclear can significantly mitigate these impacts. (Garcia-Garcia et al., 2021; Sarp et al., 2021; Ueckerdt et al., 2021). This study assumes that in a CCUS scenario, the biogenic CO_2 is delivered for conversion to methanol, followed by the production of synthetic kerosene, hereafter referred to as e-kerosene. Power-to-liquid (PtL) conversion is presented as an efficient alternative to storing CO_2 in valuable products, yielding alternative fuels that could help decarbonize the transport sector (Bellotti et al., 2017). Specifically considering aviation, PtL synthetic fuels outperform other Sustainable Aviation Fuels (SAFs) regarding long-term scalability. They possess greater supply potential due to the absence of feedstock availability restrictions and enable the production of high-energy-density fuels that are cleaner than other SAF types (KPMG, 2022). E-kerosene was chosen as it is a synthetic fuel that can be used in existing planes and engines. The conversion technology data has been taken from the studies of Hank et al. (2019) and Meunier et al. (2020). The latter describes a highly integrated capture plant within the cement production site, with a methanol production unit that uses H_2 produced by alkaline water electrolysis with 76% conversion efficiency and 99% purity. Losses along the CCUS value chain are specified in SI.1. Fig. 1 shows the value chain for the CCS and CCUS paths analysed.

2.3. *AF availability*

Refuse-Derived Fuel (RDF) constitutes more than 50% of the alternative fuel (AF) utilized in clinker production for both present-day and future scenarios examined in this paper, with the remaining portion consisting of various waste streams with high biogenic carbon content (e.g., meat and bone meal, paper sludge). This trend is consistent with Europe's cement industry, where RDF is the second most widely used fuel, accounting for approximately 5 million tons per year or 40% of the total market (de Beer et al., 2017; IEA, 2020b). RDF is a primary means of reducing fossil CO_2 emissions in cement production (CEMBUREAU, 2020), so its demand is expected to increase in the coming years. According to a report (de Beer et al., 2017), the co-processing waste rate in some European countries could rise to 90% over the next ten years. Additionally, the study found a negative correlation between the proportion of waste disposed of in landfills and fuel substitution in cement kilns. Brown (2018) suggests that the amount of landfilled waste will decrease, potentially producing 63 million tons of RDF annually, surpassing the cement industry's projected demand of around 50 million tons annually. The increase in RDF use will also result from applying European regulatory frameworks such as the EU Waste Framework Directive that establishes recycling targets and objectives for landfill reduction through instruments such as landfill bans and taxes (De Caevel et al., 2018). Likely, countries such as Denmark and the Netherlands, that already reached an overcapacity to process waste (Brown, 2018), will import the material from countries that start sorting the waste but do not have the necessary infrastructure to convert it into energy (IEA, 2020b).

The availability of alternative biogenic fuels such as meat and bone, sewage sludge, paper sludge, and wood fiber boards typically depends on local availability and is challenging to forecast. However, it is evident that there is intense competition for biogenic materials in the energy sector, and there is a limit to the global sustainable biomass resources (Lund et al., 2022).

2.4. *Prospective life cycle assessment*

2.4.1. *Foreground system and data*

A prospective cradle-to-gate LCA compares the impacts on cement clinker production followed by CCS and CCUS relative to a reference scenario where no mitigation action is undertaken. Considering that implementing CCS or CCUS does not affect the mechanical properties of cement clinker, this study disregards the downstream activities required to produce cement and concrete. Thus, the functional unit is the *production of 1 ton of grey cement clinker in 2050.* The system boundaries, as illustrated in Fig. 2, cover the operational activities of the whole value chain. This includes all the ancillary raw materials used in the clinker production and the capture plant, together with the energy requirements for operating the clinker production process, the capture plant, and the liquefaction, transportation by ship, and injection of the CO_2. Since the solvent used in the capture plant is a proprietary amine, it is impossible to know the exact type of cyclic amine used. Therefore, data to produce MEA is used instead. This implies that specific degradation products released into the air are not included due to the lack of data. Air emissions such as NO_x, SO_2, CO, and NH_3 from clinker production and

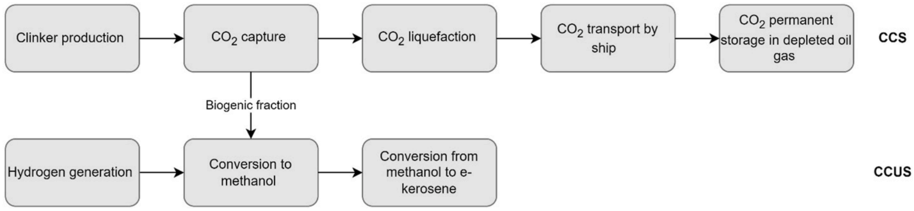

Fig. 1. Activities considered in the current study for the CCS and CCUS value chain.

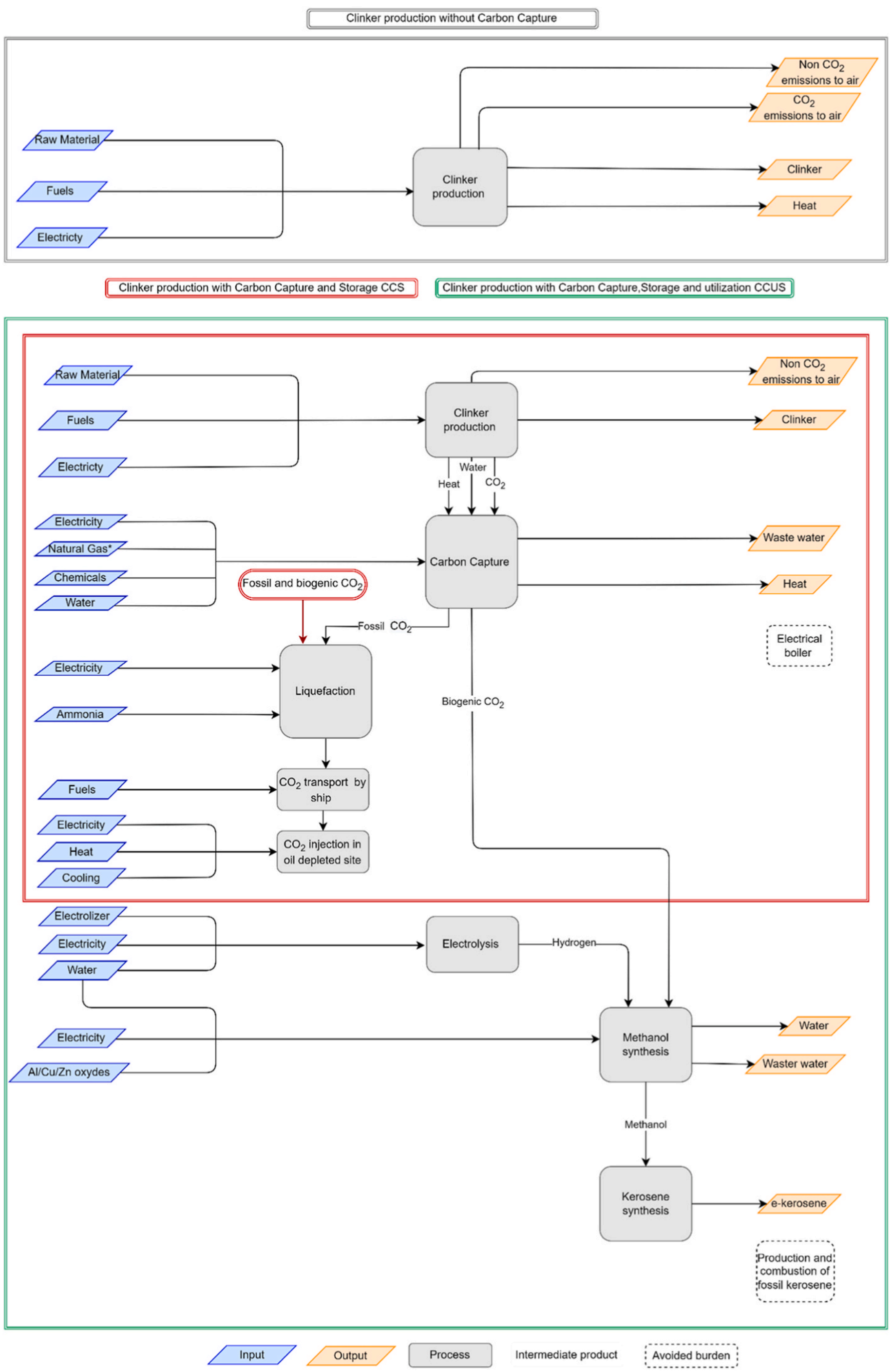

Fig. 2. System boundaries for three main scenarios. *Natural gas consumption is only relevant when the heat is generated by a gas boiler. Functional unit: 1 ton of grey cement clinker in 2050.

wastewater from the capture plant are included. The construction phase for the capture plant is disregarded as the impacts of capturing are mainly related to the operational phase (Saunier et al., 2019; Tang and You, 2018).

Besides clinker, the system also delivers excess heat to the local district heating network and e-kerosene resulting from CCUS. Following a consequential approach, the substitution method is used to model these co-products and solve the case of joint production (Von Der Assen et al., 2013), each substituting (avoiding) the supply of a marginal mix of products with identical functionality. The delivery of excess heat displaces the need for heat otherwise generated by an electrical boiler with a 99% of efficiency, a modelling choice made after discussion with the local utility company, who expected this to be the heat provider reacting to changes in supply by 2030. Due to a lack of projections, the same is assumed for a 2050 scenario. Considering the baseline scenario (no carbon capture), the cement plant delivers excess heat through district heating to the local utility company. The supply of e-kerosene is assumed to displace the need for producing and combusting fossil-based kerosene. Based on the current projections of the ReFuelEU initiative (European Comission, 2021), by 2050, 63% of aviation fuels are expected to come from SAF, and 28% of these will come from synthetic fuels. With the current limited market capacity and the constraints for European SAF production (Giannelos et al., 2021), it is sound to assume that any SAF produced by 2050 will replace fossil kerosene at the margin. It is also assumed that wind power will meet any additional demand for electricity resulting from the implementation of CCS or CCUS, according to the ENTSO (2020) projections for Denmark.

In the case of CCS, net negative CO_2 emissions are observed when the biogenic CO_2 contained in the flue gas is permanently stored. In the case of CCUS, a reduction in fossil CO_2 emissions is observed when the supply and use of e-kerosene (produced from biogenic CO_2) displaces the need for supplying and combusting fossil-based kerosene. There is no physical distinction or separation between the fossil and biogenic CO_2 at the capture unit; this separation is only virtual and done through mass balances.

Finally, RDF is generated from commercial, industrial, and municipal solid waste. Following consequential LCA principles, the supply of RDF is assumed to be constrained since an increase in demand does not lead to additional waste being made available. However, based on the market trends for RDF described in session 2.1.1, an increase in demand for RDF will likely result in less waste being treated in landfills. This will continue until the market reaches an equilibrium where the capacity for waste recovery and recycling matches the level of waste generation, effectively reducing the amount of waste that goes to landfills to zero. We assume such an equilibrium will be met in Western Europe by 2030 but with ample opportunity to source additional RDF in Central and Eastern Europe and North Africa in 2050. Hence, the distance RDF is transported will increase from 1.000 km in 2020 to 6.000 km in 2050, reflecting new but more distant RDF suppliers. To consider the effects of RDF usage, we followed the guidelines provided by Prosman and Sacchi (2018), which consider both the transportation of this material and its impact on waste treatment, in this case, landfill. Hence, the model does not reflect the competition for RDF with waste-to-energy plants since the material is sourced from markets with overcapacity for landfilling. Increasing the use of AF, particularly RDF, can lead to productivity losses compared to conventional fuels since AF has a lower calorific value due to higher moisture content. The future scenarios and their corresponding production volumes consider these losses.

Confidential data for cement production has been acquired directly with the cement company. The information regarding the energy and mass balances of the capture plant was obtained from a desk concept study conducted by the Danish consultancy engineering company COWI and commissioned by the cement company within the project Greencem (https://greencem.dk/). The study involved process simulations and covered a capture plant retrofitted to one production line at the cement facility. Information about energy consumption for transport and storage activities was collected from communications with an oil company developing research to inject CO_2 in their depleted oil fields. The inventories for the capture plant, transport, injection, and conversion of CO_2 can be found in the SI.2 (spreadsheet file). The technical parameters used both for modelling the capture process and the storage activities are supported by values reported in literature and thus sufficiently reliable for the study.

2.4.2. *Background system and data*

The database ecoinvent (Wernet et al., 2016) models the supply of commodities and services to the foreground system, such as fuels, raw materials, road, rail, and sea transport. The study uses a prospective version of the ecoinvent LCA database supplied by the library *premise* (Sacchi et al., 2022). This tool integrates projections from IAM scenarios into the ecoinvent LCA database. This prospective database is used to characterize the system's performance in the future. To do that, several considerations are made regarding expected changes in the fuel and electricity mixes, kiln efficiency, and hydrogen production, but also regarding secondary commodities and services modelled in ecoinvent. This tool is key for the current case study since it includes the expected transformations in energy-intensive sectors such as power, cement, steel, transportation, and supply of alternative fuels over time and across different climate scenarios. The prospective database allows accounting for the improvements in the energy efficiency for processes directly related to the carbon intensity of implementing CCUS by 2050, such as heat generation, electricity consumption, or the fuel blend used by trucks. Projections from the IAM model REMIND (Baumstark et al., 2021) were used to build the prospective database, following the socio-economic development scenario SSP2 "Middle of the road" (Riahi et al., 2017), combined with two climate scenarios equivalent to RCP 6 and RCP 1.9. The former represents a worst-case narrative, where anthropogenic emissions of GHG lead to an increase in the global atmospheric temperature of approximately 3.5 °C by 2100, with respect to pre-industrial levels. The latter limits that increase to 1.5 °C, equivalent to a peak cumulative emission of 900 GtC. A recent study shows that current and planned policies increase the global mean surface temperature to 2–3 °C (Pielke et al., 2022). The ReCiPe, 2008 v.1.13 method, is used in the life cycle impact assessment phase, and the results presented here focus on the midpoint impact category of global warming.

2.5. *Scenarios under assessment and variation test*

Three main scenarios were considered: clinker production without Carbon Capture as a baseline and clinker production with CCS and CCUS. The study aims to assess both value chains for CCS and CCUS to understand which path delivers the desired reduction goals and test if these two scenarios are complementary. In the CCUS case, permanent storage of the fossil and process CO_2 and delivery of the biogenic CO_2 to a Power-to-methanol plant is considered. Given that the energy penalty for capturing CO_2 highly depends on the technology that generates the LPS, three sources for this are included: E-boiler (Eb), Gas boiler (Gb), and Heat Pump (Hp). The results are presented for the +3.5 °C and +1.5 °C climate pathways and both conservative and ambitious use of AF as

Table 2
Overview of main scenarios under assessment.

Case	Sources of steam	Climate path	Use of AF
Clinker production with CCS Clinker production with CCUS	E-boiler	+3.5 °C + 1.5 °C	Conservative (77% AF) Ambitious (100% AF)
	Gas Boiler Heat Pumps		
Clinker production without CC	NA		

described in Table 1. A baseline scenario in 2020 is included to allow comparisons. Table 2 lists the scenario parameters considered, yielding 32 scenario cases.

During the development of the model, certain assumptions were required regarding specific parameter values and scenarios. Given that results can vary significantly depending on these assumptions, a variation test was conducted to understand the system's environmental performance comprehensively. Results were calculated for all feasible combinations of the most relevant input parameters, as outlined in Table 3. These distributions are not probability-based, as they merely show how the combinations of parameters (probable or not) distribute with respect to the carbon footprint per ton of cement clinker produced. The set of reasonable parameters' values was selected after communication with stakeholders such as the cement company, the utility company, and experts on CO_2 conversion and storage participating in the Greencem project. In the case of CCS, the excess heat supplied to the district heating network is assumed to displace the heat generated in electric boilers powered by wind energy. The electricity to operate the capture and liquefaction of CO_2 is deemed from wind power at the margin. The transport distance corresponds to the storage site at the depleted oil field in Danish waters, and the CO_2 is considered to be injected in an existing oil well. In the case of CCUS, in addition to the assumptions made for CCS, the hydrogen originates from water electrolysis powered with wind-based electricity, and the synthetic fuel substitutes fossil-based kerosene.

3. Results and discussion

3.1. Global warming potential (100 y)

While this section focuses on the GWP results, changes in other impact categories are presented in SI. Fig. 3 shows the carbon footprint per ton of cement clinker produced in 2020‡ and 2050, for which the contribution of the life-cycle phases in +3.5 °C and +1.5 °C scenarios are displayed. Table S2 in SI.1 shows results for the 32 scenarios. The model simulation results show that reaching climate neutrality for clinker production contribution to global warming is possible when implementing CCS or CCUS as long as a 100% alternative fuel mix with a 57% biomass content is used for clinker production. In fact, without any mitigation action undertaken other than increasing the use of alternative fuels, the cement plant is expected to lower its carbon footprint by 7–18% in 2050 relative to today, as shown when comparing the first 4 bars under "business as usual" of Fig. 3. Thus, the reduction in carbon footprint mainly stems from the use of alternative fuels, while the recovery of heat, equal approximately to 1 GJ per ton of clinker produced, contributes only minimally to reducing GHG emissions since it is expected to displace municipal electrical boiler – a source of heat already largely decarbonized.

The implementation of CCS in clinker production by 2050 shows reductions in GHG emissions of 89–91% (77% AF) and 105–106% (100% AF) relative to the performance of cement plant in 2020. This corresponds to a carbon footprint ranging from −57.5 to 97.4 kg CO_2eq/ton clinker depending on the level of AF usage, which is to be opposed to 902.2 kg CO_2eq/ton clinker produced in 2020 (table SI2 of the supplementary information). The extensive use of biomass-based AF allows for sequestering an amount of biogenic CO_2 sufficient to compensate for the fossil-based CO_2 emissions along the supply chain, despite the various losses considered during capture, liquefaction, transport, and storage. A fuel mix mostly made of AF with a high share of biogenic carbon leads to a lower, or possibly negative, carbon footprint, confirming the findings of Cavalett et al. (2022).

The use of AF is the determining parameter among the three scenarios for CCS presented in Fig. 3. When the marginal electricity mix includes mainly wind power, the means used to produce the LPS to regenerate the solvent (i.e., heat pump, natural gas, or electrical boiler) are not determining as the three options have a very similar carbon footprint. Therefore, choosing among the three options to generate the LPS depend on other variables such as costs and plant size, which for the case of the gas boiler is 16% greater than the alternative options. Also, the difference in results between the two climate scenarios remains limited, as most emitting processes occur within the boundaries of the cement plant.

The implementation of CCUS (Fig. 3) also allows reducing the carbon footprint of clinker production by 86–88% (77% AF) and 99–106% (100% AF) relative to the baseline scenario in 2020. The carbon footprint of clinker ranges from −5.6 to 124.3 kg CO_2eq/ton clinker when accounting for the avoided production and combustion of 40–70 L of fossil-based kerosene for each ton of clinker produced. Results in Fig. 3 also show that CCUS leads to slightly higher GHG emissions than the CCS option. This is because the biogenic CO_2 used to produce the fuel goes through several additional processing steps compared to the CCS option, leading to 10% higher gas losses. Any biogenic CO_2 gas loss along the e-kerosene supply chain is an amount that cannot be converted to fuel, resulting in fewer fossil-based emissions displaced. The electrolysis process is also energy-intensive and relatively inefficient: only 76% of the electricity energy entering the electrolyzer becomes hydrogen (Meunier et al., 2020). Additionally, 6% of hydrogen energy is lost in successive steps (i.e., H_2 and CO_2 to MeOH and MeOH-to-kerosene), and additional electricity and heat inputs are needed to convert methanol into kerosene. Overall, almost 1.7 MJ of final electrical and thermal energy are required per MJ of e-kerosene produced.

3.2. Variation test

The results in section 3.1 correspond to the scenarios described in Table 2, which are represented in red vertical lines of Fig. 4; these bars are positioned around the center of the distributions. This means they are neither overly optimistic nor very pessimistic regarding their carbon footprint. The figure also shows that particular sets of parameters can yield results with minor benefits of CCS and CCUS. In the case of CCS, this happens in the Baseline background climate scenario (+3.5 °C), combined with minimum usage of AF at the cement plant (60%, i.e., situation in 2020), and if the amine is regenerated using an electric boiler. The compression and liquefaction of CO_2 are powered by natural gas-based electricity, and this would yield a carbon footprint of 735 kg CO_2-eq. per ton of clinker produced, only 20% lower than the performance of the cement plant in 2020. In the case of CCUS, the worst results are, paradoxically, obtained using the highest share of AF (100%) but combined with the most carbon-intensive electricity. Under this scenario, a high percentage of AF implies a maximized production of e-

Table 3
Input parameters permuted to generate the variation test results.

Background scenario	Baseline (3.5 °C)	<1.5 °C	–	–
Use of alternative fuels	Baseline (year 2020)	77%	100%	–
Electrical Mix	Marginal mix (100% wind)	Average mix (90% renewable)	Natural Gas	Solar PV
Heat marginal technology	El boiler operated by wind	El boiler used with natural gas	–	–
Heat source	Electrical boiler	Gas boiler	Heat pump	–
Storage site	Northern Lights	Greensand	–	–
Hydrogen production pathways	Electrolysis	SMR of nat. Gas	SMR of biomethane	–
Marginal fuel supply for heavy transport	Kerosene	Gasoline	–	–

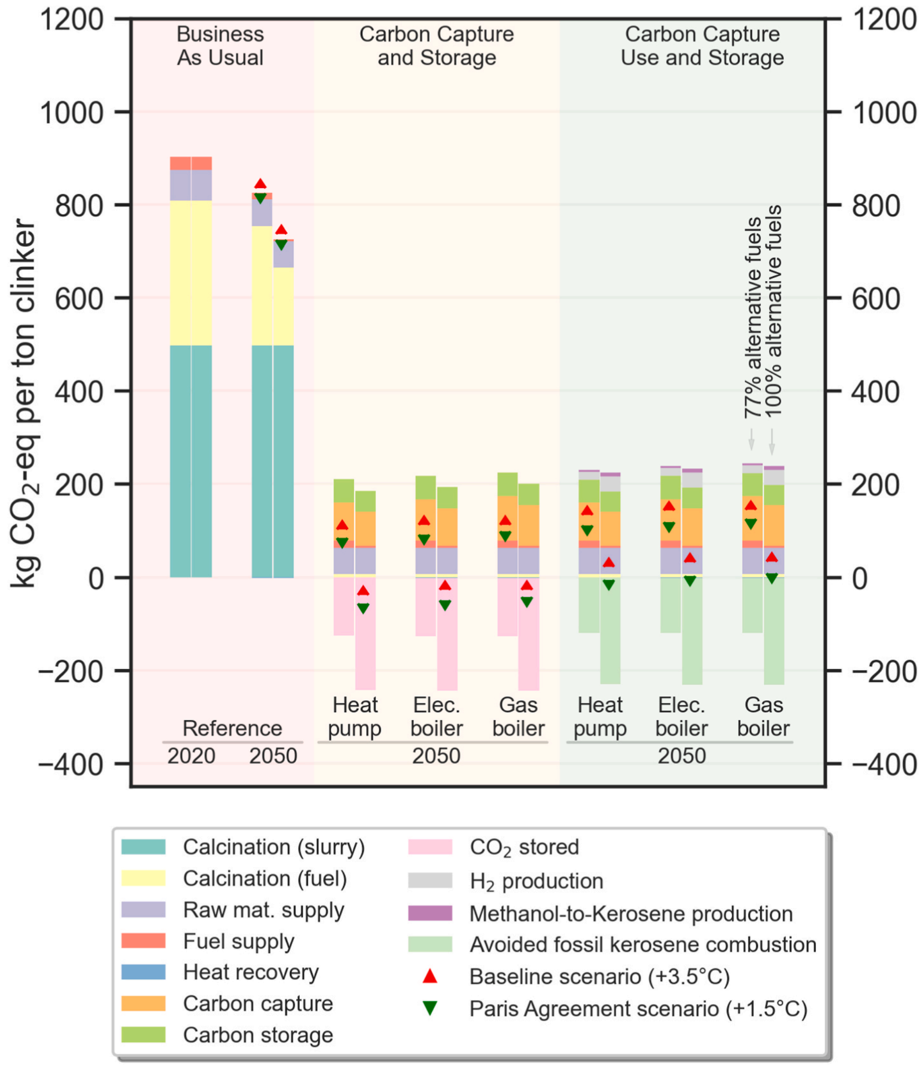

Fig. 3. Results for global warming for the baseline year (2020) and 2050 for a Business-as-Usual scenario (no carbon capture), for CCS and CCUS. All given for the three sources of steam and for the conservative (77%) and ambitious (100%) use of alternative fuels. The red and green arrow heads show the total global warming values for the +3.5 °C and +1.5 °C background climate scenarios.

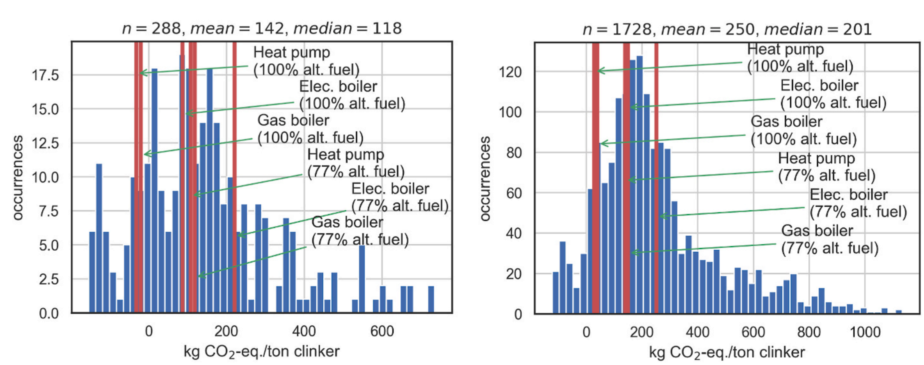

Fig. 4. Results for variation test for CCS (left figure) and CCUS (right figure).

kerosene since biogenic CO_2 is available in larger quantities. Suppose the e-kerosene is obtained via electrolysis powered by natural gas-based electricity. In that case, the additional GHG emissions associated with electricity production are far superior to the emission reductions from avoiding the combustion of fossil-based kerosene.

Results of the variation test indicate that the lowest carbon footprint for CCS is achieved in a 1.5 °C background climate scenario using 100% of AF and a gas boiler to generate the steam. Moreover, the heat displaced by heat recovery is natural gas-based, and the electricity comes from solar panels; in the case of CCUS, the same holds regarding the PA scenario, use of AF, source of electricity, and technology to generate the steam. But in this case, the lowest GWP is reached when the marginal heat supply is supplied by an electrical boiler operating with wind electricity; the results are roughly the same regardless of the means to synthesize the hydrogen.

Data from Fig. 3 suggests that the results are more sensitive to some input parameters than others. To visualize this, a Pearson correlation analysis across the unique sets of parameter combinations used in Fig. 4 is presented in section 7 of SI. The analysis reveals that in the case of CCS, the input parameters most correlated with the carbon footprint are, in order of importance and with the corresponding correlation factor: the electricity mix (0.69), the share of AF in the fuel mix, and the underlying share of biomass (0.48), the technology used to generate the LPS (0.36), as well as the heat supplier affected at the margin by the co-supply of excess heat (0.28). In the case of CCUS, those remain strongly correlated parameter inputs but in a slightly different order. For example, the electricity mix with a correlation coefficient of 0.76 seems to matter more than the share of AF used (correlation coefficient 0.17). This is because the pessimistic scenario in terms of AF considers 60% of the fuel mix. The worst case in terms of electricity carbon intensity is more impacting: natural gas-based electricity, with a carbon intensity of 490 g CO_2-eq./kWh. These parameters not only correlate with the system's carbon footprint but are the direct cause of it.

3.3. Variation of global warming to changes in the electricity and fuel mix

Fig. 5 characterizes the sensitivity of the carbon footprint of clinker production to the two most correlated model input parameters for both CCS and CCUS, namely the share of biomass in the fuel mix and the carbon intensity of electricity. Fig. 5 suggests that the percentage of biomass, via the use of AF in the fuel mix, and the carbon intensity of the electricity are two strong drivers that can help clinker production align with the Paris Agreement objectives. Its carbon footprint may become negative when the shares of AF in the fuel mix and the share of renewable electricity are close to 100%. A reduction in 116% and 107% of GHG emissions is reached for CCS and CCUS, respectively, relative to the GHG emissions per ton of clinker observed in 2020. These results are in line with the ones reported by Schakel et al. (2018) and Cavalett et al. (2022), where the authors find that the use of biogenic fuels combined with capture and storage technologies can result in very low and even negative emissions in clinker production. The result also confirms previous findings that the production of methanol and e-kerosene from the hydrogenation of CO_2 can only generate benefits if the electricity to produce the hydrogen is not carbon intensive (Artz et al., 2018; Pontzen et al., 2011; Safari and Dincer, 2018; Thonemann, 2020). Previous studies on Power-to-X plants have concluded that electricity's carbon intensity affects the system's environmental performance to a high degree (Koj et al., 2019). Hence, it is important to stress that a high share of

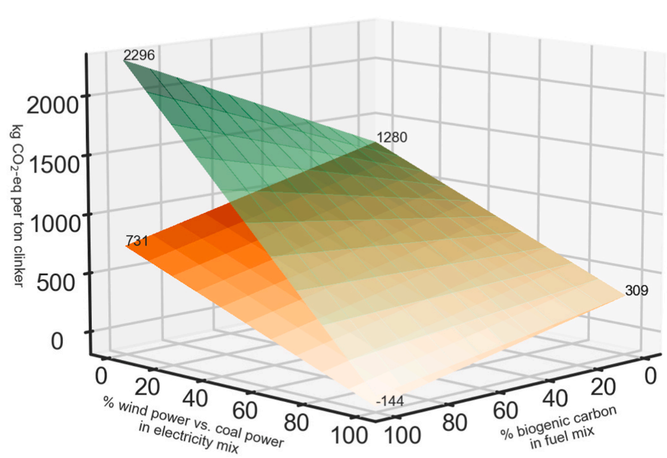

Fig. 5. Global warming values for changes in the carbon intensity for electricity and the biomass content in the fuel mix. The orange area shows the results for CCS and the green one for CCUS. All the values are given for the Baseline climate scenario (+3.5C by 2100) and the year 2050.

biomass in the fuel mix alone is not enough to reach neutrality, as seen in the previous section as well as in Fig. 5. For instance, CCUS with a high share of AF but carbon-intensive electricity leads to a net increase in GHG emissions. The CCS option is less sensitive to the carbon intensity of electricity than the CCUS option. For example, storing only the fossil CO_2 with low-carbon electricity is preferable to storing the biogenic CO_2 with carbon-intensive electricity.

3.4. Model limitations

The presented results are based on the assumptions to model the LCA for a potential CCS or CCUS scenario. The capture plant and storage activities data were obtained from a conceptual study, while literature data was used to model the utilization paths. To comply with the industrial needs to protect sensitive data, inventories about cement production remain confidential. The accuracy of the inventories for the capture plant and other parts of the value chain could be improved by using experimental data. Nevertheless, the calculations correspond to already-known processes based on proven thermodynamic principles. The capture technology is one of the few processes that has reached a commercial level and a high TRL. The model proposed here is thus a reasonable approximation of a technology that has not been implemented yet. The results can adequately support decisions on pursuing the implementation of such technology. However, data is missing for the emissions related to the production and transport of the Shell solvent, its degradation, and associated air emissions at the capture plant, and the burden from treating the brine water generated. This information will result in different environmental performances of the capture plant in impact categories such as ecotoxicity. A testing campaign should be established to obtain this data, which was outside the scope of the study and is recommended for further research. The study is also limited to the specific geographical location and distinct clinker production process (i.e., semi-wet process). Although it shows relevant findings for the cement industry, the result cannot easily be generalized to other sectors or cement plants. The sources of electricity correspond to future European energy scenarios up to 2050 presented in the ENTSO, 2020 report; these scenarios correspond not to forecasts but to potential projections. They are nevertheless a reasonable depiction of how the future energy systems may look like.

Regarding the LCA method, it should be noted that while a consequential approach is used for modelling the foreground system, the background system is modelled using a prospective *attributional* database due to the lack of a *consequential* counterpart. Since most impacts are located within the foreground model boundaries, using a consequential prospective background system would likely not change the conclusions drawn in this study. A better understanding and forecasting of the availability of AF, including RDF and biogenic sources in Denmark, would also improve the system modelling.

3.5. Have CSS and CCUS the potential to bring cement production to climate neutrality?

Fig. 3 shows that it is possible to reach climate neutrality in clinker production when implementing CCUS and to reach negative emissions when implementing CCS. The pre-conditions for this are: alternative fuels must be biomass-rich and represent close to 100% of the kiln fuel mix, the source of electricity must have a low carbon intensity, and other sectors supporting directly or indirectly the production of cement should preferably also decarbonize. Moreover, out of the 1.3 Mt of CO_2 captured, the percentage of biogenic origin ranges from 17% to 30%, depending on the share of AF in the fuel mix. Such an amount of biogenic CO_2 would allow producing between 50′000 and 90′000 tons of e-kerosene annually. Extrapolating the demand for kerosene in Denmark to 2050 based on the last 40 years of consumption (EIA, n.d.) – excluding the dip in demand caused by COVID restrictions – this supply of e-kerosene would already represent 4–8% of the 1′185′000 tons of kerosene that may be needed that year. It also represents 14%–28% of the minimum share of e-kerosene required in 2050 by the newly adopted ReFuelEU Aviation Initiative (European Comission, 2021) (i.e., 28% of the fuel blend). This supports the claim that it is imperative in Denmark to convert residual biomass resources, CO_2, and electrolytic hydrogen through Power-to-X technologies to reach a fully decarbonized society (Lund et al., 2022). Suppose the capture of CO_2 and the production and supply of e-kerosene rely on wind electricity and prevent the production and combustion of fossil-based kerosene. In that case, it is possible to avoid emitting 136′000 to 263′000 tons of fossil CO_2 annually.

It is worth noting that securing an equivalent amount of non-fossil CO_2 using direct air capture (DAC) instead would increase the amount of electricity needed. Capturing such an amount of biogenic CO_2 from the flue gases of the cement plant would necessitate between 96′000 and 238′000 MWh using a high-temperature heat pump or electric boiler, respectively. On the other hand, using a solid sorbent-based DAC system with a high-temperature heat pump with a *CoP* of 2.9 would instead require 394′000 MWh, based on the performance described in Terlouw et al. (2021).

Finally, despite its potential to reduce carbon emissions, implementing CCS or CCUS will depend on the business case and market instruments available to promote implementing these technologies in the cement industry (Stokke and Kvellheim, 2020).

3.6. Is 100% AF a realistic scenario for cement production?

The cement industry in Europe is currently the second largest consumer of AF. Various roadmaps to decarbonize the sector have identified this as a critical measure to continue reducing the carbon footprint of cement production. Nevertheless, using biomass-rich alternative fuels for clinker production is associated with several technical, regulatory, and sector competition obstacles that must be overcome. First, such biomass must originate from residual sources such as household or industrial waste, forestry residues, and dried sewage sludge to avoid additional emissions. According to the IEA (2020b), some cultivated species from fast-growing crops, such as grass and some wood, are useable from a technical perspective. Still, they are not economically viable for the cement industry and can lead to additional GHG emissions via direct or indirect land use change.

Nevertheless, the use of waste implies that the supply of such fuel would come from limited sources that cannot adjust the amount of waste fuel available to fluctuations in demand, except by reducing the amount of waste going to landfills. For example, if all the available AF on the market are already used (meaning that no more waste is being sent to landfill), obtaining an additional amount would eliminate the possibility of another consumer purchasing it. Such indirect and cascading market adjustments should be considered in sourcing alternative fuels for which supply is constrained, as shown in Prosman and Sacchi (2018).

Second, not all available biomass is suitable to burn in all cement kilns as these need fuels with a minimum calorific value of 20–22GJ/ton, which contrasts with 10–18GJ/ton for typical organic materials (ECRA, 2017). Nevertheless, the pre-calciners in the kiln run at a lower temperature and can accommodate up to 60% of low-calorific fuels.

Third, using alternative fuels can increase the thermal energy demand due to a high moisture content and more important air requirements, so heat recovery systems are needed to improve the overall energy efficiency of the process (GCCA, 2021). Suppose the thermal energy demand cannot be increased. In that case, this can lead to productivity and economic losses or the import of clinker, which may cancel out the initial GHG emissions reduction from using AF (Prosman and Sacchi, 2018). Another concern about using AF is that their composition is highly variable and, to some extent, uncertain. Therefore, special attention is focused on generating volatile S, Cl, Na, and K elements in the kilns due to incomplete combustion (Cortada Mut et al., 2015) and high concentrations of chlorine or heavy metals such as mercury and cadmium.

Fourth, waste regulation can also affect the availability of AF for clinker production, as is currently the case in Denmark, where most of the waste is treated in waste-to-energy plants, accentuating the competition for biomass-rich waste fuel. Ultimately, a cost-competitive supply of sustainable biomass is crucial to reach zero emissions in clinker production with CCS or CCUS, and this is a significant challenge as biomass-rich AF would be difficult to secure in the future due to high demands from other sectors such as bioenergy (Mortensen et al., 2020).

4. Conclusion

This study identified that a supply of alternative fuel with a high biomass content and a supply of low-carbon electricity are two necessary conditions for CCS and CCUS technologies to deliver climate neutrality in cement production. Green electricity can be secured via the grid or power purchase agreements or by directly investing in new renewable energy capacity (Bjørn et al., 2022). If a source of low-carbon electricity cannot be supplied, using a natural gas boiler is a suitable alternative with low impact. The study also shows that a specific combination of measures to reach a Paris Agreement path of 1.5 °C can lead to negative emissions. One of the main key messages of this study is that currently proposed measures for reducing the carbon footprint of the cement industry are interdependent and should all be met. For instance, a low-carbon electricity supply must be secured before undertaking CCS or CCUS to avoid increasing life cycle GHG emissions. The decision to use the biogenic CO_2 in a Power-to-Methanol plant (i.e., CCUS) rather than storing it underground (i.e., CCS) will not only depend on the environmental performance of the value chain but also the viability of the business case. Indeed, biogenic CO_2 could become a valuable commodity in the future, and which price might be influenced to some extent by the allowance price for fossil CO_2 given by the European Emissions Trading Scheme (European Union, n.d.). Another advantage of implementing CCUS is that 8% of the nationwide annual demand for kerosene could be met, substituting the need for the supply and combustion of fossil kerosene and avoiding the additional emission of 236′000 tons of CO_2 – despite being emitted into the atmosphere after being used as a fuel in the aviation sector.

These results were obtained using a comprehensive model that combines prospective LCA to address the temporary dimension of the technology and consequential LCA to address multi-functionality and the provision of alternative fuels. The study investigates a case that allows working with primary data for clinker production and specific conditions of the capture plant and downstream activities for transport and storage. The study handles the uncertainties of future conditions by using scenario and co-relation assessment instead of providing a single value. Nonetheless, there are some limitations to this choice of modelling. For instance, regarding background data, there is a need to modify the consequential database to use for prospective modelling and improve consistency. In terms of foreground data, it would be ideal to have data from a pilot test of the capture technology at the cement plant to gain a deeper understanding of the interactions between the amines and the flue gas and have primary information on the energy consumption and degradation products that influence air emissions. Even though the results correspond to specific conditions of the case study, the main conclusions can be extended to other cement plants. The results of this work should be compared with studies that use a capture technology with a similar TRL.

The findings of this study hold significance for cement companies presently contemplating CCUS deployment, as well as policymakers advocating for its technological adoption. Furthermore, it is of interest to researchers and professionals seeking to comprehend the potential of CCUS as a means of reducing CO_2 emissions and achieving the goals outlined in the Paris Agreement. Ultimately, Aalborg Portland can leverage these findings to incorporate downstream activities involved in cement and concrete production, enabling them to evaluate how these changes impact the environmental performance of their product portfolio.

A follow-up to this study could entail using experimental data from a pilot unit that tests the performance of the capture technology with the real flue gas. Also, incoming prospective databases could be used to improve the background data and to assess other impact categories better. Finally, the results of this study should be complemented with the assessment of different aspects, such as the business case and the social aspects, to have a more comprehensive view of CCUS implementation at Aalborg Portland.

Foot notes

‡The global warming potential for the baseline scenario corresponds to emissions of producing one ton of grey clinker in 2020 calculated by the model and it does not represent or is comparable with the verified CO_2 emissions reported in the European Union Emission Trading System- ETS by the company.

CRediT authorship contribution statement

Juanita Gallego Dávila: Conceptualization, Methodology, Validation, Investigation, Formal analysis, Resources, Data curation, Writing – original draft, and, Project administration. **Romain Sacchi:** Methodology, (modelling), Software, Validation, Formal analysis, Investigation, Resources, Data curation, Writing – review & editing, and, Visualization. **Massimo Pizzol:** Conceptualization, Validation, Writing – review & editing, and, Supervision.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Juanita Gallego Davila reports financial support was provided by Manufacturing Academy of Denmark. Romain Sacchi reports financial support was provided by Swiss Federal Office of Energy. Juanita Gallego Davila reports a relationship with Aalborg Portland that includes: funding grants and non-financial support. Romain Sacchi reports a relationship with Aalborg Portland that includes: previous employment. Corresponding author JGD pursues her doctoral degree at Aalborg University within a project in close collaboration with Aalborg Portland, her work is fully financed by Manufacturing Academy of Denmark (entitiy that receives a partiall contribution of funding from Aalborg Portland). Co-author RS is a former employee of Aalborg Portland and his work is funded by the Swiss Federal Office of Energy. Finally, co-author MP has no affiliation to Aalborg Portland.

Data availability

The authors have shared the data that is not confidential.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2023.138935.

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