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Assessing deployment pathways for greenhouse gas emissions reductions in an industrial plant – A case study for a complex oil refinery



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HIGHLIGHTS

- Method to assess pathways for greenhouse gas emissions reductions for industrial plants.
- Method successfully demonstrated for a large, complex oil refinery in Europe.
- We examined energy efficiency, carbon capture and storage, biomass gasification and pyrolysis.
- · Pathway with energy efficiency and BIG-CCS is most cost effective and shows deep emissions reductions.
- However, ranking of pathways in terms of costs depends strongly on energy prices.

ARTICLE INFO

Keywords: Industry Refinery Techno-economic Energy efficiency CCS Biomass

ABSTRACT

This study developed an integrated method to identify deployment pathways for greenhouse gas emissions reductions in an industrial plant. The approach distinguishes itself by assessing the techno-economic performance of combinations of mitigation options at the level of core processes of an industrial plant. Thus, synergies between mitigation options like economies of scale and negative interactions, such as overlap in emission reduction potential, are incorporated, resulting in more realistic insights into costs and associated risks. The method was successfully applied to a large petroleum refinery (~4.1 MtCO₂/y) in northwest Europe. The studied mitigation routes are: energy efficiency measures, carbon capture and storage, fast pyrolysis of woody biomass to produce infrastructure-ready transportation fuels, and gasification of torrefied wood pellets to produce electricity, hydrogen and/or Fischer-Tropsch fuels. Four deployment pathways were examined, all starting with energy efficiency measures and followed by (1) oxyfuel combustion capture, (2) post-combustion capture, (3) biomass gasification, or (4) biomass gasification with carbon capture and storage. Pathway 4 is most cost-effective under medium assumptions, regardless of the emissions reduction target, and allows for deep emissions reductions (6.3 MtCO₂-eq/y; 154% reduction compared to the 2012 base case). For a 75% emissions reduction target, the average avoidance cost of pathway 4 is around - 25 €2012/tCO2-eq. In comparison, the second most cost-effective pathway (1) was evaluated at average avoidance cost of -5€2012/tCO2-eq. However, the ranking of the pathways in terms of avoidance cost depends heavily on future energy prices.

1. Introduction

To meet the targets set at the climate summit in Paris, the European Union (EU) plans to reduce greenhouse gas (GHG) emissions in the manufacturing industry and petroleum refineries (hereafter jointly referred to as 'industrial sector') by 83–87% below the

1990 level in 2050 [1]. To this end, a broad portfolio of GHG mitigation options needs to be considered [2]. To date, many studies have investigated the GHG reduction potential at the aggregate or sector level by means of energy efficiency measures (EEMs), e.g. [3–6] carbon capture and storage (CCS), e.g. [8,9], switching to low-carbon energy and raw material supply sources, e.g. [7], or renewable energy

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Nomenc	lature	BM	biomass
		CAC	catalytic cracking
α	annuity factor (–)	CCS	carbon capture and storage
Be	benefit in terms of energy savings (€/y)	CHP	combined heat and power
C _e	cost for energy efficiency measures (ξ/y)	Combi	combination of mitigation options
Ca	GHG avoidance cost (€/tCO ₂ -eq)	DCC	direct contact cooling
$\Delta C_{O\&M}$	net change in O&M cost (€/y)	DCCI	downstream capital costs index
$\Delta E_{\rm e}$	net change in electricity consumption (GJ_e/y)	DPC	drying purification cooling
$\Delta E_{\rm f}$	net change in fuel consumption (GJ_f/y)	EEM	energy efficiency measure
ΔE_{ng}	net change in natural gas consumption (GJ_{ng}/y)	EU	European Union
GHG _a	avoided GHG emissions (tCO ₂ -eq/y)	F	furnaces
	wnstream net change in annual downstream GHG emissions	FT	fischer-tropsch
ΔGIIGdov	(tCO_2-eq/y)	GEA	global energy assessment
ACUC .		GEA	gasifier
ΔGΠG _{pla}	ntnet change in annual GHG emissions from the industrial	GHG	
ACHC	plant (tCO ₂ -eq/y)		greenhouse gas
допо _{пря}	stream net change in annual upstream GHG emissions (tCO ₂ -	GT	gas turbine
т	eq/y)	HHV	higher heating value
I	upfront investment cost (€)	HP	high-pressure
I _i	investment cost of component $i \in \mathbb{C}$	IEA	international energy agency
I _{i,ref}	investment cost of component i in reference system (\mathfrak{C})	IPCC	intergovernmental panel on climate change
LT	economic lifetime (y)	LHV	lower heating value
N _{units, i}	number of parallel process trains per system for compo-	LPG	liquid petroleum gas
	nent i	LT	economic lifetime
N _{units, i, 1}	$_{\text{ref}}$ number of parallel process trains for component i in the	MD/G	middle distillate/gasoline
	reference system of trains	MEA	solvent: monoethanolamine
PBP	simple pay-back period (y)	MT	medium term
P_{e}	price of electricity (€/GJ _e)	NG	natural gas
P_f	price of fuel (\mathbb{C}/GJ_f)	NGCC	natural gas combined cycle
P_{ng}	price of natural gas (€/GJ _{ng})	OXY	oxyfuel combustion capture
PPC	process plant cost (€)	PC	pulverized coal
r	real discount rate (%)	PCC	post-combustion capture
SF_i	economic scaling factor for component i (–)	p-oil	pyrolysis oil
SF_n	economic scaling factor for multiple trains <i>n</i> (–)	POST	post-combustion capture
s_{i}	capacity of single process train i (unit: component de-	ppm(v)	parts per million (by volume)
	pendent)	PRE	pre-combustion capture
$s_{i,ref}$	capacity of single process train <i>i</i> in reference system (unit:	PSA	pressure swing adsorption
	component dependent)	RG	refinery gas
TCR	total capital requirement (€)	RTS	Reference Technology Scenario of the IEA
TPC	total plant cost (€)	SCR	selective catalytic reduction
		SER	specific energy requirement
Abbrevia	tions	SR	steam reformer
		SRU	sulphur recovery unit
2DS	2 °C scenario of the IEA	ST	short term
ADIP-X	solvent: methyldiethanolamine mixed with piperazine	TOPS	torrefied wood pellets
AGR	acid gas removal	Vent	CO ₂ emissions vented to the atmosphere
ASU	air separation unit	WEO	world energy outlook publication of the IEA
ATR	autothermal reforming	WH	waste heat
BF	biofuels	WGS	water-gas shift
BIG	biomass gasification	11 00	acoz ou outit
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sources, e.g. [10,11]. Only few studies have assessed the GHG emissions reduction potential for a combination of mitigation options. Fais et al. [12] assessed the emission reduction potential for different portfolios of options for the UK industry, but did not give any insights into suitable strategies and associated investment costs. Boulamanti and Moya [13] analyzed the potential of various low-carbon energy

technologies, including EEMs, CCS and bioenergy, in terms of energy and GHG emissions reductions in the chemical and petrochemical industry of the EU up to 2050. They found that with the adoption of best available and innovative technologies as much as 72.5 MtCO $_2$ -eq and 225 PJ can be saved annually by 2050, compared to a baseline scenario. Johansson et al. [14] assessed the CO $_2$ reduction potential

for the European petroleum refining industry up to the year 2050 for EEMs, CCS and fuel switching. Although they acknowledged that different mitigation options affect each other's individual CO₂ reduction potential when implemented in tandem, Johansson et al. [14] did not assess the CO₂ reduction potential of the mitigation options together, nor did they determine deployment pathways.

Next to high level assessments, numerous studies have investigated the techno-economic performance and CO2 reduction potential at the industrial plant level by means of EEMs, e.g. [15] and CCS [16]. Others studies have assessed the feasibility of biomass gasification and fast pyrolysis to produce energy carriers and chemicals, which could be used as mitigation options in industrial plants [17.18]. However, to the authors' knowledge, the implementation of a combined portfolio of mitigation options at the industrial plant level has not yet been evaluated. More insight is required into how to deploy a portfolio of available mitigation options over time (i.e. with low CO₂-eq avoidance cost, high GHG emissions reduction, and/or low upfront investment cost). Promising deployment pathways for an industrial plant will depend heavily on, among others, the industrial sector, key processing technologies, (future) plant configuration, fuel and feedstock mix, and energy prices. Several studies have demonstrated that marginal abatement cost curves can be a useful tool to evaluate and identify costeffective GHG mitigation strategies (e.g. [19-21]). However, marginal abatement cost curves suffer from a number of limitations, such as the non-consideration of interactions between mitigation options, a possibly inconsistent baseline, and double counting [22]. This tool should therefore be used in a broader methodological framework when identifying and assessing effective strategies for industrial plants. The method should consider the main process components and allow for assessing different process configurations via different deployment pathways for industrial plants across all sectors.

The objective of this paper is to develop this method based on a bottom-up analysis and to illustrate the method by identifying deployment pathways in the refinery sector up to the year 2030. The time period was limited to 2030 given the large degree of uncertainty in sector developments and costs and performance of technologies in the long term.

1.1. Oil refining industry

Within the EU, the oil refining industry accounts for 8% (155 MtCO₂/y) of industrial sector emissions and has a combined processing capacity of around 770 Mt/y of crude oil [14]. Today, there are over one hundred refineries in the EU27 countries, which range in size from small, simple refineries with low conversion capacity (only naphtha reforming), and therefore few output products (so-called topping or hydroskimming refineries) to large, complex refineries with deep conversion capacity and high flexibility, meaning many different output products [14]. Johansson et al. [14] identified 18 complex refineries in the EU, which have both hydrocracking and catalytic cracking processing units and sometimes also gasification and/or coking capacity to convert heavy fractions into lighter product, power and/or steam. In this study, a large, complex petroleum refinery in North-West Europe was selected as a case study to demonstrate the method. Large, complex refineries are particularly interesting for this study considering their high annual CO2 emissions, variety in point sources, and opportunities for a mixture of mitigation options.

In this paper, the term CO_2 emissions is used to refer to the refinery (sector), CCS and CO_2 prices, whereas GHG emissions refer to emissions reduction potentials (tCO₂-eq/y) and avoidance costs (ε /tCO₂-eq).

2. Method

2.1. General approach

The general approach is divided in two parts: the assessment of the individual mitigation options and the identification of promising deployment pathways.

For the assessment of the individual mitigation options, the next steps were followed:

- 1. Inventory of key parameters of industrial plant of core process in the industrial plant (e.g. CO₂ emissions, capacity, energy flows);
- Projection of baseline GHG emissions by analyzing future trends (e.g. product demand, product quality requirements);
- 3. Identification and data collection of GHG emission mitigation options:
- Identification of interactions between mitigation options (i.e. a decrease in GHG reduction potential, cost synergies, economies of scale, lock-in effect);
- Assessment of GHG reduction potential and GHG avoidance costs of individual and combi mitigation options;
- 6. Sensitivity analysis of key parameters.

The mitigation options were divided in short-term measures (i.e. the next 5 years) and medium-term measures (i.e. between 5 and 15 years). Moreover, the mitigation options were assigned to one of the following categories, each with an increasing impact on the current plant layout:

- Add-on measures, whereby the mitigation options do not affect the core processes and throughput of the industrial plant;
- Retrofit measures, which involve modifications to either the core process units, the energy supply units (e.g. furnaces), or both;
- Replacement measures, which imply the replacement of existing process units with advanced units with a lower carbon footprint;
- New concepts that replace the core processes of the industrial plant altogether and have a lower carbon footprint than the reference processes.

After determining the techno-economic performance of the individual mitigation options, several promising deployment pathways were identified that embody different combinations of mitigation options. This allows for the exploration of a palet of pathways that differ with respect to several criteria: average $\rm CO_2$ avoidance cost, GHG emission reduction potential, technological maturity of the mitigation options and their impact on the core processes of the industrial plant. The last two criteria are relevant in terms of risk: technologies that are not yet fully developed may not become available (on time) or may not have the expected performance, while technologies that are closely integrated with the core processes could significantly affect the reliability of these processes. Mitigation options with very poor technoeconomic performance or extremely high uncertainties in the input data are excluded from the pathways.

The interactions between the mitigation options (i.e. economies of scale, cost synergies, decrease in each other's GHG reduction potential, lock-in effect) are quantified where possible. Economies of scale and cost synergies are accounted for in the cost calculations by using economic scaling factors and specific cost data for the combined application of mitigation options (e.g. biomass and CCS). The cost advantage compared to small scale and single application of mitigation options is made explicit. For the deployment pathways, the overlap in the GHG reduction potential as well as the effect on the avoidance costs of the mitigation options, was determined in two steps: (i) the GHG emission reduction and avoidance cost of the mitigation option that is implemented first is computed: (ii) the GHG emission reductions of the second mitigation option is computed by first subtracting the emission reductions of the first mitigation option from the base case emissions, and subsequently by computing the GHG emission reduction and avoidance cost of the second mitigation option. Lastly, possible lock-in effects of mitigation options are described in a qualitative manner.

2.2. System boundaries and performance indicators

The analysis incorporates the costs and GHG emissions related to the net mass and energy flows, i.e. the difference in energy and mass flows between the base case and mitigation options. The feedstock input and main industrial plant product output remain constant.¹

The annually avoided GHG emissions GHG_a (tCO₂-eq/y) is the main technical indicator in this study. GHG_a is expressed using Eq. (1):

$$GHG_a = \Delta GHG_{upstream} + \Delta GHG_{plant} + \Delta GHG_{downstream}$$
 (1)

where ΔGHG_{planb} ΔGHG_{up} , and ΔGHG_{down} are the net change in annual GHG emissions (tCO₂-eq/y) of the industrial plant, and upstream and downstream processes, respectively. Life cycle GHG emissions related to a change in the production or consumption of energy carriers were included in the analysis.

The GHG avoidance cost C_a ($\mathfrak{C}/\mathsf{tCO}_2$ -eq) is the main economic indicator in this study. C_a is expressed using Eq. (2):

$$C_{a} = \frac{\Delta E_{ng} * P_{ng} + \Delta E_{e} * P_{e} + \Delta E_{f} * P_{f} + \alpha * I + \Delta C_{O\&M}}{\Delta GHG_{a}}$$
(2)

where ΔE_{ng} , ΔE_e and ΔE_f are the net change in annual natural gas (GJ_{ng}/y), electricity (GJ_e/y), and fuel consumption (GJ_f/y), P_{ng} , P_e and P_f are the prices of natural gas (\mathfrak{E}/GJ_{ng}), electricity (\mathfrak{E}/GJ_{e}) and fuel (\mathfrak{E}/GJ_{f}), respectively. I is the upfront investment cost (\mathfrak{E}), α is the annuity factor, and ΔC_{ORM} is the net change in O&M cost (\mathfrak{E}/y).

The upfront investment $\cos I$ ($\mathfrak C$) are the total capital requirement (TCR), which comprises cost for equipment, installation, engineering fees, contingencies, owner cost and interest during construction. The TCR is derived by multiplying the process plant cost (PPC), i.e. equipment and installation costs, with typical percentages for the other cost components (see Table 1).

The annualized capital cost is calculated by multiplying the upfront investment cost with an annuity factor (see Eq. (2)). The annuity factor

is a function of the real discount rate r (%) and economic lifetime LT (years) of the technology (see Eq. (3)).

$$\alpha = \frac{r}{1 - (1+r)^{-LT}} \tag{3}$$

Only limited recent data on investment costs of EEMs was found in literature; instead, numbers on simple pay-back periods for EEMs are presented more often. Hence, the equation for the simple pay-back period was used to calculate the total capital investment costs:

$$PBP = \frac{I}{B_e - C_e} \tag{4}$$

where I is the total capital investment cost (\mathfrak{C}) , B is the annual benefit in terms of energy savings (\mathfrak{C}/y) , and C is the annual cost (\mathfrak{C}/y) . The annual costs (C) in the equation were assumed to be zero in this study (see Section 3.3.1). Literature data on simple pay-back periods and energy savings were used to derive the investment costs. Subsequently, the investment costs and energy savings were used to compute the GHG avoidance cost using Eq. (2).

2.3. Standardization of key parameters

To enable a fair comparison of the technologies, several underlying parameters were standardized. The following procedure as proposed by Kuramochi et al. [8] was adopted:

- Indexation. All cost figures were converted to €₂₀₁₂. Costs that are reported in other currencies were first converted to Euro using the year-average exchange rate data of OANDA [23] for the year the cost data are reported, and were then escalated to the year 2012 using the Downstream Capital Costs Index (DCCI) [24].
- Normalization of capital cost figures. As not all studies include the same cost components, fixed percentages were used to correct for the differences. The Total Capital Requirement consists of various components:
 - Process Plant Cost (PPC) comprising equipment and installation cost;
 - Total Plant Cost comprising PPC, engineering fees and contingencies:
 - Owner costs (i.e. costs for pre-production, royalties, inventory capital, land and site preparation) and interest during construction.
- 3. Scaling of capital cost figures. The capital cost of a system component depends mainly on two factors: the size of the component and the number of parallel components. A generic scaling relation was applied to the capital costs to account for this effect (Eq. (5)). The investment cost of a component *i* (*I_i*: €) is expressed as proposed by Larson et al. [25]:

$$I_{i} = I_{i,ref} * \left(\frac{N_{units,i}}{N_{units,i,ref}}\right)^{SF_{n}} * \left(\frac{S_{i}}{S_{i,ref}}\right)^{SF_{i}}$$

$$(5)$$

where $I_{i,ref}$ is the reference capital investment (\mathfrak{C}) , $N_{units,i}$ is the number of parallel process trains per system, $N_{units,I,ref}$ is the number of parallel process trains in the reference system, S_i is the capacity of a single process train (unit: component dependent), S_{ref} is the reference capacity of a single process train (unit: component dependent), SF_n is the scaling factor for multiple trains, and SF_i is the economic scaling factor for component i.

Table 1 provides an overview of the general parameters used in this study. The value ranges of these parameters used for the sensitivity analysis are presented in Table 7 in Section 3.5.

¹ In case of EEM, mainly natural gas and electricity are saved, which are unrelated to the feedstock (i.e. crude oil) input. However, deep energy savings may lead to savings of process fuel gases, which are a by-product of the core refinery processes and thus related to the crude oil feedstock. In principle, these saved process gases could be used as feedstock to produce valuable output products, thereby reducing the required feedstock input. However, this reduction in required feedstock level will be very small. Furthermore, for the sake of consistency, it was assumed that the feedstock input level remains constant and the saved fuel gases are sold to third parties.

 Table 1

 General techno-economic input parameters used in this study.

	Unit	Value	References
Real discount rate	%	10	Own value
Total Plant Cost ^a	%-PPC	130	[68]
Total Capital Requirement ^a	%-TPC	110	[68]
Calorific value natural gas	MJ_{LHV}/m^3	31.7	[69]
Industrial energy price			
Transport fuels ^b	ϵ /GJ _{LHV}	16	[2]
Natural gas ^c	ϵ /GJ _{LHV}	10	[2]
Electricity ^d	€/GJ _e	22	[70]
Steam (LHV) ^e	€/GJ _{th}	12	Own value
Torrefied wood pellets (TOPS) ^f	ϵ /GJ _{LHV}	8.0	[42,71]
Woody biomass for pyrolysis	ϵ /GJ _{LHV}	3.5	[44]
CO ₂ emission factor			
Natural gas	${\rm kgCO_2/GJ_{LHV}}$	56.7	[72]
Life cycle GHG emissions			
Electricity grid production ^g	kgCO ₂ -eq/GJ _e	63	[73]
Torrefied wood pellets (TOPS)h	$kgCO_2$ -eq/ GJ_{LHV}	7.9	[74–76]
Woody biomass for pyrolysis	$kgCO_2$ -eq/ GJ_{LHV}	1.9	[77]
Fossil gasoline	kg CO ₂ -eq/GJ _{LHV}	79.4	[52]
Fossil diesel	kg CO ₂ -eq/GJ _{LHV}	80.3	[52]
Natural gas	kg CO ₂ -eq/GJ _{LHV}	66.3	[52]
Industrial furnace efficiency ⁱ	%	80	[78]
Max. NGCC-CHP efficiency (LHV)	%	90	[79,80]
Power equivalent factor LP steam		0.28	[80]

^a Process plant cost (PPC) comprises equipment and installation costs. Total plant cost (TPC) comprises PPC, engineering fees and contingencies. Total capital requirement (TCR) comprises TPC, owner costs and interests during construction. The values are within observed ranges for power plant construction [8].

- ¹ In case efficiencies for industrial furnaces were not indicated, an average efficiency of 80% was assumed.
- ^j The gas turbine electrical efficiency is assumed to be two-thirds of the NGCC efficiency in condensing mode.

3. Case study

3.1. Complex oil refinery

A large, complex oil refinery in North-West Europe was selected as a case study due to its large processing capacity (over 20 Mt crude oil in

2012), high $\rm CO_2$ emissions (~4 MtCO₂/y in 2012), variety in size and spatial distribution of point sources, and large potential for various GHG mitigation options. The key characteristics of the refinery are shown in Table 2. The refinery is capable of processing a high number of crude oil types and shows high flexibility in the conversion routes and portfolio of final output products. The key processes of the refinery

b The transport fuel price was based on projected developments in the crude oil price as reported in the Energy Technology Perspectives publication of the IEA [2]. A price range of 60-100 €/bbl was observed for the period 2020-2040 in the Reference Technology Scenario (RTS) and 2 °C Scenario (2DS), which reflect varying assumptions on energy and climate policy, technology deployment and economic and demographic changes. The method of Knoope et al. [18] was adopted to derive the costs of the transport fuel price for the year 2025. Using a conversion factor of 6.12 GJ_{HHV}/bbl [81] and a HHV/LHV ratio of 1.06 [82], the ETP prices translate to a crude oil price range of 11-18 €/GJ_{LHV}. The costs for crude oil refining to obtain gasoline and diesel were indicated by Larson et al. [25] to be 27 €/m³ diesel. With a volume of 0.159 m³/bbl, this equates to a margin of 0.8 €/GJ_{LHV}. Using the crude oil price range of 81-96 €/bbl, the costs of fossil diesel and gasoline production are projected to be 12-19 €/GJ_{LHV}. A price of 16 €/GJ_{LHV} was used as medium value in this study. Infrastructure-ready green transport fuels were assumed to fetch similar prices on the transport fuel market as their fossil counterparts.

c This was based on projected natural gas price developments as reported in the Energy Technology Perspectives publication [2]. A price range of 7-11 €/GJ was observed for the period 2020–2040 in Europe in the Reference Technology Scenario (RTS) and 2 °C Scenario (2DS). A conservative price of 10 €/GJ_{LHV} was used as medium value for natural gas. As the country in which the case study is based has to significantly reduce natural gas production volumes over the coming decades, this relatively high natural gas price is deemed to be realistic.

d It was assumed that in a world where GHG mitigation measures like CCS and biomass technologies are deployed for industrial processes, the North-West European power sector is already largely decarbonised. Therefore, projected electricity prices were taken from Brouwer et al. [70], who modelled several low-carbon power systems for Western Europe with varying shares of variable renewable electricity and fossil power plant CCS using hourly power system simulation software. They found electricity prices of around 80 €/MWh (22 €/GJ) for different power system configurations. The main reason for the high electricity price, compared to today's prices, is the high investment needs required for the large-scale deployment of low-carbon technologies across the power sector, which need to be recuperated by investors through an electricity price increase.

e It was assumed that the onsite steam production costs in an industrial boiler equal the steam price. Based on a natural gas price of $10 €/GJ_{LHV}$, boiler capital costs of 85 €/kW [83] and boiler O&M costs of 2% of the total investment costs, the production costs of high-pressure steam were calculated to be $11.8 €/GJ_{LHV}$. This figure is in line with steam prices $(11.1 €/GJ_{LHV})$ indicated for industry [84].

f The method of Meerman et al. [42] was adopted to derive the medium value for the TOPS price. Albeit TOPS are not produced commercially today, Meerman et al. [42] used biomass pellet prices as a proxy for TOPS prices, assuming that the additional production costs of torrefaction compared to biomass pellets are compensated by the reduction in transportation costs. Assuming an energy density of 17 GJ_{LHV}/t [85], biomass pellet prices fluctuated between 6.9 and 8.8 €/GJ_{LHV} during the period 2007–2013 [42.71]. A medium value of 8.0 €/GJ_{LHV} was used for this study.

g This is the CO₂ emission factor for the electricity mix in 2020–2030 as modelled in the Grand Coalition scenario by van den Broek et al. [73], which is based on the premise that a large share of the CO₂ is reduced through renewable energy technologies and other CO₂ mitigation options. This scenario was assumed to be a precondition for large scale deployment of GHG mitigation options at industrial processes.

h Taken from Batidzirai et al. [74–76] who presented data on supply chain GHG emissions of TOPS made from eucalyptus and switchgrass in Mozambique, eucalyptus in Brazil, and agricultural residues in South Africa. The supply chain includes biomass production, chipping, drying, torrefaction, milling, pelletizing, and transport by truck, train and sea ship.

Table 2
Key characteristics of the case refinery in 2012.^a

	Unit	Value	References
Crude oil throughput	Mt/y	~ 20	[26]
Annual CO ₂ emissions	Mt/y	4.1	[67]
Estimated primary energy input CHPb	PJ _p /y	~16	[26,67]
Estimated electricity production CHP ^b	PJ _e /y	~5	[26,67]
Estimated steam production CHP ^b	PJ _{th} /y	~9	[26,67]
Estimated hydrogen production SR ^c	PJ _{H2} /y	~7	[26,67]
Number of stacks		18	[67]
Volumetric CO ₂ concentration ^d			
Gasifier	%-vol	99	[86]
SR WGS-PSA ^e	%-vol	30-60	[37]
SR furnace stack	%-vol	5-10	[37]
Catalytic cracker ^f	%-vol	10-20	[87]
Furnaces	%-vol	8-12	[86]
Gas turbines	%-vol	4	[86]

^a The process heat demand in the refinery is not presented for reasons of confidentiality.

are crude oil distillation, conversion of oil fractions in the catalytic reformer, (hydro, catalytic and thermal) crackers and gasifier, and desulphurization of the intermediate and final products. The refinery has been expanded, upgraded and rejuvenated several times during its lifetime [26]. A schematic overview of the refinery and $\rm CO_2$ emissions is presented in Figs. 1 and 2, respectively. The $\rm CO_2$ emissions are vented via 18 stacks and two flares, which have different mass flows and volumetric $\rm CO_2$ concentrations. The majority of the $\rm CO_2$ emissions are attributed to the process furnaces, followed by the gas turbine/CHP units, 2 and gasifier. The gas turbine, CHP units, steam reformer (SR), and gasifier on the refinery site will hereafter be referred to as *in situ* technology to distinguish them from newly built CHP units, reformers and gasifiers, which are required for the mitigation options.

3.2. Future trends

Market trends were analyzed to make projections for the baseline GHG emissions for the short and medium term. These trends were analyzed by reviewing trend forecasts studies from international organizations and academia [27,28] and industry organizations [29,30]. The review, analysis and data underlying the $\rm CO_2$ emission projections are described in detail in the Appendix. The following trends are relevant for the case refinery:

 There will be a global shift in demand from heavy (i.e. fuel oils and marine fuels) and light distillates (i.e. gasoline and naphtha) to middle distillates (i.e. diesel and jet fuel). Despite the projected EU legislation on fuel quality (mainly sulphur) will become more stringent in the future. Stricter quality specifications will result in higher specific energy requirement and concomitant CO₂ emissions in EU refineries.

Based on quantitative projections of the CONCAWE association [29,31], the case refinery CO_2 emissions were estimated to increase with $0.3\,\mathrm{MtCO}_2/\mathrm{y}$ in the short term as a result of the increasing middle distillates/gasoline (MD/G) ratio. Another increase of $0.3\,\mathrm{MtCO}_2/\mathrm{y}$ was projected for the medium term. The planned stricter quality regulations are expected to increase refinery CO_2 emissions with 11% in the short term $(0.5\,\mathrm{MtCO}_2/\mathrm{y})$ compared to 2012 levels. Additional legal changes may be introduced in the medium term. These changes could result in an increase of 16% $(0.6\,\mathrm{MtCO}_2/\mathrm{y})$ in the refinery's CO_2 emissions compared to 2012 levels.

The economic performance of the mitigation options are compared with a CO₂ price range of 20–75 €/tCO₂, which is based on CO₂ price projections for the short and medium term in three scenarios (Current Policies, New Policies and Sustainable Development) of the IEA's World Energy Outlook (WEO) 2017 [32], each founded on varying assumptions on energy and climate policy, technology deployment and economic and demographic changes. The Current Policies scenario represents a development in which there are no changes in the established policies. In the New Policies scenario, it is assumed that a broad set of the policy commitments and plans that have been announced by countries to reduce GHG emissions will come into effect. The Sustainable Development scenario takes into account all ambitious policies and measures needed to achieve the 450 ppmv target, while also reducing air pollutant emissions and achieving universal energy access. More information about the scenarios can be found in WEO 2017 [32].

3.3. GHG mitigation options and data collection

The selected mitigation options are: energy efficiency measures, CO_2 Capture and Storage and the switch of natural gas to biomass, both as feedstock and as fuel. The following mitigation options were excluded from the analysis due to a lack of detailed data required to perform the analysis:

- Switch to lighter crude oil fractions to reduce the energy use and concomitant CO₂ emissions in the processing steps.
- Utilization of waste heat (WH) that cannot be used in the core refinery processes. Previous research has shown that process integration with adjacent industries as well as using low-temperature WH for post-combustion solvent regeneration, district heating, or biomass drying could result in significant CO₂ emissions reductions [14,33].
- The use of geothermal energy for process or fuel heating.

The selected mitigation options are described in the following subsections. A more detailed description of the technologies and the assumptions and data underlying the calculations can be found in the Appendix.

3.3.1. Energy efficiency measures

Several opportunities for EEMs were identified for the refinery based on the current plant configuration. Data on energy savings, payback periods and economic lifetime were taken from the Energy Guide on available EEMs for petroleum refineries [34]. The data are mainly based on case studies from U.S. refineries around the year 2010.

^b Based on a total *in situ* CHP input capacity of 560 MW, an operation time of 8000 h/y, and an assumed average electric and thermal efficiency of 33% and 56%, respectively. The internal consumption and production of electricity were assumed to be equal, i.e. no electricity import or export takes place.

^c Natural gas was assumed to be the main fuel and feedstock for the *in situ* SR to produce hydrogen. Based on the fuel input capacity, a capacity factor of 56%, a calorific value of 121 MJ_{LHV}/tH₂, a SR efficiency of 75% [88] and a ratio between the natural gas used for fuel and feedstock of around 1:2.9 [88], the hydrogen production was estimated to be around 7 PJ/y. The capacity factor was derived from the SR furnace capacity and annual fuel gas input.

 $^{^{\}rm d}$ The values for the volumetric CO₂ concentrations are based on generic data, which do not apply specifically to the case refinery.

^e WGS and PSA stand for watergas shift and pressure swing adsorption, respectively.

 $^{^{\}rm f}$ The high-temperature flue gas from the catalytic cracker contains typically around 10–20 vol% CO₂ when running on a full combustion mode (excess air in the regenerator) [87]. A conservative value of 12 vol% was used in this study.

lower crude oil demand in the EU, the specific refinery energy consumption and ${\rm CO_2}$ emissions are expected to rise due to higher demand for middle distillates.

² The refinery has four gas turbines; three of them are CHP units.

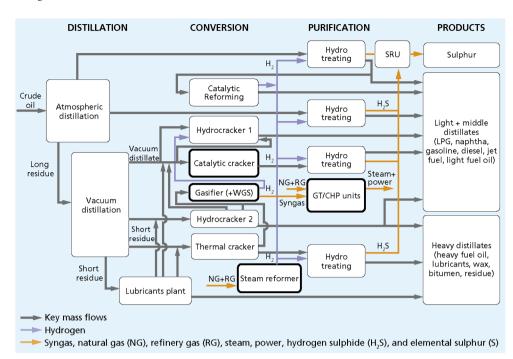


Fig. 1. Simplified process layout of the case refinery. The main CO_2 emission sources in the refinery are depicted in bold. The other two CO_2 emission sources (furnaces and flares) are not displayed. GT, CHP, LPG, SRU, WGS stand for gas turbine, combined heat power, liquid petroleum gas, sulphur recovery unit, and watergas shift, respectively.

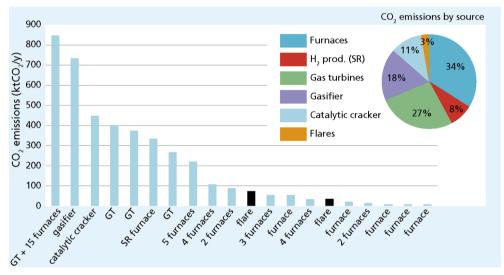


Fig. 2. CO₂ point sources (bar chart) and breakdown of CO₂ emissions by source (pie chart), both for the year 2012 [67]. The process units related to the stacks are presented on the horizontal axis; each bar represents one stack (blue bar) or flare (black bar). GT stands for gas turbine.

Table 3

Primary energy reduction potential for the refinery for the short and medium term. The data are based on Worrell et al. [34]. The reductions are related to the total primary energy use in the refinery in the year 2012, thus excluding growth in emissions due to future trends.

Short term EEMs	PJ/y	%	Medium term EEMs	PJ/y	%
Energy management & control	1.7	3.0	Advanced desulphurization	5.8	10.0
Heat integration distillation units	1.7	3.0	Advanced separation systems	3.9	6.8
Motors & pumps	1.4	2.4	Turbine pre-coupling	1.5	2.5
Steam distribution system	1.2	2.0			
Heat integration & waste heat recovery	1.2	2.0			
Fouling mitigation	0.6	1.0			
Improved furnace performance	0.6	1.0			
Hydrogen management & recovery	0.4	0.7			
Flaring	0.3	0.5			
Total	9.1	15.6	Total	11.2	19.3

Changes in the annual costs of the process units due to the implementation of EEMs were neglected in this study.³

For the short term, nine EEMs were identified (see Table 3). The largest primary energy reductions can be achieved via heat integration between the distillation units (\sim 3%) and improvements in energy management & control (\sim 3%). The joint primary energy reduction of short term EEMs amounts up to 15–16%. For the medium term, three EEMs were identified, with advanced desulphurization systems showing the largest primary energy reductions (\sim 10%). In total, an estimated reduction in primary energy use of 19–20% was computed for the medium term. Whereas the short term EEMs can be implemented by retrofitting process units in the current refinery layout, two of the medium term EEMs (advanced separation and desulphurization systems) have a higher impact on the refinery layout as it requires the replacement of current core process units (distillation and hydrotreating units). Ideally, these units would be installed by the time the current process units have to be replaced.

3.3.2. CO2 capture and storage

Three first generation CO_2 capture technologies are assessed: post, pre- and oxyfuel combustion capture. While post-combustion capture is an add-on technology, pre- and oxyfuel combustion are considered to be retrofit technologies that require additional equipment and adjustments to the existing refinery infrastructure [35]. The key data of the CO_2 capture technologies can be found in Table 4.

Post-combustion capture is applied to the main CO_2 sources on the refinery site: furnaces, catalytic cracker, in situ CHP unit, and the in situ hydrogen plant. Oxyfuel and pre-combustion technology are applied to all these sources, except to the in situ hydrogen plant (no oxyfuel combustion) and catalytic cracker (no pre-combustion), respectively, due to technological constraints. Although some gas turbines display operational difficulties when fired with nearly pure oxygen or hydrogen [36], it is assumed these issues will be solved in the medium term.

Table 4
Techno-economic parameters for CO₂ capture and storage.

required. The *in situ* hydrogen plants are also retrofitted with chemical scrubbers (ADIP-X) to capture CO₂. CO₂ capture from the *in situ* refinery gasifier, which processes catalytic cracker residue, does not require a specific technology as the separation of the CO₂ is already an inherent part of the gasification process. In all capture routes, the CO₂ is cooled, dried, purified and compressed to 110 bar before it is transported to a storage site. An economic lifetime of 20 years was used for all three capture technologies [37,38].

3.3.3. Biomass utilization

Balat et al. [39] and Faaij [40] distinguished three main thermochemical conversion routes for biomass: gasification, pyrolysis and combustion. As the refinery units are mainly fueled with refinery gases, (solid) biomass combustion does not seem to be a realistic option. Therefore, only the first and second options were considered in this study. Both options are new concepts that differ from the core refinery process.

3.3.3.1. Biomass gasification. The first biomass route is the production of syngas through biomass gasification (BIG) from torrefied wood pellets (TOPS) with its favorable properties of a high heating value and low moisture content. Next, syngas is converted to steam and electricity in the BIG-CHP sub-route, to hydrogen in the BIG-H₂ sub-route, and to Fischer-Tropsch (FT) fuels in the BIG-FT sub-route. In BIG-CHP the hydrogen completely replaces the natural gas for the *in situ* gas turbine and CHP units in the refinery and in BIG-H₂, it replaces the hydrogen from the *in situ* SR. In BIG-FT, the FT-fuels are produced in addition to the fossil gasoline and diesel made in the refinery, and is assumed to replace fossil diesel and gasoline in other less efficient refineries. In this sub-route the upgrading process of the FT-process occurs in a new hydrocracker and distillation columns as it is unknown whether the existing units on the refinery site have spare throughput capacity. Moreover, integration of the FT-process with the existing units

	Unit	Post-combustion	Oxyfuel combustion	Pre-combustion	References
Technical					
CO ₂ capture ratio	%	87	87	95 ^a	[45]
Spec. electricity use	GJ _e /tCO ₂	1.0-1.3 ^b	0.5	0.4	[37,89–93]
	GJ_e/tO_2	_	0.7	_	[94]
Spec. heat use ^b	GJ _{th} /tCO ₂	3.5-4.0 ^b	_	1.97 ^a	[37,95]
Natural gas use	GJ _p /tH ₂	-	-	165	[88]
Economics					
Specific capital cost ^c	M€/MtCO ₂ /y	101 (4 vol%);	42	130	[45]
•		75 (12–14 vol%)°			
	M€/ktO ₂ /y	_	53	_	[45]
Specific O&M cost	€/tCO ₂	4.8	9.0	7.1	[45]
•	€/tH ₂	_	_	4.5	[45]
Maintenance cost	% of TPC	3.8	_	_	[96]
CO_2 transport and storage costs	€/tCO ₂	16	16	16	[61,62]

^a This applies to CO₂ capture from the high-pressure process gas after the PSA unit.

Deploying pre-combustion capture across the refinery would require additional SR's as the *in situ* SR plants will most likely not have sufficient production capacity to deliver the large amounts of hydrogen

would require detailed data on the refinery processes, which is not publicly available. The BIG-FT option was limited to $1000\,\mathrm{MW_{HHV}}$ of TOPS feed input because larger plant scales would hardly lower production costs [41].

Finally, a fourth sub-route, BIG-combi, combines the other 3 BIG

^b Regeneration heat and electricity needed for flue gas with volumetric CO₂ concentrations in the range of 4–16%; the specific regeneration energy (GJ_{th}/tCO₂; GJ_e/tCO₂) decreases with higher volumetric CO₂ concentrations.

^c This excludes capital cost for required modifications to the flue gas stacks of 0.1 M€/stack [78].

 $^{^3}$ The sensitivity analysis shows the effect of a change in the annual energy savings and natural gas price on the CO₂-eq avoidance costs. Considering the large uncertainty in the gas price and annual energy savings, the effect of a change in annual costs due to the implementation of an EEM (typically \pm 5% of the investment costs [34]) on the CO₂-eq avoidance costs will be minor.

 $^{^4}$ The $1000\,MM_{HHV}$ input TOPS results in an FT output capacity of 458 MW $_{FT}$, which corresponds to around 13 PJ/y ($\sim\!2.9*10^5$ toe/y; 1% of the refinery's crude oil input).

Table 5Input data for biomass gasification sub-routes. Based on NEA [26,67] and Meerman et al. [42,43].

	Unit	REF	BIG-CHP		${\rm BIG\text{-}H}_2$		BIG-FT		BIG-comb	oi
			Vent	CCS	Vent	CCS	Vent	CCS	Vent	CCS
Input refinery										
Natural gas ^a	MW_{LHV}	1167	607	607	560	560	1167	1167	0	0
TOPS	MW_{LHV} (MW_{HHV})	-	473	511	401	401	937	937	1499	1499
							(1000)	(1000)		
Output refinery										
Steam	MW_{th}	314	314	314	406 ^b	406 ^b	314	314	314	314
Elec. CHP	MW_e	182	72	51	156	149	308°	286°	86	86
Hydrogen	MW_{H2}	255	255	255	255	255	255	255	255	255
FT-fuels	MW_{FT}	_	_	_	_	_	458	458	458	458
CAPEX	M€	_	266	328	264	268	571	582	765	783
OPEX	M€/y	_	16	19	16	16	34	34	45	46

^a Natural gas used for the production of steam, electricity and/or hydrogen.

sub-routes and all sub-routes can be implemented without CCS (vent) and with CCS. The key input data of the eight BIG sub-routes were based on process simulations developed by Meerman et al. in the AspenPlus® software [42,43] and adapted for this specific refinery case (see Table 5). We assumed the BIG technology to become available in the medium term and to have an economic lifetime of 20 years.

3.3.3.2. Fast pyrolysis. The second biomass route is the production of pyrolysis oil from biomass and its upgrading to infrastructure-ready gasoline and diesel. A typical process for fast pyrolysis oil production comprises several steps: drying of feedstock (pre-treatment), thermal decomposition of the biomass (pyrolysis), upgrading of the pyrolysis oil using hydrogen gas (hydrotreating), and the separation of stable pyrolysis oil into different fractions, including gasoline and diesel (hydrocracking).

Due to potential issues with corrosion and limited availability of spare throughput capacity in existing refinery process units, the pyrolysis fuels were assumed to be produced in new processing units in addition to the refinery product output mix. For the same reason, the required hydrogen was assumed to be produced in a new SR due to possible capacity constraints in *in situ* SMR plants. We assumed that pyrolysis technology has an economic lifetime of 20 years and will become available in the medium term. The size of the pyrolysis train was limited to 1000 MW_{HHV} feed input. A PNNL study [44], which presents recent detailed and publicly available data coming from several US research institutes, was used for the analysis. The key data of the fast pyrolysis route can be found in Table 6.

3.4. Interactions among mitigation options

The mitigation options EEM, CCS and biomass can interact in several ways. The EEMs affect each other's GHG reduction potential (see also Section 3.3.1). Likewise, fewer emissions can be reduced via CCS in the case where EEMs are already implemented. However, the interaction between EEMs and BIG is minor. Although the three CO₂ capture technologies can be implemented side by side, this was found to be rather expensive in earlier research by the authors [45], due to of the high capital and operational costs per tonne CO₂-eq avoided, and thus undesirable. Significant economies of scale can be achieved in both the combi CCS routes in which CO₂ is captured from multiple sources, and in the BIG-combi sub-route in which multiple output products (steam, electricity, hydrogen and FT-fuels) are produced. CCS and BIG are

Table 6
Input data for fuel production via fast pyrolysis, hydrotreating and hydrocracking. All weights pertain to dry woody biomass. The abbreviations p-oil, bf and bm stand for pyrolysis oil, biofuel and biomass, respectively. Based on PNNL [44].

	Unit	Quantity
Energy content biomass	MJ_{LHV}/kg_{bm}	18.6
Fast pyrolysis		
Capacity pyrolysis unit ^a	kt _{bm} /d	2.0 ^a
Mass yield	t p-oil/t _{bm}	0.62
Overall energy yield	GJ_{bf}/GJ_{bm}	0.70
Energy content biofuel	MJ_{bf}/L_{bf}	32
Electricity	$\mathrm{MJ_e/L_{bf}}$	1.2
Water	$L/L_{ m bf}$	1.5
Natural gas	MJ/L_{bf}	4.7
Costs		
Process Plant Cost	€/L _{bf} /day	385
OPEX	ϵ /L _{bf}	0.2

 $^{^{\}rm a}$ The reference size of the pyrolysis unit was 2.0 kt/d of dry biomass. For 1000 MW biomass input (5.1 kt/d), three pyrolysis units were assumed to be required.

complementary technologies that can deliver net negative emissions. BIG and pyrolysis can compete with respect to the production of green fuels.

3.5. Sensitivity analysis

The impact of the input parameter values was assessed by doing a sensitivity analysis. The parameters included in this analysis are: energy prices, annualized capital cost, grid electricity GHG emission factor, biomass supply chain GHG emissions, $\rm CO_2$ transport & storage costs, process energy use, and NGCC(-CHP) efficiency. The variations in the input parameters are presented in Table 7. The combined effect of the parameters on the results was assessed by calculating an extreme pessimistic and extreme optimistic case, in which all parameters are set at their least and most favorable values, respectively. The impact of the varied input parameters is indicated with error bars in the GHG reduction potential and avoidance costs.

^b Excess heat in the gasification train is used for the production of steam.

^c The waste gases coming from the gasification process are combusted in a newly built steam turbine to produce electricity.

Table 7Input parameters and ranges used for sensitivity analysis.

	Unit	Medium value	Range for sensitivity analysis	References
Generic				
Industrial energy price 2025				
Transport fuel ^a	\in /GJ _{LHV}	16	9–26	[2]
Natural gas	\in /GJ _{LHV}	10	7–13	[2]; own estimations
Electricity	€/GJ _e	22	15–29	[70]; own estimations
Steam (LHV) ^b	€/GJ _{th}	12	8–15	Own value
Torrefied wood pellets (TOPS) ^c	\in /GJ _{LHV}	8.0	3.4-8.8	[42,71]
Woody biomass for pyrolysis	\in /GJ _{LHV}	3.5	2.6-5.2	[44]
Life cycle GHG emissions				
Electricity grid production ^d	kgCO ₂ -eq/GJ _e	63	16–110	[73]
Torrefied wood pellets (TOPS)	kgCO ₂ -eq/GJ	7.9	4.9–10.9	[74,75,97]
Woody biomass for pyrolysis	kgCO2-eq/GJ	1.9	1.2-2.6	[77]
CAPEX		_e	\pm 30% for total annualized capital cost	[38]
EEMs				
Primary energy savings	GJ/y	_e	± 30%	[98]
CCS				
Energy use CO ₂ capture	GJ/tCO ₂	_e	± 30%	[38]
HP steam from waste heat of the SR	GJ_{th}/tCO_2	25	20–30	Own values
Max. NGCC efficiency (LHV)	%	_f	45–60	Own values
Max. NGCC-CHP efficiency (LHV)	%	90	75–100	Own values
CO ₂ transport & storage costs	€/tCO ₂	16	7–26 ^g	[61,62]
BIG				
Energy efficiency	%-pt	_h	$+15/-5^{i}$	Own values
Pyrolysis				
Energy efficiency	%	70	62–72	[44]

a The transport fuel price was based on projected developments in the crude oil price. Considering the large uncertainty in the crude oil price, which is driven by many factors (e.g. market conditions, economic events and forecasts, geopolitics, oil reserves), we assumed extreme crude oil prices of 50 and 150 €/bbl for the sensitivity analysis; this translates to a transport fuel price of 9 and 26 €/GJ_{LHV}, respectively. A similar price range was used for green transport fuels.

4. Results

4.1. Mitigation options

Fig. 3 presents the GHG reduction potential and GHG avoidance cost of all individual mitigation options (hereafter called 'sub-routes') and of several sub-routes together (hereafter called 'combined sub-routes'). 5 These GHG reduction potentials were based on the 2012 base case emissions of 4.1 MtCO2-eq. However, as shown by the black bars in the figure, base case emissions may increase to 4.8 MtCO2-eq/y in the medium term as a result of the higher MD/G ratio, and possibly to 5.9 MtCO2-eq/y due to stricter regulations on product quality change

(see Section 3.2). If these higher projected emissions were taken as basis for the analysis, the absolute reduction potentials for the medium term would have been higher. Most error bars of the CO_2 avoidance costs are asymmetric due to uneven uncertainty ranges of the input parameters, and the fact that optimistic changes in input parameters can have a larger effect on the avoidance costs than pessimistic changes, and vice versa.

In the EEM combi sub-route with short and long term EEMs GHG emissions are reduced by 1.1 MtCO $_2$ -eq/y, which corresponds to 28% of the base case emissions, and avoidance cost is $-133\,\text{€/tCO}_2$ -eq on average. The EEM with the lowest avoidance cost is improved energy management & control, followed by the optimization of the steam distribution systems and fouling mitigation (medium values: -172 to $-166\,\text{€/tCO}_2$ -eq). As the avoidance costs are well below the CO $_2$ price range for the short-medium term (20–75 $\,\text{€/tCO}_2$), the EEMs can be considered non-regret options.

^b The steam production costs were varied with the range in natural gas price.

^c The method of Meerman et al. [42] was adopted to derive the TOPS price range (see also footnote e in Table 1). A high biomass pellet price of $8.8 \, \epsilon_{2012}$ /GJ_{LHV} was used as a high-end value for the TOPS price. Several literature studies showed that TOPS prices could decrease to $3.0 \, \epsilon_{2008}$ /GJ_{HHV} [99–101]. Assuming a HHV/LHV ratio of 1.06 for TOPS [43], this translates to $3.4 \, \epsilon_{2012}$ /GJ_{LHV}, which was used as a low-end value for the TOPS price.

 $^{^{\}rm d}$ A large uncertainty range of \pm 75% was assumed to examine the impact on the GHG reduction potential.

^e The medium value for the CAPEX and energy use differs for each technology and is therefore not indicated in the Table. The \pm 30% range was based on typical uncertainty ranges indicated by several detailed techno-economic studies on CO₂ capture technologies [56,89,96] that were used as input for Berghout et al. [38]. Ranges of \pm 30% were used for EEM, BIG and pyrolysis as well.

f The medium value for the gas turbine electrical efficiency depends on the size of the installation and is therefore not indicated in the Table. Usually values of 50–55% were calculated (see [80]).

^g The presented cost range is mainly due to the various transport conditions (on- and offshore pipelines, volumes (2.5–20 Mt/y) and distances (180–1500 km)) as well as storage conditions (on- and offshore, depleted gas/oil reservoirs and saline aquifers, field capacity and well injectivity, new and existing wells, liability transfer costs), and to a lesser degree to uncertainty in the cost elements [61,62].

^h The TOPS input energy demand differs for each BIG sub-route and is therefore not indicated in the Table.

i An asymmetric uncertainty range (+15%/-5%) was used for the BIG technology, as the positive error, partly due to the improvement potential in the BIG conversion efficiency for the medium term compared to the year 2012, is expected to be larger than the negative error.

⁵ In this section, a clear distinction is made between 'sub-routes', like 'Energy management & control' or 'post-combustion capture from gas turbines', and 'combined sub-routes', like all post-combustion sub-routes together, to avoid confusion. Combined sub-routes were explored to examine possible interactions between different sub-routes.

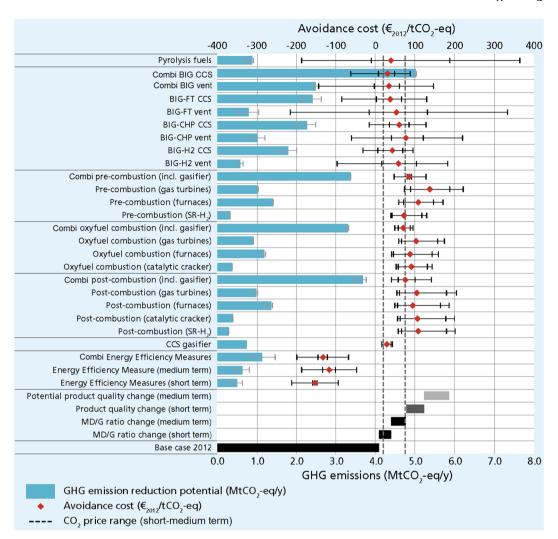


Fig. 3. CO₂ emission projections, GHG reduction potential and GHG avoidance cost of the sub-routes. The top blue bar and diamond in each main route represent the GHG reduction potential and *average* avoidance cost of all sub-routes together (also called the combi sub-routes). The error bars show the sensitivity of the results to the uncertainty ranges in the input parameters. Whereas the outer error bar for the avoidance costs presents the sensitivity of the results to all parameter variations, the inner error bar excludes variations in natural gas, transport fuel, woody biomass and TOPS prices. For CO₂ capture from the *in situ* gasifier, it was cheaper to purchase electricity from the grid rather than installing a new CHP unit. For CO₂ capture from the CHP unit, post-combustion is also used in the pre-combustion route resulting in lower costs than the use of pre-combustion capture.

The CCS routes show significantly higher GHG reduction potentials than the EEMs, with up to 90% of the base case emissions (3.7 MtCO $_2$ -eq/y) in the post-combustion route. In the other CCS routes less CO $_2$ is avoided, because CO $_2$ cannot be captured from the SR in the oxyfuel route and from the catalytic cracker in the pre-combustion route. CO $_2$ capture from the *in situ* gasifier shows the lowest avoidance costs (29 €/tCO $_2$ -eq) as this process involves only the compression of CO $_2$ before it is sent off for transport and storage. For all three CCS-combi routes in which CO $_2$ is captured from all suitable refinery emission sources, the *average* avoidance cost is lower than when CO $_2$ is captured from individual CO $_2$ emission sources, due to economies of scale and the low costs for CO $_2$ capture from the *in situ* gasifier. Costs of the oxyfuel and post-combustion capture combi sub-routes are the lowest with 76 €/tCO $_2$ -eq and 71 €/tCO $_2$ -eq), respectively. However, the costs for the post-combustion combi sub-route increase to 91 €/tCO $_2$ -eq when rev-

enues and avoided GHG emissions related to export of excess electricity production are excluded 7 from the analysis.

The individual BIG vent and BIG CCS sub-routes (only steam/electricity, H_2 or FT-fuel production) display GHG reduction potentials of 0.6–1.0 MtCO₂-eq (14–25%) and 1.8–2.4 MtCO₂-eq (44–59%), respectively. The low avoidance costs are mainly because the natural gas price (medium value NG price: $10 \, \text{€/GJ}$) is higher than the TOPS price (medium value: $8.0 \, \text{€/GJ}$), which has a dampening effect on the avoidance costs. The larger uncertainty in the values of the BIG vent compared to BIG CCS sub-routes is mainly caused by the lower amount of avoided GHG emissions, which makes these sub-routes more sensitive to changes in key input parameters, particularly energy prices. The BIG-combi CCS sub-route shows a very large GHG reduction potential (5.0 MtCO₂-eq; 123% reduction compared to 2012 base case emissions),

⁶ For CO₂ capture from the gasifier, electricity was purchased from the grid rather than generated in a newly installed CHP unit as this showed lower costs.

 $^{^7}$ When excluding credits for electricity export, the monetary value and (indirect) CO_2 emissions of both the exported electricity and natural gas related to this electricity, which was determined on an exergy basis (electricity: 1; heat: 0.28) (see Table 1), were subtracted from the total emissions and costs.

which can be achieved at avoidance costs of $31 \, \text{C/tCO}_2$ -eq. These net negative emissions are the sum of the avoided fossil fuels (crude oil as feed in refinery and natural gas both as feed and fuel for H_2 production), CO_2 captured from the biomass, and CO_2 credits for export of excess electricity produced.

The pyrolysis route shows a GHG reduction potential of 0.9 MtCO₂-eq (21%) and avoidance costs of 39 €/tCO₂-eq. The small GHG reduction potential is a direct result of the 'limited' size assumed for the pyrolysis train (1000 MW_{HHV} feed input), which enabled us to compare the performance with the BIG-FT sub-route. The results show that avoidance costs and GHG reduction potential of the pyrolysis and BIG-FT CCS sub-route are comparable.

The large uncertainty ranges, especially for the BIG vent and

pyrolysis routes, are due to stacking of (independent) uncertainties in the underlying input parameters. The high uncertainties for the BIG vent and pyrolysis routes are mainly because these routes depend on both biomass and natural gas prices, whereas EEMs and CCS depend predominantly on either natural gas or electricity prices. As the uncertainty ranges are most likely normally distributed, the extreme values will be less likely than the medium values.

4.2. Deployment pathways

Four promising pathways were identified based on the performance of the individual mitigation options, the technological maturity of these options and their potential impact on the core refinery processes. In all

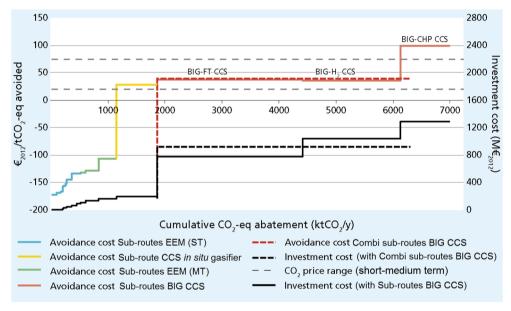


Fig. 4. Marginal abatement cost curve for pathway EEM-BIG-CCS.

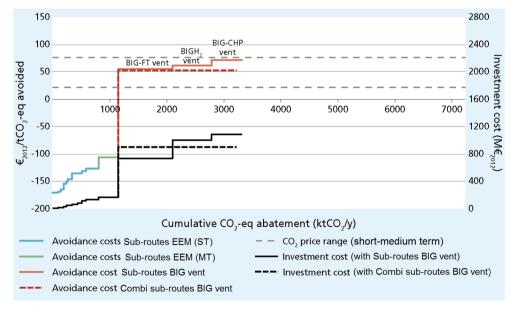


Fig. 5. Marginal abatement cost curve for pathway EEM-BIG-vent.

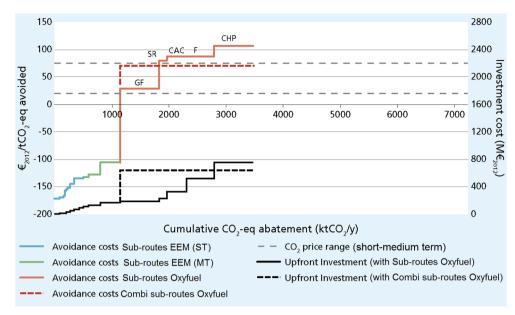


Fig. 6. Marginal abatement cost curve for pathway EEM-OXY. GF, SR, CAC, F and CHP stand for gasifier, steam reformer, catalytic cracker, furnaces and combined heat power, respectively.

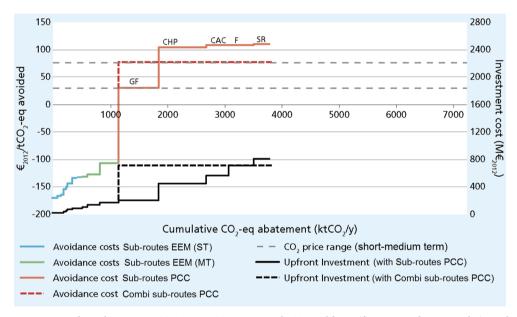


Fig. 7. Marginal abatement cost curve for pathway EEM-POST. GF, SR, CAC, F, CHP and PCC stand for gasifier, steam reformer, catalytic cracker, furnaces, combined heat power and post-combustion capture, respectively.

four pathways, EEMs are implemented first due to their negative avoidance costs. Albeit implementing EEMs first may create a lock-in of the current refinery infrastructure, fossil-based refined products will most likely continue to play a significant role up to the year 2030, thus ensuring an outlet for these products in the coming decades. The first pathway, EEM-BIG-CCS, has the lowest *average* avoidance cost and highest emission reductions, due to the favorable performance of the BIG-CCS option. BIG vent and CCS were also considered separately in three other pathways to explore the possibility that the other technology, CCS or biomass gasification, would not become available in the

future (on time) (lower risk pathways). The CCS technologies chosen are oxyfuel combustion (lowest cost) and post-combustion (most technologically mature and lowest potential impact on core refinery processes). For the biomass utilization pathway, BIG was chosen over pyrolysis due to the large uncertainty in the input parameters of the latter technology. Furthermore, the BIG options are complementary and provide opportunities for synergies.

To summarize, the following four deployment pathways were investigated:

Table 8

GHG emissions reduction potential and costs of applying one individual mitigation category to the refinery (column 2, as also presented in Fig. 3) and of the four studied pathways in which the interactions among mitigation categories are taken into account (see column 3). The EEMs are the first mitigation category to be implemented in all four pathways and are therefore similar to the values as presented in column 2. Column 4 presents the difference between the reduction potential and costs of the individual mitigation category and the pathway. The costs and GHG emissions reduction potentials of CCS and BIG apply to the combi cases. The abbreviation %pt stands for percentage point.

	Unit	NO INTER Individual	RACTION mitigation category	WITH INTERACTION Pathway		COMPARISON Difference pathway mitigation category	Difference pathway with individual	
	Oilit							
Pathway EEM-BIG-CCS		EEM	BIG CCS	EEM	CCS in situ gasifier + BIG CCS	Total	BIG CCS	Total
GHG emissions avoided	MtCO2-eq/y	1.1	5.0	1.1	5.2	6.3	0.1	1.3
Reduction base case 2012	%	28%	140%	28%	126%	154%	-14%pt	14%pt
Upfront investment cost	·10 ³ M€	0.2	0.8	0.2	0.8	0.9	0.0	0.2
Average GHG avoidance cost	€/tCO ₂ -eq	-133	31	-133	39	12	8	-19
Pathway EEM-BIG-vent		EEM	BIG vent	EEM	BIG vent	Total	BIG vent	Total
GHG emissions avoided	MtCO2-eq/y	1.1	2.5	1.1	2.1	3.2	-0.4	0.7
Reduction base case 2012	%	28%	61%	28%	51%	79%	-10%pt	18%pt
Upfront investment cost	·10 ³ M€	0.2	0.8	0.2	0.7	0.9	0.0	0.1
Average GHG avoidance cost	€/tCO ₂ -eq	-133	34	-133	52	-13	18	-48
Pathway EEM-OXY		EEM	Oxyfuel	EEM	Oxyfuel	Total	Oxyfuel	Total
GHG emissions avoided	MtCO2-eq/y	1.1	3.3	1.1	2.3	3.5	-1.0	0.2
Reduction base case 2012	%	28%	81%	28%	57%	85%	-24%pt	4%pt
Upfront investment cost	·10 ³ M€	0.2	0.8	0.2	0.6	0.7	-0.2	0.0
Average GHG avoidance cost	€/tCO ₂ -eq	-133	71	-133	71	4	0	-67
Pathway EEM-POST		EEM	Post-combustion	EEM	Post-combustion	Total	Post-combustion	Total
GHG emissions avoided	MtCO2-eq/y	1.1	3.7	1.1	2.6	3.8	-1.0	0.1
Reduction base case 2012	%	28%	90%	28%	64%	92%	-26%pt	2%pt
Upfront investment cost	·10 ³ M€	0.2	0.7	0.2	0.6	0.8	0.0	0.1
Average GHG avoidance cost	€/tCO ₂ -eq	-133	76	-133	77	14	1	-62

- Pathway EEM-BIG-CCS Consecutive implementation of EEMs and BIG CCS.⁸
- Pathway EEM-BIG-vent Consecutive implementation of EEMs (short and medium term) and BIG vent;
- Pathway EEM-OXY the consecutive implementation of EEMs (short and medium term) and the CCS technology with the lowest GHG avoidance cost (i.e. oxyfuel combustion);
- Pathway EEM-POST Consecutive implementation of EEMs (short and medium term) and the CCS technology with the lowest impact on the core refinery processes (i.e. add-on technology post-combustion capture);

Marginal cost abatement curves were constructed for all four pathways, starting with the route with the lowest avoidance cost (see Figs. 4–7). These curves show the medium values of the computed GHG reduction potentials and *marginal* avoidance costs, i.e. the costs per tonne of CO₂-eq avoided of the last implemented sub-route. The colored lines denote the *marginal* avoidance costs of the individual sub-routes, whereas the black lines represent the cumulative upfront investment costs. In all pathways, the abatement curves are similar for the deployment of the EEMs (blue and green lines). The dashed red lines represent the avoidance cost of the combined sub-routes, whereas the dashed black lines show the total cumulative upfront investment cost of the EEMs and combined cases. An overview of the results on the pathways and the combined sub-routes is presented in Table 8.

Pathways EEM-BIG-CCS and EEM-BIG-vent show GHG reduction potentials of $6.3\,\mathrm{MtCO_2}$ -eq/y (154%, i.e. net negative emissions) and $3.2\,\mathrm{MtCO_2}$ -eq/y (79%), respectively. When comparing the GHG reduction potentials of the pathways with the projected refinery $\mathrm{CO_2}$ emissions in 2030, the reduction potentials are reduced to 108% (EEM-BIG-CCS) and 55% (EEM-BIG-vent), respectively. Compared to the CCS routes, the EEMs and BIG routes reduce each other's emissions

reduction potential to a smaller extent (BIG CCS: -14%pt; BIG vent: -10%pt), because most EEMs are heat related and do not interact with BIG (see Table 8). The EEMs interacting with the BIG-CHP sub-route are energy management & control, motors & pumps and steam distribution system. These EEMs reduce primary energy input of the gas turbines/ CHP's with 30%, which decreases the mitigation potential of the BIG-CHP sub-route. Similarly, the BIG-H2 sub-route interacts with the EEM hydrogen management & recovery, which lowers the natural gas input of the hydrogen production process by 2%9; this leaves a smaller reduction potential for the BIG technology. 10 The reduction in average avoidance costs of the pathway compared to applying only BIG (EEM-BIG-CCS: -19 €/tCO₂-eq; EEM-BIG-vent: -48 €/tCO₂-eq) is not as large as for the CCS pathways, but still considerable. However, the uncertainties in average avoidance costs for both pathways (EEM-BIG-CCS: -93 to $81 €/tCO_2$ -eq; EEM-BIG-vent: -180 to $100 €/tCO_2$ -eq) are relatively high, mainly due to their high dependence on energy prices. The marginal avoidance costs of the combined sub-routes (dashed red and black lines) are lower than for most individual BIG sub-routes (solid red and black lines), and are around the upper end (75 €/tCO₂) of the CO2 price range. The difference in the GHG reduction potential of the BIG-combi CCS sub-route (6.3 MtCO₂-eq/y) and individual BIG subroutes (7.0 MtCO₂-eq/y) in the EEM-BIG-CCS pathway is because of the lower amount of CO2 captured in the combi sub-route. 11 The GHG

 $^{^8}$ Low-cost CO_2 capture from the *in situ* gasifier is included in this pathway.

 $^{^9}$ In reality, the interaction between the EEMs hydrogen management & recovery and the BIG-H $_2$ sub-route will be smaller as not all the energy savings are related to the SR unit, but also to the hydrogen distribution system, which does not show any interactions with the BIG-H $_2$ sub-route.

 $^{^{10}\,\}rm It$ was assumed that the EEM turbine pre-coupling, which uses the exhaust gas of a newly built gas turbine to supply heat to the *in situ* SR furnace, can be used together with the BIG-H $_2$ sub-route as well without any interaction between both sub-routes.

 $^{^{11}}$ In the BIG-FT CCS and BIG-combi CCS facilities, the CO_2 capture unit is placed after the sour WGS and AGR units. However, the syngas needs to be further shifted in a sweet WGS unit for the production of hydrogen, power and

reduction potential of the pathway with the BIG-CCS combi sub-route could be enhanced to $7.0\,MtCO_2$ -eq/y as well by designing the BIG facility differently, which would result in slightly higher avoidance costs. However this option was not examined in this study.

The total GHG reduction potentials of pathways EEM-OXY and EEM-POST are 3.5 MtCO₂-eq/y (85%) and 3.8 MtCO₂-eq/y (92%), respectively. When comparing the GHG reduction potentials to the projected refinery CO2 emissions in 2030, the potentials are reduced to 59% (EEM-OXY) and 64% (EEM-POST). After the EEMs, pathways EEM-OXY and EEM-POST both start with CO₂ capture from the in situ gasifier (30 €/tCO₂-eq). Although CO₂ capture from the high-pressure process gas of the in situ SR is not an oxyfuel technology, this sub-route was included in pathway EEM-OXY - second point source from which CO₂ is captured - to maximize emissions reductions. The EEMs and CCS routes show large interactions: the more emissions are reduced via EEM, the less emissions can be reduced via CCS. In pathways EEM-OXY and EEM-POST, 24%pt and 26%pt, respectively, are reduced less than when CCS is applied without EEMs (see Table 8). In total, only 4%pt and 2%pt emissions, respectively, are reduced more in the pathways than when only CCS is applied. Nevertheless, the average avoidance costs of the entire pathways are substantially lower (EEM-OXY: -67 €/tCO₂-eq; EEM-POST: -62 €/tCO₂-eq) than when applying only CCS due to the negative avoidance cost of the EEM. The marginal avoidance costs of the combined CCS sub-routes (dashed red lines) are lower than for most individual CCS sub-routes (solid red lines), mainly due to economies of scale of the capture equipment (and CHP plants).

Under full deployment, the cumulative upfront investment costs (dashed black lines) for the BIG pathways (0.9 billion euros) are higher than for EEM-OXY (0.7 billion euros) and EEM-POST (0.8 billion euros), but all four pathways show large uncertainty ranges of \pm 0.3-0.4 billion euros. Significant reductions in investment costs (0.1-0.3 billion euros) can be achieved by combining the CCS and BIG sub-routes in one combi CCS or BIG facility (solid black lines). Note that up to an emissions reduction of 45%, the performance of the pathways is rather similar as they all start with EEMs and three of them follow with CO₂ capture from the in situ gasifier. The avoidance costs of the pathways start to differ for higher emissions reductions. Although the average avoidance cost of pathway EEM-BIG-CCS is higher than for EEM-OXY and EEM-BIGvent under full deployment, the average and marginal avoidance costs are lowest up to 6.1 MtCO₂/y avoided. However, next to the techno-economic performance, the feasibility of the pathways also depends on the availability of a sustainable biomass supply chain and/or a CCS infrastructure as well as the operator's willingness to use novel technologies and (partly) abandon conventional operational processes. While the EEM-POST pathway involves relatively little change in the process units, the EEM-OXY pathway already requires more modifications. The BIG pathways are new concepts that completely differ from the core refinery process.

The method has proven useful to evaluate emissions reduction strategies in an industrial plant based on criteria such as GHG avoidance cost and GHG reduction potential. It can also be applied to other refineries and industries to identify such strategies, provided sufficient data on core processes is available. However, there is scope to improve the method and ensure that complete optimization of

(footnote continued)

steam. The CO_2 formed during this reaction is not captured but vented to the atmosphere, resulting in a lower total amount of CO_2 captured than in the individual BIG facilities together. This effect is reinforced by the fact that the syngas composition in the BIG-combi facility is geared towards a low H_2 :CO ratio, which is necessary for the production of the FT fuels, but sub-optimal for the production of the hydrogen, power and steam. Consequently, more CO_2 is created in the sweet WGS unit. This issue can be tackled by installing an additional CO_2 capture unit after the sweet WGS unit, which would increase the avoidance cost of the BIG-combi CCS facility. This option would still be cheaper than implementing three individual BIG facilities separately.

emissions reduction pathways is achieved (see discussion section). Nevertheless, the results described above provide the refinery owner with information about the costs and GHG reduction potential of various mitigation options and deployment pathways, their ranking in terms of costs, their uncertainty and the key parameters affecting their performance. As a result, the demonstrated method in this study can help inform companies about the risks related to investments in certain technologies. The findings may also serve as valuable input for plant owners when doing detailed follow-up analyses with inhouse data to reduce uncertainties and refine the strategy. These evaluations should also consider drivers and barriers to low-carbon energy technology investment. For instance, a potential driver for the implementation of energy efficiency measures and use of biomass is lower sensitivity to fluctuations in fossil fuel prices. Conversely, investments in CCS may be hampered by high risks related to the availability of CO2 transport and storage infrastructure, public acceptance, the liability of geologically stored CO₂.

5. Discussion

This section provides a comparison of the current results and findings of other studies (Section 5.1). Subsequently, the context and main limitations of the analysis are described (Section 5.2).

5.1. Comparison with other studies

The results were compared with findings in other studies by converting these findings to €2012 (see Table 9). Note that other dissimilarities (i.e. input parameters and system boundaries) were not considered unless otherwise indicated. The total GHG reduction potential of the EEMs (20-36%) was found to be in line with figures in other studies investigating the EEMs in US (20-30%) and European $(20-50\%)^{12}$ [46] refineries. However, lower values (10-20%) were found by Szklo and Schaeffer [47]. This can be explained by: (i) the refinery-specific GHG reduction potential; and (ii) the exclusion of particular EEMs from their analysis due to unfavorable economics and/or practical issues. The avoidance cost ranges indicated by Ecofys [46] are similar to values found in this study; however, less optimistic values were reported by Holmgren and Sternhufvud [48]. A more detailed analysis into the costs and practical issues related to the EEMs in the studied refinery is therefore recommended. For the CCS cases, only the capture costs were compared, i.e. the costs for the CO2 transport and storage step were subtracted from the avoidance costs found in this study. The average avoidance costs for post-combustion capture are in agreement with Johansson et al. [17], but are lower than the findings of Kuramochi et al. [8], which can be partly explained by the large economic scale effects and high revenues for electricity sale in the current study. The lower emissions reductions in the other two studies are due to the fact that CO₂ is not captured from the NGCC-CHP [8], or from all possible available point sources [17]. The emissions reductions and avoidance costs of the oxyfuel and precombustion cases are in line with other studies [8,38]. The avoidance costs of the BIG fuel case of Meerman et al. [18] are similar to our own findings.

Finally, due to lack of data on avoidance costs for pyrolysis fuel production with a similar production process as used in this study – which was based on NREL data [44] – we compared the production costs of the first production step, the actual pyrolysis, which is similar in most studies covering fast pyrolysis. A generic equation presented by Bridgwater [49] was used to compute the bio-oil production cost (214 €/t bio-oil); this figure is in agreement with the cost found in this study (206 €/t bio-oil).

 $^{^{\}rm 12}\,\rm The$ high end value refers to old and inefficient refineries with an extremely high reduction potential.

Table 9
Comparison of results with other studies. The results from the present study are highlighted in bold. The results for CCS exclude costs for CO₂ capture from the gasifier and CO₂ transport & storage for the sake of comparison.

	GHG reduction potential (%)	Geographical scope	Aggregation level	Economics (€ ₂₀₁₂ /tCO ₂ -eq)	Reference
EEMs	20–36	NW Europe	Refinery	-226 to -33	
	20-50	Europe	Sector	-150 to -102	[46]
	20-30	US	Sector	N/A	[34]
	8–22	Sweden	Refinery	-98 to -27	[48]
	10-20	Brazil	Sector	N/A	[47]
	5–20	Europe	Sector	N/A	[14]
Post-combustion capture (combined)	66-75 ^a	NW Europe	Refinery	45-167	
•	61–77 ^a	Not indicated	Sector	40-130 ^b	[17]
	59	Worldwide	Sector	104-168	[8]
Oxyfuel combustion (combined)	62-63	NW Europe	Refinery	53-106	
•	77	Worldwide	Sector	52-81	[8]
Pre-combustion (combined)	65	NW Europe	Refinery	54-152	
	64–82	Netherlands	Refinery	87-90 ^c	[38]
BIG-FT vent	11–25	NW Europe	Refinery	64-123 ^d	
	N/A	NW Europe	Plant	82-137	[18]
BIG-FT CCS	48–60	NW Europe	Refinery	41-65	
	N/A	NW Europe	Plant	38-66	[18]

^a The GHG reduction potential of this study and Johansson et al. [17] include CO₂ capture from the NGCC-CHP, whereas this is not included in Kuramochi et al. [8].

5.2. Discussion of method, data quality and results

Four main points of discussion have been identified.

First, this study aimed to develop a method to identify emissions reduction strategies. Although this method has proven useful, further efforts are required to achieve complete optimization. Several aspects should be considered in further detail, such as biomass supply chains, CCS infrastructure, age and decommission of existing capital stock and the willingness of industries to use novel technologies. Furthermore, the calculations on the industrial plant need to be better linked to the temporal dynamics of the energy systems in which it is embedded, including changes in industrial demand, energy prices and the availability of hydrogen. A more sophisticated sensitivity analysis with a better characterization of uncertainty ranges of the input parameters would improve the quality of the results and be useful input for strategic planning [50].

The second issue concerns paucity of data and assumptions on refinery processes and mitigation options. Limited information on core refinery processes made it necessary to make assumptions on the refinery hydrogen production output as well as on capacity factors and efficiencies of process units. However, the effect of these assumptions on the final results is expected to be minor. For the same reason, the pyrolysis and FT-fuel production routes were examined as additional output products (processed in new equipment) rather than substitutes of fossil gasoline and diesel, which were kept constant in the analysis. For the pyrolysis fuel route, utilizing existing process units could result in significantly lower avoidance cost [51]. Also, heat integration of the FT-process in the refinery could enhance the GHG reduction potential and economic performance of this mitigation option [52]. However, at the same time, the GHG reduction potential and avoidance cost of several EEMs would deteriorate, once green fuel production replaces part of the crude oil throughput, due to a lower fossil energy saving potential. Follow-up research is needed to determine at which level of replacement implementing EEMs remains beneficial, and when it is better to omit EEMs and implement the green fuel technologies directly. For the pre-combustion route, using an Autothermal Reformer (ATR) with CO2 capture is probably economically preferable over a SR with CO2 capture from both the high-pressure process gas and fuel gas stream [53,54]. More detailed and reliable data on this mitigation option is needed. Also, possible improvements in post-combustion solvents, and the oxygen and hydrogen production efficiency could lower the avoidance costs of the capture technologies. Several mitigation options (e.g. low-grade WH utilization for district heating and geothermal energy) were not investigated in this study, but could be promising from a GHG reduction perspective. Golombok and Beintema [55] report that under favorable conditions, utilizing geothermal energy for process heating could have a high financial reward for a large scale refinery, but risks are currently too high and further research is required before this process can become economically viable. A follow-up study for the case refinery would require more detailed plant data as well as a total site analysis to assess the energy recovery by the different site processes.

Third, the cost estimates of the mitigation options involve some degree of uncertainty. Although key techno-economic data underlying the mitigation options stem from studies covering multiple data sources, these figures are not tailored to the specific configuration of the case refinery. Due to the lack of detailed data on investment costs for EEMs, costs were derived using the simple payback period equation. More reliable and detailed data is needed on investment costs for EEMs to eliminate the large uncertainty in the avoidance costs. All the input cost data pertain to technologies that are commercially mature, assuming the costs of advanced EEMs, CCS and biomass routes will decline over time as a result of technological learning. Yet, this will depend strongly on the deployment of these technologies over the coming decades. Moreover, it should be noted that several factors affecting the practical feasibility and economic performance of the mitigation options have been neglected, such as the creation of space for new process equipment and biomass storage, (production loss due to) retrofit and lower process reliability. A rough calculation by the authors indicates that a storage dome with a 40 m diameter and 50 m height would be required to run the combined BIG facility for one week. 13 Considering

b The range only covers the cases with an NGCC-CHP as energy plant for post-combustion capture as presented in Johansson et al. [17].

^c These are medium values; the sensitivity of these results were explored by doing a sensitivity analysis for each input parameter. However, no cumulative cost uncertainty range is available.

d We recalculated our values by using the natural gas price (6 €/GJ), TOPS price (6.7 €/GJ) and crude oil price (73 €/bbl) of Meerman et al. [18].

 $^{^{13}}$ Calculation based on a calorific value of 19–23 MJ $_{\rm LHV}/kg$ [4], a bulk TOPS density of 750–850 kg/m 3 [4], and a dome-shaped storage facility with a diameter of 63 m, a height of 50 m and a volume of 110,000 m 3 [7].

the space availability on the case study refinery site this should be practically feasible. Furthermore, (part of the) biomass could be stored in river barges until unloading to the storage domes. Another issue not covered in this study is that hydrogen and/or FT-fuel production via TOPS gasification will result in lower O&M expenses, but possibly also undepreciated capital costs for the conventional hydrogen and fuel production routes; these costs have not been taken into account. Lastly, limited water availability in the future could become a critical factor for water intensive technologies like post-combustion capture technology [56]. Although most of these neglected factors probably fall within the uncertainty ranges used for the most important input parameters underlying the analysis, more research is desirable to acquire more accurate results. The avoidance costs of some mitigation options (e.g. post-combustion capture) include economic revenues and avoided GHG emissions related to the sale of excess electricity or steam produced in the process. One might argue whether these revenues should be taken into account. When excluding electricity sale, the avoidance cost of the combined post- and pre-combustion mitigation options would increase from 40 to $142 \in /tCO_2$ -eq (post) and $46-129 \in /tCO_2$ -eq (pre) to 49–158 €/tCO₂-eq (post) and 50–144 €/tCO₂-eq (pre). For the BIG-FT vent and BIG-FT CCS cases, the medium avoidance costs rise with 54 €/ tCO₂-eq and 13€/tCO₂-eq, respectively, when excluding electricity sale. Finally, it should be mentioned that the computed avoidance costs also depend on the regional scope. The high European natural gas price used in this study (10 €/GJ) results in high avoidance costs for the precombustion route; in the US, however, pre-combustion (and postcombustion) capture will perform much better due to the lower natural gas price, which was on average around 3 €/GJ in 2012 [57].

The fourth issue is related to the CO2 emission projections for the refinery. The results show significant increases in the business-as-usual CO₂ emissions projections until 2030 (16%) as a result of the increasing MD/G ratio, assuming the heavy fuel oil and total refinery product output in the case refinery remain constant. These constant output levels are based on the premise that the studied complex refinery will be more likely to survive in the market at the expense of simple refineries, which require more investments in new processing capacity to accommodate for the changes in product demand. Furthermore, the increase in CO2 emissions was based on the CONCAWE model, for which several assumptions were made inter alia regarding the share of gasoline vehicles in the total EU car fleet (50%) and the alternative fuel vehicles growth (15% in the total car fleet) for 2030 [29]. The projected CO₂ emissions are, therefore, uncertain. The GHG reduction potentials of the deployment pathways investigated in this study will be higher due to the projected increase in future CO2 emissions of the refinery. The combined effect of an increasing MD/G ratio and stricter product quality regulations will increase the hydrogen demand, while the output of the catalytic cracker, which is geared towards a high gasoline output, will decline. As a result, the potential for CO2 capture from the high-concentration SR may be higher than indicated in the results, whereas the opposite is true for the catalytic cracker. This effect has not been examined in this study due to limited data available on the core refinery processes that is needed to allocate the future emission increases to the specific CO2 point sources.

6. Conclusions

The objective of the paper was to design and test an integrated method to assess deployment strategies for deep GHG emission reductions in industrial plants. Important steps in the method are the identification of GHG emission mitigation options and the design of pathways combining different options. Next, the techno-economic

performance of both individual options and pathways are evaluated. The evaluation accounts for interactions between options such as the impact on each other's GHG reduction potential, cost synergies, economies of scale, and lock-in effects. The method was demonstrated for a large, complex petroleum refinery in North-West Europe with emissions of around 4.1 MtCO₂ in 2012.

For the short term, energy efficiency measures were identified as mitigation options, while for the medium term additional efficiency measures, carbon capture and storage (CCS), biomass gasification and fast pyrolysis of biomass were assessed. The efficiency measures were found to be non-regret options due to their negative GHG avoidance costs, but have a limited GHG reduction potential of 28% in total. Further GHG emission reductions can be realized in four identified pathways which all start with efficiency measures and are followed by either oxyfuel or postcombustion capture, or BIG with or without CCS. Under medium conditions, a pathway of efficiency measures and biomass gasification with CCS was found to be most cost-effective due to cost synergies between the latter two options. Furthermore, such a pathway has the largest GHG reduction potential of 154% compared to the 2012 base case, resulting in negative emissions. This pathway can be realized with on average negative costs of -25 €/tCO₂-eq when 75% of the emissions are reduced due to the revenues from efficiency measures, while this would be $-5 \in /$ tCO2-eq in a CCS pathway based on oxyfuel combustion. In a sensitivity analysis, it was found that costs are highly dependent on uncertainties in especially energy prices, but also techno-economic parameters. When CCS is fully deployed, marginal avoidance costs vary between 44 and 115€/ $\ensuremath{\mathsf{tCO}_2}\xspace\text{-eq},$ while full deployment of biomass gasification with CCS results in a range of -64 to $98 €/tCO_2$ -eq.

The method has proven useful to evaluate emissions reduction strategies in an industrial plant based on criteria such as GHG avoidance cost and GHG reduction potential. It provides insight in the potential of various mitigation options, their uncertainty and the main parameters affecting their performance, which is also useful information for plant owners when doing detailed follow-up analyses and determining risks related to potential investments in these technologies. The method can also be applied to other refineries and industries to identify deployment pathways, but requires detailed techno-economic data on core processes as well as the inclusion of other mitigation options, such as electrification of processes, use of secondary materials and geothermal energy. Furthermore, the method could be extended to ensure that complete optimization of emissions reduction pathways is achieved. Given the overlap in cost ranges, evaluation of pathways should consider decommissioning of existing capital stock and the deployment of biomass supply chains and CCS infrastructure. Furthermore, temporal dynamics of the energy systems in which the industrial plant is embedded, such as changes in energy prices and the availability of cheap hydrogen, should be integrated in the analysis in a more sophisticated manner. Lastly, potential barriers to low-carbon technology investment should be considered, especially those related to CCS and biomass gasification which have struggled to attract serious levels of commitment and investment to date.

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Appendix A

Future trends were analyzed to make projections for the baseline GHG emissions for the year 2020 and 2030, which reflect the short and medium term, respectively. Johansson et al. [14] and Rootzén and Johnsson [58] analyzed scenarios on future market trends and trends for fuel quality in the EU refinery sector by reviewing studies from government and industry organizations [31,59], and by collecting data from the EU statistical database. This approach was adopted for the present study and updated using additional studies [29,30]. Furthermore, the approach was extended to the global level based on the World Energy Outlook (WEO) 2013 [27], Energy Technology Perspective [60], and Global Energy Assessment [28]. The following trends have been observed:

- The global refining sector will be re-shaped in the coming decades owing to a declining oil demand in the OECD countries alongside an increasing demand in non-OECD areas like Asia and the Middle East. Following liquid fuel demand, future refinery capacity additions are expected in non-OECD countries. The future global refinery capacity additions, however, will depend strongly on the crude oil demand increase, which ranges from 0.0 to 1.0 Gt/v (-3) to 42 EJ/v)¹⁴ for the year 2030 compared to 2012 in the different scenarios of the WEO. For 2012, an excess global crude oil refinery capacity was calculated of around 0.2 Gt/y (11 EJ/y). At the same time, new refinery projects with a total crude oil capacity of over 0.5 Gt/y (22 EJ/y) have been announced by several countries [27]. This overhang will lead to more competition for internationally traded crude and result in capacity cuts, especially in the United States and the EU where crude oil demand is expected to decline and refineries are older and less sophisticated than in other parts of the world. Strains on the oil refining industry will be amplified by a growing share of fuels (biofuels and oil supply coming from natural gas and coal) bypassing the refining system. Another notable trend is the global shift in demand from heavy (i.e. fuel oils and marine fuels) and light distillates (i.e. gasoline and naphtha) to middle distillates (i.e. diesel and jet fuel). In the EU, a constant increase in the middle distillates/gasoline (MD/G) ratio can be observed as a result of transport and fiscal policies that favor diesel use over gasoline. If there will be no reversal in policies, significant investments are required in new processing capacity - mainly hydrocracking, coking and desulphurization - to accommodate these throughput trends. The EU demand for fossil-based refined products is expected to decline over the coming decades from 709 Mt/y (30 EJ/y) in 2008 to 603 Mt/y (26 EJ/y) in 2030, caused in large part by legislative mandates to improve vehicle efficiency and increase the share of alternative fuels (e.g. biofuels, electricity, hydrogen) [29]. Despite the fall in total energy use in the EU refinery sector, the specific refinery energy consumption and CO₂ emissions are expected to rise due to higher demand for MD products. 15 The overall EU refining capacity is projected to drop with 0.4 Mt/d (6 EJ/y) up till 2035 owing to the combination of declining North Sea crude oil output, increased global competition, and loss of gasoline export markets [27].
- Legislation on fuel quality (mainly sulphur and aromatic levels) has been tightened over the last decades in several parts of the world, including the EU, and will become more stringent in the future. In the EU, more stringent standards will follow for marine fuels in the coming decade. Stricter quality specifications require significant investments in additional fuel purification capacity, resulting in higher specific energy requirement (SER) and concomitant CO₂ emissions.

Both trends are expected to partly affect the CO₂ emission profile of the case refinery. As indicated by Johansson et al. [14], complex refineries can rather easily diversify and change the product output mix, which enables them to cope with changes in fuel demand, and are thus more likely to survive in a competitive environment than simple hydroskimming refineries with low conversion capacity that need to make large investments in new processing capacity. Although the overall EU demand for refined product declines, it was assumed that the current product output level of the case refinery will be maintained. Nevertheless, the SER and CO₂ emissions in the refinery were assumed to increase as a result of the increasing MD/G ratio. Quantitative projections of future CO₂ emissions in the EU refining industry as a result of changes in the MD/G ratio and fuel quality requirements were made based on the refining model of the CONCAWE association [29,31]. Based on a projected increase in the MD/G ratio from 2.4 in 2012 to 2.8 in 2020 and 3.3 in 2030 in the CONCAWE base case scenario, the specific refinery CO₂ emissions (tCO₂/t product) were found to increase with 8% in 2020 and 17% in 2030 compared to 2012. Assuming the relative change in total CO₂ emissions for the EU refining sector applies to the individual refinery as well, this corresponds with additional CO₂ emissions of 0.3 MtCO₂/y in 2020, increasing to 0.6 MtCO₂/y in 2030. ¹⁶ Furthermore, as a result of planned stricter quality regulations, the case refinery's CO₂ emissions were found to increase with 11% up till 2020 (0.5 MtCO₂/y) compared to 2012. The planned tightening in regulations applies particularly to marine fuels. Additional legal changes applying to other refinery products (e.g. gasoline, jet fuel, diesel, heating oil and heavy fuel oil) might be introduced in the period 2020–2030. These potential product quality changes could result in an increase of 16% (0.6 MtCO₂/y) in the refinery's CO₂ emissions compared to 2012.

Appendix B

B.1 Energy efficiency measures

The largest primary energy reductions in the short term can be achieved via heat integration between the distillation units (\sim 3%) and improvements in energy management & control, such as the implementation of organization-wide energy management programs, energy monitoring and control systems (\sim 3%). Other short term EEMs are: the replacement, adjustment and upgrading of pumps and electric motor systems; improved insulation of steam distribution systems and steam traps; fouling mitigation (e.g. online cleaning of heat exchangers); and improved furnace performance. The latter can be achieved, for instance, via improved maintenance, inflow air preheating and/or the use of low-NOx burners, which reduces the need for energy consumption in the downstream selective catalytic reduction (SCR) unit. The last two short term EEMs are efficiency improvements in hydrogen production & management as well as more efficient flaring systems, such as flare gas recovery systems and/or the use of

¹⁴ The original values in the underlying sources were given in million barrels per day or EJ/y. For crude oil, an energy content of 6.12 GJ/bbl and a density of 0.14 t/bbl crude oil were assumed to compute the values presented in this study.

¹⁵ As MD production requires much higher amounts of hydrogen than gasoline production, and hydrogen production is an energy-intensive process, the overall balance tends towards increased energy use and CO₂ emissions at high MD/G ratios.

 $^{^{16}}$ Ideally, the additional CO₂ emissions at the refinery level would be computed on basis of the mass flows and additional hydrogen demand. However, this was impossible due to limited data availability on the core refinery processes. The CO₂ projections are therefore indicative in nature.

electronic ignition system. The joint primary energy reduction of short term EEMs amounts up to 15-16% (see Table B1).

For the medium term, large primary energy reductions are expected to come from advanced desulphurization systems, such as advanced hydrotreating or oxidative desulphurization (\sim 10%). Energy savings can also be realized through advanced separation systems; for example, dividing wall columns, which combine two conventional distillation columns into one, show lower heat losses due to better heat transfer. Lastly, a newly built gas turbine can be pre-coupled to the *in situ* SR so as to utilize the hot exhaust gas of the gas turbine to supply heat to the SR furnace. In total, an estimated reduction in primary energy use of 19–20% was computed for the medium term (see Table B1). A more detailed description of the EEMs can be found in Worrell et al. [34].

The EEMs improved energy management & control and heat integration & waste heat recovery apply to the overall refinery site and show interaction with each other and with the other 'isolated' EEMs: steam distribution system, fouling mitigation, and improved furnace performance. Assuming that heat integration & waste heat recovery will be implemented after the 'isolated' EEMs, and followed by energy management & control, the primary energy reductions of these two refinery wide EEMs are reduced from 2.0% to 1.9% and 3.0% to 2.8%, respectively.

Table B1
Input data for energy saving measures identified for the refinery for the short and medium term. The data are based on Worrell et al. [34]; the lifetimes are based on an expert's estimations [98]. The reductions are related to the total heat, electricity, and primary energy use in the refinery in the year 2012, thus excluding growth in emissions due to future trends.

	Reduction final energy use ^a			Reduction pr	Reduction primary energy use		Life time	Invest-ment ^b	
	Heat PJ/y	Electricity % PJ/y %		y %	РЈ/у %		- У	y	M€
at			.,				-		
Short term measures									
Energy management & control ^c	1.1	3	0.1	3	1.7	3.0	0.25	5	4.1
Heat integration distillation units	1.4	4	-	-	1.7	3.0	5	15	87
Motors & pumps ^d	-	_	0.1	3	1.4	2.4	3	10	42
Steam distribution system ^e	0.9	3	-	-	1.2	2.0	1	20	12
Heat integration & waste heat recovery	0.9	3	_	_	1.2	2.0	5	15	56
Fouling mitigation ^f	0.5	1	_	_	0.6	1.0	1	10	6
Improved furnace performance ⁸	0.5	1	_	_	0.6	1.0	2.5	15	14
Hydrogen management & recovery ^h	0.3	1	_	_	0.4	0.7	3	15	12
Flaring ⁱ	0.2	1	_	_	0.3	0.5	2.5	20	7
Total	5.8	16	0.2	6	9.1	15.6			240
Medium term measures									
Advanced desulphurization ^j	4.6	13	_	_	5.8	10.0	10	25	580
Advanced separation systems ^k	3.2	9	_	_	3.9	6.8	7	25	276
Turbine pre-coupling ¹	1.2	3	_	_	1.5	2.5	6	20	87
Total	9.0	25	0	0	11.2	19.3	-	-	942

^a A furnace efficiency of 80% and a single-cycle gas turbine CHP with an electric efficiency of 27% and heat efficiency of 56% were assumed to compute the heat and electricity savings, respectively. The primary energy reduction is assumed to result in natural gas savings. In case refinery fuels are saved, it was assumed that these can be used elsewhere in the refinery where they replace natural gas, or in case where this is not possible, to be sold to other parties against the same economic value per energy unit and carbon footprint as natural gas.

- b The investment costs were calculated using the simple payback period equation as presented in Section 2.2. The annual costs C (M€/y) were assumed to be zero.
- ^c E.g. the implementation of organization-wide energy management programs, energy teams or energy monitoring and control systems.
- ^d The replacement, adjustment and upgrading of pumps and electric motor systems.
- ^e E.g. improved insulation and steam trap improvement.
- ^f E.g. online cleaning of heat exchangers.
- g E.g. better furnace maintenance, inflow air preheating and/or the use of low-NO_x burners. The latter reduces the need for energy consumption in the selective catalytic reduction unit.
- h E.g. hydrogen network integration to match different hydrogen sources with uses based on the quality of the hydrogen streams.
- ⁱ E.g. implementing flare gas recovery systems and/or the use of electronic ignition system, which only burns fuel gas when needed, instead of flare systems with a continuously burning pilot flame.
 - ^j E.g. advanced hydrotreating or oxidative desulphurization.
 - k E.g. a dividing wall-column, which combines two conventional distillation columns into one resulting in a better heat transfer and lower capital cost.
 - 1 E.g. a newly built gas turbine can be pre-coupled to the in situ SR so as to utilize the hot exhaust gas of the gas turbine to supply heat to the SR furnace.

B.2 CO2 capture and storage

Three first generation CO₂ capture technologies are assessed: post-, pre- and oxyfuel combustion capture. While post-combustion capture has already been demonstrated on a commercial scale, pre- and oxyfuel combustion are not expected to be deployed in t. The studied concepts are:

- Post-combustion capture, in which CO₂ is separated from a flue gas stream. In this study, the CO₂ is extracted using the chemical solvent monoethanolamine (MEA).¹⁷ Post-combustion is an add-on technology as no drastic changes have to be made to the process units [8].
- Oxyfuel combustion, in which fuel is burned with nearly pure oxygen (instead of air), thereby creating an almost pure CO₂ off-gas. The oxygen is produced in a cryogenic Air Separation Unit (ASU). As several adjustments and additional equipment (e.g. piping, ducting, flue gas recirculation

¹⁷ Although improved solvents with lower specific energy requirements (SER) than MEA are available today, no reliable data was found on these solvents' SER for flue gases with different volumetric CO₂ concentrations.

fan) are needed to convert boilers, furnaces and gas turbines to oxyfuel mode, oxyfuel combustion is considered to be a retrofit technology [8].

• Pre-combustion capture, in which the fuel is decarbonized prior to the combustion step. In this study, methane and refinery gas are converted to hydrogen in a newly built SR. The CO₂ is captured from the high-pressure (HP) process gas between the watergas shift (WGS) reactor and Pressure Swing Adsorption (PSA) unit using the mixed solvent ADIP-X (methyldiethanolamine mixed with piperazine). Pre-combustion is also considered to be a retrofit technology as several adjustments need to be made to refinery infrastructure [35].

The key data of the CO_2 capture technologies can be found in Tables B2-B4. The CO_2 transport and storage costs (16 €/tCO_2) were taken from the Zero Emissions Platform [61,62]. Electricity and steam required for the post- and pre-combustion routes are produced in a newly built NGCC-CHP unit from which the CO_2 is also captured. Electricity demand for the oxyfuel route is purchased from the grid as electricity production in a newlybuilt gas turbine was found to be more expensive. As the newly built CHP units are dimensioned towards the heat demand, often excess electricity is produced, which is sold to the grid. Additional equations, assumptions and data used to calculate the techno-economic performance of the CHP units and CCS configurations can be found in Berghout et al. [38,45,63].

Table B2
Techno-economic parameters for post-combustion capture. The economic data pertain to a post-combustion system capturing annually $1.0\,\mathrm{MtCO_2}$ from flue gas streams of both 4 and $12-14\,\mathrm{vol}\%\,\mathrm{CO_2}$ using one absorber and one stripper. Based on: [56,89,96].

	Unit	Value	References
Technical			
CO ₂ capture ratio	%	87	[89]
Maximum processing capacity per unit			
Absorber	ktCO ₂ /d	3	[102,103]
Stripper	ktCO ₂ /d	10	[102,103]
CO ₂ compressor	ktCO ₂ /d	8	[104]
Electricity flue gas blower	GJ_e/tCO_2	0.35	[92]
Regeneration heat ^b	GJ_{th}/tCO_2	3.5-4.0	[95]
CO ₂ capture (pumps and fans) ^b	$\mathrm{GJ_e/tCO_2}$	0.1-0.3	[90,91]
CO ₂ treatment & compression	$\mathrm{GJ_e/tCO_2}$	0.6	[89]
CAPEX			
Flue gas ducting	M€/MtCO ₂ /y	12 ^c	[92]
Modifications to stacks	M€/stack	0.1	[92]
CO ₂ capture equipment ^d	M€/MtCO ₂ /y	75 (4 vol%); 49	[56,89,96]
2 1 1 1	. 2.	(12–14 vol%)	
CO ₂ treatment & compression ^d	M€/MtCO ₂ /y	14	[56,89,96]
Economic scaling factor	•		
Amine absorber		0.67	[105,106]
Stripper		0.67	[25,106]
Compressor		0.67	[104,105]
OPEX			
Labor	€/tCO ₂	0.2^{e}	[56,89,96]
Administration & overhead	% of labor	28	[96]
	cost		
Taxes & Insurances	€/tCO ₂	2.0	[96]
Maintenance	% of TPC	3.8	[96]
Water usage	€/tCO ₂	1.4	[96]
MEA	€/tCO ₂	0.5	[96]
Activated carbon	€/tCO ₂	0.1	[96]
Ammonia	€/tCO ₂	0.2	[96]
Corrosion inhibitor	€/tCO ₂	0.0	[96]
SCR catalyst ^f	€/tCO ₂	0.1	[96]
Other chemicals	€/tCO ₂	0.2	[96]

^a The parameters used for economic costs are based on three detailed studies [56,89,96], which describe post-combustion capture at a pulverized coal-fired (PC) power plant ($12-14 \text{ vol}\% \text{ CO}_2$ concentration) and at a natural gas fired combined cycle (NGCC, $3-4 \text{ vol}\% \text{ CO}_2$ concentration). Since the CO₂ concentration in the flue gases can change significantly among and within industrial plants, data from both the capture systems applied at the PC and NGCC power plants were used to account for this factor.

^b Regeneration heat and electricity needed for flue gas with volumetric CO₂ concentrations in the range of 4–16%; the specific regeneration energy (GJ_{th}/tCO₂; GJ_e/tCO₂) decreases with higher volumetric CO₂ concentrations. It is assumed there is no effect of scale on the specific energy consumption, which is concluded from a literature review [107].

^c A total flue gas ducting system of 5 km was assumed for the refinery.

^d All capital costs were standardized to and corrected for the amount of CO₂ captured of 1.0 MtCO₂ per year using one absorber and stripper. Scaling factors were used to account for differences in CO₂ capture capacity. The cost category CO₂ capture equipment represents the direct contact cooler (DCC), absorber and stripper; the cost category CO₂ treatment & compression comprises a CO₂ drying, purification, cooling (DPC) and compression unit.

e The average amount of labor hours were taken from CESAR [89], IEA GHG [56] and NETL [96], and multiplied with a European wage for an operator of 60,000 €/FTE/y [56].

f SCR stands for selective catalytic reduction.

Table B3 Techno-economic parameters for oxyfuel combustion capture.

	Unit	Value	Reference
Technical			
CO ₂ capture ratio	%	87	[108,109]
Maximum processing capacity			
Air Separation Unit	ktO ₂ /d	6	[110]
Oxygen production	GJ_e/tO_2	0.7^{a}	[94]
Stoichiometric O ₂ :CO ₂ combustion ratio (weight basis) ^b			
Cokes (catalytic cracker)		0.73	
Natural gas		1.43	
Refinery gas furnaces		1.45	[78,108]
Excess oxygen use	%	3	[111]
Fuel savings furnaces ^b	%	8.3	[108,109]
CO ₂ treatment & compression ^c	$\mathrm{GJ_e/tCO_2}$	0.5	[93]
CAPEX			
Furnace modification	M€/MtCO ₂ /y	1^d	[108,109]
Air Separation Unit (ASU)	M€/ktO ₂ /y	53 ^e	[42,108,109,112,113]
Cooling water system	M€/MtCO ₂ /y	12	[108,109]
Flue gas gathering system	M€/MtCO ₂ /y	7	[108,109]
CO ₂ treatment & compression	M€/MtCO ₂ /y	23	[113]
Economic scaling factor ASU		0.63	[41,105,113]
OPEX			
Labor	€/tCO ₂	1.0 ^f	[108,109]
Administration & overhead	€/tCO ₂	0.1	[108,109]
Maintenance	€/tCO ₂	3.0	[108,109]
Taxes & Insurances	€/tCO ₂	3.0	[108,109]
Water	€/tCO ₂	1.6	[108,109]
Consumables	€/tCO ₂	0.3	[108,109]

^a The current SER for cryogenic oxygen production was found to be in the range of $0.6-0.8\,\mathrm{GJ_e/tO_2}$ ($160-220\,\mathrm{kWh_e/tO_2}$) [114].

Techno-economic parameters for pre-combustion capture.

	Unit	Value	References
Technical			
Feed natural gas needed for H2 production	GJ_p/tH_2	122	[88]
Fuel natural gas needed for H ₂ production	GJ_p/tH_2	43	[88]
Electricity needed for H ₂ production	GJ _e /tH ₂	2.2	[88]
CO ₂ produced			
Production process (CO ₂ in process gas)	tCO_2/tH_2	6.9	[88]
SR furnace (CO ₂ in flue gas)	tCO_2/tH_2	3.2	[88]
HP steam from waste heat of the SR ^a	GJ_{th}/tH_2	25	[116]
CO ₂ capture ratio process gas	%	95	[37]
Maximum processing capacity			
Steam reformer	ktH ₂ /d	10	[117]
Heat for regeneration ADIP-X	GJ_{th}/tCO_2	1.97	[37]
Power for regeneration ADIP-X	GJ_e/tCO_2	0.04	[37]
Power for CO ₂ treatment and compression	$\mathrm{GJ_e/tCO_2}$	0.3	[37]
CAPEX			
SR plant (incl. WGS, PSA, SCR)	M€/MtCO ₂ /y	87	[88,118-120]
CO ₂ capture equipment	M€/MtCO ₂ /y	19	[88,117-120]
DPC & Compression	M€/MtCO ₂ /y	24	[88]
Modified burners	M€/MtCO ₂ /y	0.1	[78]
Economic scaling factor SR plant		0.67	[105]
OPEX			
Labor	€/tCO $_2$	0.4 ^b	[88]
			(continued on next page

 $^{^{\}rm b}$ Assumptions were made on the stoichiometric ${\rm O}_2$: ${\rm CO}_2$ combustion ratio due to insufficient information on the fuel composition in the refinery. In the catalytic cracker, oxygen is used to burn the coke that is deposited on the surface of the catalyst $(C + O_2 \rightarrow CO_2)$, resulting in a molar ratio of 1:1, which translates to a mass ratio of 0.73 (32/44). The mass ratio for the combustion of natural gas in the in situ gas turbine/CHP units (1.43) was derived from the volumetric O₂:CO₂ ratio as presented by C + B [115]. The oxygen requirement for the refinery gases was based on two studies performing detailed analyses into oxyfuel combustion of refinery fuel gases. The mass ratio derived from these studies was 1.44 [108] and 1.49 [78]. A value of 1.45 was used in this study. The high stoichiometric O2:CO2 mass ratio for refinery fuel gas is mainly due to the high methane and hydrogen concentrations in the fuel streams. However, as fuel gas compositions tend to vary considerably (also within refineries), the impact of the oxygen demand was indirectly examined by varying the energy use for oxygen production in the sensitivity analysis.

The treatment unit cools, purifies and dries the hot, wet CO₂ coming from the process units.

d Average values calculated from the total furnace modification costs in Allam et al. [108,109]. It was assumed there is no scaling effect for furnace modifications considering the small scale of most furnaces.

e Average value based on Allam et al. [108,109], Meerman et al. [42], Spero [112] and IEA GHG [113] and valid for a cryogenic ASU and the oxygen compressor to 40 bar.

f No data were available on the amount of operating manpower hours in Allam et al. [108,109]; only monetary values were presented.

Table B4 (continued)

	Unit	Value	References	
Administration & overhead	% of labor costs	20	[88]	
Taxes & Insurances	€/tCO ₂	3.7	[88]	
Maintenance	€/tH ₂	4.5	[88]	
Demineralized water	€/tCO ₂	0.9	[88]	
Chemicals ^c	€/tCO ₂	2.0	[88]	

^a The SR generates a lot of high-pressure steam (75 bar, 350°C) in a waste heat recovery boiler. In theory, $30 \, \text{GJ}_{\text{th}}/\text{tH}_2$ of WH would become available. Considering heat losses, it is assumed that $25 \, \text{GJ}_{\text{th}}/\text{tH}_2$ of steam can be produced.

B.3 Biomass gasification

The first biomass route is the production and processing of syngas through biomass gasification (BIG), which is subsequently converted into electricity, chemicals and/or transportation fuels. The BIG route has been investigated in numerous studies over the years, e.g. [43,52]. Gasification is a thermo-chemical process that converts carbonaceous materials (i.c. biomass) into syngas (mixture of mainly CO and H₂) carried out at high temperatures (> 900 °C) and by using a sub-stoichiometric amount of oxygen [64]. Next, a purification and optimization step is needed before the syngas can be converted into the final products. Four sub-routes are investigated for the case study, which are all preceded by a sour water-gas shift reactor and acid gas removal (AGR) process: (1) the production of steam and electricity by combusting the syngas/H₂ and combustible waste streams (diluted with N₂) in the CHP units (BIG-CHP); (2) the production of hydrogen by shifting the syngas in a sweet water-gas shift reactor and extracting the hydrogen from the resulting product gas using a pressure swing adsorption (PSA) unit (BIG-H₂); (3) the production of Fischer-Tropsch (FT) fuels by converting the syngas to linear alkanes and 1-alkenes, followed by a hydrocracking process to upgrade the intermediate products to FT-syncrude, and subsequently by a distillation process to separate the FT-fuels into FT-gasoline and FT-diesel (BIG-FT); and (4) all three output products combined (BIG-combi). The acid compounds are converted to liquid sulphur in the Claus/SCOT unit. All four sub-routes can be combined with CCS. CO₂ capture after the acid gas removal step is particularly interesting for two reasons. First, the process already requires the removal of CO₂ from the process gas. Second, the high partial pressure of CO₂ makes capture using a physical solvent relatively cheap.

Table B5 shows there is excess electricity production in the BIG-FT sub-route, excess steam production in the BIG-H₂ sub-route and a shortage of electricity production in the BIG-CHP, BIG-H₂ and BIG-combi sub-routes. These deviating electricity and steam flows are due to an assumed fixed heat-power ratio for the CHP units (see footnote c in Table B5), and excess heat and waste gas streams, which were assumed to be utilized for additional steam and electricity production, respectively. Excess steam and electricity were assumed to be sold to third parties and the grid, respectively, whereas an electricity shortage was compensated by purchasing electricity from the grid.

Table B5
Input data for biomass gasification sub-routes. Based on NEA [26,67] and Meerman et al. [42,43].

	Unit	REF	REF BIG-CHP		BIG-H ₂		BIG-FT		BIG-combi	
			Vent	CCS	Vent	CCS	Vent	CCS	Vent	CCS
Input refinery										
Natural gas ^a	MW_{LHV}	1167	607	607	560	560	1167	1167	0	0
TOPS	MW_{LHV} (MW_{HHV})	-	473	511	401	401	937 (1000)	937 (1000)	1499	1499
	t _{dry} /h	-	84	91	71	71	167	167	267	267
	tC/h	-	166	179	141	141	329	329	527	527
Output refinery ^b										
Steam ^{c,e}	MW_{th}	314	314	314	406 ^d	406 ^d	314	314	314	314
Elec. CHP ^{c,e}	MW _e	182	72	51	156 ^f	149 ^f	308 ^h	286 ^h	86	86
Hydrogen ^{e,g}	MW_{H2}	255	255	255	255	255	255	255	255	255
FT-fuels	MW_{FT}	-	-	-	-	-	458	458	458	458
Δ output REF										
Steam	MW_{th}	0	0	0	92	92	0	0	0	0
Electricity	MW_e	0	-110	-131	-26	-33	126	104	-61	-96
GHG emissions										
Emitted	tCO ₂ -eq/h	512	385	229	425	287	390	193	201	-116
Captured	tCO ₂ /h	_	_	162	_	139	_	202	-	324
FT (carbon)	tCO ₂ /h	_	_	_	_	_	107	107	107	107
Avoided	tCO ₂ -eq/h	_	127	283	87	225	122	319	311	627

(continued on next page)

b The average amount of labor hours were taken from CESAR [89], IEA GHG [56] and NETL [96], and multiplied with a European wage for an operator of 60,000 €/FTE/y [56].

^c SR and WGS catalysts, PSA sorbent, HP process gas solvent, MEA, SCR catalyst.

Table B5 (continued)

	Unit	REF	BIG-CHP		${\rm BIG\text{-}H}_2$		BIG-FT		BIG-combi	
			Vent	CCS	Vent	CCS	Vent	CCS	Vent	CCS
CAPEX OPEX ⁱ	M€ M€/y	-	266 16	328 19	264 16	268 16	571 34	582 34	765 45	783 46

^a Natural gas used in the base case for the production of steam, electricity and hydrogen.

- f In the BIG-H₂ sub-route electricity is consumed for the production of hydrogen, which is purchased from the grid.
- ^g The in situ SR unit is assumed to operate at an efficiency of 75%.
- ^h The waste gases coming from the gasification process are combusted in a newly built steam turbine to produce electricity.
- ⁱ Meerman et al. [42,43] assumed the O&M cost to be 4% of the total capital investment cost.

B.4 Fast pyrolysis

The second biomass route is the production and upgrading of pyrolysis oil from biomass as a means for generating infrastructure-ready renewable gasoline and diesel. At present, fast pyrolysis is the preferred process mode for the production of transport fuels as it exhibits high liquid yields [39]. Several advanced pyrolysis routes (e.g. catalytic pyrolysis) show promising economics, but are currently still at the R&D or pilot stage [65]. A typical process for pyrolysis oil production comprises several steps [51]:

- Pre-treatment: drying and size reduction (pre-treatment) of lignocellulosic biomass.
- Fast pyrolysis: thermal decomposition of carbonaceous material in the absence of oxygen in a reactor to produce raw pyrolysis oil, which is a liquid rich in oxygenated hydrocarbons and pyrolytic lignin.
- Hydrotreating: upgrading of the raw, unstable pyrolysis oil to a conventional hydrocarbon fuel by deoxygenating the oxygenated hydrocarbons and pyrolytic lignin by means of pressurized hydrogen gas at moderate temperatures (< 400 °C) in a fixed bed reactor. The co-produced off-gas is sent to a PSA unit for recovery of hydrogen gas. The PSA tail gas is routed to the SR for hydrogen production and partly to the furnace that preheats the feed for the hydrotreater.
- Hydrocracking and product separation: the separation of stable pyrolysis oil into light and heavy fractions, the hydrocracking (~425 °C; 90 bar) of the heavy fractions into light fractions, and the separation of the light fractions into gasoline and diesel. The off-gas is sent to the SR.

Direct use of pyrolysis oil in existing hydrotreaters and hydrocrackers on the refinery site would require complete deoxygenation and a low acid number to prevent corrosion, assuming the existing processing units are not made of dedicated stainless steel. Furthermore, the integration of these flows in the refinery would require additional information on the spare capacity of the process units as well as on the core process streams to determine the effect on the other refinery flows. Therefore, the pyrolysis fuels were assumed to be produced in new processing units and in addition to the refinery product output mix, which was kept constant in this study. For the same reason, the required hydrogen was assumed to be produced in a new SR. An economic lifetime of 20 years was assumed. Although the fast pyrolysis process is already commercial, the upgrading step to transport fuels requires further development. Hence, we assume this technology to become available in the medium term. Fast pyrolysis and upgrading to transport fuels have been investigated in numerous studies, e.g. [13,66]. The techno-economic input data in the referenced studies varies considerably and is based on several assumptions regarding fuel yield, fuel energy content and process conditions. A PNNL study [44], which presents recent, detailed and publicly available data coming from several US research institutes, was used for the analysis. Similar to the BIG-FT sub-route, the size of the pyrolysis train was limited to 1000 MW_{HHV} feed input, which allows for a fair comparison of both options.

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^b The capacity factors of the CHP units (power/steam production) and SR (H₂ production) are 91% and 56%, respectively. To meet the current power and steam demand, a similar capacity factor was assumed for the BIG-CHP sub-route. The BIG-H₂ sub-route was designed in such a way that the current H₂ demand can be met when operating the BIG train with a capacity factor of 91% (8000 h/y). For the BIG-FT sub-route, also a capacity factor of 91% was assumed.

^c The model was set to replace 560 MW of natural gas used as fuel for the *in situ* CHP units in sub-routes 1 (BIG-CHP) and 4 (BIG-combi). The CHP units were assumed to be dimensioned towards steam output (314 MW_{th}), as this is often the case in the refining industry. The BIG trains produce large amounts of WH, which is converted to medium and high-pressure steam. As a result, the steam demand from the CHP units is lower than in the base case. Assuming a fixed heat-power ratio, this results in less electricity produced (2 PJ_e/y) compared to the base case (5 PJ_e/y). The deficit electricity was assumed to be purchased from the grid. The fixed heat-power ratio was assumed, because of missing information on the CHP characteristics and steam quality.

d Excess heat in the gasification train is used for the production of excess steam.

^e The fossil fuel replaced in the BIG-CHP and BIG-H₂ sub-routes was considered to be natural gas. The small amount of refinery fuels entering these units in the base case were assumed to be used elsewhere in the refinery where they replace natural gas, or in case where this is not possible, to be sold to other parties against the same economic value per energy unit and carbon footprint as natural gas. In light of other uncertainties in this study, the effect of this assumption on the final results is expected to be minor.

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