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Techno-economic performance and challenges of applying CO_2 capture in the industry: A case study of five industrial plants

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ABSTRACT

To date, literature often presents generic results on the techno-economic performance of CO_2 capture in industry. Insufficient knowledge is available on the impact of site-specific factors on the feasibility of CO_2 capture at industrial plant level. This article presents a techno-economic analysis and an inventory of potential implementation and operational challenges related to the three main CO_2 capture technologies applied at industrial plant level for the short term (2020–2025) and long term (2040–2050). Five industrial plants from various industrial sectors (a medium and large sized petroleum refinery, a small and medium sized chemical plant, and a large hydrogen plant) in the Netherlands were used for this study.

The results show the lowest CO_2 avoidance costs for the refineries (24–57 €/t) and chemical plants (37–124 €/t) when operated in oxyfuel combustion mode, both for the short and long term, although post-combustion is economically preferable for the smallest chemical plant (117 €/t) in the short term. For the hydrogen plant, avoidance costs (67 €/t) are lowest when capturing CO_2 solely from the high-pressure process gas. For the short term cases, spatial constraints on existing plant sites could increase the indicated CO_2 avoidance costs, especially for post-combustion capture; for the long term cases, new-built capture ready process units, plant integration and optimized utilities are expected to lower the avoidance costs for all three capture technologies.

1. Introduction

Industrial energy contributes to roughly one third of the total CO_2 emissions worldwide (about 12 $GtCO_2$ in 2007) (Gielen et al., 2008). According to the reference case of the World Energy Outlook 2009 (IEA, 2009a), worldwide industrial energy consumption is projected to grow in primary terms from 185 EJ to 259 EJ in 2030. Several studies have looked at options to mitigate CO_2 emissions in the industrial sector. Many of these studies focus on the potential of increasing energy efficiency (e.g. Gielen et al., 2002; Phylipsen et al., 2002; Wang et al., 2007). However, forecast exercises have indicated that additional saving measures are necessary if stabilization targets of around 450 ppm(v) or lower are to be achieved. A combined portfolio of mitigation options including energy and material efficiency, feedstock substitution, and carbon capture and storage (CCS), is necessary to achieve larger reduction targets. Scenario work made by the IEA (Blue Map Scenario exploring the portfolio of measures needed to achieve 450 ppm(v) stabilization targets) concluded that CCS can contribute about one fifth to the required CO_2 emission reductions, of which almost half would take place in industry and fuel transformation sectors (IEA, 2009b).

To date, the techno-economic performance of CO_2 capture in the industrial sector has been assessed at the aggregate, sectoral and industrial plant level. The first category is mainly exploratory in nature and tends to take average industrial figures for CO_2 concentrations in flue gases (Damen et al., 2009; IEA, 2009b), energy and capital requirements for CO_2 capture (Damen et al., 2009), or only considered large and high concentration emission sources (IEA GHG, 2002). Saygin et al. (2013), on the other hand, carried out a top-down analysis for the Dutch industry, and distinguished between CO_2 concentrations in gas streams, specific energy and capital requirements for CO_2 capture and included the emission sources from less CO_2 emission and energy intensive sectors as well (e.g. food, paper). The second category focussed on usually one industry, e.g. the cement (IEA GHG, 2008) or iron and steel industry (Oda et al., 2007), and did not specify or examine the three main CO_2 capture technologies: post-, oxyfuel and pre-combustion capture. Up to now, only Kuramochi et al. (2012) made an extensive techno-economic assessment and comparison of the main CO_2 capture

Abbreviations: ASU, air separation unit; CCS, carbon capture and storage; CHP, combined heat and power; CT, combined technology case; DPC, drying purification cooling; FGD, flue gas desulphurization; GT, gas turbine; HP, high-pressure; LT, long term; MEA, monoethanolamine; ITM, ion transport membrane; ppm(v), parts per million (by volume); PSA, pressure swing adsorption; SCR, selective catalytic reduction; SR, steam reformer; ST, short term, single technology case; t, metric tonne; WGS, water-gas shift.

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Table 1
Overview of case studies investigated in this study.

Industry	Refinery I	Refinery II	Chemical plant I	Chemical plant II	Hydrogen plant
	Oil refining		Petrochemical		H_2 production
Mt CO_2/y	4.1	2.2	0.4	<0.1	0.8
Space availability	Low	Low	Low	Medium	Low
Point sources[a]	17[b]	8[b]	4	2	1

Based on: DHV (2009), Esso (2011a,b), RAP (2011), ROP (2011), Shell (2010, 2011).
[a] Flares and point sources smaller than 5 kt/year were not considered in this study.
[b] These point sources are scattered across the industrial plant site.

technologies for industrial processes in various sectors. In the third category, several bottom-up studies have been conducted to assess the techno-economic performance of CO_2 capture at industrial plants (e.g. Allam et al., 2005a,b; IEA GHG, 2000, 2008; van Straelen et al., 2009). Apart from the IEA GHG (2000, 2008) reports, in which one industrial plant was investigated, all the bottom-up studies examined only one type of CO_2 capture technology. So far, the techno-economic consequences of applying different types of CO_2 capture technologies in real industrial plants, in contrast to standardized models of an industrial process, have not been analyzed and compared. Therefore, insufficient knowledge is available on how the different types of CO_2 capture technology at specific industrial plants and how site-specific factors (e.g. scale effects and amount of point sources) can affect the CO_2 avoidance costs.

Furthermore, although several studies state that retrofitting industrial boilers and furnaces with CO_2 capture equipment is technically feasible (Allam et al., 2005a,b; IEA GHG, 2000; Wilkinson et al., 2003), no overview exists of additional challenges which may arise during implementation and operation of CO_2 capture equipment at industrial plants. It may, for example, be challenging to install flue gas ducting and CO_2 capture equipment on an already congested industrial site (Hurst and Walker, 2005; IEA GHG, 2008; van Straelen et al., 2009). More insights can help to evaluate whether such challenges could impede the adoption of CO_2 capture altogether, or to what extent they would affect the implementation and operation costs, and/or the technical performance of an industrial plant.

This is why the objective of this study is twofold, namely to investigate and compare the techno-economic performance of the three main CO_2 capture technologies in the industry, and to make an inventory of potential implementation or operational challenges related to these technologies. For this purpose, five industrial plants from various industrial sectors (a medium and large sized petroleum refinery, a small and medium sized chemical plant, and a large hydrogen plant) have been selected as case study. The variety in case studies allows for a comparison of different industrial sectors, plant sizes and plant configurations. All plants are located in the Dutch industrial area Rijnmond, which is appointed by the local government as a potential area for large scale CO_2 capture (RCI, 2011). Based on the results, efforts will be made to draw general lessons for CO_2 capture at industrial plants.

This paper is structured as follows. Section 2 describes the methodology and data used in this study, while Section 3 presents the results on the energy use, CO_2 emissions and costs for different CO_2 capture configurations. The research methodology, results, and uncertainties involved are discussed in Section 4. Lastly, the main conclusions are reported in Section 5.

2. Methodology and data

The bottom-up analysis allows for a detailed assessment of the techno-economic performance of different CO_2 capture technologies (post-, pre- and oxyfuel combustion) at industrial plant level. The analysis is based on five case studies for which several CO_2 capture configurations are considered. Additionally, a literature review and interviews with experts are conducted to identify and assess issues and remaining knowledge gaps related to the implementation and operation of CO_2 capture at industrial plants.

2.1. Case studies

Table 1 shows the five case studies investigated in this study, namely two petroleum refineries, two petrochemical plants, and a steam reforming hydrogen plant. These case studies were selected in order to assess how different characteristics of industrial plants can affect the techno-economic performance and challenges encountered during the implementation and operation of CO_2 capture equipment. The case studies vary with respect to industrial sector, annual CO_2 emissions (large vs. small emitters), space availability for capture equipment, and the distribution of the point sources. Two petroleum refineries were chosen to examine the effect of plant size and plant specific factors on CO_2 capture. The Shell refinery Pernis (refinery I) is one of the largest refineries worldwide with an annual throughput of around 21 million tonnes of crude oil, whereas the Esso refinery (refinery II) processes less than half the amount of crude oil per year (~10 million tonnes). Moreover, the refineries have low space availability on the plant site and a high number of dispersed point sources. The Rotterdam Aromatic Plant (RAP, chemical plant I) and Rotterdam Oxo-alcohol Plant (ROP, chemical plant II) are both small CO_2 emitters and have less point sources. Finally, the steam reforming hydrogen plant HyCO4 of Air Products is a medium sized CO_2 emitter that represents an attractive opportunity to capture CO_2 from high-pressure process gas, which requires relatively little energy, rather than only from the atmospheric pressure flue gases emanating from the stacks.

2.1.1. Core processes and data sets

Fig. 1 shows simplified flow diagrams of the core processes (demarcated by the blue boxes) in the industrial plants. The main mass and energy flows are indicated with arrows. Although both refineries (Fig. 1a) differ significantly in daily throughput, amount of point sources and process units (i.e. boilers, furnaces, catalytic crackers, CHP plants, hydrogen plants and gasifiers), the core processes are in general rather similar. A relevant difference is that the more extensive refinery I has four cracking installations and a separate gasifier, whereas refinery II has only one hydrocracker and a combined thermal gasification/cracking unit (Flexicoker). The process units from which the CO_2 is captured are indicated by the hatched boxes. The core processes of the chemical plants are presented in Fig. 1b and c. Unlike the refineries, the CO_2 is only captured from the furnace(s) (and boilers). As the CO_2 capture configurations are principally similar for all industrial plants, the dashed boxes were omitted in Fig. 1b and c. The hydrogen plant is not shown separately in Fig. 1 as the core processes are similar to the pre-combustion configuration in box III. In order to perform detailed and plans-specific calculations, data sets for the refineries and chemical plants (DHV, 2009; Esso, 2011a,b; RAP, 2011;

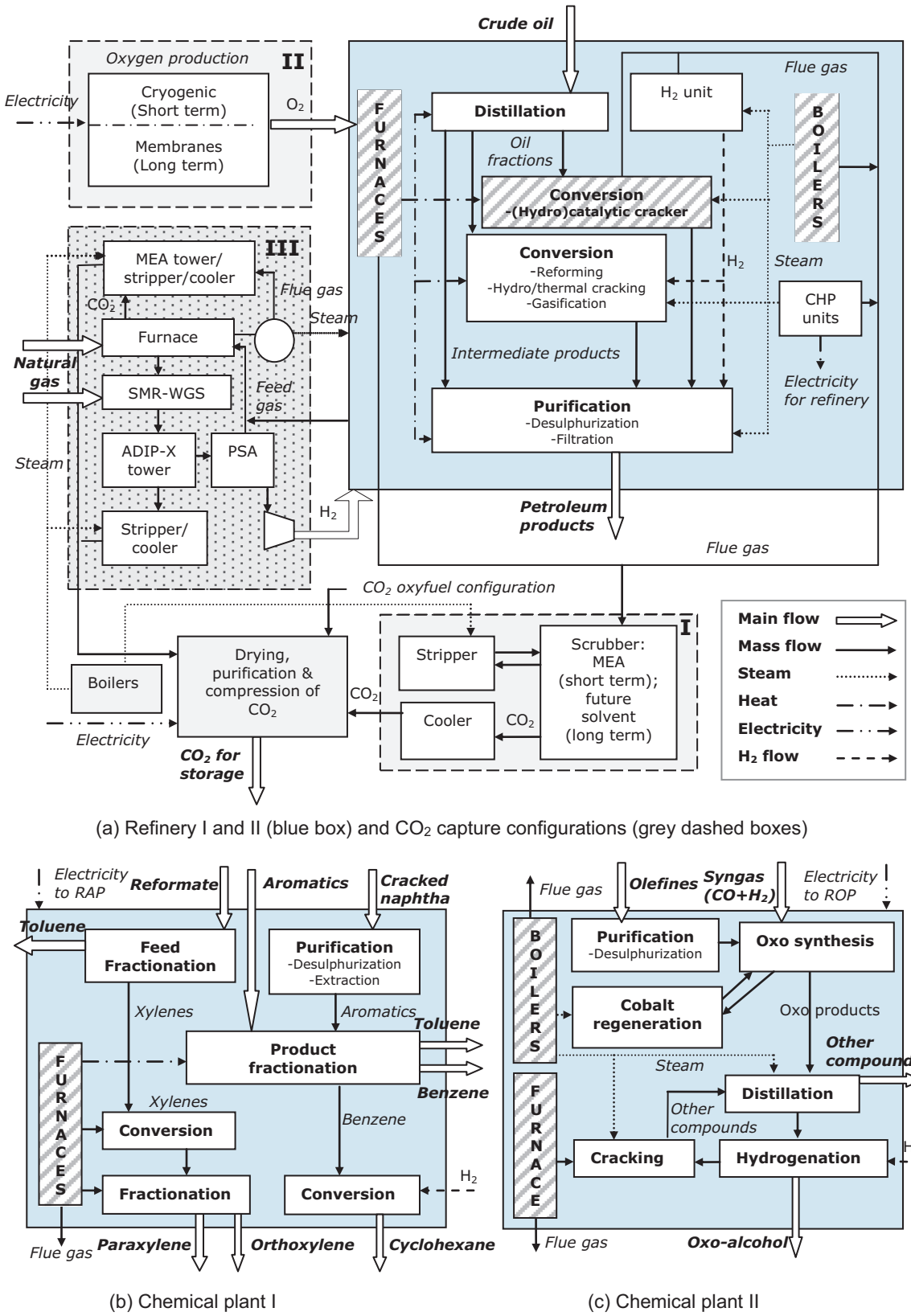

Fig. 1. Simplified flow diagrams of the core processes in the industrial plants (blue boxes) and the basic CO_2 capture configurations (dashed boxes) for post-combustion (I), oxyfuel (II) and pre-combustion technology (III). Box III (dashed-dotted box) also represents the flow diagram of the existing steam reforming hydrogen plant of Air Products with CO_2 capture. The hatched boxes represent the process units from which the CO_2 is captured. The CO_2 drying, purification and compression step is shared for all three CO_2 capture configurations. The long-term pre-combustion configuration by, for example, using a steam methane reformer in combination with a Sorption Enhanced Water Gas Shift Reactor or Water Gas Shift Membrane Reactor is not analyzed in this study due to limited data availability and thus omitted from the flow diagrams. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 2
General input parameters standardized for technical and economic performance calculations in this study.

Parameter	Unit	Nominal value		Sensitivity analysis	References
		Short term	Long term		
Real discount rate	%	10	10	5–15	Own value
Economic lifetime	Years	20	20	15–25	Own value
Total Plant Cost[a]	%-PPC[a]	130	130		van Horssen et al. (2009)
Total Capital Requirement[a]	%-TPC	110	110		van Horssen et al. (2009)
Calorific value natural gas	MJ_{LHV}/m_3	31.7	31.7		Rabou et al. (2006)
Heat content high-pressure steam	GJ_{th}/t	2.79	2.79		Rabou et al. (2006)
Industrial energy price					ECN/PBL (2010), IEA (2010), CBS (2011); own estimations
Natural gas[b]	€/GJ_{LHV}	9.3	14.0	±30%	
Electricity[b]	€/GJ_e	18.5	21.5	±30%	
CO_2 emission factor					
Dutch electricity production[c]	kg CO_2/GJ_e	88.9	9.3	±30%	van den Broek et al. (2011)
Natural gas	kg CO_2/GJ_{LHV}	56.7	56.7		Agentschap NL (2010)
Industrial boiler efficiency[d]	%	85	85		IEA GHG (2000)
Industrial furnace efficiency[d]	%	80	80		IEA GHG (2000)
Capital costs boiler	€/kW_{th}	78	78		Grahn et al. (2007)
O&M costs boiler	%	2	2		Assumption
Scaling factor boiler		0.57[e]	0.57[e]		DOE/NETL (2002)

[a] As the indirect costs were not specified or similarly defined in the underlying studies, standard percentages were used to account for these costs. Process plant cost (PPC) comprises equipment and installation costs. Total Plant Cost (TPC) comprises PPC and engineering fees and contingencies. Total Capital Requirement (TCR) comprises TPC, owner costs and interests during construction. The values are within the ranges observed for power plant construction (Kuramochi et al., 2012; van Horssen et al., 2009).

[b] Ranges are assumed based on variations of quarterly energy prices within individual years studied over a period 1997 and 2008 for the small electricity (2–3 MWh/yr) and natural gas (500–150,000 m^3/yr) consumers in the Netherlands (CBS, 2011).

[c] It was assumed that in the industrialized world where CCS is also deployed for industrial processes, a large share of the worldwide CO_2 emissions is already reduced though renewable energy technologies, power plant CCS, and other CO_2 mitigation options. Therefore, the CO_2 emission factors for the Dutch electricity mix in 2020–2025 (short term) and 2040–2050 (long term) as modelled by van den Broek et al. (2011) in MARKAL was used, which assumed a scenario for which around 50% of the CO_2 is reduced worldwide.

[d] In case efficiencies were not indicated in the environmental reports or monitoring plans, average efficiencies of 80% for industrial furnaces and 85% for stand-alone boilers are assumed to compensate for missing data.

[e] This figure is derived from the data reported for boilers of 4–111 MW_{th} scale generating steam of 18.3 bar, 263 °C (DOE/NETL, 2002).

ROP, 2011; Shell, 2010, 2011) were obtained from environmental reports (Dutch: Milieujaarverslagen) and monitoring plans commissioned by the Dutch Emissions Authority on an annual basis. These data sets contain, among others, detailed key data on energy flows, fuel consumption, fuel composition, CO_2 emissions, capacities, and capacity and load factors of the process units. For the recently built hydrogen plant, the submitted license application is used instead, because no environmental reports were available yet. The data sets are not presented in this study for the sake of brevity.

2.2. CO_2 capture configurations and data

For the refineries and chemical plants, the techno-economic performance of the post- and oxyfuel configurations are calculated for the short term (2020–2025) and long term (2040–2050), whereas only a short term configuration is calculated for the pre-combustion configuration (see Section 2.2.3). The configurations are schematically shown in Fig. 1 and described in the next sub-sections. The short term configurations are characterized by commercially available, first generation CO_2 capture technologies, whereas the long term configurations are based on advanced, second generation capture technologies. Three configurations are devised for the steam reforming hydrogen plant: (i) CO_2 capture from the atmospheric pressure stack flue gases (post-combustion), (ii) CO_2 capture from the high-pressure (HP) process gas between the water gas shift (WGS) reactor and Pressure Swing Adsorption (PSA) unit, and CO_2 capture from both the HP process gas and flue gas (iii) (see box III, Fig. 1). Since CHP plants still display operational difficulties when fired with nearly pure oxygen or hydrogen (Kvamsdal et al., 2006), two sub-configurations are devised for the oxyfuel and pre-combustion configurations of the two refineries, which both have one or more CHP units installed on the industrial plant site. In the first sub-configuration, CO_2 is captured from all process units except the CHP plants by using oxyfuel or pre-combustion technology (single technology (ST) configuration); in the second sub-configuration, the CO_2 emanating from the CHP plant(s) is also captured by using post-combustion technology, thereby combining either oxyfuel or pre-combustion technology with post-combustion technology (combined technology (CT) configuration). The latter option allows for a higher CO_2 emission reduction rate. All configurations require additional energy for the CO_2 capture process. Additional heat is provided by an extra boiler (of which the CO_2 is also captured), whereas the additional electricity is purchased from the grid.

Although plant modifications will probably take place to some degree, the main plant characteristics and layouts are assumed to stay roughly the same in the short term. The recent data sets of the industrial plants are, therefore, assumed to represent the short term conditions of the industrial sites. The short term capture technologies will likely be applied by means of retrofit under non-optimal conditions (e.g. space limitations and existing process units), resulting in several issues with respect to the implementation and operation of process units at industrial plant level. A literature review and interviews with experts are conducted to identify and assess these issues and knowledge gaps. The literature encompasses reports and papers on CO_2 capture technologies and industrial case studies, mostly refineries. Interviews will be held with ten experts working at large oil companies, an oil and gas contractor, an industrial gas producer, and furnace and boiler manufacturers. All experts have profound knowledge and/or are doing specific research on industrial facilities (mostly refineries) in combination with CO_2 capture. The names, organizations and affiliations of the experts are not mentioned for the sake of confidentiality.

The data sets are also used for the long term, despite higher chances of changes in the plant composition regarding process units, energy flows and CO_2 emissions due to core process

Table 3
Technical and economic performance parameters for post-combustion capture. The economic data pertain to a post-combustion system capturing annually 1.0 Mt CO_2 from 4 vol% CO_2 flue gas and 12–14 vol% CO_2 flue gas using one absorber and one stripper. Based on: CESAR (2011), IEA GHG (2010), NETL (2010).[a]

Technical	Unit	Nominal value		References
		Short term	Long term	
CO_2 capture ratio	%	90[b]	90[b]	CESAR (2011), IEA GHG (2000), NETL (2010)
Maximum processing capacity				
Absorber	kt/d	3	8	Kuramochi et al. (2010), Rao et al. (2004), Yagi et al. (2004)
Stripper	kt/d	10	10	
Electricity consumption flue gas blower	GJe/t CO_2	0.35[b]	0.35[b]	Hurst and Walker (2005)
Regeneration heat[c]	GJth/t CO_2	3.5–4.0	2.3–2.6	TNO (2012), Feron (2005), Peeters et al. (2007)
CO_2 capture (pumps and fans)[c]	GJe/t CO_2	0.1–0.3[b]	0.1–0.3[b]	
CO_2 treatment and compression	GJe/t CO_2	0.6[b]	0.6[b]	CESAR (2011)
Technological learning				
CO_2 capture equipment	%	45		
CO_2 treatment & compression	%	0[b]		van den Broek et al. (2009)
O&M costs	%	18		
CAPEX		4 vol%	12–14 vol%	
Gas gathering system	€/Mt CO_2/yr	12	12	Hurst and Walker (2005)
Modifications to stacks	M€/stack	0.1	0.1	Hurst and Walker (2005)
CO_2 capture equipment[d]				
Materials/equipment	€/Mt CO_2/yr	46	22	CESAR (2011), IEA GHG (2010), NETL (2010)
Construction	€/Mt CO_2/yr	26	14	
CO_2 treatment & compression[d]				
Materials/equipment	€/Mt CO_2/yr	7	7	CESAR (2011), IEA GHG (2010), NETL (2010)
Construction	€/Mt CO_2/yr	3	3	
Economic scaling factor				
Amine absorber		0.67	0.67	Kreutz et al. (2005), Sipöcz et al. (2011).
Stripper		0.67	0.67	Larson et al. (2005), Sipöcz et al. (2011)
Compressor		0.7	0.7	Kuramochi et al. (2010)
OPEX		4 vol%	12–14 vol%	
Fixed costs				
Labour	M€/yr	0.2[e]	0.1[e]	See footnote
Admin & overhead	% of labour	27.5	27.5	NETL (2010)
Maintenance	% of TPC	3.8	3.8	NETL (2010)
Variable costs				
Water usage	M€/yr	0.9	0.9	
Other chemicals	M€/yr	0.1	0.2	
MEA	M€/yr	0.5	0.5	NETL (2010)
Activated carbon	M€/yr	0.1	0.1	
Ammonia	M€/yr	0.2	0.1	
Corrosion inhibitor	M€/yr	0.0	0.0	
SCR catalyst	M€/yr	0.1	0.1	

[a] The parameters used for economic costs are based on three detailed studies (CESAR, 2011; IEA GHG, 2010; NETL, 2010), which describe post-combustion capture at a pulverized coal-fired (PC) power plant (high CO_2 concentration) and at a natural gas fired combined cycle (NGCC, low CO_2 concentration). Since the CO_2 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] These values were assumed to remain constant for the long term. The development potential of CO_2 compression is expected to be marginal, because the compression train is based on mature and optimized technology (IEA GHG, 2006; Peeters et al., 2007; van den Broek et al., 2009). The same reasoning applies to the electricity consumption of the blower, fans and pumps.

[c] Regeneration heat and electricity needed for flue gas with CO_2 concentrations in the range of 4–16%. It is assumed there is no effect of scale on the specific energy requirement, which is concluded from a literature review (Kuramochi et al., 2013).

[d] All capital costs were standardized to and corrected for the amount of CO_2 captured of 1.0 Mt CO_2 per year using one absorber and stripper. Scaling factors were used not only to account for differences in CO_2 capture capacity but also for the number of absorbers and strippers. The cost category CO_2 capture equipment represents the direct contact cooler, absorber and stripper; the cost category CO_2 treatment & compression comprises a CO_2 drying, purification and compression unit.

[e] The average amount of labour hours were taken from CESAR (2011), IEA GHG (2010) and NETL (2010), and multiplied with the average European wages of 60,000 €/FTE/yr (IEA GHG, 2010).

modifications, replacements of process units, plant expansion, energy efficiency measures, or other reasons. It should, therefore, be emphasized strongly that the long term results are more indicative than the short term results. The long term technologies will probably capture CO_2 under more optimal conditions in terms of space availability and capture ready process units. The impact of more optimal conditions on the techno-economic performance of CO_2 capture will also be discussed in this paper.

The relevant formulae used to calculate the technical and economic performance of the CO_2 capture configurations can be found in the Appendix. Table 2 gives an overview of the general parameters used in this study. All costs are reported in $€_{2010}$. Costs data found in the literature are converted with the European power capital cost index (EPCCI) (IHS CERA, 2011) to $€_{2010}$, thus taking into account cost increases of energy, steel and engineering services over the last few years (van Straelen et al., 2009). Costs that are reported in US$ are first converted to Euro using the exchange rate data (year average) from OANDA (2011), and are then updated to $€_{2010}$.

2.2.1. Post-combustion

The post-combustion technology is used to capture CO_2 from the flue gases (4–16% CO_2 concentration in the industrial plants) by using (an) absorber(s) with an aqueous solvent with a mass fraction of 30% monoethanolamine (MEA), and a regeneration tower (stripper). MEA was chosen because it is already widely used in industry and sufficient detailed data are available for this solvent. No flue gas desulphurization (FGD) and Selective Catalytic Reduction (SCR) units were installed prior to the CO_2 capture process since the SO_2 and NO_x concentrations in the flue gases were found

Table 4
Technical and economic performance parameters for oxyfuel combustion capture.

General	Unit	Nominal value		References
		Short term	Long term	
CO_2 Capture Ratio (CR)	%	87	87	Allam et al. (2005a,b)
Oxygen production (cryogenic distillation)	$MJ_e/t\ O_2$	684[a]	376	ZEP (2011); Fogash (2007)
Fuel savings furnaces	%	8.3	5.9[b]	Allam et al. (2005a,b)
Fuel savings boilers	%	4.2	9.6[b]	Allam et al. (2005a,b)
CO_2 treatment and compression[c] energy requirement	GJ_e/tCO_2	0.53	0.53	IEA GHG (2005)
CAPEX				
Boiler modification	M€/$MtCO_2$/yr	3[d]	3[d]	Allam et al. (2005a,b)
Furnace modification	M€/$MtCO_2$/yr	1[d]	1[d]	
Air Separation Unit (ASU)	M€/ktO_2/day	75[e]	21	Allam et al. (2005a,b), IEA GHG (2008), Spero (2008)
Cooling water system	M€/$MtCO_2$/yr	11	14	Allam et al. (2005a)
CO_2 treatment and compression	M€/$MtCO_2$/yr	18	18	IEA GHG (2008)
Economic scaling factor ASU		0.62	0.62	See footnote[f]
OPEX				
Maintenance	M€/yr	2.2	2.6	Allam et al. (2005a,b)
Operator manpower	M€/yr	0.7[g]	0.7[g]	
Water	M€/yr	1.2	2.2	
Consumables	M€/yr	0.2	0.2	

[a] The electricity consumption for oxygen production depends on the extent of heat and cycle integration (e.g. with a gas turbine). However, this study assumed a non-integrated ASU. The effect of the production capacity on the Specific Electricity Requirement (scale effect) was considered to be negligible.
[b] ITM oxygen production (long term) shows higher fuel savings than cryogenic oxygen production due to the higher oxygen delivery temperature of this technology.
[c] The treatment unit cools and dries the hot, wet CO_2 coming from the boilers and furnaces.
[d] Average values calculated from the total boiler and furnace modification costs in Allam et al. (2005a,b). The scaling factor for boiler and furnace modification are assumed to be 1 because the modification takes place on many small-medium scale boilers and furnaces so that the total modification cost can be considered to be proportional to the total CO_2 emissions of the industrial plant in question.
[e] Average value based on Allam et al. (2005a,b), IEA GHG (2008) and Spero (2008).
[f] An average value was used based on Hamelinck and Faaij (2002), IEA GHG (2008), Kreutz et al. (2005), Larson et al. (2005), Tijmensen et al. (2002).
[g] No data were available on the amount of operating manpower hours in Allam et al. (2005a,b); only monetary values were presented.

to be lower than the maximum allowed level for MEA scrubbing (<10 ppmv for SO_x and <10 ppmv for NO_x) as indicated by Hurst and Walker (2005). The specific heat and electricity requirements for the regeneration of MEA were calculated for different CO_2 concentrations using process simulation software Aspen Plus® (TNO, 2012). According to Feron (2005) and Peeters et al. (2007), the thermal energy of the regeneration process for future solvents could be improved by approximately 35% compared to the level in 2025. This study assumes a similar improvement for the long term. After the stripper(s), the high-temperature CO_2 flow is routed to a cooling tower, and subsequently, to the Drying, Purification and Cooling (DPC) units and compressors in order to make it ready for transport. CO_2 compression to 110 bar was assumed to ensure that the CO_2 flow retains its dense phase during transport (IEA GHG, 2004; Koorneef et al., 2008). The compression is performed in two steps. First, the CO_2 is compressed up to the critical pressure of 73.8 bar by using a multistage compressor. Second, the dense CO_2 is further compressed by using a pump (Kuramochi et al., 2013). For the long term post-combustion case, cost reductions due to technological learning were assumed to take place for the CO_2 capture equipment and O&M costs. The cost reductions were derived by using the learning rates as reported in van den Broek et al. (2009) in combination with the projected cumulative amount of installed capacity in 2040–2050 as indicated in the Technology Roadmap of the IEA (2009) (see Table 3). A simplified overview of the post-combustion configuration is given in Fig. 1a, box I. The technical and economic performance parameters for the post-combustion capture configurations are presented in Table 3.

2.2.2. Oxyfuel combustion

In the oxyfuel configurations fuel gases are fired with nearly pure oxygen (instead of air) in the boilers, furnaces and catalytic cracker, to obtain a flue gas consisting mainly of water and CO_2. Subsequently, the water vapor is condensed by cooling the flue gas, after which the flue gas is treated and compressed (see Fig. 1a, box II). For the short term, oxygen is assumed to be produced in a cryogenic air separation unit (ASU). According to UIG (2010), on-site oxygen production via ASU is preferable over import when using more than 100 t/d oxygen. For the long term, Ion Transport Membranes (ITM) are expected to become the most cost efficient oxygen production technology (Allam et al., 2005b). The boilers, furnaces and catalytic cracker need to be adjusted to avoid air-in leakage and enable flue gas recirculation. Hot flue gas is partly recirculated to control combustion temperatures, thus saving fuel flow to the boilers, furnaces and catalytic cracker. Additional pipelines are needed to route the oxygen to the process units and CO_2 to the DPC units and compressors. Two different sub-configurations were devised for the refineries for both the short and long term technologies: a single technology and a combined technology configuration. Table 4 shows the technical and economic performance parameters for oxyfuel combustion capture used in this study.

2.2.3. Pre-combustion

In the pre-combustion concept CO_2 is captured through decarbonization of the fuel gases in order to obtain a nearly pure CO_2 and H_2 stream (see Fig. 1a, box III). The steam reforming decarbonisation process involves the reaction of a fuel gas with steam to produce syngas composed of carbon monoxide and hydrogen. Subsequently, the carbon monoxide is reacted with steam in a water gas shift (WGS) reactor to produce CO_2 and hydrogen (IPCC, 2005). The high partial pressure CO_2 and hydrogen are separated after the WGS by using the solvent methyldiethanolamine mixed with small amounts of piperazine (3 mass%). This mixed solvent (ADIP-X) was indicated by Meerman et al. (2012) to be preferable over other solvents (high loading capacity per time unit, chemical stability and low energy requirement, corrosiveness and vapour pressure) to capture CO_2 from the high-pressure process gas in a steam methane reformer (SMR). The hydrogen in the process gas is extracted using a pressure swing adsorption (PSA) unit. Next, the hydrogen is fired in the boilers, furnaces and catalytic cracker.

Table 5
Technical and economic performance parameters for pre-combustion capture.

General	Unit	Nominal value	References
Feed natural gas needed for H_2 production	$GJ_p/t\ H_2$	132	NREL (2009)
Fuel natural gas needed for H_2 production	$GJ_p/t\ H_2$	46	
Electricity needed for H_2 production	$GJ_e/t\ H_2$	2.2	
CO_2 produced			
Production process (CO_2 in process gas)	$t\ CO_2/t\ H_2$	6.9	
SR furnace (CO_2 in flue gas)	$t\ CO_2/t\ H_2$	3.2	
Waste heat (HP steam)[a]	$GJ_{th}/t\ H_2$	30	Appl (1997)
CO_2 capture ratio process gas	%	95	
Heat for regeneration ADIP-X	$GJ_{th}/t\ CO_2$	1.97	Meerman et al. (2012)
Power for regeneration ADIP-X	$GJ_e/t\ CO_2$	0.04	
Power for CO_2 treatment and compression	$GJ_e/t\ CO_2$	0.30	
CAPEX			
SMR-WGS plant	M€/Mt CO_2/yr	118	NREL (2009)
Balance of Plant	M€/Mt CO_2/yr	47	
Drying & Compression	M€/Mt CO_2/yr	35	
SCR NO_x control on stack	M€/Mt CO_2/yr	23	IEA GHG (2000)
Modified burners	k€/Mt CO_2/yr	80	IEA GHG (2000)
Economic scaling factor SR plant		0.67	Kreutz et al. (2005)
OPEX			
Fixed costs			
Labour	€/t_{CO_2}	0.4[b]	
Administration & overhead	% of labour costs	20	NREL (2009)
Maintenance	% of TPC	2	
Variable costs			
Demineralized water	€/t_{CO_2}	0.8	NETL (2010)
Chemicals[c]	€/t_{CO_2}	1.8	

[a] High-pressure steam of 75 bar and 350 °C. It was assumed that all waste heat is transferred to produce steam, thereby ignoring possible heat losses.
[b] The average amount of labour hours were taken from CESAR (2011), IEA GHG (2010) and NETL (2010), and multiplied with average European wages 60,000 €/FTE/yr (IEA GHG, 2010).
[c] SMR and WGS catalysts, PSA sorbent, HP process gas solvent, MEA, deNO_x catalyst.

The CO_2 in the atmospheric pressure flue gas, produced during the combustion of fuel gases in the reforming furnace, is captured by using MEA. The waste heat coming from the process can be used for steam production using heat exchangers. The captured CO_2 coming from the high-pressure process gas and flue gas are directed to the DPC units and compressors. Similar to the oxyfuel configurations, two different sub-configurations are devised for the refineries, i.e. a single and combined technology configuration. All fuel gases are assumed to be used as feed and fuel for the Steam Reformer (SR) facility. Table 5 shows the technical and economic performance parameters for pre-combustion capture used in this study.

There are several studies (e.g. Middleton et al., 2005; Gazzani et al., 2013; Manzolini et al., 2013) which investigated promising future pre-combustion technologies (e.g. the Sorption Enhanced Water Gas Shift process and the Water Gas Shift Membrane Reactor). However, the techno-economic performance data presented in these studies were too aggregated for the purposes of this study. Efforts to retrieve more detailed data were not successful for reasons of confidentiality. Therefore, the pre-combustion configuration is not included in the long term portfolio.

3. Results

3.1. Techno-economic performance

The results of the techno-economic analysis will be discussed for the two refineries (Section 3.1.1), two chemical plants (Section 3.1.2), and the hydrogen plant (Section 3.1.3). Hereafter, the CO_2 avoidance costs will be presented for each individual point source within refinery I.

3.1.1. Refineries

The techno-economic performance of the CO_2 capture configurations of refinery I is presented in Table 6 and Fig. 2.

The results show that the heat production is high for the post-combustion cases (MEA regeneration) compared to the other cases, whereas the oxyfuel cases require larger amounts of electricity, especially for oxygen production. The post-combustion cases require extra boilers with capacities of 448 MW_{th} (short term) and 291 MW_{th} (long term). The pre-combustion cases show no additional heat production due to the availability of waste heat for

Table 6
Energy requirements for CO_2 capture configurations in refinery I. CT and ST stand for combined technology and single technology configuration, respectively.

	Base case	Post-combustion		Oxyfuel combustion				Pre-combustion	
		Short term	Long term	Short term		Long term			
				CT	ST	CT	ST	CT	ST
Energy (PJ/yr)									
Heat	17.3	27.6	24.0	17.3	17.3	17.3	17.3	17.3	17.3
Electricity	0.9	2.4	2.4	4.3	4.3	3.5	3.5	3.2	3.0
CO_2 (Mt/yr)									
Produced	4.1	5.0	4.6	4.4	4.0	4.0	3.7	5.4	5.4
Captured	–	4.3	4.1	3.6	2.6	3.7	2.6	4.9	3.9
Emitted	4.1	0.7	0.6	0.8	1.3	0.4	1.0	0.5	1.5
Avoided	–	3.4	3.6	3.3	2.8	3.7	3.1	3.6	2.6
CO_2 reduction (%)	–	83	87	80	68	89	75	87	64

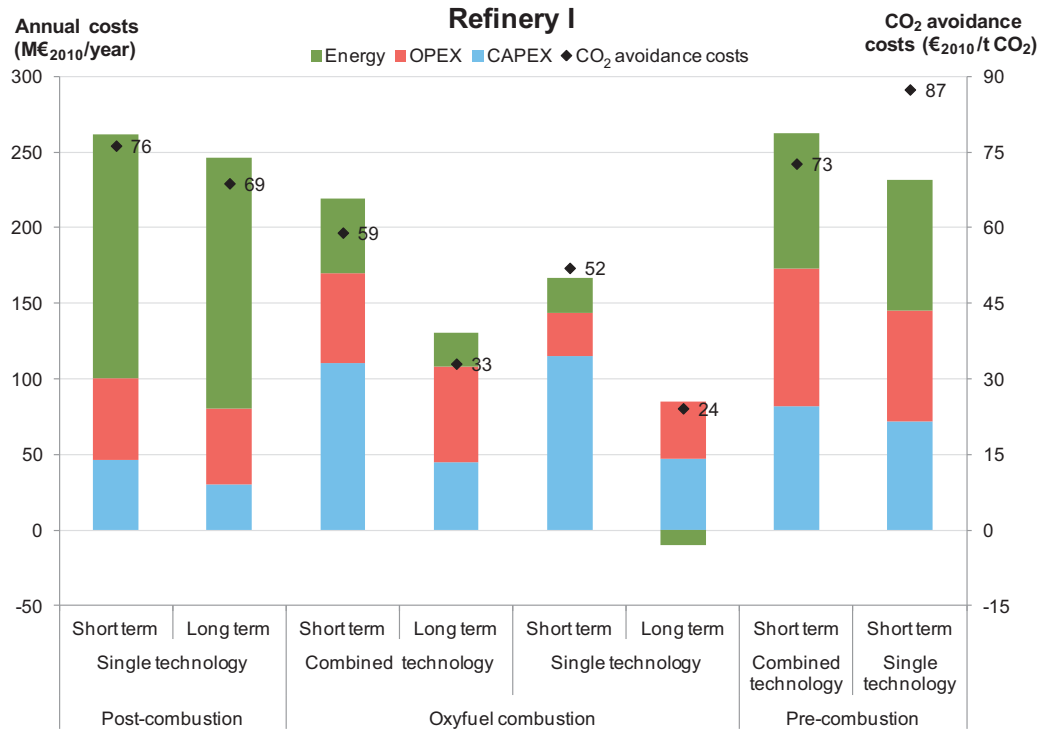

Fig. 2. Economic costs and CO_2 avoidance costs of CO_2 capture configurations for refinery I.

solvent regeneration; however, significant amounts of natural gas are needed for hydrogen production. The pre-combustion cases also show the highest CO_2 production. The short term single technology (ST) and long term oxyfuel cases show even lower amounts of produced CO_2 than in the base case due to fuel savings when firing fuels in oxyfuel mode. Note that the amount of CO_2 avoided is lowest for the ST oxyfuel (2.8–3.1 Mt/y) and pre-combustion cases (2.6 Mt/y). The long term combined technology (CT) oxyfuel case shows the highest amount of avoided CO_2 emissions (3.7 Mt/y).

Fig. 2 shows that the CT pre-combustion and short term post-combustion case display the highest annual costs, whereas the long term ST oxyfuel case shows the lowest annual costs. Note that the CT cases display higher CAPEX than the ST cases, since multiple CO_2 capture technologies have to be installed. Furthermore, the energy costs of the oxyfuel cases are much lower than for the post- and pre-combustion cases, mainly due to fuel savings. The long term ST oxyfuel case even displays negative fuel costs, implying that the cost savings related to the boiler and furnace fuel reductions outweigh the additional electricity costs for oxygen production using ion transport membranes. Despite significant reductions in solvent regeneration heat (35%) for the long term compared to the short term post-combustion cases, the energy costs remain about the same, because of projected increases in natural gas price. The lower CO_2 avoidance costs for the long term compared to the short term post-combustion case result from technological learning, which is reflected in the decrease in CAPEX.

For the short term, the ST oxyfuel case displays the lowest CO_2 avoidance costs (66 €/t CO_2), followed by the CT oxyfuel case (69 €/t CO_2), pre-combustion case (73 €/t CO_2), post-combustion case (76 €/t CO_2), and ST pre-combustion case (87 €/t CO_2). The main reason that the ST pre-combustion case shows lower costs for the CT case is because of synergies between the post- and pre-combustion technologies. After all, as the MEA absorbers and strippers are already built to capture CO_2 from the stack flue gas of the Steam Reformer, additional absorber and stripper capacity lowers the average CO_2 avoidance costs due economies of scale.

Also for the long term, the ST oxyfuel case displays the lowest CO_2 avoidance costs (24 €/t CO_2), followed by the CT oxyfuel case (39 €/t CO_2) and the post-combustion case (69 €/t CO_2).

The techno-economic performance of the CO_2 capture configurations of refinery II is presented in Table 7 and Fig. 3.

Refinery II shows similar results as refinery I in terms of additional heat and electricity production for the different CO_2 capture configurations. The capacities of the extra boilers were calculated to be 218 MW_{th} and 134 MW_{th} for short and long term post-combustion cases, respectively. The amount of CO_2 avoided is lowest for the ST oxyfuel cases (1.6 Mt/y). The short term CT pre-combustion case shows the highest amount of avoided CO_2 emissions (2.1 Mt/y).

Fig. 3 shows that the order of CO_2 avoidance costs from high to low is almost similar as for refinery I. The only difference is that the short term post-combustion case is now cheaper (80 €/t) than the ST pre-combustion case (84 €/t). The cost levels, however, are different for the two refineries; the reason for this difference is threefold. First, as refinery I emits almost twice as much CO_2 in the base case (refinery I: 4.1 Mt/y; refinery II: 2.2 Mt/y), refinery I enjoys significant economic scale effects, implying lower average CO_2 avoidance costs. Second, refinery I has a higher number of utility facilities (three CHP plants and one gas turbine), whereas refinery II has only one CHP plant. As a result, refinery I enjoys more economic scale effects for post-combustion capture at the CHP plants in the CT cases. After all, more CO_2 is captured from the CHP plants and gas turbine. Third, refinery I has a substantial amount of CO_2 (848 kt/y) coming available from the gasifier in nearly pure form (99% pure CO_2), which saves significant energy expenditures and capital costs compared to refinery II.

For refinery I, the CAPEX of the pre-combustion cases are still lower than the CAPEX of the short term oxyfuel cases. However, for refinery II, the CAPEX of the pre-combustion cases are about similar as for refinery I. On top of that, the energy costs (mostly for natural gas) are very high. The post-combustion cases show low CAPEX, but high energy costs; the oxyfuel cases show high CAPEX, but

Table 7
Energy requirements for CO_2 capture configurations in refinery II. CT and ST stand for combined technology and single technology configuration, respectively.

	Base case	Post-combustion		Oxyfuel combustion				Pre-combustion	
		Short term	Long term	Short term		Long term			
				CT	ST	CT	ST	CT	ST
Energy (PJ/yr)									
Heat	17.3	23.6	21.2	17.3	17.3	17.3	17.3	17.3	17.3
Electricity	0.9	1.9	1.8	2.9	2.8	2.5	2.3	2.0	1.8
CO_2 (Mt/y)									
Produced	2.2	2.7	2.4	2.3	2.2	2.2	2.1	2.2	2.2
Captured	–	2.3	1.9	1.9	1.6	1.9	1.6	2.1	1.8
Emitted	2.2	0.4	0.4	0.4	0.6	0.3	0.6	0.1	0.4
Avoided	–	1.8	1.7	1.8	1.6	1.9	1.6	2.1	1.8
CO_2 reduction (%)	–	81	85	81	70	86	74	96	82

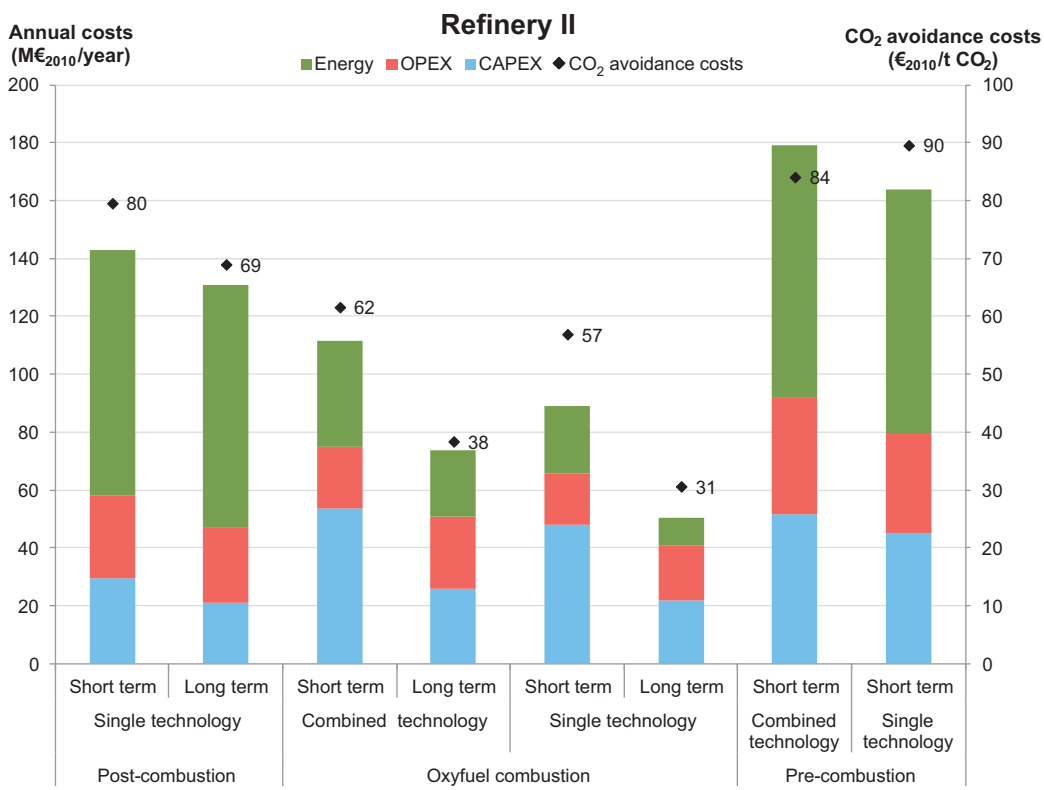

Fig. 3. Economic costs and CO_2 avoidance costs of CO_2 capture configurations for refinery II.

low energy costs; however, the pre-combustion cases show both high CAPEX, especially when the amount of avoided CO_2 becomes smaller, and high energy costs.

The pre-combustion cases display the highest CO_2 avoidance costs (84–90 €/t), followed by the post-combustion cases (80–69 €/t), and the oxyfuel cases (31–62 €/t). The long term ST oxyfuel case shows the lowest CO_2 avoidance costs (31 €/t).

3.1.2. Chemical plants

The combined technology configurations do not apply for the two small chemical plants since these plants only have significant amounts of CO_2 emissions coming from boilers and/or furnaces. The techno-economic performance of the CO_2 capture configurations of chemical plant I is presented in Table 8 and Fig. 4.

Table 8 shows a similar pattern in the additional energy demand for CO_2 capture as for the refinery cases. The capacities of the extra boilers were calculated to be 48 MW_{th} and 30 MW_{th} for short and long term post-combustion cases, respectively. The main difference with the refineries is the considerably lower amount of avoided CO_2, which is lowest for the post-combustion cases (0.3–0.4 Mt/y), followed by the short term oxyfuel case (0.4 Mt/y), the pre-combustion case (0.4 Mt/y), and the long term oxyfuel case (0.4 Mt/y).

Fig. 4 shows that the lower amount of CO_2 captured is reflected in the high CO_2 avoidance costs compared to the refineries, resulting from lower economies of scale. This is also illustrated by the smaller cost difference between the short term post-combustion (92 €/t CO_2) and short term oxyfuel combustion (80 €/t CO_2) cases, which is due to the relatively high capital expenditures for the Air Separation Unit. Similarly, the pre-combustion case becomes rapidly more expensive with lower amounts of CO_2 avoided (114 €/t). The long term oxyfuel case shows by far the lowest CO_2 avoidance costs (37 €/t).

The techno-economic performance of the CO_2 capture configurations of chemical plant II is presented in Table 9 and Fig. 5.

Table 8
Energy requirements for CO_2 capture configurations in chemical plant I. Some categories show figures of zero owing to rounding.

	Base case	Post-combustion		Oxyfuel combustion		Pre-combustion
		Short term	Long term	Short term	Long term	Short term
Energy (PJ/y)						
Heat	4.3	5.6	5.1	4.3	4.3	4.3
Electricity	0	0.2	0.2	0.5	0.4	0.2
CO_2 (Mt/y)						
Produced	0.4	0.5	0.5	0.4	0.4	0.4
Captured	–	0.4	0.4	0.4	0.4	0.4
Emitted	0.4	0.1	0.1	0.0	0.0	0.0
Avoided	–	0.3	0.4	0.4	0.4	0.4
CO_2 reduction (%)	–	81	85	87	96	90

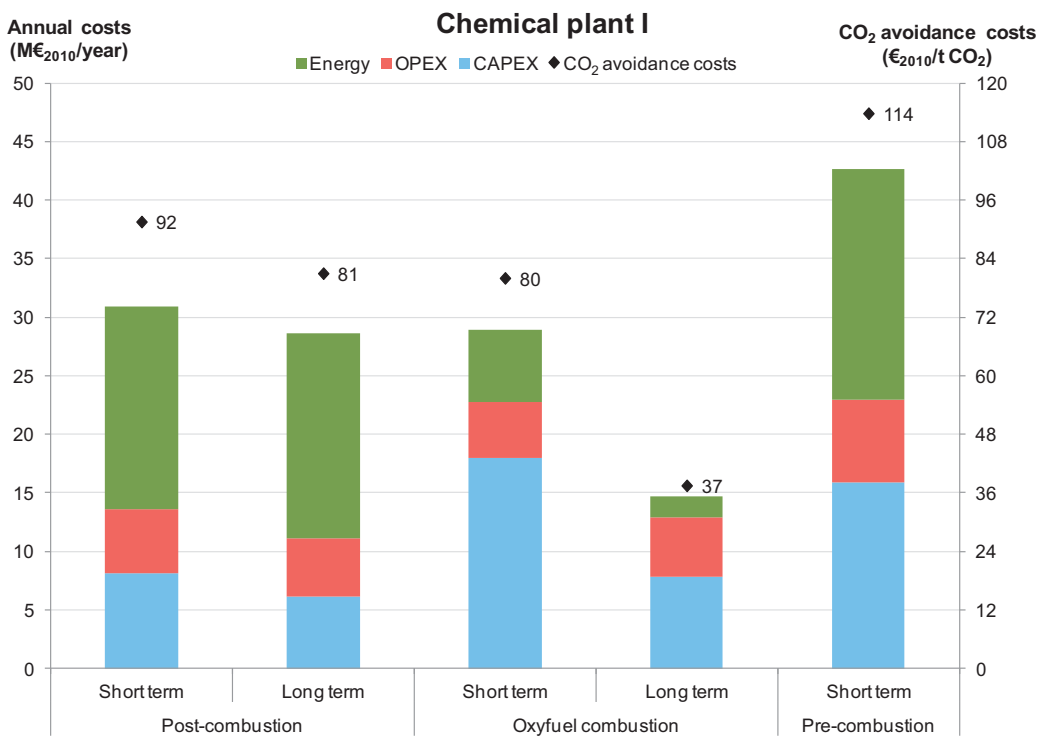

Fig. 4. Economic costs and CO_2 avoidance costs of CO_2 capture configurations for chemical plant I.

Table 9
Energy requirements for CO_2 capture configurations in chemical plant II. Some categories show figures of zero owing to rounding.

	Base case	Post-combustion		Oxyfuel combustion		Pre-combustion
		Short term	Long term	Short term	Long term	Short term
Energy (PJ/y)						
Heat	1.2	1.4	1.3	1.2	1.2	1.2
Electricity	0.0	0.0	0.0	0.1	0.1	0.0
CO_2 (kt/y)						
Produced	53	67	60	59	48	57
Captured	–	56	53	56	42	58
Emitted	53	11	7	2	6	−2
Avoided	–	42	46	50	47	55
CO_2 reduction (%)	–	79	86	88	98	102[a]

[a] The CO_2 reduction ratio of more than 100% is due to steam export.

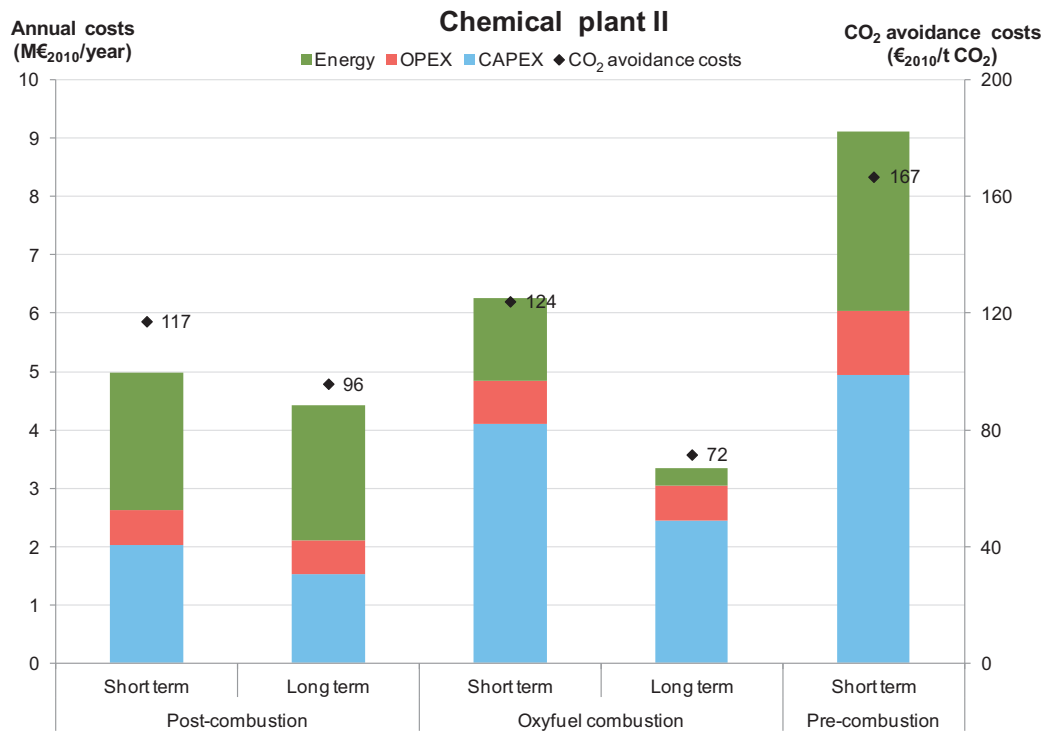

Fig. 5. Economic costs and CO_2 avoidance costs of CO_2 capture configurations for chemical plant II.

Table 10
Energy requirements for CO_2 capture configurations in the steam reforming hydrogen plant. Some categories show figures of zero owing to rounding.

	Base case	Post-combustion		Capture HP process gas	Capture HP process gas + post-combustion
		Short term	Long term		
Energy (PJ/y)					
Heat	6.8	9.2	8.5	7.8	8.5
Electricity	0.0	0.9	0.9	0.2	0.4
CO_2 (Mt/y)					
Produced	0.8	1.0	0.9	0.9	1.0
Captured	–	0.8	0.8	0.5	0.9
Emitted	0.8	0.2	0.1	0.4	0.1
Avoided	–	0.6	0.7	0.4	0.7
CO_2 reduction (%)	–	71	82	54	84

Chemical plant II emits annually the lowest amount of CO_2 (53 kt/y). The main difference with chemical plant I is the amount of CO_2 emissions captured per year. Although the core processes of the chemical facilities are very different, the energy production units, i.e. boilers and furnaces, are similar. The capacities of the extra boilers were calculated to be 6 MW_{th} and 4 MW_{th} for the short and long term post-combustion cases, respectively. Table 9 and Fig. 5 show that the CO_2 avoidance costs are higher for chemical plant II due to the smaller amount of CO_2 captured. Moreover, the declining trend in the relative CO_2 avoidance costs of the post-combustion cases (117–96 €/t) compared to the short term oxyfuel case (124 €/t) is continued. The CO_2 avoidance costs were calculated to be very high: 142 €/t. Despite its lowest CO_2 avoidance costs, the long term oxyfuel case shows significantly higher avoidance costs (72 €/t) than for the refineries and chemical plant I (24–37 €/t). The pre-combustion case shows the highest CO_2 avoidance costs (167 €/t).

3.1.3. Hydrogen plant

The techno-economic performance of the CO_2 capture configurations of the steam reforming hydrogen plant is presented in Table 10 and Fig. 6.

The short term post-combustion case requires the highest amount of heat and electricity of all four cases due to the high amounts of CO_2 captured using a solvent (MEA) with a high heat requirement per tonne CO_2 captured. The case with CO_2 capture from the high-pressure process gas shows significantly lower heat requirements due to the low specific energy requirements for the solvent ADIP-X. The capacity of the extra boilers needed for heat production was calculated to be 89 MW_{th}, 62 MW_{th}, 85 MW_{th}, and 43 MW_{th} for the short term post-combustion case, long term post-combustion case, WGS-PSA case, and WGS-PSA + post-combustion case, respectively. The long term post-combustion case and WGS-PSA + post-combustion case show the highest amount of CO_2 avoided.

The WGS-PSA case shows the lowest CO_2 avoidance costs (67 €/t) due to the low CAPEX and low energy consumption. However, only a limited amount of CO_2 is avoided (54%). Although the WGS-PSA + post-combustion case shows higher CO_2 avoidance costs (87 €/t), significantly more CO_2 can be reduced (84%).

Fig. 6 shows that the energy expenses for the post-combustion cases are highest of all four cases.

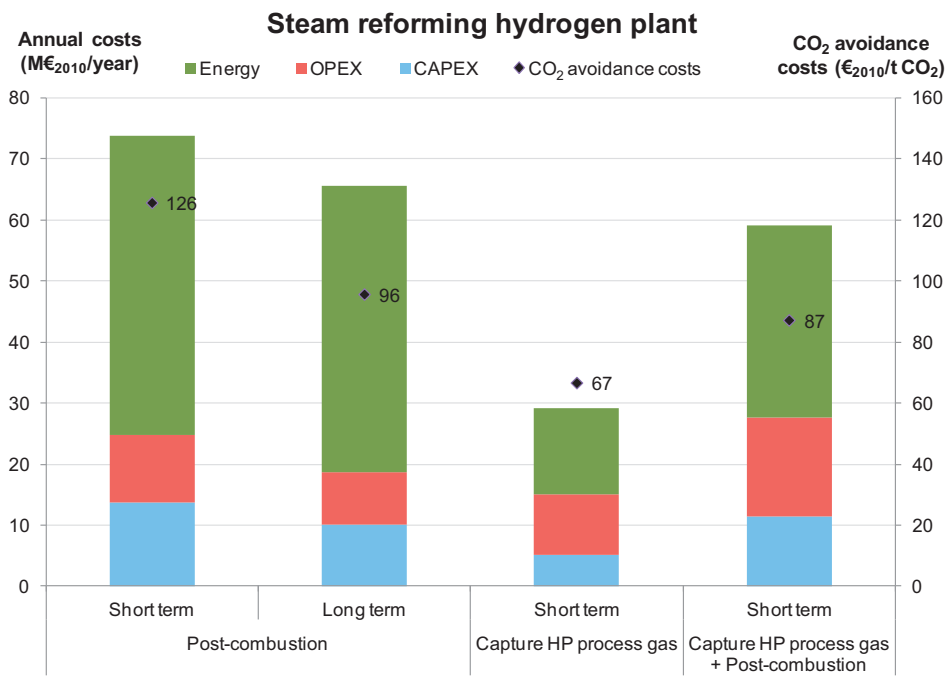

Fig. 6. Economic costs and CO_2 avoidance costs of CO_2 capture configurations for the steam reforming hydrogen plant.

3.1.4. *Marginal abatement cost curve refinery I*

Fig. 7 shows CO_2 abatement cost curves for the short term post-combustion configuration in refinery I. The dashed line denotes the avoidance costs when capturing CO_2 from each individual point source alone, starting with the lowest avoidance costs (28 €/t CO_2) for the gasifier on the left side of the graph and ending with the highest avoidance costs (1221 €/t CO_2) for a furnace stack emitting less than 1 kt annually (right side of the graph, not visible in Fig. 7), which would never be captured alone. The blue, red and green areas show the respective *marginal* capital, operational and energy costs per tonne CO_2 avoided when capturing CO_2 from the next cheapest point source in the refinery. The upper limit of the green area shows the *marginal* total CO_2 avoidance costs. The solid line denotes the average avoidance costs of the cumulative amount of CO_2 avoided up till that point.

Fig. 7 shows that for refinery I the avoidance costs can differ significantly with cumulative amounts of avoided CO_2 and varying CO_2 concentrations in the flue gases. The low CO_2 avoidance costs for the gasifier are a result of the low capital and energy costs, mainly because the chemical absorption step is not required (99% pure CO_2 stream). A big step in capital and energy costs can be observed for the second point source (H_2 plant furnace), which is due to the relatively small scale of the absorber and stripper used for this step. With higher cumulative amounts of avoided CO_2 the capital costs per tonne CO_2 avoided decrease slightly up till the point that an additional absorber and/or stripper is needed

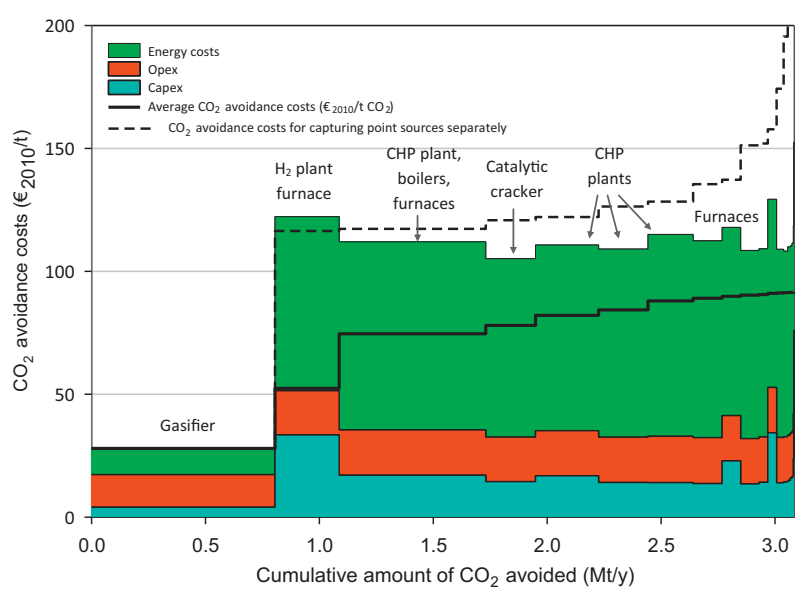

Fig. 7. CO_2 abatement cost curve for post-combustion capture in refinery I.

(absorber: 1 Mt/y; stripper: 3.3 Mt/y), which is reflected in the three bumps in the blue area (as of 2.0 Mt CO_2/y). This illustrates that the impact of economies of scale of post-combustion capture is rather limited. Nonetheless, especially the small point sources (on the right of the graph) benefit greatly from the scale effects as can be seen by the widening gap between the dashed line representing the avoidance costs of each separate point source and the solid line representing the average avoidance costs. Whereas the energy costs fluctuate to some extent per point source, due to varying CO_2 concentrations in the flue gases, the operational costs per tonne CO_2 avoided (red area) remain rather constant. Note that the *marginal* avoidance costs remain quite similar for all point sources after capturing CO_2 from the gasifier. The *average* avoidance costs increase with higher cumulative amounts of avoided CO_2.

3.1.5. Overview and comparison of results

Table 11 gives an overview of the CO_2 reductions and CO_2 avoidance costs of the five case studies. The implications of our study are many. As expected, the results show that for all cases the CO_2 avoidance costs are lower for the long term than for the short term, mainly as a result of the lower regeneration energy requirements for future solvents and future oxygen production technology. Also, the single technology (ST) cases for the refineries display lower CO_2 avoidance costs than the combined technology (CT) cases (including CO_2 capture from the CHP plants and gas turbine using post-combustion), because of the relatively lower CAPEX and OPEX per tonne CO_2 avoided for the ST cases.

Table 11 also shows that CO_2 avoidance costs increase across the board for smaller emitter size (annual CO_2 emissions), due to economies of scale. Next to emitter size, the refineries have a gasifier (refinery I) and hydrogen plants from which the CO_2 can be captured at low costs, thereby lowering the average CO_2 avoidance costs. As post-combustion technology seems to be less affected by emitter size, the economic competiveness compared to the oxyfuel and pre-combustion cases improves for smaller emitter sizes. Notwithstanding, the long term ST oxyfuel case using ion transport membranes for oxygen production remains the most competitive option from an economic point of view.

Note that the pre-combustion cases shows structurally higher CO_2 avoidance costs than the post- and oxyfuel combustion cases, mainly because of high natural gas and capital costs. We should emphasize that the CO_2 avoidance costs of the post- and pre-combustion cases are significantly affected by the natural gas price, whereas the oxyfuel cases are predominantly affected by the CAPEX. When comparing the ST cases, the CO_2 reduction ratios depend heavily on the amount of gas turbines and CHP plants present at the facilities' sites. For example, the CO_2 reduction rates of the ST pre-combustion and ST oxyfuel cases for the refineries are significantly lower than for the chemical plants, which have no CHP plants or gas turbines on their premises.

For the steam reforming hydrogen plant, the CO_2 avoidance costs are highest for the post-combustion cases (short term: 114 €/t; long term: 98 €/t), followed by the case combining capture from both the process gas and flue gas (94 €/t), and the case capturing CO_2 solely from the process gas (75 €/t). However, the latter case reduces considerably less CO_2 (54%) than the other cases (71–84%). The case combining capture from both the process gas and flue gas shows the highest CO_2 reduction rate (84%).

3.2. Challenges related to implementation and operation of CCS in the industry

Several issues related to the implementation and operation of CO_2 capture in the industry were identified and assessed in this analysis. Some general findings, which are important to consider before discussing the specific issues, are itemized below.

- Most experts emphasized that capture-ready greenfield facilities (i.e. new-built industrial plants) involve less spatial and reliability issues than brownfield plants (i.e. existing industrial plants).
- Retrofitting existing process units with CO_2 capture technology is usually preferred over replacement with new process units already equipped with capture technology from a cost perspective, unless the process units are at the end of their lifetime. Investments made in existing process units (frozen capital) pose a barrier for replacement with new process units equipped with capture technology.
- The reliability of the core processes is a leading principle when considering CO_2 capture. A drop-out would have a significant impact on the economics of the industrial plant. CO_2 capture installations will most likely be designed in such a way that the CO_2 capture equipment can be shut down and operations can continue without capturing CO_2.

Table 11
Overview of the CO_2 avoidance costs for the CO_2 capture configurations of all five industrial plants. CT and ST stand for combined technology and single technology configuration, respectively.

	Unit	Post-combustion		Oxyfuel combustion				Pre-combustion	
		Short term	Long term	Short term		Long term		CT	ST
				CT	ST	CT	ST		
Refinery I (4.1 Mt CO_2/yr)									
CO_2 reduction	%	83	87	80	89	68	75	87	64
Avoidance costs	€/t CO_2	76	69	59	33	52	24	73	87
Refinery II (2.2 Mt CO_2/yr)									
CO_2 reduction	%	81	85	81	86	70	74	96	82
Avoidance costs	€/t CO_2	80	69	62	38	57	31	84	90
Chemical plant I (0.4 Mt CO_2/yr)									
CO_2 reduction	%	81	85	n/a	n/a	87	96	n/a	90
Avoidance costs	€/t CO_2	92	81	n/a	n/a	80	37	n/a	114
Chemical plant II (54 kt CO_2/yr)									
CO_2 reduction	%	79	86	n/a	n/a	94	87	n/a	102[a]
Avoidance costs	€/t CO_2	117	96	n/a	n/a	124	72	n/a	167
	Unit	Post-combustion		Capture HP process gas			Capture HP process gas + post-combustion		
Hydrogen plant (0.8 Mt CO_2/yr)									
CO_2 reduction	%	71	82	54			84		
Avoidance costs	€/t CO_2	114	98	67			87		

[a] The CO_2 reduction ratio of more than 100% is due to steam export.

Table 12
Overview of issues related to the implementation and operation of industrial process units with oxyfuel, post-, and pre-combustion technology. The performance of the capture technologies for the issues is indicated as follows: possible (++); probably possible (+); neutral (0), probably not possible (−); not possible (−−).

	Post-combustion	Oxyfuel combustion	Pre-combustion
Implementation			
Technically feasible			
Furnaces	++[a]	+[b]	+[c]
Boilers	++[a]	+[b]	+[c]
Catalytic cracker	++[a]	+[d]	n/a
CHP plant/GT	++[a]	+[e]	+[e]
H_2 plant (SR)	++[a]	n/a	n/a
Spatial limitations			
Around process units or stack	+[f]	0[g]	++[h]
Overall industrial site	0[g]	0[g]	+[h]
Operational issues			
Chance of operational failure in process unit	Small[i]	Unknown[k]	Unknown[k]
Effect of operational failure on core processes	Small[j]	Unknown[k]	Unknown[k]

[a] All experts and literature (e.g. Hurst and Walker, 2005; IEA GHG, 2000; van Straelen et al., 2009) state that retrofitting industrial process units with post-combustion is technically feasible.
[b] Boilers and furnaces can, in principle, be retrofitted to oxyfuel combustion (e.g. Allam et al., 2005a,b); however, it depends on the space availability on site (experts).
[c] Consensus exists in literature (e.g. Thomas and Kerr, 2005) and among experts that this is possible; however, it still needs to be proven in practice.
[d] Pilot plant runs show no operational difficulties (de Mello et al., 2009); however, the technology still needs to be proven on a commercial scale.
[e] Although oxyfuel and pre-combustion capture is, in principle, technically feasible for CHP plants, these technologies will most likely face a high degree of operational challenges (Kvamsdal et al., 2006).
[f] Space is often available around the stacks; however, this differs for each case (experts).
[g] Depends on space availability on specific industrial site and on the amount of point sources from which CO_2 is captured. After all, more ducting and piping is required to capture CO_2 from the small point sources (van Straelen et al., 2009).
[h] The existing fuel distribution network can be used for hydrogen distribution (experts; Lowe et al., 2010). The spatial footprint of the Steam Reformer is relatively small compared to post-combustion absorbers.
[i] The implementation of post-combustion technology does not have a serious impact on the reliability of the process units, i.e., the chance of an operational failure is nearly the same as when operating without CO_2 capture.
[j] A drop-out of the CO_2 capture equipment requires only the opening of a valve to vent the flue gas or CO_2-rich stream to the atmosphere (expert). This can be done on a short notice.
[k] None of the experts had any idea on the chance of an operational failure, nor on the impact it would have on the core processes, nor on the time needed to restore normal operation of the process units.

An overview of the specific findings of the issue inventory is presented in Table 12. In addition, several findings are described in more detail for the technical feasibility of retrofitting, spatial limitations and operational issues. Most findings of the inventory were based on experiences with refinery furnaces and boilers, but they probably also hold for installations in the chemical industry because of large similarities in boiler and furnace design.

3.2.1. Technical feasibility

- All experts and literature (e.g. Hurst and Walker, 2005; IEA GHG, 2000; van Straelen et al., 2009) indicate that retrofitting process units to post-combustion technology is technically feasible, because it is an add-on technology, relatively simple process and has been applied in the natural gas sector for years.
- The technical difficulties and costs of retrofitting furnaces and boilers to oxyfuel combustion depend on their configuration. Experts indicate that natural draft furnaces are more costly and difficult to retrofit than forced draft furnaces, since the former was originally not designed for forced draft operation and requires, therefore, additional modifications (piping, ducting, flue gas recirculation fan, burners) to operate in oxyfuel mode. Also, furnaces differ from boilers in a number of respects: a wider range of furnace designs and different techniques used to construct them, often results in greater air in-leakage (1); hydrocarbons are often present in furnace tubes, presenting additional hazards (2); usually, burner types are used without automatic control of air/fuel ratio and draft. Retrofitting of furnaces may, therefore, raise operational issues not encountered with boilers (Kuramochi et al., 2012; Wilkinson et al., 2003).
- Metallurgical limitations of boiler and furnace walls due to higher oxyfuel combustion temperatures do not seem to be a problem as flue gases can be looped back to the inlet of the process unit to simulate air combustion conditions (Scheffknecht et al., 2011). Although the flame characteristics and temperature distribution across the radiative and convective sections of the process unit will change, the flue gas recycle ratio can be used as a tuning parameter to control those parameters (Andersson et al., 2008). Problems with fuel ignition and flame instability can be solved by special oxygen injection or by making modifications to the burner geometry itself to ensure good fuel-oxygen mixtures (Scheffknecht et al., 2011).
- In recent years, a number of laboratory and pilot-scale tests have been carried out to shed light on corrosion mechanisms taking place in oxyfuel gas atmospheres. To date, no distinct prove has been provided that oxyfuel gas conditions, i.e. a combination of high partial pressure CO_2 and O_2 gas as well as large amounts of water, induce more corrosion than air-fired conditions (Otsuka, 2012). Some studies report that the oxyfuel gas atmosphere can actually have a counteractive effect on corrosion (Otsuka, 2012). Nevertheless, problems with corrosion may occur if fuel gases contain high levels of sulphur, chlorine and/or heavy metals. In case these contaminant levels are too high, the flue gases need to be condensated first to extract the contaminants before recirculating the gases back to the process unit. The aforementioned research results on corrosion should, however, be placed in perspective since the underlying lab and pilot-scale tests were carried out in synthetic atmospheres and with exposure times below a thousand hours (Scheffknecht et al., 2011). Further research on corrosion in large-scale applications is therefore needed.
- The retrofit of catalytic crackers to oxyfuel combustion is deemed possible. Pilot plant tests run by Petrobras showed no significant changes from normal operation of a fluid catalytic cracker when operated in oxyfuel mode. The product profile, stability of operation and effectiveness of coke burn did not show any abnormalities (de Mello et al., 2009).

- For pre-combustion, Thomas and Kerr (2005) report that no modifications of furnaces and boilers are needed for the firing of nearly pure hydrogen. Nevertheless, Lowe et al. (2010) recommend that upgrading of materials is advisable to ensure an extended lifetime of the walls and burner, because the different hydrogen flame geometry (compared to refinery and natural gas) could change the heat flux distribution in the convection and radiation sections and expose materials to higher temperatures.
- The replacement of pre-mixed burners (if used) for raw gas burners is needed due to the danger of flash-backs in pre-mixed burners as a result of the high flame speed of hydrogen. Installed raw gas burners do not need to be adjusted as the Wobbe index of hydrogen and natural gas is rather similar (Lowe et al., 2010).
- For hydrogen firing, more maintenance on burners may be required, especially burner tips need to replaced more often.
- Several experts recommend the installation of a downstream deNO_x unit for hydrogen firing to ensure NO_x emission thresholds are not exceeded.
- Retrofit of boilers and furnaces to oxyfuel and pre-combustion mode, and the hook-up to oxygen/hydrogen supply and CO_2 processing equipment can be executed during plant turn around.

3.2.2. Spatial limitations

- All experts indicated that spatial issues for industrial plants are very site-specific.
- Spatial constraints regarding post-combustion concentrate specifically on the CO_2 capture equipment (especially absorbers, cooling towers and blowers) and large diameter ducting, whereas for oxyfuel combustion the spatial issues are predominantly related to the space directly around the boilers, heaters and catalytic cracker. Pre-combustion seems to have the least spatial constraints. Another advantage of pre-combustion is that the additional hydrogen plant and CO_2 compressor do not have to be installed in close proximity to the process units where space is often limited. Several refinery experts indicated the following capture technology ranking in terms of spatial limitations from most to least preferable: pre-combustion (1); oxyfuel combustion (2); post-combustion (3).
- Next to site-specific conditions, the amount of captured CO_2 also determines the extent of the space problem for post- and oxyfuel combustion capture. After all, in case of ambitious CO_2 emission reductions targets, more ducting and piping is required to capture CO_2 from the small and scattered point sources where space is often limited (van Straelen et al., 2009; Lowe et al., 2010). Pre-combustion capture does not have this problem, since the existing fuel system can most likely be used for hydrogen distribution. A preliminary assessment of the metallurgy of the existing fuel distribution system in a Chevron refinery showed that no piping material modifications are required when shifting from fossil gas to hydrogen (Lowe et al., 2010).
- Several experts noted that infrastructural modifications and replacement of existing installations (e.g. storage tanks) on the plant site is unavoidable to make space for large diameter post-combustion absorbers. However, other experts refuted this statement and claimed space can always be found.

3.2.3. Operational issues

- The process units when operated in oxyfuel or pre-combustion mode are slightly less reliable than when operated in air- or fossil fuel combustion mode. An operational failure might occur as a result of the changed combustion process or, more likely, due to a drop-out in the upstream oxygen or hydrogen supply.
- None of the experts were able to indicate the chance of a drop-out of the upstream oxygen and hydrogen production units. Furnaces and boilers will always retain the option of switching back from oxygen to air combustion, and from hydrogen to fossil gas fuelling. However, experts were also not able to indicate the time required to this.
- Although oxyfuel and pre-combustion capture is, in principle, technically feasible for CHP plants, these technologies will face a high degree of operational challenges. Both technologies require the recirculation of exhaust gases, which makes the system vulnerable to start-up, shut-down and load changes. It involves a high number of subsystems, recirculation streams and heat exchanging streams, what makes the control of such plants very challenging (Kvamsdal et al., 2006).

3.3. Sensitivity of CO_2 avoidance costs

The CO_2 avoidance costs of the different CO_2 capture configurations, which were presented in Section 3.1, may change due to uncertainties in key input parameters, but can also be affected by the implementation and operational challenges dealt with in Section 3.2. This section discusses both effects. The spider diagrams in Fig. 8 show the sensitivity of the CO_2 avoidance costs of refinery I related to the following key variables: natural gas price, electricity price, grid electricity CO_2 emission factor, annuity factor, CAPEX, OPEX, and energy use of CO_2 capture. The uncertainty ranges of the general input data and the technical and economic parameters of CO_2 capture were taken from Table 2 and the underlying data sets, respectively. The centres (100%) of each diagram are the nominal values used in this study. The sensitivity analysis of the CT oxyfuel configurations, and the other industrial plants were not presented, since virtually similar spider diagrams were produced as presented in this paper.

The implementation and operational issues associated with the short term configurations revolve mainly around retrofitting CO_2 capture to existing plant sites and process units. Although space limitations are not directly reflected in CO_2 avoidance costs, site-specific conditions can necessitate the replacement of existing pipelines and installations, thereby incurring higher costs. Also, the costs of retrofitting process units might turn out higher than assumed for this study due to different boiler and furnace types. Although the higher retrofit capital costs are only minor considering its small share in the overall CAPEX, the avoidance costs of the oxyfuel configurations will be influenced most, due to the high capital expenditures per tonne CO_2 avoided (see Fig. 8). Finally, although the chance and the effect of an operational failure of the process units when operated in oxyfuel or pre-combustion mode is still unknown, it could impact the core processes of the industrial plant and have significant economic consequences.

If CCS is to be deployed on a larger scale, retrofit issues are expected to play a less prominent role for the long term. Instead, expectations are that the concept of CO_2 capture will become increasingly incorporated in industrial plant process design by replacing old process units by new-built capture-ready process units, minimum spatial restrictions in (new-built) plant configurations and capture technologies highly integrated with core processes, thereby avoiding costs of retrofit, space consuming piping around the process units, and lower operational issues due to optimal process unit design for CO_2 capture. Although difficult to quantify, these factors will have a damping effect on the projected long term CO_2 avoidance costs. The installation or expansion of a (existing) CHP plant could have a lowering effect on the CO_2 avoidance costs, because of economic scale effects and high electric efficiencies of large-scale CHP plants (compared to smaller scales), thereby mitigating the impact of a higher natural gas price.

Both for the short and long term cases, the availability of waste heat coming either from the own core processes or nearby facilities, could significantly lower the CO_2 avoidance costs of the post- and pre-combustion configurations, due to lower energy costs for

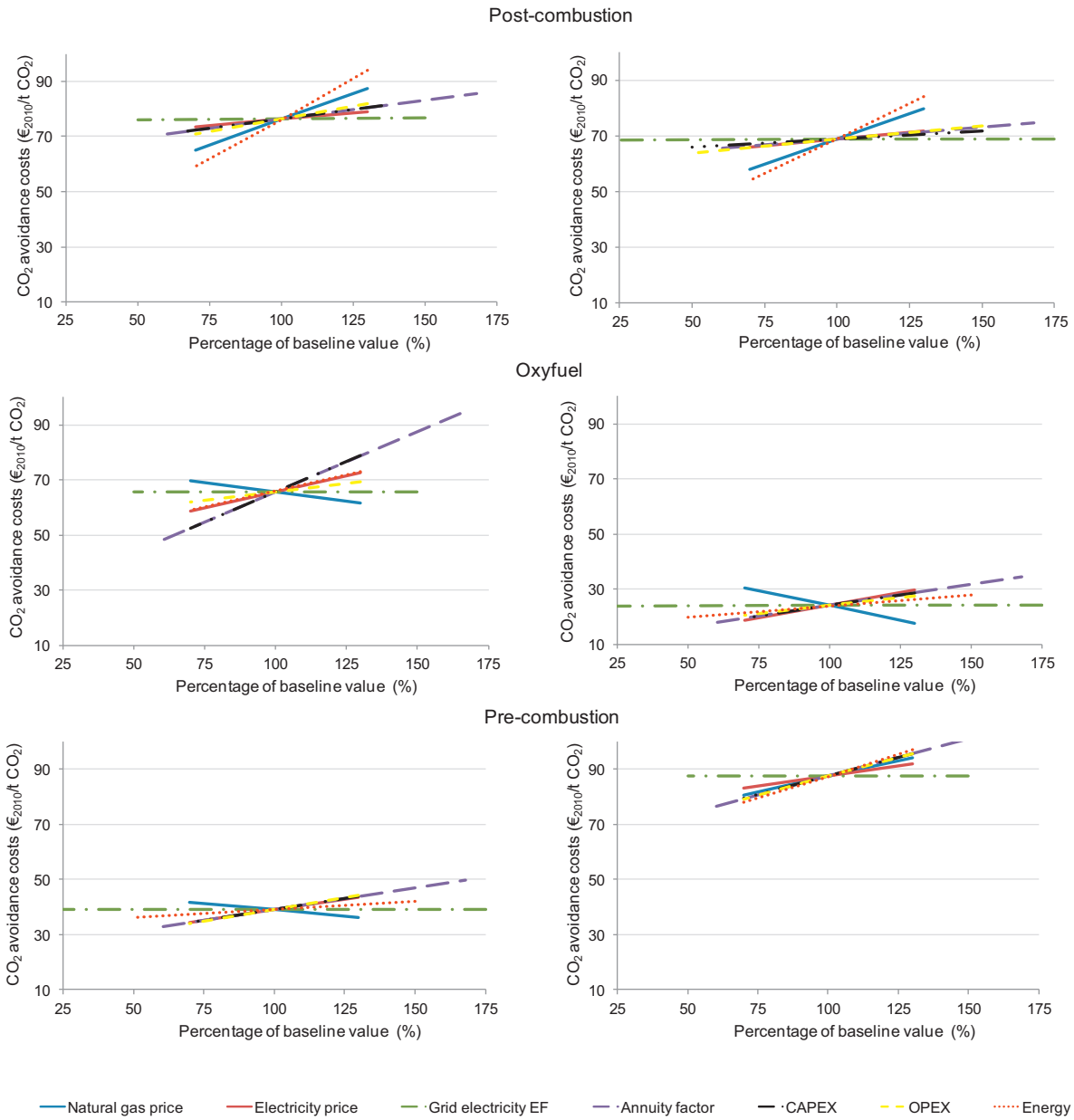

Fig. 8. Sensitivity of CO_2 avoidance costs of refinery I for the short term (left) and long term (right) of the post combustion and oxyfuel configurations. The lower two spider diagrams depict the combined technology (left) and single technology (right) pre-combustion configurations.

solvent regeneration. The high impact of energy use and natural gas price on the avoidance costs of the aforementioned configurations is illustrated in Fig. 8. A variation of ±30% in energy use already results in around ±23% and ±11% change in the CO_2 avoidance costs of the post- and pre-combustion configurations, respectively. By varying the natural gas price with ±30%, a deviation of around ±16% and ±8% can be observed in the CO_2 avoidance costs of the respective post- and pre-combustion configurations. Given the unpredictable nature of natural gas prices, CO_2 avoidance costs can, therefore, differ significantly. Note that a 30% lower short term natural gas price, or 30% higher short term electricity price, would render post-combustion more cost efficient than oxyfuel combustion in the short term. However, as natural gas and electricity prices are to some extent correlated (Yang and Blyth, 2007), future natural gas and electricity prices will probably show similar trends, thereby making such a scenario unlikely.

The CO_2 avoidance costs of the oxyfuel configurations also show high sensitivity to the annuity factor. By using an uncertainty range of ±5 years for the economic lifetime and ±5% for the real discount rate results in a deviation of minus 26% to plus 45% in the CO_2 avoidance costs. The energy use and energy prices show lower impacts given its smaller contribution in the overall costs. The negative correlation between natural gas price and CO_2 avoidance costs results from the fossil fuel savings in the process units when operated in oxyfuel mode. Note that electricity prices and OPEX show higher impacts on the long term cases, because of the higher share in the overall costs. The two spider diagrams at the bottom of Fig. 8 show the sensitivity of the CO_2 avoidance costs of the CT (left) and ST (right) pre-combustion configurations. For these configurations, both the energy use and annualized capital costs have high impacts on the CO_2 avoidance costs. The energy use shows the largest impact on the CO_2 avoidance costs of the CT configuration.

4. Discussion

The main strength of this study's methodology is the high level of detail at plant level based on various case studies. The underlying data is both specific and reliable in nature, especially regarding the characteristics of the process units and energy and CO_2 flows. Nevertheless, this study has several limitations.

4.1. CO_2 capture technologies and clustering of industrial plants

This study investigated the techno-economic performance of pre-, post- and oxyfuel combustion technologies. However, several alternative solvents, capture technologies and process configurations may show different economics and challenges related to the implementation and operation of CO_2 capture equipment. More research on these technologies (e.g. membranes and chilled ammonia process) is needed to provide a more comprehensive overview of the technological possibilities, constraints and avoidance costs of CO_2 capture in industry. Also, it would be useful to extend the analysis with a regional case study in which the techno-economic implications of clustering industrial facilities are evaluated. Especially the economic competitiveness of the pre-combustion configurations might benefit from such clusters, due to economies of scale.

4.2. Data limitations

The plant-specific data underlying this study represents the current situation in the industrial plants and does, therefore, not reflect future plant configurations. Process units, energy flows and CO_2 emissions may change considerably in the future as a result of changes in the core processes, replacement of process units, energy efficiency measures, or other reasons. It should, therefore, be emphasized that the long term results are more indicative than the short term results. Also, cost data uncertainty, especially for the long term technologies, is a caveat that should be addressed in future work. The sensitivity analysis has shown that the capital expenditures can have a significant impact on the CO_2 avoidance costs. Hence, more reliable (pilot plant) data is needed on the investment costs of future technologies, particularly for the long term oxyfuel configuration. Other uncertainties are related to energy prices and energy use.

4.3. Heat integration and CHP plant

More research is needed to determine the potential for cost reductions by achieving a higher level of heat integration between the post-combustion capture unit and the industrial plants, particularly the refineries. Johansson et al. (2012) showed that the utilization of excess heat for post-combustion CO_2 capture in the process industry can drive down avoidance costs to the same cost range as reported for oxyfuel combustion in industry (30–60 €/t CO_2 avoided (Kuramochi et al., 2012)), and post-combustion capture in the power sector (30–40 €/t CO_2 avoided; ZEP, 2011). The advent of solvents with low regeneration temperatures could result in even further cost reductions. Another option that can have a significant impact on the economic performance is the use of an additional CHP plant rather than a boiler (Kuramochi et al., 2012). This economic impact is even larger when the cogenerated steam is used to replace that of the existing boilers, due to economic scale effects and high electric efficiencies of a large-scale CHP plant. Especially by clustering industrial facilities, CHP plants may become interesting as capital costs will be lower per tonne of CO_2 avoided. Finally, more research is needed to determine the techno-economic feasibility and challenges related to the implementation and operation of oxyfuel and pre-combustion capture at utility and CHP plants. Although operational difficulties are still present today (Kvamsdal et al., 2006), oxyfuel and pre-combustion capture might be promising options for the long term.

4.4. Implementation and operation of CO_2 capture equipment

In this study a first overview is given of the remaining challenges and knowledge gaps related to the implementation and operation of CO_2 capture equipment at industrial plant level. Industrial experts stressed the importance of identifying these challenges since related uncertainties could impede, or at least delay, the adoption of CO_2 capture in industry. The next step would be to quantify these findings by collecting data on spatial footprints, heat integration, retrofitting and replacement costs for a wider array of furnace and boiler designs, and more (pilot plant) tests and modelling to determine chances, impacts and recovery time of operational failures in both process units and upstream H_2/O_2 supply facilities. By quantifying and extending knowledge on these issues, the overall impact on the CO_2 avoidance costs for both the short and long term can be made more explicit.

5. Conclusions

This study did a techno-economic analysis and made an inventory of potential implementation or operational challenges related to the three main CO_2 capture technologies applied to five industrial plants from various industrial sectors in the Netherlands.

The results show that CO_2 capture at the boilers, furnaces, catalytic crackers and gasifier of the refineries result in CO_2 reductions of 64–75% (including CO_2 capture of additional heat production) for oxyfuel and pre-combustion technology. For the post-combustion configurations, CO_2 was also captured from the hydrogen plants, CHP plants and gas turbine, resulting in CO_2 reductions of 81–87%. CO_2 capture from these process units using oxyfuel or pre-combustion technology was not deemed realistic due to operational difficulties reported in literature. By combining oxyfuel and pre-combustion with post-combustion technology (combined technology configuration, CT) for the hydrogen plants and utilities, significantly higher CO_2 reductions (80–96%) were calculated for the refineries. Whereas combining oxyfuel and post-combustion technology results in higher CO_2 avoidance costs, the combination of pre- and post-combustion technology displays lower CO_2 avoidance costs (−6–17%), due to economies of scale of shared absorbers and strippers. As the chemical plants do not have cogeneration or hydrogen units, only single technology (ST) configurations were devised.

The CO_2 avoidance costs increase for smaller CO_2 emitter size (i.e. annual CO_2 emissions) for all CO_2 capture configurations, due to economies of scale. For the short term ST cases, the results show the lowest CO_2 avoidance costs for the oxyfuel configurations for the refineries (both 52–57 €/t) and medium sized chemical plant (80 €/t) cases, partly because of fuel savings, resulting from the operation of the process units in oxyfuel mode. For the small sized chemical plant, the post-combustion configuration shows the lowest CO_2 avoidance costs (117 €/t), due to the relatively low capital expenditures for post-combustion technology. The pre-combustion configurations show structurally higher CO_2 avoidance costs (ranging from 73 €/t for the large refinery to 167 €/t for the small chemical plant) than the post- and oxyfuel combustion configurations, due to high natural gas costs and high capital expenditures.

The study shows that CO_2 avoidance costs can also differ significantly within industrial plants, not only because of economies of scale, but also due to varying CO_2 concentrations in flue gases. Implementation issues associated with short term configurations

revolve mainly around retrofitting process units. Although retrofitting is technically feasible for all three capture technologies, it still needs prove on a commercial scale. Limited space availability for capture equipment is a potential problem, especially for post-combustion, but is a very site-specific issue. Although not quantified in this study, site-specific conditions could lead to higher costs than indicated in this study coming from the replacement of existing equipment, retrofit of process units, and although still unknown, higher chances of operational failure in the process units (or upstream H_2/O_2 supply facilities) when operated in oxyfuel or pre-combustion mode. More research into the latter issue is, therefore, imperative to eliminate uncertainties for plant operators.

The long term results are more indicative than the short term results, due to cost data uncertainty and the long time frame in which possible plant layout changes may take place. For the long term ST cases, CO_2 avoidance costs are lowest for the oxyfuel configurations (large refinery: 24 €/t; small chemical plant: 72 €/t), followed by the post-combustion configurations (large refinery: 69 €/t; small chemical plant: 96 €/t). The pre-combustion configuration was not examined for the long term due to data limitations; more detailed and robust techno-economic input data is required to construct a long-term pre-combustion case. Expectations for the long term are that the focus will shift from retrofit issues to the replacement of old process units with new-built capture-ready process units. Furthermore, long term configurations will probably have minimal spatial constraints in (new-built) plant lay outs, capture technologies that are highly integrated with core processes, and optimized utility plants. These factors are expected to have a damping effect on the projected long term CO_2 avoidance costs.

For the hydrogen plant case, the CO_2 avoidance costs (67 €/t) are lowest for the configuration for which 54% of the CO_2 is captured solely from the high pressure process gas between the water gas shift reactor and pressure swing adsorption unit. Yet, by also capturing CO_2 from the furnace stack flue gas, a total CO_2 reduction of 84% is achieved with a CO_2 avoidance cost of 87 €/t. Post-combustion capture appears to be more expensive for both the short and long term.

Both for the short and long term cases, the availability of waste heat could significantly lower the CO_2 avoidance costs of the post- and pre-combustion configurations as well as for the capture configuration of the hydrogen plant.

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Appendix A

The relevant formulae used to calculate the technical and economic performance of the CO_2 capture configurations are described below.

A.1. Technical parameters

Base case

Most of the heat[1] and electricity required for the core processes of the industrial facility are produced by on-site CHP plants, boilers and furnaces. The total heat, electricity and CO_2 emissions produced in n process units in the base case (without CO_2 capture) were calculated by using Formulae (1)–(3), respectively.

$$E_{th,bc} = \sum_{i=1}^{n} C_{th,i} * N_{th,i} * CF_{th,i} * 3.6 * 8.76 \quad (1)$$

$$E_{e,bc} = \sum_{i=1}^{n} C_{e,i} * N_{e,i} * CF_{e,i} * 3.6 * 8.76 \quad (2)$$

$$Y_{bc} = \sum_{i=1}^{n} \frac{FC_i * OF_i * EF_i}{10^9} \quad (3)$$

where $E_{th,bc}$ and $E_{e,bc}$ are the respective total annual heat (TJ_{th}) and electricity (TJ_e) production in the industrial plant in the base case. C_i, CF_i and η_i are the respective capacity (MW), capacity factor, and efficiency of process unit i, Y_{bc} is the total annual CO_2 emissions in the industrial plant in the base case (Mt/y). FC_i, OF_i and EF_i of process unit i are the respective annual fuel consumption (GJ_i), oxidation factor, and emission factor (kg CO_2/GJ_p).

Energy consumption for CO_2 capture process

The additional energy required for CO_2 capture using post-combustion technology is given by Formulae (4) and (5).

$$\Delta E_{th,post} = \sum_{i=1}^{n} Y_{bc,i} * CR_{post} * SHR_{cc,post} \quad (4)$$

$$\Delta E_{e.post} = \sum_{i=i}^{n} Y_{bc,i} * CR_{post} * (SER_{fg\ blower} + SER_{CC} + SER_t) \quad (5)$$

where ΔE_{th} and ΔE_e are the respective total additional annual heat (TJ_{th}/y) and electricity production (TJ_e/y) required for the post-combustion capture configurations. $SER_{fg\ blower}$, SER_{cc}, and SER_t are the Specific Electricity Requirement for flue gas transport, CO_2 capture[2], and CO_2 treatment[3] (GJ_e/t CO_2), respectively. $SHR_{cc,post}$ is the Specific Heat Requirement for solvent regeneration (GJ_{th}/t CO_2). CR_{post} is the CO_2 capture ratio. $Y_{bc,i}$ are the CO_2 emissions for process unit i.

The heat savings for the oxyfuel configuration $\Delta E_{th,oxy}$ (GJ_{th}/y) are calculated by Formula (6), using the base case fuel consumption for heat production for m boilers $E_{p,b,bc,j}$ (GJ_p/y) and n furnaces $E_{p,f,bc,i}$, and the Fuel Reduction Rates for boilers FRR_b and furnaces FRR_f.

$$\Delta E_{th,oxy} = \sum_{i=i}^{n} (-E_{p,f,bc,i} * FRR_f) + \sum_{j=i}^{m} (-E_{p,b,bc,j} * FRR_b) \quad (6)$$

The additional electricity production for the oxyfuel case ΔE_e (GJ_e/y) is calculated by using Formula (7). SER_{O2} is the Specific Electricity Requirement for the amount of oxygen required for the firing of the fuels (GJ_e/t O_2). Fi (t/y) is the amount of fuel i consumed per year. The amount of oxygen needed for oxyfuel combustion was estimated by multiplying the fuel consumption per hour with the stoichiometric amount of oxygen needed for the combustion of fuel i $M_{O_2,i}$ (t O_2/t fuel), which in turn is multiplied with an Excess Oxygen Factor (EOF) of 1.03 (3% excess oxygen). In addition, the annual CO_2 emissions in the oxyfuel case Y_{oxy} (Mt/y) were multiplied with the capture ratio CR_{oxy} and the Specific Electricity Requirement for the CO_2 treatment and compression $SER_{t,oxy}$ (GJ_e/t CO_2).

$$\Delta E_{e,oxy} = \sum_{i=1}^{n} \left(F_i * SER_{O_2} * M_{O_2,i} * EOF\right) + \left(Y_{oxy} * CR_{oxy} * SER_{t,oxy} * 10^6\right) \quad (7)$$

[1] This study uses the term *heat* for direct heat (low and high temperature) as well as indirect heat (low, medium, high pressure steam).

[2] The electrical requirements are a result of the various pumps, fans, etc. in the amine process and cooling tower system.

[3] The category CO_2 treatment & compression comprises a CO_2 drying, purification and compression unit.

The hydrogen required to operate the boilers and furnaces in the pre-combustion configuration is produced in a Steam Reformer (SR). It was assumed that the input energy requirement (natural gas) is proportionally related to the hydrogen output. Hence, the amount of input energy required $E_{ng,pre}$ (GJ_p/t natural gas) for the SR plant was obtained by multiplying the hydrogen in terms of energy $E_{H_2,pre}$ (GJ_p/t H_2) – which is required as input fuel for the furnaces and boilers in the industrial plant in the pre-combustion configuration – with the ratio between the natural gas requirements $E_{ng,ref}$ (GJ_p/t natural gas) and the hydrogen output $E_{H_2,ref}$ (GJ_p/t H_2) as indicated in literature (see Formula (8)). Similarly, the annual CO_2 emissions of the SR plant Y_{pre} (Mt/y) are calculated by multiplying the hydrogen in terms of energy with the ratio between the annual CO_2 emissions in the reference case Y_{ref} (Mt/y) and the hydrogen production in the reference case (see Formula (9)).

$$E_{ng,pre} = E_{H_2,pre} * \frac{E_{ng,ref}}{E_{H_2,ref}} \tag{8}$$

$$Y_{pre} = E_{H_2,pre} * \frac{Y_{ref}}{E_{H_2,ref}} \tag{9}$$

To achieve high reduction levels, the CO_2 is captured from both the high-pressure process gas (~17 bar) and from the SR furnace stack flue gas (~1 bar). In both processes, the captured CO_2 is stripped, cooled, dried, purified and compressed to make it ready for transport. The CO_2 capture from the process gas is done by using the solvent ADIP-X. As the (partial) pressure and CO_2 concentration of the process gas is constant, one value was used for the Specific Electricity Requirement $SER_{ADIP\text{-}X}$ (GJ_e/t CO_2) and Specific Heat Requirement $SHR_{ADIP\text{-}X}$ (GJ_{th}/t CO_2) of the regeneration step of ADIP-X. The CO_2 capture from the flue gases was done by using the solvent MEA.

The additional heat $\Delta E_{th,cc,pre}$ (GJ_{th}/y) and electricity $\Delta E_{e,cc,pre}$ (GJ_e/y) required for the CO_2 capture process is calculated by multiplying the specific heat and electricity requirements with the CO_2 emissions and capture ratios (see Formulae (10) and (11)).

$$\Delta E_{th,cc,pre} = [Y_{pg} * CR_{pg} * 10^6 * SHR_{ADIP-X}] + [Y_{fg} * CR_{post} * 10^6 * SHR_{MEA}] \tag{10}$$

$$\Delta E_{e,cc,pre} = \left[Y_{pg} * CR_{pg} * 10^6 * SER_{ADIP-X}\right] + \left[Y_{fg} * CR_{post} * 10^6 * SER_{MEA}\right] \tag{11}$$

The additional electricity requirement $\Delta E_{e,pre}$ (GJ_e/t CO_2) and heat requirement $\Delta E_{th,pre}$ (GJ_{th}/t CO_2) were calculated by using Formulae (12) and (13), where $SER_{t,pre}$ (GJ_e/t CO_2) is the CO_2 treatment and compression step, $SER_{H_2C,pre}$ (GJ_e/t CO_2) is the hydrogen compression step, $SHRF$ is the Specific Heat Recovery Flow from the SR per tonne hydrogen (GJ_e/t H_2) and M_{H_2} is the amount of hydrogen per tonne CO_2 captured (t H_2/t CO_2).

$$\Delta E_{th,cc,pre} = \Delta E_{th,cc,pre} + [(Y_{pg} * CR_{pg} + Y_{fg} * CR_{fg}) * 10^6 * (M_{H_2} * SHRF + SHR_t)]. \tag{12}$$

$$\Delta E_{e,pre} = \Delta E_{e,cc,pre} + \left[(Y_{pg} * CR_{pg} + Y_{fg} * CR_{fg}) * 10^6 * (SER_{t,pre} + SER_{H_2C,pre})\right]. \tag{13}$$

Amount of CO_2 captured and avoided

The annual amount of CO_2 produced in the capture configuration $Y_{p,cc}$ (Mt/y) is calculated by multiplying the additional primary energy requirements ΔE_p (GJ_p/y) with the specific CO_2 emissions factors for the fuels used in the industrial plants EF_{fuel} (kg/GJ_p) and adding this to the annual CO_2 emissions in the base case Y_{bc} (Mt/y), and annual amount of CO_2 related to the production of the imported electricity Y_{im} (Mt/y) (see Formula (14)). Y_{im} (Mt/y) is calculated by multiplying the additional imported electricity $\Delta E_{e,im}$ (GJ_e/y) with the average short or long term CO_2 emission factor of the Dutch electricity mix EF_{grid} (kg/GJ_e) (see Formula (15)).

$$Y_{p,cc} = Y_{bc} + \frac{\Delta E_p * EF_{Fuel,i}}{10^9} + Y_{im} \tag{14}$$

$$Y_{im} = \frac{\Delta E_{e,im} * EF_{grid}}{10^9} \tag{15}$$

The annually captured CO_2 emissions Y_c (Mt/y) are calculated by multiplying the sum of the total base case CO_2 emissions Y_{bc} (Mt/y) and total CO_2 emissions of the extra boiler (s) Y_{bc} (Mt/y) with the capture ratios of the CO_2 capture technique applied (see Formula (16)). The amount of remaining CO_2 emissions Y_e (Mt/y) and avoided CO_2 emissions Y_a (Mt/y) in the capture configuration are calculated by using Formulae (17) and (18), respectively.

$$Y_c = (Y_{bc} + Y_{ep}) * CR \tag{16}$$

$$Y_e = Y_{bc} - Y_c \tag{17}$$

$$Y_a = Y_{bc} - Y_e \tag{18}$$

A.2. Economic parameters

The annualized capital costs (CAPEX) are calculated by multiplying the investment costs I (M€) with an annuity factor α (see Formula (19)). The annuity factor is obtained by using the real discount rate r (%) and lifetime LT (y) of the investment (see Formulae (20)).

$$CAPEX = \alpha * I \tag{19}$$

$$\alpha = \frac{r}{1-(1+r)^{-LT}} \tag{20}$$

The capital cost of a system component depends mainly on two factors: the size of the process trains and the number of parallel process trains. A generic scaling relation is applied to the capital costs taken from literature to account for this effect (see Formula (21)). SF is the typical scaling factor for a particular capture equipment.

$$\frac{Cost_A}{Cost_B} = \left(\frac{Scale_A}{Scale_B}\right)^{SF} \tag{21}$$

The installed cost per train will be somewhat lower for a multi-train system than that for a single-train system as the multiple units may share some auxiliary equipment, and the required labour and special machinery costs per train generally becomes lower (Larson et al., 2005). The capital investment of a component i (I_i: M€) is expressed as proposed by Larson et al. (2005) (Formula (22)):

$$I_i = I_{i,ref} * \left(\frac{N_{units}}{N_{units,ref}}\right)^{SF_n} * \left(\frac{S}{S_{ref}}\right)^{SF_i} \tag{22}$$

where $I_{i,Ref}$ is the reference capital investment (M€), N_{units} is the number of parallel process trains per system, $N_{units,ref}$ is the number of parallel process trains in the reference system, S is the capacity of a single process train, 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 scaling factor for component i.

The operational costs (OPEX) comprise the costs for the additional energy use, materials, maintenance, and miscellaneous costs (overhead, insurance, taxes and owner costs). The additional energy costs can be calculated by multiplying the additional heat and electricity production with the natural gas and electricity prices. As it is assumed that the extra boiler(s) will use merely natural gas as feedstock, the Dutch natural gas and electricity prices for the industry P_{ng} (€/GJ_p) and P_e (€/GJ_e), the extra boiler(s) efficiency η, and the additional heat (ΔE_{th}/y) and electricity (ΔE_e/y) can be used for

the calculation of the additional electricity C_e (M€/y) and natural gas C_{ng} (M€/y) costs (see Formulae (23) and (24)).

$$C_e = \Delta E_e * P_e \tag{23}$$

$$C_{ng} = \left(\frac{\Delta E_{th}}{\eta_{boiler}}\right) * P_{ng} \tag{24}$$

The annual material costs C_m (M€/y) are calculated by multiplying the demand of material i M_i (t/t CO_2 captured) with the price of material i P_i (unit: material and capture case dependent). Next, the costs of all materials are summed up (see Formula (25)).

$$C_m = \sum_{i=1}^{n} M_i * P_i \tag{25}$$

The total operational costs (M€/y) are obtained by adding up the annual costs for materials, fuel, electricity, maintenance C_{ma} (M€/y) and labour C_l (M€/y) (see Formula (26)).

$$OPEX = C_m + C_{ng} + C_e + C_{ma} + C_l \tag{26}$$

The total annual cost (C_{tot}: M€/y) are obtained by adding up the OPEX and CAPEX (see Formula (27)).

$$C_{tot} = OPEX + CAPEX \tag{27}$$

The CO_2 captured C_c and CO_2 avoidance costs C_a (€/t CO_2) are calculated by dividing the total annual costs by the annually captured (Y_c) and annually avoided CO_2 emissions (Y_a), respectively (see Formulae (28) and (29)).

$$C_c = \frac{C_{tot}}{Y_c} \tag{28}$$

$$C_c = \frac{C_{tot}}{Y_a} \tag{29}$$

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