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Cost and environmentally efficient design of an absorption-based post-combustion carbon capture unit for industry applications

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ABSTRACT

Carbon capture and storage technologies are currently considered promising solutions to mitigate CO_2 emissions of hard-to-abate industries. Among those, the cement industry accounts alone for around 6–7 % of the global anthropogenic CO_2 emissions and necessitates the integration of carbon capture and storage to substantially reduce its CO_2 emissions. This work focused on the design and analysis of a cost-efficient and low-emitting cement plant integrated with an absorption-based Post-Combustion Carbon Capture (PCCC) system using an aqueous water-amine solution. An innovative system layout is presented to minimize CO_2 emissions and energy consumption. Furthermore, the sizing of the absorber and stripper of the PCCC unit was performed to minimize the cost of the avoided CO_2. The environmental and economic performances of the overall process are evaluated under different boundary conditions, encompassing both gate-to-gate and cradle-to-gate perspectives. A reduction of the CO_2 emissions of around 66–87 % was obtained, depending on the fuel used to produce the steam needed in the PCCC unit and the analysis boundary considered. The primary energy consumption per avoided CO_2 was estimated at 5.5–7.3 MJ/kg_{CO2}. The specific cost per avoided CO_2 also varies with the fuel choice and the analysis boundary. Through the optimal sizing of the PCCC unit, costs of 56–57 €/t_{CO2} were obtained for the integration of carbon capture in the plant. When a cradle-to-gate boundary is considered, costs for CO_2 transportation and storage need to be accounted for, and costs of equivalent avoided CO_2 ranging from 74.5 €/t_{CO2} to 124 €/t_{CO2} are estimated, depending on the transportation and storage scenario considered.

1. Introduction

Industry decarbonization represents one of the major challenges to the pursuit of climate change mitigation. In 2020, GHG emissions from all the manufacturing industries totaled around 8.4 Gt, with the heavy industries being the largest contributors [1]. Among those, the cement sector is one of the largest energy consumers and CO_2 emitters, accounting for around 1.4 Gt of CO_2, i.e. 6–7 % of the total GHG anthropogenic emissions [2,3]. Cement production, furthermore, is a complex and hard-to-abate process where the production of clinker (the main constituent of cement) represents the most energy-intensive and CO_2-emitting step. The clinker is produced in the dedicated kiln where over 60 % of its CO_2 emissions come from the calcination of limestone (the principal raw material), while only the remaining 40 % come from the combustion of fossil fuels (usually coal) [2,4]. Carbon Capture and Storage (CCS) is, therefore, considered one of the most promising strategies for the mitigation of CO_2 emissions coming from cement production and is expected to play a critical role in developing an environmentally sustainable cement industry in the coming years [3]. Post-Combustion Carbon Capture (PCCC) techniques, indeed, can abate both process and combustion emissions and can be used to retrofit the existing plants with minimal influence on the existing production process. For these reasons, PCCC integration into the cement industry is attracting a lot of attention in the scientific community, and studies on absorption, adsorption, and membrane separation systems are already present in the literature [5–7]. Absorption systems, and in detail systems based on aqueous solutions of amines, are the only ones that already reached the commercial scale. The first cement plant integrated with a large-scale carbon capture facility (Norcem Brevik plant, Norway) is installing an absorption-based PCCC system. The plant operation is expected to start in 2024 [8].

These systems can greatly reduce the CO_2 emissions of the plant at the expense of a significant increase in energy consumption. The

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Nomenclature

Acronyms and abbreviations

AC	Avoided CO_2 emissions
ASR	Avoided-Stored CO_2 ratio
BEC	Bare Erected Cost
BREF	European best available technique reference document
$C_{T\&S}$	Annual CO_2 transport and storage costs
CAC	Cost of Avoided CO_2
CAPEX	Capital expenditure
$CAPEX_{ann.}$	CAPEX annuity
CCS	Carbon Capture and Storage
CHP	Cogeneration heat and power unit
CtG	Cradle-to-gate boundary
DCC	Direct Contact Cooler
EC	Equipment Cost
EPCC	Engineering, Procurement, and Construction Cost
GtG	Gate-to-gate boundary
HRSG	Heat Recovery Steam Generator
KPI	Key Performance Indicator
LCA	Life cycle analysis
LCOC	Levelized Cost of Cement
MEA	Monoethanolamine
OPEX	Operative Expenditure
PCCC	Post-Combustion Carbon Capture
SFC	Specific Fuel Consumption per CO_2 avoided
SNCR	Selective non-catalytic reduction
SPECCA	Specific Primary Energy Consumption per equivalent CO_2 Avoided
T&S	Transportation and Storage
TPC	Total Plant Cost
TOC	Total Overnight Cost
TASC	Total As-Spent Cost

Symbols

F_{BM}	Bare module factor
M_{cem}	Annual plant cement production
N	Plant operating lifetime
f_{EPC}	EPC cost factor
$f_{process}$	Process Contingency factor
$f_{project}$	Project Contingency factor
f_{OC}	Owner's cost factor
e	Specific CO_2 emissions
$m_{CO2,stored}$	Specific CO_2 stored underground
ped	Specific production process primary energy consumption
q	Specific plant fuel consumption
r	Discount rate

Subscripts

GtG	Parameter estimated with a Gate-to-Gate boundary
CtG	Parameter estimated with a Cradle-to-Rate boundary
upstream	Parameter referred to upstream processes
ref	Parameter referred to the reference unabated process

Table 1
Overview of previous techno-economic works on CCS integration in the cement industry focused on absorption-based post-combustion carbon capture systems (from 2017 to 2023).

Reference			System layout	Process modeling Main units modeled	Upstream, downstream processes	Absorber/stripper sizing
Jaffar et al.	2023	[12]	No heat recovery	Flowsheet model: Cement plant, PCCC, CO_2 compression	Upstream (Only electricity)	Not specified
Tanzer et al.	2023	[11]	No heat recovery	Black box model with constant ratios	Upstream, downstream (raw materials, fuels, electricity, CO_2 transp. and storage, concrete production, disposal, other)	No
Calin C. Cormos	2022	[13]	No heat recovery	Flowsheet model: PCCC, steam generation, CO_2 compression	Upstream (Only electricity)	Not specified
Cormos et al.	2020	[14]	No heat recovery	Flowsheet model: PCCC, steam generation, CO_2 compression	Upstream (Only electricity)	Not specified
Nwaoha et al.	2018	[15]	No heat recovery	Flowsheet model: PCCC, CO_2 compression	No	Assumed stages size
Gardarsdottir et al.	2019	[5]	Heat recovery from cement plant	Flowsheet model: PCCC, CO_2 compression	Upstream (Only electricity)	Not specified
Cormos et al.	2017	[16]	No heat recovery	Flowsheet model: PCCC, steam generation	Upstream (Only electricity)	Assumed size
Gerbelova et al.	2017	[17]	No heat recovery	Flowsheet model: Cement plant, PCCC, CO_2 compression	No	Not specified
Roussanaly et al.	2017	[9]	Heat recovery from cement plant	Flowsheet model: PCCC, CO_2 compression	Upstream (Only electricity)	Colum design software (SULCOL)
This work			Heat recovery from cement plant and CO_2 compression Flue gas from steam generation treated in the PCCC unit	Flowsheet model: Cement plant, PCCC, Steam generation, CO_2 compression	Upstream (raw materials, fuels, electricity, CO_2 transp. and storage)	Optimal size (costs sensitivity)

regeneration of the CO_2 solvent, indeed, requires a considerable amount of heat, which can only be partially supplied through heat recovery from the cement production process. The state-of-the-art cement plant is already a very energy efficient system with a limited amount of residual waste heat. The larger part of the heat duty for carbon capture needs, therefore, to be supplied from a dedicated steam production plant leading to an overall increase in fossil fuel consumption. The produced CO_2 for steam generation is usually assumed to be released into the

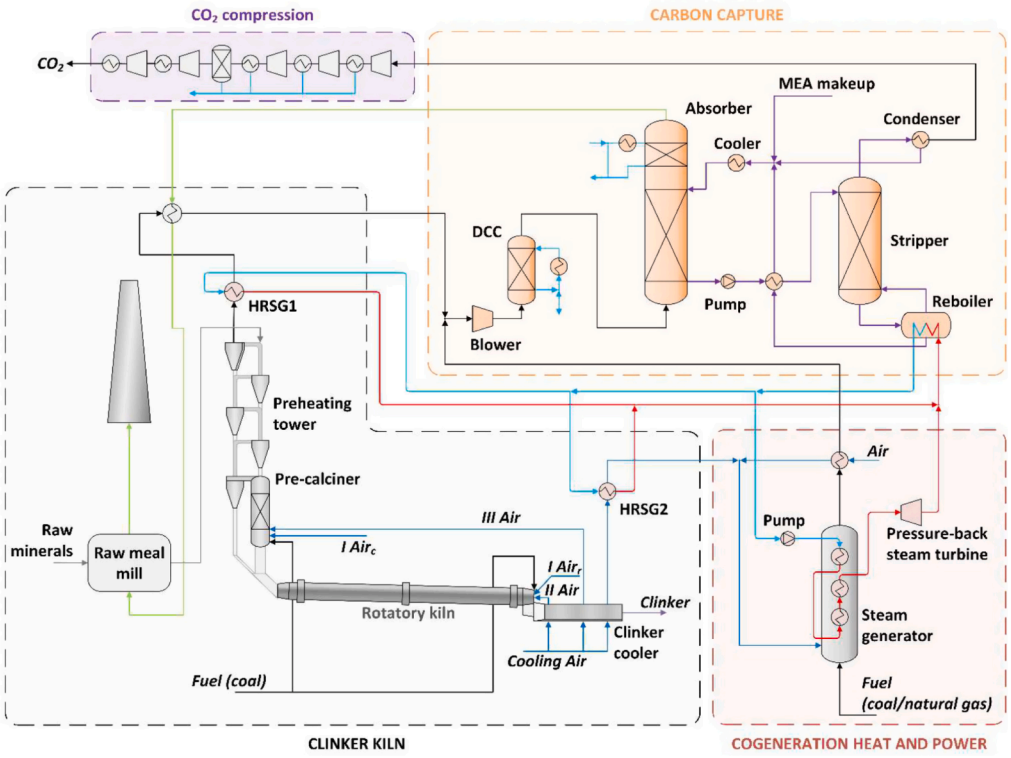

Fig. 1. System layout of the PCCC integrated cement plant.

atmosphere reducing the amount of CO_2 emissions that are truly avoided [6,9]. In this work, however, we consider an innovative plant layout where the flue gas produced from the steam generation system is treated together with the cement plant flue gas in the PCCC unit. This solution allows to reach very low plant CO_2 emissions. To the best of our knowledge, a similar solution was previously proposed only by De Lena et al. [10], where the authors performed, however, only an energy analysis of the system.

The focus of this work is to delineate a simple and replicable methodology for the optimal design of a thermally integrated PCCC unit within the production process, considering energy, environmental, and economic performance metrics. The findings reported were obtained through comprehensive process analyses, economic analyses, and carbon footprint analyses performed with a Life Cycle Analysis (LCA) approach. The mass and energy balance of the overall integrated system was estimated through a detailed process modeling work, which included all the main sections of the plant: cement plant, PCCC unit, steam generation system, and CO_2 compression unit. To minimize the plant fuel consumption, thermal integration between the PCCC unit, the cement plant, and the CO_2 compression unit was assessed. Additionally, a simple sizing procedure for the absorber and the stripper columns of the PCCC unit is outlined.

Table 1 provides a literature review of recent techno-economic publications on PCCC (absorption systems) integration in a cement plant highlighting key distinctions between this work and existing literature. While several authors have conducted detailed process analyses using various software, few have compared their results with a comprehensive carbon footprint analysis. Only Tanzer et al. [11] performed both an economic analysis and a detailed cradle-to-grave LCA analysis. These authors, however, analyzed the process with a simple black box model with assumed constant performance efficiencies and did not address the design of the integrated process. This paper, instead, addresses critical aspects often overlooked or described in different studies, mainly: (i) system layout optimization, i.e. thermal integration into the production process with heat recovery from cement plant and CO_2 compression and treatment of the flue gas produced during steam generation; (ii) sizing of the main components of the PCCC system (absorber and stripper column); (iii) environmental assessment, with CO_2 emissions accounting and primary energy consumption estimation; (iv) economic analysis for calculation of the cost per net avoided CO_2 considering all the CCS value chain.

2. Process description

Fig. 1 shows the layout considered in this work of a state-of-the-art cement plant integrated with a post-combustion carbon capture system. The plant can be divided into four different main sections:

i. Cement production: obtained by mixing clinker with other minerals (gypsum and limestone) [3]. Clinker is produced in a state-of-the-art kiln as described in the European Best Available Technique (BREF) document for the cement industry [18].
ii. Steam production unit: composed of two heat recovery steam generators (HRSG1 and HRSG2) and a dedicated fuel-fired heat and power cogeneration unit (CHP).
iii. PCCC unit: state-of-the-art absorption-based CO_2 capture system employing a monoethanolamine (MEA) water solution as CO_2 solvent and treating both the flue gas produced by the clinker kiln and the CHP.
iv. CO_2 compression unit: system required to increase the pressure of the CO_2 obtained to the pressure level needed for long distance transportation by pipeline.

The layout proposed in this work is characterized by the following points: (i) the carbon capture unit is placed on the flue gas line downstream of the preheating tower before the raw meal mill. The flue gas exits the preheater with a CO_2 concentration of around 30 vol% at around 330 °C and is normally recirculated to the raw meal mill and used in the drying of the raw minerals. This last component, however, is characterized by very large air in-leakages that drastically lower the CO_2

concentration in the flue gas. Placing the PCCC immediately downstream of the preheating tower, where the flue gas CO_2 concentration is higher, reduces, therefore, the heat requirement for carbon capture. (ii) The steam required in the PCCC unit is produced from heat recovery and a CHP employing a pressure-back steam turbine. This allows on-site production of electricity reducing the electricity demand of the plant (coming from the grid) and thus lowering the associated upstream CO_2 emissions. (iii) The flue gas produced by the CHP is treated, together with the clinker kiln flue gas, in the PCCC unit.

2.1. Cement production

Cement production comprises several steps: raw material extraction, grinding and mixing; clinker production; cement mixing, and milling [4]. The raw minerals consist mainly of limestone, clay, sand, and iron ore [19] that are usually extracted near the cement plant [18] and are then milled, dried, and mixed to produce a mixture called "raw meal". The raw meal is then sent to the clinker kiln (typical capacity of around 2,825 t_{clk}/day) that, according to the BREF [18], is composed of a preheating tower, a pre-calciner, a rotatory kiln, and a clinker cooler. In the preheating tower, the raw meal is heated in countercurrent flow by the upward kiln flue gas in five cyclones placed in series. The cooled flue gases are then usually recirculated to the raw meal mill [20], while the solids are sent to the pre-calciner placed at the bottom of the preheating tower. Here, almost all the $CaCO_3$ contained in the minerals undergoes calcination, a process that requires the combustion of around 2/3rd of the overall coal consumption of the entire kiln and produces a large amount of CO_2. The calcined raw meal is then fed to the rotatory kiln where is burned with coal to reach sintering temperatures. The remaining $CaCO_3$ is completely calcinated and the CaO reacts with SiO_2, Al_2O_3, and Fe_2O_3. The obtained product (clinker) is composed mainly of dicalcium silicate (belite), tricalcium aluminate, tetracalcium aluminoferrite, and tricalcium silicate (alite) [4]. The clinker, after being cooled down with ambient air in the clinker cooler, is finally mixed with gypsum and additional limestone to produce cement. In this paper a cement composition of 65 % clinker [3], 5 % gypsum [19], and 30 % limestone [3].

2.2. Post-combustion carbon capture

Absorption-based systems, and in particular systems employing an aqueous MEA solution (30 wt% MEA), are the PCCC technology with the highest technology readiness level and are thus commonly considered as the reference solution regarding carbon capture in industry [5,21]. In state-of-the-art absorption PCCC systems the flue gases are first cooled down to around 40 °C in a Direct Contact Cooler (DCC) and are then sent to a packed absorber column [6,9], divided into a bottom absorption section and a top washing section [9]. The flue gas flows from the bottom to the top of the column in countercurrent with the CO_2 liquid solvent. In the absorber section, the solvent reacts with the CO_2 thus reducing the CO_2 content of the flue gas. In this regard, a CO_2 capture efficiency of the absorber equal to 90 % was assumed. The CO_2-lean gas is then washed with water in the washing section of the column to reduce the system MEA losses. A 5.5 m tall washing section was estimated to be able to recover almost all the solvent contained in the treated flue gas. The CO_2-rich solvent, exiting from the bottom of the absorber column, is then pumped to 1.8 bar and is regenerated in a packed stripper column equipped with a reboiler and a condenser. Nearly pure gaseous CO_2 is produced in the stripper condenser and is then sent to the compression unit. The regenerated solvent, exiting from the reboiler bottom, is then recirculated to the absorber after the addition of fresh solvent [21,22]. Regarding the condensed liquid exiting the stripper condenser, two different possible configurations are reported in the literature. In the first one, the liquid is recirculated at the top of the stripper column [9], while in the other one, the liquid is mixed with the regenerated solvent exiting the stripper reboiler and sent to the absorber [22,23]. In this work, the second configuration is applied. Such solution, indeed, reduces the heat required in the stripper reboiler [22]. The captured CO_2 is then compressed to 120 bar, which is the pressure needed for long-distance pipeline transportation [24]. This unit is usually a five-stages inter-refrigerated compressor system equipped with a dehydration TEG unit placed between the third and fourth stage [6,24].

It should be noted that the NO_x and the SO_x contaminants (usually present in combustion flue gases) may poison the MEA solvent and should be lowered to acceptable levels before being sent to the absorber. The necessary NO_x reduction can be achieved through the SNCR process, a technique already commonly employed in cement plants. The SCNR process consists of the injection of an aqueous NH_3 solution (25 wt% NH_3) in the flue gas stream. As reported in the literature, high NO_x removal rates can be reached with the injection of the NH_3 solution in over-stoichiometric amounts (1.5 times) [25]. Systems dedicated to SO_x removal, instead, are not normally installed in cement plants, this contaminant, indeed, reacts almost completely with the CaO forming $CaSO_4$. The untreated flue gas, therefore, is characterized by a SO_x content lower than the regulatory emissions limits. The gas entering the absorber column, however, should have a very low SO_x (less than 10 ppm) content, a value lower than the typical SO_x emissions of a cement plant. This level can be guaranteed with the injection of an aqueous NaOH solution (50 wt% NaOH) in the direct contact cooler in a stochiometric amount [25]. Consumptions of 5 kg/t_{clk} of NH_3 solution and 1 kg/t_{clk} of NaOH solution were assumed for the cleaning of the clinker kiln flue gas [5], and the additional consumptions for the CHP flue gas cleaning were calculated from the gas NO_x and SO_x content (estimated though process modeling).

2.3. Steam generation

The heat requirement of the stripper reboiler is supplied by condensation of steam at 2.7 bar (saturation temperature of 130 °C, considering a minimum temperature difference of 10 °C). Part of the steam can be produced from heat recovery from the clinker kiln. In detail, the kiln flue gas exits the pre-heating tower at around 330 °C and can be cooled down to 120 °C (our assumption to avoid condensation phenomena) producing around 126 kWh/t_{clk}, some of the obtained heat (84 kWh/t_{clk}) is used in a countercurrent gas/gas heat exchanger for the heating of the CO_2 lean flue gas exiting the absorber column, which will then be sent to the raw meal mill, the remaining available heat (42 kWh/t_{clk}) is recovered in the exchanger HRSG1 for production of steam. 37 kWh/t_{clk} is also recovered in the HRSG2 from the cooling of the clinker cooler gas exhaust. The remaining amount of steam is produced in the CHP equipped with a pressure-back steam turbine. The condensed water from the reboiler is first compressed to around 60 bar in a pump. Superheated steam at around 540 °C is produced in a traditional steam boiler and then expanded in a pressure-back steam turbine [26]. No pressure losses were considered in the steam boiler. Regarding the fuel consumed in the CHP, two different scenarios were considered: in the first one, the CHP is fired with coal, which is still the most used fuel in state-of-the-art clinker kilns. Being coal a very carbon intensive fuel, its use in the CHP, however, will impact the overall energy and environmental performance of the PCCC system. For this reason, a second scenario with a natural gas (NG) fired CHP is considered.

2.4. CO_2 transport and storage

In order to achieve a reduction in the process's overall GHG emissions, the CO_2 captured from the cement plants must, of course, be permanently sequestrated. CO_2 transportation and storage (T&S) constitute, therefore, a crucial step in the CCS value chain and need to be considered when a cradle-to-gate approach is adopted [11].

Currently, transportation via pipeline is recognized as one the most promising technological solutions for long-distance CO_2 transportation [27,28], while injection in deep underground geological reservoirs

Table 2
CO_2 transport and storage assumptions: upstream emissions, electricity consumption, and unitary costs (reference year 2019).

	Value	Unit	Reference
Pipeline CO_2 losses	0.026	%/1000 km	[36]
Electricity consumption for recompression	5	kWh/t_{CO2}	[36]
CO_2 recompression losses	0.05	%	[36]
Electricity consumption for underground CO_2 injection	7	kWh/t_{CO2}	[36]
CO_2 injection losses	0.1	%	[36]
T&S base-case			
Transportation distance	680	km	
Offshore	180	km	[30]
Onshore	500	km	[30]
Onshore transport unitary cost	0.0158	€/(km•t_{CO2})	[27]
Offshore transport unitary cost	0.0237	€/(km•t_{CO2})	[27]
CO_2 transportation costs	12.2	€/t_{CO2}	
Offshore underground CO_2 injection	18.1	€/t_{CO2}	[27]
T&S best-case			
Transportation distance (onshore)	100	km	[33]
Onshore transport unitary cost	0.0429	€/(km•t_{CO2})	[27]
Offshore transport unitary cost	0.0765	€/(km•t_{CO2})	[27]
CO_2 transportation costs	4.3	€/t_{CO2}	
Onshore underground CO_2 injection	7.3	€/t_{CO2}	[27]
T&S worst-case			
Transportation distance	980	km	
Offshore	180	km	[30]
Onshore	800	km	[36]
Onshore transport unitary cost	0.0158	€/(km•t_{CO2})	[27]
Offshore transport unitary cost	0.0237	€/(km•t_{CO2})	[27]
CO_2 transportation costs	16.9	€/t_{CO2}	
Offshore underground CO_2 injection	18.1	€/t_{CO2}	[27]

(saline formations and depleted oil and gas fields) is the prevailing technique for permanent CO_2 storage, with numerous projects operational worldwide (over 20 projects with size between 0.3–8.4 Mt_{CO2}/year, including also enhanced oil recovery projects) [29]. Although, long-distance gas transportation with pipeline is a well-established practice, estimating the cost for CO_2 transport remains a complex issue, with limited literature focusing on this topic. Various factors such as CO_2 flow rate, transportation distance, pressure level, pipe wall thickness, and land use influence the costs associated with the implementation of a pipeline network [30–33]. Storage costs are also highly variable and depend on the site location (onshore or offshore), type of reservoir (depleted Oil&Gas field or saline formation), and geological characteristics (depth, porosity, permeability) [29,32]. The data reported in the literature, indeed, range from very low values (1–7 €/t_{CO2} [32]) for onshore depleted Oil&Gas fields to much higher values (2–12 €/t_{CO2} [32]) for onshore saline formation. Furthermore, according to the literature, offshore underground CO_2 injection is around 2.5 times more expensive than onshore injection [27]. It should be noted that the ranges reported in the literature are often authors' own estimations, often tailored around particular case studies, and average costs resulting from a detailed statistical analysis are still missing [32]. Therefore, considering that geological characteristics also highly influence storage costs, it should be expected that poor storage site candidates may present costs above the typical ranges reported in the literature. For further information, Table S13 and Table S14 of supplementary material report T&S unitary costs ranges recurring in the literature.

Ideally, to reduce transportation costs, the transportation distance should be minimized while favoring large-size pipelines over small-size ones [30]. Moreover, injection costs should not be overlooked, and the storage site should be thoroughly selected [31]. A T&S focused study should, therefore, consider the geographical characteristics of the case study and a possible optimized topology for the T&S network [27,28,31]. In this study, a simplified, nonetheless conservative approach was adopted based on specific unitary costs for pipeline transportation and underground CO_2 injection. Notably, in a European context, many studies reveal that many (over 70 %) CO_2 emissions sources (cement plants, coal power plants, refineries, steel plants) are in close proximity (<100 km to potential onshore geological storage sites) [29,33]. Under optimistic assumptions, it is conceivable to establish a low-cost network with short transportation distances and several onshore injection sites [33]. Nevertheless, numerous studies consider a conservative scenario with few CO_2 offshore storage sites located in the North Sea [27,28]. Indeed, the only large-scale CO_2 sequestration projects currently in operation in Europe involve offshore CO_2 injection in the North Sea [34]. Furthermore, many European countries are restricting the realization of onshore CO_2 injection facilities [28].

In this study, three distinct T&S scenarios are considered (Table 2): (i) a base-case with CO_2 transportation over a distance of 680 km [30] with large size pipelines and offshore underground injection, to consider transportation from northern central Europe to Rotterdam; (ii) a best-case scenario with transportation distance of 100 km in small-medium

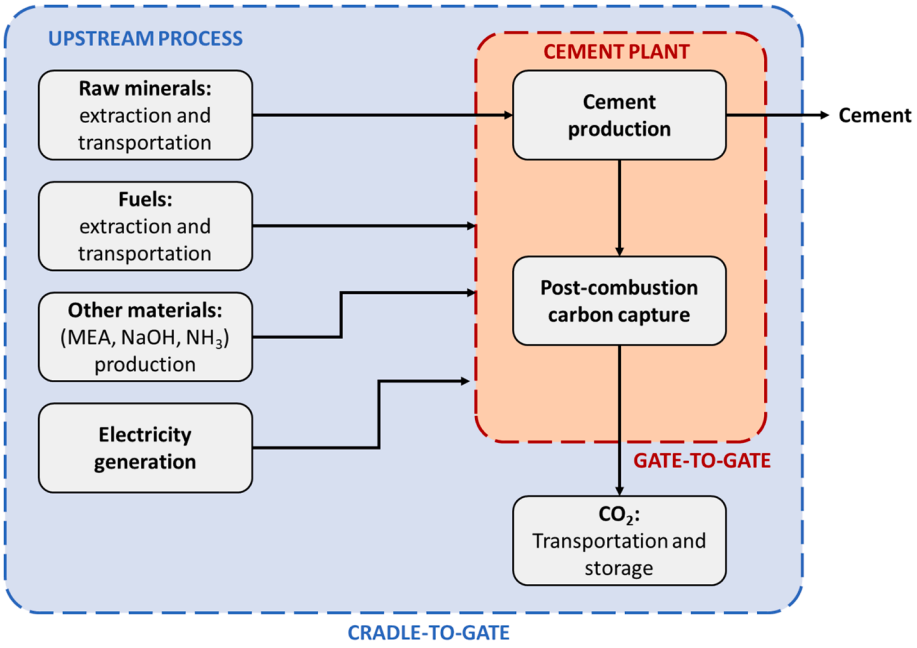

Fig. 2. Representation of the process boundaries (GtG and CtG) considered in the environmental and economic analyses.

size pipelines and onshore underground injection and (iii) a worst-case with transportation distance over 980 km in large size pipeline with offshore injection, which cover CO_2 transport from southern continental Europe to Rotterdam and the Nord Sea storage sites. Both in the base case and the worst case scenario an offshore distance of 180 km was considered [30].

As a comparison, Northern Light (Norway, commercial start-up expected in 2024 [35]) will be the first CCS project aiming to develop a large-scale CO_2 T&S network allowing different industries to capture, transport, and sequester underground their CO_2 emissions. The project is planning to capture CO_2 from industrial emitters in the Oslo-fjord region (cement plant and waste incinerator). The captured CO_2 will then be shipped to a common terminal on the Norwegian west coast, from where it will be transported by pipeline (around 100 km long) to an offshore storage facility [35].

3. Methods

The results presented in this work were obtained through different analyses. The mass and energy balance of the system was obtained through a detailed process modeling of all the main plant sections: clinker kiln, PCCC unit, CHP, and CO_2 compression. The clinker kiln and CO_2 compression were modeled considering the Peng Robison equation of state, for the CHP the IAPWS formulation 1995 (IAPWS-95) was selected, and the unsymmetric electrolyte Non-Random Two Liquid with Redlich-Kwong (NRTL-RK) equation of state was adopted for the PCCC unit. Furthermore, the absorption and the stripper columns of the PCCC unit were modeled with rigorous rate-based columns and kinetically driven reactions following the most recent literature available on the subject [22,37,38]. All the main assumptions used in the process modeling of the plant are reported in the supplementary material (section S1). The results obtained were then used in the economic analysis for the estimation of the operative (OPEX) and capital costs (CAPEX). A simplified life cycle analysis for the estimation of carbon footprint (equivalent CO_2 emissions) and total primary energy demand of the production process was also performed. In detail the following upstream processes were considered: electricity generation, extraction and transportation of the fuels and the raw materials, CO_2 transportation and storage, and production of the MEA and other (NaOH e NH_3) solvents (Fig. 2). The equivalent CO_2 emissions coming from the infrastructure realization and use were considered negligible as many times reported in the literature for energy-intensive processes [11,39]. The upstream emission and primary energy consumption (reported in supplementary material, section S2) were obtained with the open-source software OpenLCA linked with the database Environmental Footprint v3.0. Regarding grid electricity consumption the associated upstream emissions (307.8 kg_{CO2}/kWh_{el}) and primary energy demand (generation efficiency of 0.501) were obtained considering the average 2019 grid electricity mix [40]. The limestone was assumed to be extracted near the plant while for all the other raw materials transportation for 100 km by lorry was assumed. CO_2 transportation with pipelines and underground long-term storage (underground off-shore injection) was also taken into account considering fixed specific emissions, electricity consumption, and unitary costs according to the literature [27,36] (supplementary material section S2). The economic and environmental performances of the CCS integrated system were then evaluated through the estimations of key performance indicators (KPIs). In this regard, the KPIs were calculated considering two different system boundaries (Fig. 2): (i) Gate-to-Gate (GtG) boundary that considers only the material and energy balance of the plant and the CO_2 emissions at the plant stack. In this case, the costs for CO_2 transportation and storage were not considered. (ii) Cradle-to-Gate (CtG) boundary that also considers the primary energy consumption and equivalent CO_2 emissions of the upstream emissions. In this case, the costs for CO_2 transportation and storage were included.

Table 3
Main assumption used for the CAPEX estimation.

Parameter	Symbol	Value	Unit	Reference
Service costs for engineering, procurement, and construction	f_{EPC}	17.5	%	[42]
Process contingency	$f_{process}$	10	%	[42]
Project contingency	$f_{project}$	15	%	[42]
Owner's cost	f_{OC}	20	%	[42]
Construction time	–	5	years	[42]
Discount rate	r	8	%	[5]
Plant operating life	N	25	years	[5]

Table 4
OPEX assumptions.

Parameter	Value	Unit	Reference
Cement plant capacity factor	91.3	%	[43]
Coal	10.8	€/MWh_{LHV}	[44]
Natural gas (2019)	20.2	€/MWh_{LHV}	[45]
Electricity (2019)	59.4	€/MWh_{el}	[45]
Raw minerals	3.012	€/t	[44]
Water cost	1	€/m^3	[46]
MEA cost	1450	€/t	[5]
Ammonia solution for NO_x removal	130	€/t	[5]
Sodium hydroxide for SO_x removal	370	€/t	[5]
Other variables OPEX	0.8	€/t_{cem}	[9]
Fixed OPEX (cement plant)	8.7	% of TPC	Obtained from [9]
Fixed OPEX (PCCC plant)	7.7	% of TPC	Obtained from [9]

3.1. Economic assessment

The CAPEX and OPEX of the integrated plant were estimated with a detailed economic analysis. Regarding the CAPEX, the Bare Erected Costs (BEC) of the main components were calculated from Eq. (1) [41], where the Equipment Costs (EC) were obtained considering cost functions from literature and reported in the supplementary material (section S3) and the bare module factors (F_{BM}) taken from Turton et al. [41]. The EC obtained were then updated using the 2019 CEPCI index ($CEPCI_{2019} = 607.5$) and converted in euro with a dollar to euro conversion of 0.893 (2019 average). The total as-spent capital (TASC) of the plant was then estimated following the NETL guidelines [42], through the calculation of the engineering procurement and construction cost (EPCC, Eq. (2), the total plant cost (TPC, Eq. (3), and the total overnight cost (TOC, Eq. (4). Different multiplying factors (Table 3) were considered to account for the engineering, procurement, and construction service costs (f_{EPC}), process and project contingencies ($f_{process}$, $f_{project}$), owner's costs (f_{OC}), and plant construction time of 5 years (Eq. (5). The CAPEX annual ($CAPEX_{ann.}$) cost was then evaluated through the annuity method (Eq. (6) considering a discount rate of 8 % and plant operating lifetime of 25 years. The OPEX was then estimated considering the materials and energy consumptions obtained through the process modeling and the assumptions summarized in Table 4.

$$BEC = \sum BEC_i = EC_i \cdot F_{BM} \tag{1}$$

$$EPCC = BEC \bullet (1 + f_{EPC}) \tag{2}$$

$$TPC = \left(EPCC + f_{process} \bullet BEC\right) \bullet (1 + f_{project}) \tag{3}$$

$$TOC = TPC \bullet (1 + f_{OC}) \tag{4}$$

$$TASC = TOC \bullet 1.154 \tag{5}$$

$$CAPEX_{ann.} = TASC \cdot \frac{r \cdot (1+r)^N}{(1+r)^N - 1} \tag{6}$$

3.2. Key performance indicators

The environmental and economic performances were evaluated through the analysis of different KPIs, in detail:

- Specific plant fuel consumption – q_{GtG} [GJ_{LHV}/t_{cem}]: ratio between the total fuel consumption (sum of the clinker kiln and the CHP consumptions) of the plant (GtG boundary) and the cement production.
- Specific plant electricity consumption – el_{GtG} [GJ_{el}/t_{cem}]: specific net plant electricity consumption (considering the electricity produced at the CHP and used in the PCCC system and cement plant).
- Specific CO_2 GtG emissions – e_{GtG} [$kg_{CO2,eq}/t_{cem}$] specific CO_2 emissions at the plant stack.
- Specific primary energy demand – ped_{CtG} [GJ_{LHV}/t_{cem}], obtained by adding at the plant fuel consumption, q_{GtG}, the primary energy consumption of the upstream process, $ped_{upstream}$ (CtG boundary).

$$ped_{CtG} = q_{GtG} + ped_{upstream} \tag{7}$$

- Specific CtG equivalent CO_2 emissions – e_{CtG} [$kg_{CO2,eq}/t_{cem}$], obtained through the sum of e_{GtG} and the total equivalent CO_2 upstream emissions.

$$e_{CtG} = e_{GtG} + e_{upstream} \tag{8}$$

- Avoided CO_2 – AC_{GtG} [%]: reduction of the CO_2 emissions at the plant stack (e_{GtG} [kg_{CO2}/t_{cem}]) with CCS integration respect with the emissions of the unabated reference plant ($e_{GtG,ref}$ [kg_{CO2}/t_{cem}]).

$$AC_{GtG} = \frac{e_{GtG,ref.} - e_{GtG}}{e_{GtG,ref.}} \tag{9}$$

- Equivalent Avoided CO_2 – AC_{CtG} [%]: reduction of the CtG CO_2 emissions (e_{CtG} [kg_{CO2}/t_{cem}]) with CCS integration respect with the emissions of the unabated production process ($e_{CtG,rif}$ [kg_{CO2}/t_{cem}]).

$$AC_{CtG} = \frac{e_{CtG,ref.} - e_{CtG}}{e_{CtG,ref.}} \tag{10}$$

- Avoided-Stored CO_2 ratio – ASR [-]: ratio between the effective reduction in the cradle to gate emissions of the cement production process and the respective amount of CO_2 stored underground ($m_{CO2,stored}$ [kg_{CO2}/t_{cem}]).

$$ASR = \frac{e_{CtG,ref.} - e_{CtG}}{m_{CO_2,stored}} \tag{11}$$

- Specific fuel consumption per avoided CO_2 – SFC_{GtG} [MJ/kg_{CO2}] ratio between the total plant fuel consumption and the amount of avoided CO_2 emissions at the plant stack (gate-to-gate boundary).

$$SFC_{GtG} = \frac{q_{GtG} - q_{GtG,ref}}{e_{GtG,ref} - e_{GtG,eq}} \tag{12}$$

- Specific primary energy consumption per equivalent CO_2 avoided – $SPECCA_{CtG}$ [$MJ/kg_{CO2,eq}$] ratio between the increase in the cradle-to-gate primary energy consumption and the reduction in the cradle-to-gate equivalent CO_2 emissions.

$$SPECCA_{CtG} = \frac{ped_{CtG} - ped_{CtG,ref}}{e_{CtG,ref} - e_{CtG,eq}} \tag{13}$$

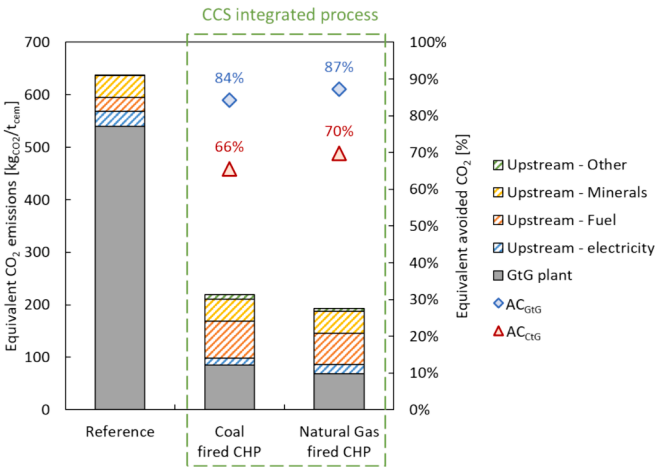

Fig. 3. Detail on the equivalent CO_2 emissions with and without CCS integration.

- Levelized gate-to-gate cost of cement – $LCOC_{GtG}$ [€/t_{cem}] cement production cost, comprehensive of CAPEX and OPEX of the integrated plant and the savings for the CHP electricity production, with the integration of PCCC unit and divided by the annual cement production (M_{cem} [t_{cem}/y]). The costs for CO_2 transportation and storage are thus not included.

$$LCOC_{GtG} = \frac{CAPEX_{ann.} + OPEX - SAVINGS}{M_{cem}} \tag{14}$$

- Levelized cradle-to-gate cost of cement production – $LCOC_{CtG}$ [€/t_{cem}]: production cost considering both the cost for the PCCC unit integration and the annual costs ($C_{T\&S}$) for transportation and storage of the captured CO_2.

$$LCOC_{CtG} = \frac{CAPEX_{ann.} + OPEX - SAVINGS + C_{T\&S}}{M_{cem}} \tag{15}$$

- Cost of avoided CO_2 [€/t_{CO2}]: ratio between the cost for the integration of the PCCC system into the plant and the GtG CO_2 emissions.

$$CAC_{GtG} = \frac{LCOC_{GtG} - LCOC_{GtG,ref}}{e_{GtG,ref} - e_{GtG}} \tag{16}$$

- Cost of equivalent avoided CO_2 [€/$t_{CO2,eq}$]: ratio between the global costs comprehensive both of the PCCC integration into the plant and the CO_2 transportation and storage divided by the reduction of the equivalent CtG CO_2 emissions.

$$CAC_{CtG} = \frac{LCOC_{CtG} - LCOC_{CtG,ref}}{e_{CtG,ref} - e_{CtG}} \tag{17}$$

4. Results and discussion

4.1. System performances

As illustrated in Fig. 3, the integration of PCCC into a cement plant yields in a substantial reduction in GHG emissions. Considering a GtG boundary, a reduction of around 84-87 % (coal fired CHP-NG fired CHP) is achieved, while a 66-70 % reduction in the equivalent CO_2 emissions is estimated for a CtG boundary. On the other hand, the heat duty of the PCCC unit is estimated at around 3.3 MJ per kg of captured CO_2 and only 7–9 % of this demand can be met with heat recovery. Consequently, its

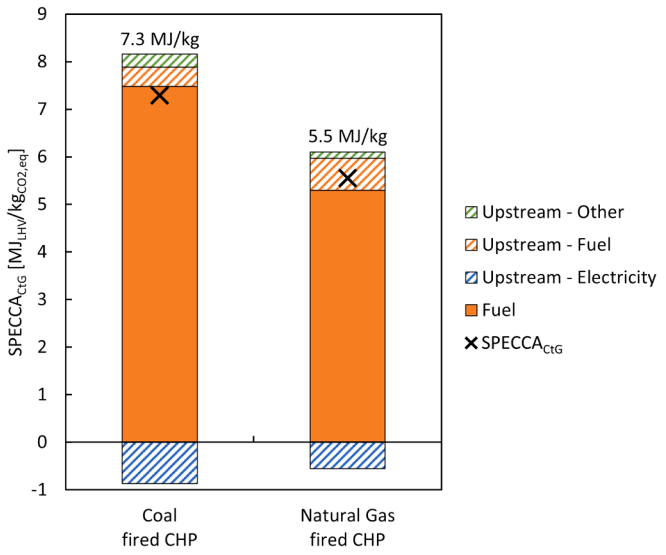

Fig. 4. Breakdown of the primary energy consumption in a cradle-to-gate approach.

integration in the production plant increases the plant fuel consumption (+160 % coal CHP and +120 % NG CHP) resulting in a SFC$_{GtG}$ of 6.9 MJ/kg$_{CO2}$ and 5.0 MJ/kg$_{CO2}$ respectively for the coal fired CHP and the NG fired CHP (Table 4).

As shown in the system carbon balance, reported in the Sankey diagrams Fig. 5 and Fig. 6, the increase in the plant fuel consumption rises, of course, also the CO_2 produced by combustion processes. Therefore, the overall GtG emissions reduction is lower than the absorber carbon capture rate (90 %). When a CtG boundary is considered, furthermore, a lower value of AC$_{GtG}$ is observed due to the influence of the upstream equivalent CO_2 emissions not abated by CCS. In this regard, the larger upstream emissions originate from the raw mineral supply and the fossil fuel supply. For all these reasons, the net amount of CtG avoided CO_2 is always lower than the amount of CO_2 captured and stored underground, where this difference mainly depends on the carbon intensity of the fuel used in the steam generation system. For example, the ASR index is 0.56 in the coal scenario, while in the NG scenario is 0.74. It is, therefore, imperative to estimate the energy and economic performance of the overall CCS value chain in relation to the net avoided CO_2 rather than the amount of captured CO_2.

Concerning the process energy balance (reported in Table 5), the primary energy consumption increases from 3.42 GJ$_{LHV}$/t$_{cem}$, obtained (for the reference plant) to 6.47 GJ$_{LHV}$/t$_{cem}$ for the PCCC-integrated plant and the coal fired CHP scenario (+90 %) or 5.89 GJ$_{LHV}$/t$_{cem}$ for the NG scenario (+70 %). SPECCA$_{CTG}$ values of 7.3 MJ$_{LHV}$/kg$_{CO2,eq}$ (coal fired CHP) and 5.5 MJ$_{LHV}$/kg$_{CO2,eq}$ (NG fired CHP) are thus obtained, where the larger contribution is represented by the consumption of fossil fuel for steam production, (Fig. 4). It should be noted that the use of the CHP setup with the back-pressure steam turbine reduces the grid electricity consumption of the plant thus reducing also the upstream equivalent CO_2 emissions and primary energy consumption.

Regarding the production costs (see Table 5), the addition of the PCCC unit in the cement plant increases the LCOC$_{GtG}$ by around +70 %: from 40.4 €/t$_{cem}$ (reference plant) to around 66–67 €/t$_{cem}$, corresponding to a cost of CO_2 avoided (CAC$_{GtG}$) of 56.7 €/t$_{CO2}$ for the coal scenario and 55.7 €/t$_{CO2}$ for the NG scenario (Fig. 7 a.). The operative costs (OPEX) constitute around 70 % of the gross costs, while the reduction of the electricity consumption of the plant allows savings of around 11–8 %. When a CtG boundary is considered (and costs for CO_2 transportation and storage are accounted for), the cement production (LCOC$_{CtG}$) cost increases to 89.0–85.0 €/t$_{cem}$ considering the base case scenario for T&S, resulting in a cost of equivalent avoided CO_2 (CAC$_{CtG}$) of 116 €/t$_{CO2}$ with coal consumption in the CHP and 100 €/t$_{CO2}$ with NG fired CHP (Fig. 7 b.). T&S costs thus account for around 41–47 % (41–55 € per tonne of avoided CO_2) of the total CAC$_{CtG}$ in the base case scenario. The CAC$_{CtG}$ index, furthermore, varies considerably depending on the T&S scenario, ranging from 75 to 82 €/t$_{CO2}$ in the best-case scenario to 107–125 €/t$_{CO2}$ in the worst-case scenario. It should be noted that, in all the cases considered, higher T&S costs (expressed as € per tonne of avoided CO_2) were associated with a coal-fired CHP due to the high ratio between the effective avoided CO_2 and the captured and stored CO_2 (ASR index) and characteristic of this case.

4.2. Optimization of absorber and stripper size

Fig. 8 shows a sensitivity analysis of the energy consumption (SFC$_{GtG}$ and SPECCA$_{CtG}$) and the cost of avoided CO_2 (CAC$_{GtG}$ and CAC$_{CtG}$) for absorber packing heights ranging from 10 m to 60 m. The diameter of

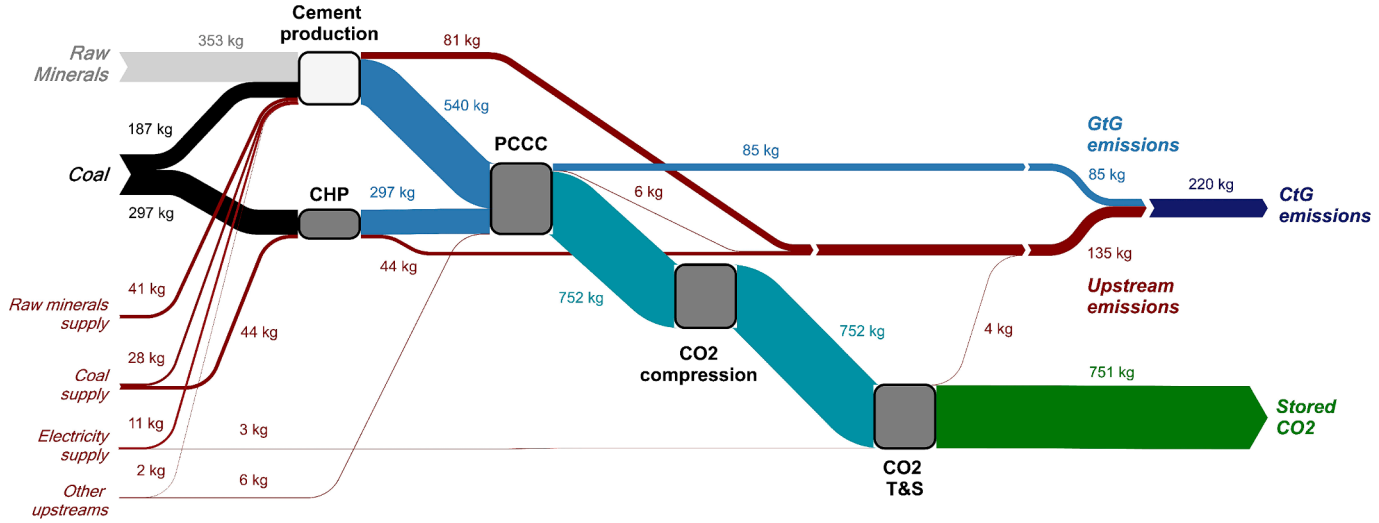

Fig. 5. Carbon Sankey of the CCS integrated production process (coal fired CHP and base-case T&S scenario). The data are expressed in kg of equivalent CO_2 per t of cement [kg$_{CO2,eq}$/t$_{cem}$].

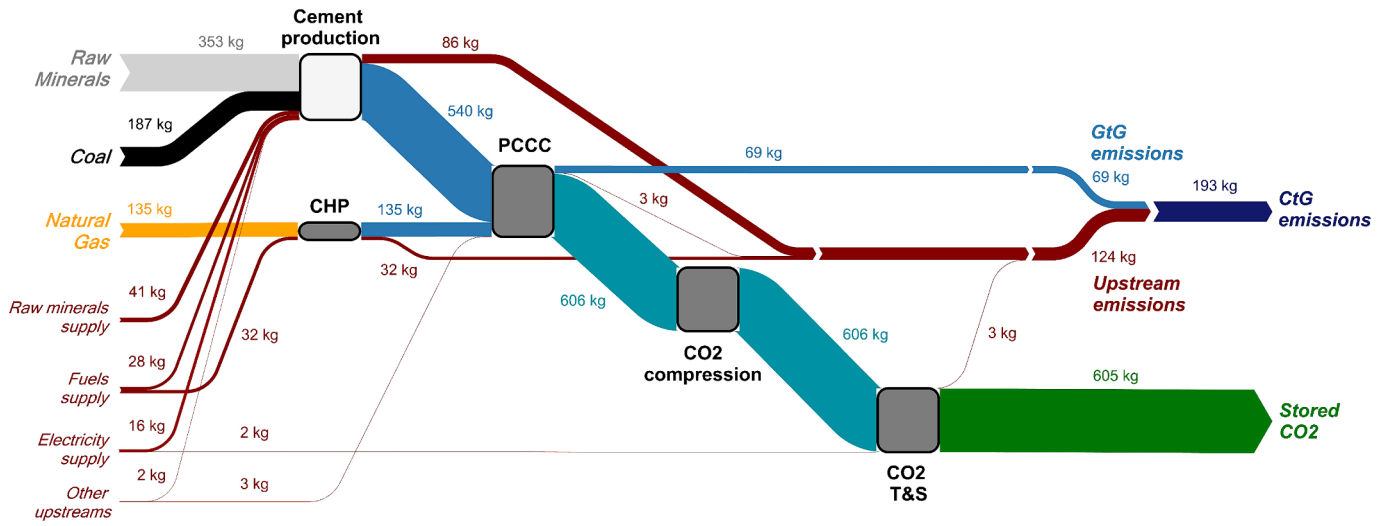

Fig. 6. Carbon Sankey of the CCS integrated production process (coal fired CHP and base-case scenario). The data are expressed in kg of equivalent CO_2 per t of cement [$kg_{CO2,eq}/t_{cem}$].

Table 5
Environmental and economic performances of the systems analyzed. Results for absorber packing height of 30 m and stripper packing height of 7 m.

Parameter	Symbol	Referenceplant	Coal fired CHP	NG fired CHP	Unit
Annual cement production	M_{cem}	1,448,336	1,448,336	1,448,336	t_{cem}/y
Coal consumption		796,685	2,056,876	796,685	MWh_{LHV}/y
Natural gas consumption		0	0	948,896	MWh_{LHV}/y
Electricity consumption		131,799	50,382	74,616	MWh_{LHV}/y
Cooling demand		0	−961,941	−851,329	MWh_{th}
Specific plant fuel consumption	q_{GtG}	1.98	5.11	4.34	GJ_{LHV}/t_{cem}
Specific plant electricity consumption	el_{GtG}	0.33	0.13	0.19	GJ_{el}/t_{cem}
Absorber Carbon Capture Rate		0	90	90	%
Captured CO_2		0	752	606	kg_{CO2}/t_{cem}
Specific CO_2 emissions at the plant stack	e_{GtG}	540	85	69	kg_{CO2}/t_{cem}
Avoided CO_2 emissions at the plant stack	AC_{GtG}	–	84.3	87.3	%
PCCC steam consumption per CO_2 captured		–	3.3	3.3	MJ/kg_{CO2}
of which supplied by heat recovery		–	0.24	0.30	MJ/kg_{CO2}
Specific fuel consumption per avoided CO_2	SFC_{GtG}	–	6.9	5.0	MJ_{LHV}/kg_{CO2}
Levelized cost of cement w/o CO_2 T&S	$LCOC_{GtG}$	40.4	66.2	66.7	€/t_{cem}
Cost of Captured CO_2		–	34.3	43.3	€/t_{CO2}
Cost of Avoided CO_2 (GtG boundary)	CAC_{GtG}	–	56.7	55.7	€/t_{CO2}
Specific primary energy consumption	ped_{CtG}	3.42	6.47	5.89	GJ_{LHV}/t_{cem}
Specific equivalent CO_2 emissions	e_{CtG}	638	220	193	$kg_{CO2,eq}/t_{cem}$
CO_2 stored underground	$m_{CO2,stored}$	0	751	605	kg_{CO2}/t_{cem}
Equivalent Avoided CO_2	AC_{CtG}	–	65.6	69.8	%
Avoided-Stored CO_2 ratio	ASR	–	0.56	0.74	–
Specific Primary Energy Consumption per equivalent CO_2 Avoided	$SPECCA_{CtG}$		7.3	5.5	$MJ_{LHV}/kg_{CO2,eq}$
BASE CASE T&S					
Levelized cost of cement with CO_2 T&S	$LCOC_{CtG}$	40.4	89.0	85.0	€/t_{cem}
Cost of Stored CO_2			64.7	73.8	€/t_{CO2}
Cost of equivalent Avoided CO_2 (CtG boundary)	CAC_{CtG}		116.0	100.2	€/t_{CO2}
BEST CASE T&S					
Levelized cost of cement with CO_2 T&S	$LCOC_{CtG}$	40.4	74.9	73.6	€/t_{cem}
Cost of Stored CO_2			45.9	55.0	€/t_{CO2}
Cost of equivalent Avoided CO_2 (CtG boundary)	CAC_{CtG}		82.1	74.5	€/t_{CO2}
WORST CASE T&S					
Levelized cost of cement with CO_2 T&S	$LCOC_{CtG}$	40.4	92.5	87.9	€/t_{cem}
Cost of Stored CO_2			69.4	78.5	€/t_{CO2}
Cost of equivalent Avoided CO_2 (CtG boundary)	CAC_{CtG}		124.7	106.7	€/t_{CO2}

the column was obtained considering a maximum column flooding of 80 %. An increase in the absorber size reduces the amount of solvent recirculating in the system, and, thus, the heat demand of the stripper reboiler (as also reported by Madeddu et al. [22]). A decrease in the stripper heat duty leads to reduced fuel consumption for steam generation and CO_2 production from combustion in the CHP. Consequently, taller absorbers result in reduced OPEX at the expense of larger CAPEX. A too small absorber may, therefore, entail high costs due to high OPEX and a too large absorber may entail high costs due to high CAPEX. In this work a sensitivity analysis was carried out to investigate the variation of the CAC_{GtG} and CAC_{CtG} values with the absorber size and a minimum value of CAC_{GtG} for a height of around 30 m was obtained for both a coal-fired CHP, Fig. 8 a., and NG-fired CHP, Fig. 8 c.

It should be noted that when a CtG boundary is considered, the optimal system configuration may involve larger absorber sizes to also reduce the T&S costs. Taller columns, indeed, entail lower values of ASR thus reducing the T&S costs per tonne of avoided CO_2. This point was observed especially when a Coal-fired CHP is assumed. In this case, an

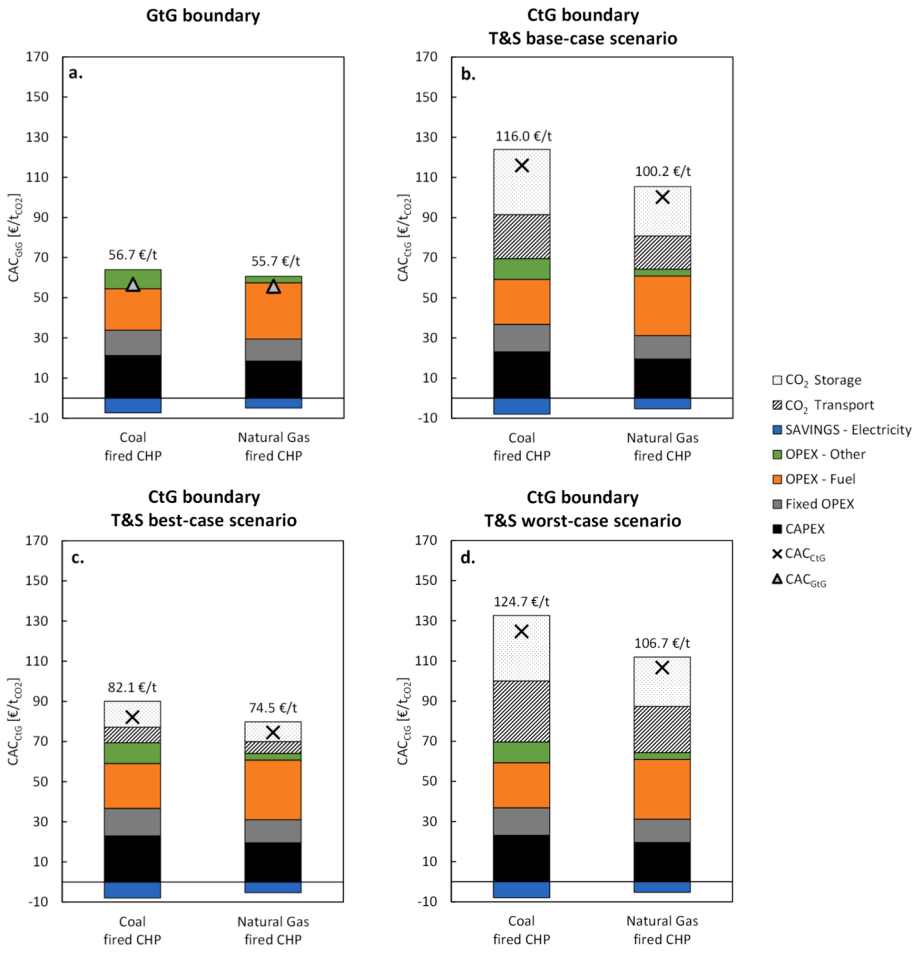

Fig. 7. Breakdown of the cost of avoided CO_2 considering a GtG (a.) and a CtG (b., c., and d.) boundaries and different T&S scenarios. Results for absorber packing height of 30 m and stripper packing height of 7 m.

optimal absorber size minimizing the CAC_{CtG} is estimated for a packing height of 40 m (base case T&S scenario), Fig. 8 b. Of course, different T&S scenarios may impact the optimal size as depicted in Table 6. Indeed, assuming a coal fired CHP, an optimal height of 30 m is observed in the best-case scenario, while, in the worst-case scenario, a size of 40 m is estimated. Notably, with an NG-fired CHP the optimal configuration is less dependent on the T&S scenario, due to the lower ASR value, and, indeed, the optimal absorber height is observed at around 30 m for all the T&S scenarios.

On the other hand, the stripper size only slightly influences the energy consumption of the system (as shown in Fig. 9). An increase in the stripper size results in an increase in the CAC_{GtG} (due to the increase in the CAPEX). In this case, the optimal stripper packing height is determined by the minimum column height-diameter ratio considered. In this work a minimum packing height of 7 m was selected, considering a minimum height-diameter ratio of 1 [22].

4.3. Heat recovery from CO_2 compression

The estimated cooling duty of the compression unit amounts to around −625 kJ/kg of captured CO_2. This amount of heat is removed from the unit intercoolers and is characterized by a low-temperature level (depending on the intercooling temperature considered, usually 25–30 °C [24,47]) and therefore is usually not recovered [6]. It is however possible to increase this temperature level by raising the intercooling temperature. In this way, this waste heat can be used to produce steam at the expense of increased electricity consumption for CO_2 compression [48]. Fig. 10 reports a sensitivity analysis of the plant energy consumption (SFC_{GtG} and $SPECCA_{CtG}$) and the costs of avoided CO_2 (CAC_{GtG} and CAC_{CtG}) for an intercooling temperature that ranges between 25 °C (reference case, no heat recovery) and 140 °C (maximum intercooling temperature considered). At increased intercooling temperature, part of the steam demand of the stripper reboiler is supplied by the heat recovery from the compression unit allowing a 20 % reduction in the fuel consumption (SFC_{GtG}) from 6.9 MJ_{LHV}/kg_{CO2} (intercooling temperature of 25 °C) to 5.4 MJ_{LHV}/kg_{CO2} (intercooling temperature of 140 °C) in the coal scenario (Fig. 10 a.) and from 5.0 MJ_{LHV}/kg_{CO2} to 4.1 MJ_{LHV}/kg_{CO2} in the NG scenario (Fig. 10 c.). On the other hand, a smaller reduction of the $SPECCA_{GtG}$ index (Fig. 10 b. and d.) is obtained due to the increase in the electricity consumption for compression. For an intercooling temperature of 60 °C, moreover, the steam production from heat recovery is not enough to compensate for the increase in primary energy consumption due to the increased electricity demand, thus resulting in a slight increase in the $SPECCA_{GtG}$ value.

This heat recovery entails not only an increase in the CAPEX, due to the installation of new heat exchangers, and a reduction in the OPEX for steam generation, but also a reduction in electricity savings. For these reasons, when a GtG boundary and an electricity cost of 59.4 €/MWh are

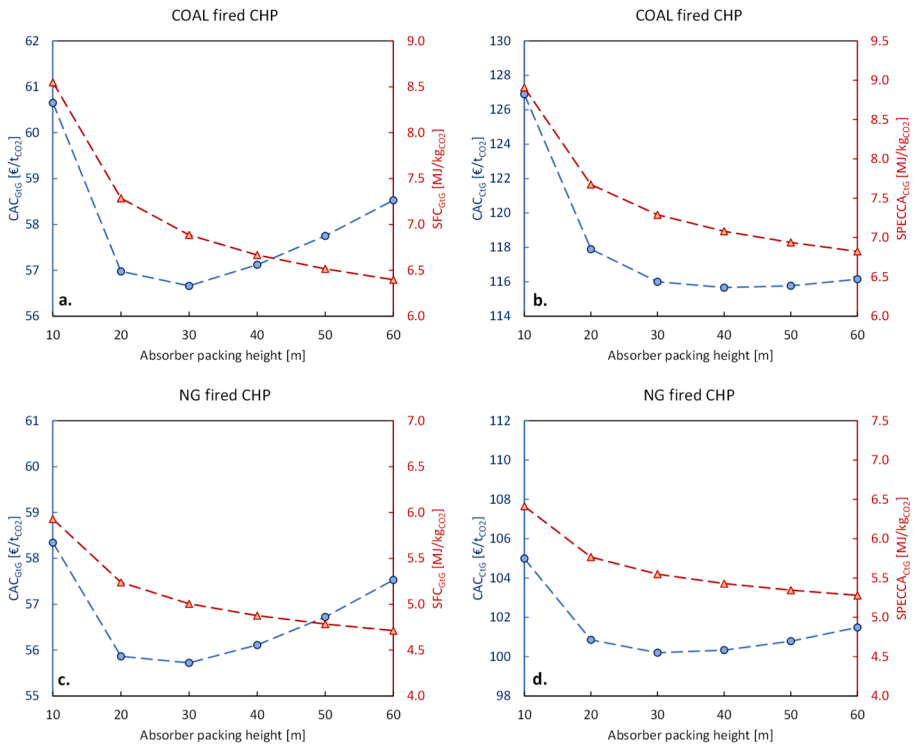

Fig. 8. Relation between the absorber packing height and the cost of avoided CO_2 with a GtG analysis boundary, CAC_{GTG} (a. e c.), and a CtG analysis boundary, CAC_{CTG} (b. and d.), in the coal-fueled CHP (a. e b.), and natural gas CHP (c. and d.). Stripper packing height fixed at 7 m.

Table 6
Cost of equivalent CO_2 avoided (CAC_{CtG}) at different absorber sizes and T&S scenarios. Stripper packing height fixed at 7 m.

Absorber packing height	T&S scenario					
	Base case		Best case		Worst case	
	Coal CHP	NG CHP	Coal CHP	NG CHP	Coal CHP	NG CHP
[m]	**[€/t_{CO2}]**	**[€/t_{CO2}]**	**[€/t_{CO2}]**	**[€/t_{CO2}]**	**[€/t_{CO2}]**	**[€/t_{CO2}]**
10	126.9	105.0	89.4	78.2	136.5	111.8
20	117.9	100.9	83.2	74.9	126.8	107.4
30	116.0	100.2	82.1	74.5	124.7	106.7
40	115.7	100.3	82.3	74.8	124.2	106.8
50	115.8	100.8	82.7	75.4	124.2	107.2
60	116.1	101.5	83.3	76.2	124.5	107.9

considered, the heat recovery from CO_2 compression does not seem to be economically convenient both for the coal scenario and for the NG scenario. When an intercooling temperature of 60 °C is considered the amount of heat recoverable is not enough to compensate for the increase in the CAPEX. At higher intercooling temperatures the CAC_{GtG} decreases without, however, reaching the value of the reference configuration. Nonetheless, for lower electricity costs, such as 40 €/MWh, the increase in the electricity consumption for compression is less impactful on the CAC_{GtG} making this heat recovery more economically attractive (Fig. 10 a. and c.).

In a CtG approach, the reduction in the CHP fuel consumption leads to a better ASR ratio and thus lowers the impact of the T&S costs on the CAC_{GtG} value. This is particularly impactful when a coal-fired CHP is assumed where this additional heat recovery allows for an increase in the ASR to 0.61 (intercooling temperature 140 °C) and a reduction in the CAC_{GtG} value from 116 €/$t_{CO2}$to 113 €/t_{CO2}.

4.4. Fuel cost sensitivity

Fig. 11 shows a sensitivity analysis of the cost of avoided CO_2 (CAC_{GtG} and CAC_{CtG}) for different fuel (coal or NG) prices (−50 % and +50 % of the reference price reported in Table 4). As already explained previously, the OPEX related to fuel consumption greatly influences the CAC_{GtG} and CAC_{CtG} values. Considering a coal-fired CHP, CAC_{GtG} values that range from 46 €/t_{CO2} to 67 €/t_{CO2} and CAC_{CtG} from 106 €/$t_{CO2,eq}$ to 127 €/$t_{CO2,eq}$ (Base case T&S scenario) are obtained. For the NG scenario, instead, the CAC_{GtG} values are estimated between 41 €/t_{CO2} and 70 €/t_{CO2} and CAC_{CtG} between 85 €/$t_{CO2,eq}$ and 115 €/$t_{CO2,eq}$ (Base case T&S scenario). Notably, very similar costs between the two system configurations studied (coal-fired CHP and NG-fired CHP) are observed, especially when a GtG boundary is considered. It should also be noted that different fuel prices may impact the optimal size of the absorber. In detail, for reduced coal or NG prices (−50 %) an optimal height of 20 m is obtained (to minimize the CAC_{GtG}).

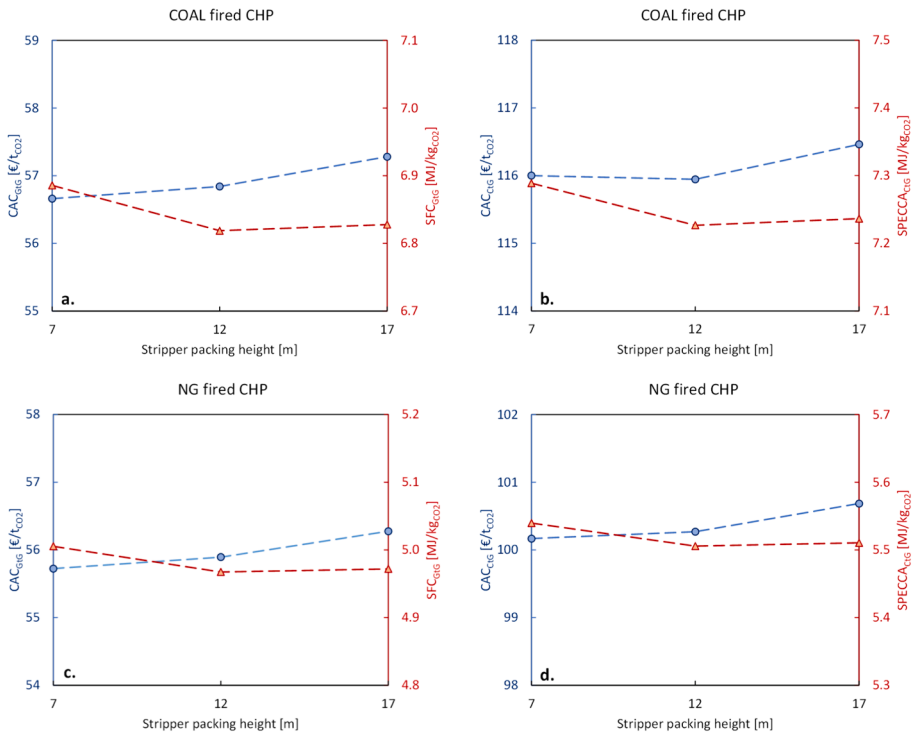

Fig. 9. Relation between the stripper packing height and the cost of avoided CO_2 with a GtG analysis boundary, CAC_{GTG} (a. e c.), and a CtG analysis boundary, CAC_{CTG} (b. e d.), in the coal-fueled CHP (a. e b.), and natural gas CHP(c. e d.). Absorber packing height fixed at 30 m.

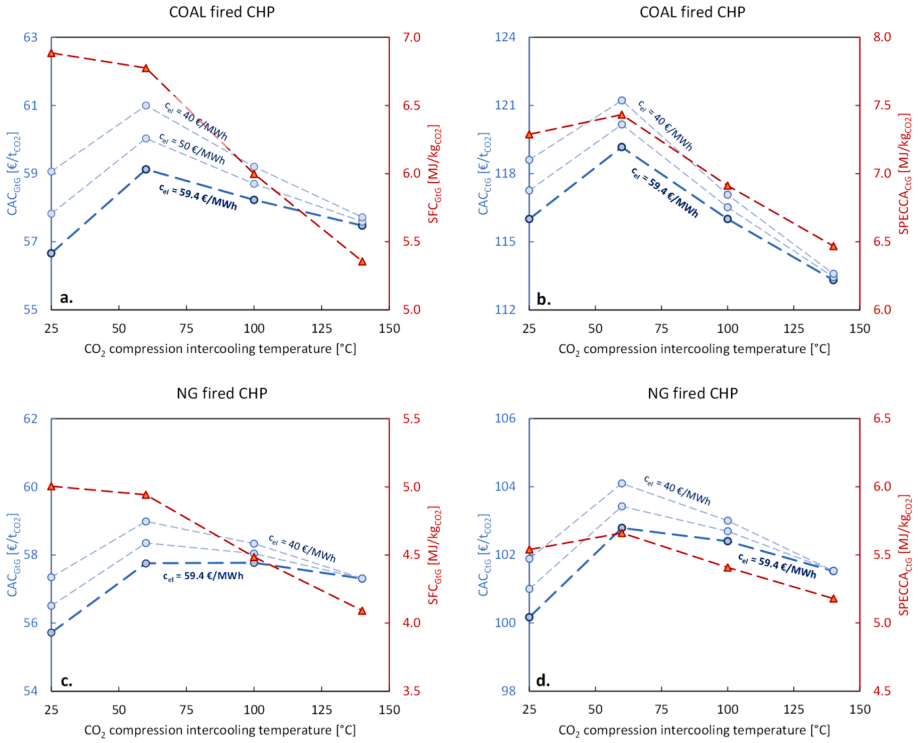

Fig. 10. Cost of avoided CO_2 (CAC_{GtG} and CAC_{CtG}) and energy consumption (SFC_{GtG} and $SPECCA_{CtG}$) with heat recovery from the CO_2 compression at different intercooling temperatures and different grid electricity prices. Results for a base case T&S scenario and a 30 m tall absorber packing and 7 m tall stripper packing.

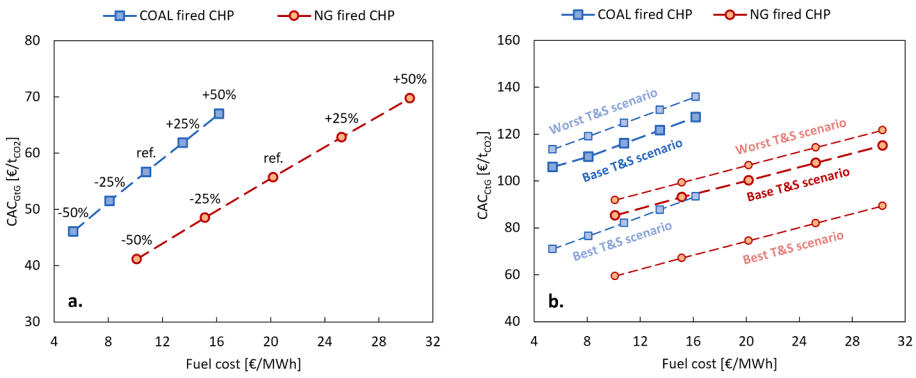

Fig. 11. Cost of avoided CO_2 at different coal and NG prices with a GtG (a.) and a CtG (b.) boundary.

5. Conclusions

In this work, a comprehensive analysis of the integration of an absorber PCCC unit in a state-of-the-art cement plant was performed. The analyzed plant layout includes the use of a CHP for steam production (needed in the stripper reboiler), the thermal integration between the cement plant and the PCCC unit, and the treatment of the CHP flue gas in the absorber. Heat recovery from CO_2 compression was also evaluated. Results obtained from the process modeling of the integrated plant were used as input for a simplified life cycle analysis and an economic analysis for the evaluation of the process carbon footprint and economic performances, considering also different possible CO_2 transport and storage scenarios (base-case, best-case, and worst-case). Results obtained considering both a GtG and a CtG boundary were compared and discussed. Furthermore, a detailed sizing of the absorber and the stripper was performed to minimize the cost of avoided CO_2. The main conclusions obtained can be summarized as follows:

- Integration of an MEA-based PCC unit in the production process can reduce the overall plant CO_2 emission by 84-87 % (GtG boundary) and the overall cement production process equivalent CO_2 emission by 66-70 % (CtG boundary) depending on the selected fuel for steam production in the CHP (i.e., coal or NG). Such emission reduction is obtained at the expense of a large increase in the plant fuel consumption between + 120 % and +160 %, where the larger increase is associated with a coal-fired CHP while the lower to an NG-fired CHP.
- The system boundary (GtG or CtG) greatly influences the estimated production costs. The levelized cost of cement is around 66–67 €/t_{cem} when a GtG boundary is considered, while it goes up to 85-89 €/t_{cem} when a CtG boundary is applied and a base case scenario for CO_2 transport and storage is considered. This difference is due to the T&S costs, which are considered only within the CtG boundary. Similarly, the costs of the avoided CO_2 are estimated around 56–57 €/t_{CO2} with a GtG boundary (CAC_{GtG}) and around 75–125 €/$t_{CO2,eq}$ with a CtG boundary (CAC_{CtG}).
- The costs of equivalent avoided CO_2 (CAC_{CtG}) are strongly influenced by the T&S scenario considered with estimated values of around: (i) 85–89 €/t_{CO2} in the base case scenario (500 km transportation and offshore injection); (ii) 75–82 €/t_{CO2} in the best-case scenario (short distance transportation and onshore injection); and (iii) 107–125 €/t_{CO2} in the worst-case scenario (long distance transportation and offshore injection).
- The absorber and stripper columns of the PCCC system can be sized to minimize the cost of avoided CO_2. In detail, an absorber packing height of 30 m and a stripper packing height of 7 m are obtained to minimize the CAC_{GtG}. The absorber can also be sized to minimize the CAC_{CtG}. In this case, taller columns may be needed to reduce the ratio between the equivalent avoided CO_2 and the stored CO_2 reducing, thus, also the cost for CO_2 transportation and storage per avoided CO_2.
- By increasing the intercooling temperature in the CO_2 compression unit is possible to use part of the heat produced in the intercoolers for steam production thus reducing the fuel consumption of the plant. This integration entails, however, an increase in the CAPEX and an increase in the grid electricity consumption. This integration may result in economic convenience with low electricity prices and when highly carbon-intensive fuels (coal) are used in steam production.

Future works stemming from this study may be several, such as: (i) investigation of the impact of alternative steam generation process on the performances of the PCCC integrated system (steam generation with electric boiler, heat pumps, waste-derived fuels); (ii) environmental and economic analysis, with a similar methodology to the one here described, of CCS integration in other heavy industries and industrial process; (iii) investigation of the integration of alternative PCCC technologies at lower TRL (i.e. calcium looping, temperature or pressure swing adsorption, membrane separation, etc.).

CRediT authorship contribution statement

Daniele Ferrario: Writing – review & editing, Writing – original draft, Visualization, Methodology, Conceptualization. **Tobias Pröll:** Writing – review & editing, Supervision, Methodology, Funding acquisition, Conceptualization. **Stefano Stendardo:** Writing – review & editing, Supervision, Funding acquisition. **Andrea Lanzini:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors 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

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cej.2024.152900.

References

[1] I.E.A. Net Zero by 2050: A Roadmap for the Global Energy Sector. 2021.
[2] C. Carbone, D. Ferrario, A. Lanzini, S. Stendardo, A. Agostini, Evaluating the carbon footprint of cement plants integrated with the calcium looping CO2 capture process, Front Sustain 3 (2022) 14, https://doi.org/10.3389/frsus.2022.809231.
[3] I.E.A. Technology Roadmap: Low-Carbon Transition in the Cement Industry. Paris: 2018.
[4] D. Ferrario, S. Stendardo, V. Verda, A. Lanzini, Solar-driven calcium looping system for carbon capture in cement plants: Process modelling and energy analysis, J Clean Prod 394 (2023) 136367, https://doi.org/10.1016/j.jclepro.2023.136367.
[5] S.O. Gardarsdottir, E. De Lena, M. Romano, S. Roussanaly, M. Voldsund, J.F. Pérez-Calvo, et al., Comparison of technologies for CO 2 capture from cement production—Part 2: Cost analysis, Energies 12 (2019) 542, https://doi.org/10.3390/en12030542.
[6] M. Voldsund, S.O. Gardarsdottir, E. De Lena, J.F. Pérez-Calvo, A. Jamali, D. Berstad, et al., Comparison of technologies for CO 2 capture from cement production—Part 1: Technical evaluation, Energies 12 (2019) 559, https://doi.org/10.3390/en12030559.
[7] M.G. Plaza, S. Martínez, F. Rubiera, CO2 Capture, Use, and Storage in the Cement Industry: State of the Art and Expectations, Energies 13 (2020) 5692, https://doi.org/10.3390/en13215692.
[8] Heidelberg Materials. Brevik CCS n.d. https://www.brevikccs.com/en.
[9] Roussanaly S, Fu C, Voldsund M, Anantharaman R, Spinelli M, Romano M. Techno-economic Analysis of MEA CO2 Capture from a Cement Kiln - Impact of Steam Supply Scenario. Energy Procedia, vol. 114, Elsevier; 2017, p. 6229–39. https://doi.org/10.1016/j.egypro.2017.03.1761.
[10] E. De Lena, M. Spinelli, A. Conversano, M.C. Romano, M. Gatti, Assessing heat pumps as heat supply option for solvent regeneration in cement plants with post-combustion CO2 capture: heat integration, energy performance and CO2 emissions, SSRN Electron J (2022), https://doi.org/10.2139/ssrn.4286141.
[11] S.E. Tanzer, K. Blok, A.R. Ramírez, Scoping cost and abatement metrics for biomass with carbon capture and storage — the example of bioCCS in cement, Int J Greenh Gas Control 125 (2023) 103864, https://doi.org/10.1016/j.ijggc.2023.103864.
[12] M.M. Jaffar, C. Brandoni, J. Martinez, C. Snape, S. Kaldis, A. Rolfe, et al., Comparative techno-economic analysis of the integration of MEA-based scrubbing and silica PEI adsorbent-based CO2 capture processes into cement plants, J Clean Prod 414 (2023) 137666, https://doi.org/10.1016/j.jclepro.2023.137666.
[13] C.C. Cormos, Decarbonization options for cement production process: A techno-economic and environmental evaluation, Fuel 320 (2022) 123907, https://doi.org/10.1016/j.fuel.2022.123907.
[14] A.M. Cormos, S. Dragan, L. Petrescu, V. Sandu, C.C. Cormos, Techno-economic and environmental evaluations of decarbonized fossil-intensive industrial processes by reactive absorption & adsorption CO2 capture systems, Energies (2020).
[15] C. Nwaoha, M. Beaulieu, P. Tontiwachwuthikul, M.D. Gibson, Techno-economic analysis of CO2 capture from a 1.2 million MTPA cement plant using AMP-PZ-MEA blend. Int J Greenh Gas, Control 78 (2018) 400–412, https://doi.org/10.1016/J.IJGGC.2018.07.015.
[16] A.M. Cormos, C.C. Cormos, Reducing the carbon footprint of cement industry by post-combustion CO2 capture: Techno-economic and environmental assessment of a CCS project in Romania, Chem Eng Res Des 123 (2017) 230–239, https://doi.org/10.1016/j.cherd.2017.05.013.
[17] H. Gerbelová, M. Van Der Spek, W. Schakel, Feasibility assessment of CO2 capture retrofitted to an existing cement plant: post-combustion vs, Oxy-Fuel Combustion Technology. Energy Procedia 114 (2017) 6141–6149, https://doi.org/10.1016/J.EGYPRO.2017.03.1751.
[18] F. Schorcht, I. Kourti, B.M. Scalet, S. Roudier, L.D. Sancho, Best Available Techniques (BAT) Reference Document for the Production of Cement, Lime and Magnesium Oxide, European Commission Joint Research Centre, 2013.
[19] D.N. Huntzinger, T.D. Eatmon, A life-cycle assessment of Portland cement manufacturing: comparing the traditional process with alternative technologies, J Clean Prod 17 (2009) 668–675, https://doi.org/10.1016/j.jclepro.2008.04.007.
[20] E. De Lena, M. Spinelli, I. Martínez, M. Gatti, R. Scaccabarozzi, G. Cinti, et al., Process integration study of tail-end Ca-Looping process for CO2 capture in cement plants, Int J Greenh Gas Control 67 (2017) 71–92, https://doi.org/10.1016/j.ijggc.2017.10.005.
[21] F. Salomone, P. Marocco, D. Ferrario, A. Lanzini, D. Fino, S. Bensaid, et al., Process simulation and energy analysis of synthetic natural gas production from water electrolysis and CO2 capture in a waste incinerator, Appl Energy (2023) 343, https://doi.org/10.1016/j.apenergy.2023.121200.
[22] C. Madeddu, M. Errico, R. Baratti, Process analysis for the carbon dioxide chemical absorption–regeneration system, Appl Energy 215 (2018) 532–542, https://doi.org/10.1016/j.apenergy.2018.02.033.
[23] Y. Le Moullec, M. Kanniche, Screening of flowsheet modifications for an efficient monoethanolamine (MEA) based post-combustion CO2 capture, Int J Greenh Gas Control 5 (2011) 727–740, https://doi.org/10.1016/j.ijggc.2011.03.004.
[24] S. Posch, M. Haider, Optimization of CO 2 compression and purification units (CO 2CPU) for CCS power plants, Fuel 101 (2012) 254–263, https://doi.org/10.1016/j.fuel.2011.07.039.
[25] Cinti G, Anantharaman R, De Lena E, Fu C, Gardarsdottir SO., Hoppe H, et al. D4.4: Cost of critical components in CO2 capture process. 2018.
[26] D.A. Snow, Plant Engineer's Reference Book, Butterworth-Heinemann, 2002 https://doi.org/978-0-7506-4452-5.
[27] F. d'Amore, M.C. Romano, F. Bezzo, Carbon capture and storage from energy and industrial emission sources: A Europe-wide supply chain optimisation, J Clean Prod 290 (2021) 125202, https://doi.org/10.1016/j.jclepro.2020.125202.
[28] F. d'Amore, M.C. Romano, F. Bezzo, Optimal design of European supply chains for carbon capture and storage from industrial emission sources including pipe and ship transport, Int J Greenh Gas Control 109 (2021) 103372, https://doi.org/10.1016/j.ijggc.2021.103372.
[29] I.E.A. ETP 2020 special report on carbon capture utilisation and storage: CCUS in clean energy transitions. 2020.
[30] Zep., The Costs of CO 2 Transport Post-demonstration CCS in the EU, Euro (2011) 1–53.
[31] T. Grant, A. Guinan, C.Y. Shih, S.M. Lin, D. Vikara, D. Morgan, et al., Comparative analysis of transport and storage options from a CO2 source perspective, Int J Greenh Gas Control 72 (2018) 175–191, https://doi.org/10.1016/j.ijggc.2018.03.012.
[32] E.S. Rubin, J.E. Davison, H.J. Herzog, The cost of CO2 capture and storage, Int J Greenh Gas Control 40 (2015) 378–400, https://doi.org/10.1016/j.ijggc.2015.05.018.
[33] E. Smith, J. Morris, H. Kheshgi, G. Teletzke, H. Herzog, S. Paltsev, The cost of CO2 transport and storage in global integrated assessment modeling, Int J Greenh Gas Control 109 (2021) 103367, https://doi.org/10.1016/j.ijggc.2021.103367.
[34] ZEP. CCS/CCU projects - Zero Emissions Platform n.d. https://zeroemissionsplatform.eu/about-ccs-ccu/css-ccu-projects/ (accessed January 30, 2024).
[35] Northern lights n.d. https://norlights.com/.
[36] Chisalita DA, Petrescu L, Cobden P, van Dijk HAJ (Eric., Cormos AM, Cormos CC. Assessing the environmental impact of an integrated steel mill with post-combustion CO2 capture and storage using the LCA methodology. J Clean Prod 2019;211:1015–25. https://doi.org/10.1016/j.jclepro.2018.11.256.
[37] C. Madeddu, M. Errico, R. Baratti, Rigorous modeling of a CO2-MEA stripping system, Chem Eng Trans 57 (2017) 451–456, https://doi.org/10.3303/CET1757076.
[38] M. Errico, C. Madeddu, D. Pinna, R. Baratti, Model calibration for the carbon dioxide-amine absorption system, Appl Energy 183 (2016) 958–968, https://doi.org/10.1016/j.apenergy.2016.09.036.
[39] C. Carbone, D. Ferrario, A. Lanzini, V. Verda, A. Agostini, S. Stendardo, Calcium looping in the steel industry: GHG emissions and energy demand, Int J Greenh Gas Control 125 (2023) 103893, https://doi.org/10.1016/J.IJGGC.2023.103893.
[40] Our World in Data. Electricity Mix n.d. https://ourworldindata.org/electricity-mix.
[41] R. Turton, J.A. Shaeiwitz, D. Bhattacharyya, W.B. Whiting, Analysis, Prentice Hall, Synthesis and Design of Chemical Processes, 2018.
[42] Theis J. Quality Guidelines for Energy Systems Studies: Cost Estimation Methodology for NETL Assessments of Power Plant Performance - Feb 2021. 2021.
[43] S. Khosravi, S. Hossainpour, H. Farajollahi, N. Abolzadeh, Integration of a coal fired power plant with calcium looping CO2 capture and concentrated solar power generation: Energy, exergy and economic analysis, Energy 240 (2022) 122466, https://doi.org/10.1016/j.energy.2021.122466.
[44] E. De Lena, M. Spinelli, M. Gatti, R. Scaccabarozzi, S. Campanari, S. Consonni, et al., Techno-economic analysis of calcium looping processes for low CO2 emission cement plants, Int J Greenh Gas Control 82 (2019) 244–260, https://doi.org/10.1016/j.ijggc.2019.01.005.
[45] Eurostat. Database - Eurostat n.d. https://ec.europa.eu/eurostat/web/main/data/database.
[46] R. Chauvy, D. Verdonck, L. Dubois, D. Thomas, G. De Weireld, D. Verdonck, et al., Techno-economic feasibility and sustainability of an integrated carbon capture and conversion process to synthetic natural gas, SSRN Electron J 47 (2021) 101488, https://doi.org/10.2139/SSRN.3811432.
[47] Voldsund M, Anantharaman R, Berstad D, Cinti G, De Lena E, Gatti M, et al. D3.2: CEMCAP framework for comparative techno-economic analysis of CO 2 capture from cement plants. 2017.
[48] L.M. Romeo, I. Bolea, Y. Lara, J.M. Escosa, Optimization of intercooling compression in CO 2 capture systems, Appl Therm Eng 29 (2008) 1744–1751, https://doi.org/10.1016/j.applthermaleng.2008.08.010.