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Quantifying carbon capture potential and cost of carbon capture technology application in the U.S. refining industry

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ABSTRACT

Carbon capture (CC) technology is receiving increasing attention as a critical technology for climate change mitigation. Most previous studies focus on the application of CC technology in the power generation sector, while fewer studies have analyzed applications in the refining industry, which is one of the largest greenhouse gas (GHG) emissions sources in the U.S. industrial sector. Unlike the power generation sector, the refining industry has highly distributed CO₂ emission sources. In this paper, bottom-up modeling and techno-economic analysis approaches are integrated to quantify the national CO₂ emission reduction potential and costs of three types of CC technologies applied to U.S. refineries: (1) pre-combustion, (2) post-combustion, and (3) oxyfuel-combustion. Two scenarios are developed to compare different design strategies for CC systems; one is a distributed design scenario for post-combustion technology, the other is a centralized design scenario for pre-combustion and oxyfuel-combustion technology. The results of the two scenarios are compared, and the trade-offs between different design strategies are highlighted. The results shown in this study provide an intuitive and quantitative understanding of the potential of CC technology to reduce CO₂ emissions from the U.S. refining industry. Such information is helpful to policymakers, oil companies, and energy/environmental analysts for strategic planning and systems design to manage future CO₂ emissions of refineries.

1. Introduction

Increasing attention has been paid to carbon capture (CC) technology as it is widely considered as a promising option for mitigating greenhouse gas (GHG) emissions (IEA, 2016). During past two decades, the performance, cost, and impacts of CC technology have been intensively studied, and most of these studies focus on the application of CC in the power generation sector (Gerdes et al., 2014; Sathre et al., 2012; Sathre and Masanet, 2012, 2013). Comparatively less attention has been paid to the application of CC technology in the petroleum refining industry, which ranks as the third largest industrial source of GHG emissions in the United States (EPA, 2014a). In 2013, the U.S. refining industry emitted approximately 177×10^6 t of CO₂-equivalent emissions (MtCO₂-eq), accounting for 2.8% of total U.S. GHG emissions (EPA, 2014b). Therefore, applying CC technology to the refining industry could contribute to significant reductions in national GHG emissions.

A few studies have evaluated the industry-wide CO₂ reduction potential and cost of CC technology in different industries other than the

power generation sector. Kuramochi et al. conducted a techno-economic assessment of CC technology applied to several key industrial sectors, including iron and steel, cement, petroleum refineries, and petrochemicals (Kuramochi et al., 2012). Several studies have discussed CO₂ emissions in refineries (Abella and Bergerson, 2012; Gunaseelan et al., 2009), while others have analyzed the CO₂ reduction potential of different measures in the refining industry, such as energy system integration and process intensification (Morrow et al., 2013). Van Straelen et al. evaluated the technical feasibility of applying post-combustion CC technology to one world-scale refinery plant and concluded that it is technically feasible to capture CO₂ emissions from refineries at the cost of 90–120 Euro/t CO₂ (van Straelen et al., 2010). Berghout et al. presented a techno-economic analysis for implementing CC in five industrial plants (including refineries and chemical plants) in the Netherlands for the short term and long term (Berghout et al., 2013). Johansson et al. analyzed different heat supply options for post-combustion CC in oil refineries and estimated the capture avoidance cost (Johansson et al., 2013). However, most of previous studies do not quantify the CO₂ reduction potentials and costs of CC technology for

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Nomenclature

CC	Carbon capture	DS_u	The quantity of steam that is directly used for the unit process (billion kg/year)
ADU	Atmospheric distillation	SMR_u	The steam requirement of SMR (billion kg/year)
VDU	Vacuum distillation unit	WT_u	Quantity of steam required for wastewater treatment systems (billion kg/year)
ADU	Atmospheric distillation unit	AGR_u	The quantity of steam required for acid gas removal systems (billion kg/year)
VDU	Vacuum distillation unit	LB_u	Steam blowdown and loss during steam generation and delivery (billion kg/year)
CKU	Coking unit	SW	Steam requirement for treating one unit of wastewater (kg steam/kg wastewater)
HCU	Gas oil hydrocracking unit	WW_u	Quantity of wastewater generated during the operation of unit process u (kg wastewater/oil barrel)
CTU	Catalytic hydrotreating unit	S_u	Quantity of solvent required to removal H_2S and CO_2 (gallon/oil barrel)
CCU	Catalytic cracking unit	f_c	Steam usage for removing one unit of acid gas (kg steam/gallon solvent)
HCU	Hydrocracking unit	F_t	Average steam loss in the refinery process (%)
DTU	High-severity distillate hydrotreating;	fb	Blowdown rate of the boilers used in the steam generation system (%)
KTU	Kerosene treating unit – low-severity distillate hydro-	DE_u	Direct electricity consumption for each unit process u (GW h/year)
	treating	UE_u	Electricity consumption of utility systems
GTU	Gasoline hydrotreating	f_{ed}, f_{eu}	Electricity usage factors
NTU	Naphtha treating unit	$VO_{u,p'}$	The volume of refinery byproduct p' that can be used as a fuel
CRU	Catalytic reforming unit	TG	Total on-site CO_2 emissions
ISU	Isomerization unit	$EF_{p'}$	Emission factors of byproduct p' (kg/oil barrel)
AKU	Alkylation unit	EF_f	Emission factors of fossil fuel (kg/oil barrel)
$VO_{u,p}$	Outputs (p) of unit process u (thousand oil barrel/calendar day, M BPCD)	$LHV_{p'}$	Lower heating value (LHV) of refinery byproducts
$VI_{u,i}$	Inputs (i) of unit process u (thousand oil barrel/calendar day, M BPCD)	CT_s	Un-levelized capital cost of CO_2 capture system for scenario s
$Y_{u,i}$	Volume yields of product p based on the input i for the unit process u	FC_s	Costs of fuel
$FC_{u,f}$	Fuel consumption for each unit process u (10^6 GJ/year) by different fuel f	OM_s	Costs of operation and maintenance (O&M) other than fuel
EC_u	Electricity consumption for each unit process u (GW h/year)	$CT_{s=post,u}$	Capital cost of post-combustion system (\$/t CO_2 PA)
EP_g	Percentage of electricity from different source g	CP_p	Capital cost (\$/t CO_2 PA) of CO_2 capture system implemented in the power plant
PF_g	The efficiency of electricity generation and distribution (%)	CF_p	CO_2 captured per year (10^6 t CO_2 PA) by the CO_2 capture system applied to different power plant type p
$ED_{u,f}$	Fuel consumption of fired heaters (10^6 GJ/year) that burn fuels and provide direct heat to each unit process u .	CR_u	Total CO_2 emitted from different unit process, steam system, and SMR system in the refinery process
$ESMR_{u,f}$	Fuel consumption of steam methane reforming systems (SMR) that produce hydrogen (10^6 GJ/year)	CE_u	Carbon capture efficiency (%)
$ES_{u,f}$	Fuel consumption of steam systems	SF	Scaling factor
$f_{u,f}$	Fuel use factor for fired heaters used in each unit process u (GJ/oil barrel)		
$f_{u,f}$	Fuel use factor of SMR, GJ/thousand standard cubic feet		
HL_u	Hydrogen requirement for each unit process u (MSCF/oil barrel)		
f_{s_f}	Average fuel consumption for steam generation (MJ/kg steam)		

different unit processes (e.g., implementing CC specifically for the atmospheric distillation process (ADU)), precluding the use of their results by refineries to develop process-specific design strategies. Unlike power plants, CO_2 in refineries is emitted from highly distributed sources, such as unit processes (e.g., catalyst regeneration), utility systems (e.g., steam systems and hydrogen production), and fired heaters associated with the individual unit process. Due to the distributed nature of CO_2 emission sources in refineries, there is no single cost for CC technology because these emission sources vary considerably with respect to their CO_2 concentration and capacity. Understanding potential costs and reduction potential of CC application in specific unit processes are critical for individual refineries to develop specific application strategies based on their process configurations.

In this study, we integrate bottom-up unit process modeling and techno-economic analysis approaches to address the three research questions that have not been fully answered by previous studies – (1) By how much could CO_2 emissions be reduced by applying CC to the entire U.S. refining industry? (2) How much do different CC systems cost and

where are the associated uncertainties based on tested industrial applications? (3) What are the future directions for promoting CC application moving forward? Answers to these three questions will provide a quantitative understanding of the potential trade-offs and barriers to implementing CC technology in U.S. refineries, while the results of this work could serve as a transparent data source for decision makers interested in developing CC technology strategies at refineries with the largest reduction potential and least cost. Furthermore, there are many cost-effective measures available for improving the energy efficiency of refineries (e.g., energy integration), and some of them have the potential to be integrated with CC technology (Morrow et al., 2013). Although this study does not include the analysis of an integrated approach, the results of this study could provide a foundation for future analysis to explore the potential of coupling CC with other process improvement measures. Emerging technologies (e.g., membrane) are also excluded in this study because this analysis does not intend to predict the feasibility of technology integration and adoption in the future. Although the focus of this work is U.S. refineries, the modeling

approach presented in this work has the potential to be used to quantify the costs and CO₂ mitigation potential of CC applications in other industry sectors.

2. Materials and methods

The system boundary of this work is gate-to-gate, including all refinery operations from crude oil inputs to refined product outputs, and major unit processes used in refineries and CC facilities installed within refineries. The analysis was performed in three steps. First, on the basis of a previously developed model (Morrow et al., 2015), a bottom-up unit process modeling approach was used to develop a representative model for typical operations in U.S. refineries. Refineries vary regarding their process configurations, feedstocks, and product slate. Because process-level data for individual refineries are not publicly available, it is unrealistic to build models for each of the 137 refineries in the United States (EIA, 2015b). However, the typical unit processes used in the refining industry are similar (Gary et al., 2007; Lyons and Plisga, 2011). The representative model simulates the energy and mass balance of typical unit processes in the refineries based on the data for average operational conditions, process configurations, feedstocks, product yields, and energy consumption for the U.S. refining sector collected from the literature (Gary et al., 2007; Morrow et al., 2015) and from national statistics (EIA, 2015a, 2015b, 2015c; Oil and Gas Journal Research Center, 2014). Because operational data for refineries used in this work are based on the data from U.S. Energy Information Administration (EIA) in 2014, all costs data and analysis are harmonized to

2014 as the base year. However, the transparent modeling framework presented allows future updates for use by a range of decision makers (e.g., policymakers) and energy/environment analysts in future macroeconomic analyses of industry-wide CC technology applications. Alternatively, the general method and results in this work can also be applied by refinery stakeholders for decision making around CC in specific refineries using refinery-specific data.

Second, the CO₂ emissions from different unit processes and utilities were estimated based on average emission factors for different fuels and the energy balance simulated in the previous step. The points of emissions include fired heaters related to each unit process, utility systems such as steam generation systems and steam reforming systems (SMR) for hydrogen production, and unit processes directly emitting CO₂ such as the catalytic cracking unit (CCU). Third, emissions estimated were used in the techno-economic analysis to quantify national-level CO₂ reduction potential and costs of different CC systems implemented for various unit processes and utility systems. Two scenarios were developed to represent different design strategies. One is a post-combustion scenario using the point-by-point source approach, in other words, post-combustion CC system is implemented as distributed systems associated with major CO₂ emission points. The other is a pre-combustion scenario coupled with oxyfuel-combustion for CCU, which is a centralized design. In this study, we assumed that CC systems are sized based on the CO₂ emissions generated in the refineries, and emissions from energy supply of CC systems are not captured, which is a standard assumption for CC analysis.

Fig. 1. Process flow of a representative U.S. refinery.

2.1. Refining industry

The typical unit processes in refineries are shown in Fig. 1, depicted as boxes. Both solid and dash lines represent material flows, dash lines are used to avoid confusion when two lines cross. Each colored cube (e.g., SMR and hydrogen recovery) represent the consumption of utilities from a specific system. For example, the ADU has three colored cubes, representing the consumption of utilities from steam systems, refinery gas processing and flare systems, and water treatment and delivery systems. Due to a large number of intermediate and byproducts generated in each unit process, only the major products or the products that are used as inputs to other unit processes are specifically labeled in Fig. 1.

In the ADU, crude oil is separated into various groups of hydrocarbons based on their boiling points, the bottom products are sent to the vacuum distillation unit (VDU) for further separation (EIA, 2012). In the VDU, the atmospheric reduced crude is separated into light gas oil, heavy gas oil, and residual left in the bottom (EIA, 2012). In the coking unit (CKU), the residual from the VDU is converted to petroleum coke and other intermediate chemicals and light products such as naphtha, gas oil, methane, and ethane (Morrow et al., 2015). The gas oil from the ADU, VDU, and CKU is catalytically treated in the catalytic treating unit (CTU) to remove contaminants (Lyons and Plisga, 2011). The treated gas oil is sent to the CCU to be converted to high-value products such as gasoline and olefins (Sadeghbeigi, 2000). The gasoline hydrotreating unit (GTU) is used to remove contaminants in the CCU-produced gasoline and to reduce the quantity of olefins and aromatics (Speight and Ozum, 2001). Sulfur in CCU-produced products is removed by high-severity distillate hydrotreating (DTU) (Morrow et al., 2015). The olefins produced from the CCU and isobutane are converted to alkylate, a high-octane component (EIA, 2013a), through the Alkylation Unit (AKU). In the hydrocracking unit (HCU), low-quality heavy gas oil from the ADU, VDU, and CKU is converted to high-quality products such as gasoline, kerosene, and diesel (EIA, 2013c). The low-severity distillate hydrotreating unit (KTU) is used to remove sulfur in product streams from the HCU and ADU (Morrow et al., 2015). The catalytic reforming unit (CRU) uses naphtha to produce reformate that is a high-octane gasoline blending component (EIA, 2013b). Straight-run naphtha produced from ADU is treated in the naphtha treating unit (NTU). The light naphtha from the HCU and NTU is upgraded in the isomerization unit (ISU) to achieve a desirable octane value and to reduce benzene content (Valavarasu and Sairam, 2013).

2.2. Material balance

A bottom-up unit process model was developed to simulate the material balance (material inputs and outputs) of each unit process typically operated in the U.S. refining system, as depicted in Fig. 1. The bottom-up material balance is the foundation of the energy and CO₂ emission analysis described in the next section, which ultimately estimates quantities of CO₂ from different unit processes and utility systems.

The materials balance model was constructed according to Eq. (1):

$$VO_{u,p} = Y_{u,i,p} \times VI_{u,i} \quad (1)$$

Where $VO_{u,p}$ and $VI_{u,i}$ are the outputs (p) and inputs (i) of unit process u (thousand oil barrel/calendar day, M BPCD). $Y_{u,i,p}$ is the volume yields of product p based on the input i for the unit process u . Using volume instead of mass as the basis for the material balance is a standard practice used in the refining industry (EIA, 2015c).

Recent data on aggregated capacities and utilization of different unit processes in U.S. refineries were collected from the EIA (EIA, 2015a, 2015b, 2015c) and 2014 Worldwide Refining Survey (Oil and Gas Journal Research Center, 2014). These data, which are provided in SI Table 1, were used to establish the average feed rates of major unit processes in the bottom-up model. Data on product yields for each unit

process were collected from the literature (Gary et al., 2007; Morrow et al., 2015). All of the data were collected for the year 2014, but the model developed in this study can be updated when more recent data become publicly available. If more recent data is not available, the method is still valid and this work can be used for decision making in CC application in specific refineries.

2.3. Energy and CO₂ emission analysis

Based on the material balance analysis discussed above, energy and CO₂ emission analysis were then conducted to estimate the amount of CO₂ from different unit processes and utility systems, as well as quantify the industry-wide CO₂ emissions from the U.S. refining industry. The overall approach includes two steps. First, energy consumption of each unit process and utility system was estimated based on the material balance and fuel consumption factors collected from the literature. Second, CO₂ emissions were estimated based on the emission factors of different fuels and energy consumption obtained from the first step.

The total primary energy consumption (TE : 10⁶ GJ/year) is estimated by the equation below.

$$TE = \sum_u \sum_f FC_{u,f} + \sum_u \sum_g EC_u \times EP_g / PF_g \times 3.6 / 10^3 \quad (2)$$

Where $FC_{u,f}$ is the fuel consumption of different fuel f for each unit process u (10⁶ GJ/year), inclusive of all the unit processes shown in Fig. 1. Fuel f includes fossil fuels and byproducts produced in the refining process that can be used as fuels (e.g., hydrogen, ethane, and methane). EC_u is the electricity consumption for each unit process u (GWh/year). EP_g is the percentage of electricity from different power sources g , which includes grid purchased electricity and on-site generation through combined heat and power (CHP). PF_g is the overall efficiency of electricity generation, transmission, and distribution (%).

According to a recent analysis from the U.S. Department of Energy (DOE), 34% of the electricity used in the U.S. refining industry is produced on-site using CHP (Brueske et al., 2012). Therefore, $EP_{g=CHP}$ is assumed to be 34%. The overall efficiency of electricity generation, transmission, and distribution for grid-purchased electricity is assumed to be 33%, based on analysis from U.S. EIA (EIA, 2016a) and the literature (Yao et al., 2016, 2015). The efficiency of electricity generation of CHP used in the U.S. refineries is 69% based on the recent U.S. DOE analysis (Brueske et al., 2012). $3.6 / 10^3$ is unit conversion for GWh to 10⁶ GJ.

The fuel consumption of each unit process is estimated by Eq. (3).

$$FC_{u,f} = ED_{u,f} + ESMR_{u,f} + ES_{u,f} \quad (3)$$

Where $ED_{u,f}$ is the fuel consumption of fired heaters (10⁶ GJ/year) that burn fuels and provide direct heat to each unit process u . $ESMR_{u,f}$ is the fuel consumption of SMR that produce hydrogen (10⁶ GJ/year). $ES_{u,f}$ is the fuel consumption of utility steam systems that generate and deliver steam used in the reboiler, compressor steam drivers, SMR systems, and acid gas removal systems. Because the SMR and utility steam systems are commonly built as centralized facilities, the fuel consumption $ES_{u,f}$ calculated here can be viewed as the centralized facilities' energy consumption allocated to each unit process u .

The fuel consumption of direct fired heaters for each unit process is calculated by Eq. (4).

$$ED_{u,f} = f_{u,f} \times \sum_i VI_{u,i} \times T / 10^3 \quad (4)$$

Where $f_{u,f}$ is the fuel use factor for fired heaters used in each unit process u (GJ/oil barrel). T is the number of calendar days in a year (365 days/year). 10^3 is used to convert 10³ GJ to 10⁶ GJ. The fuel use of the SMR is calculated by Eq. (5).

$$ESMR_{u,f} = fr_{u,f} \times HI_u \times \sum_i VI_{u,i} \times T/10^3 \quad (5)$$

Where $fr_{u,f}$ is the fuel use factor of the SMR (GJ/10³ standard cubic feet (MSCF)). HI_u is the hydrogen requirement for each unit process u (MSCF/oil barrel). Hydrogen is consumed at the following unit processes: CTU, HCU, DTU, CCU, KTU, NTU, ISU, and GTU.

The fuel use of steam systems allocated to each unit process is calculated based on the steam requirement of each unit process and utility systems that consume steam, as shown in Eq. (6).

$$ES_{u,f} = fs_f \times (DS_u + SMR_u + WT_u + AGR_u + LB_u) \quad (6)$$

Where fs_f is the average fuel consumption for steam generation (MJ/kg steam). DS_u is the quantity of steam that is directly used for unit process u (billion kg/year), such as steam used for stripping in the ADU and steam used for steam turbine driving air compressors. SMR_u is the steam requirement of the SMR (billion kg/year). WT_u and AGR_u are the quantities of steam required for waste water treatment systems and acid gas removal systems (billion kg/year), respectively. LB_u is the steam blowdown and loss during steam generation and delivery (billion kg/year). The calculation procedure for each of those terms are discussed in details in the SI.

Note that the steam requirement for some unit process (e.g., acid gas removal) are negative due to steam recovery. The fuel consumption factor for steam generation, fired heaters, and other unit processes discussed above are derived from the literature (Gary et al., 2007; Maples, 2000) and the authors' previous publications (Morrow et al., 2015, 2013).

The total on-site CO₂ emissions (TG: 10⁶ t/year) are estimated using Eq. (7) which calculates CO₂ emissions coming from burning fossil fuels and refinery byproducts.

$$TG = \sum_u \left(\sum_{p'} EF_{p'} \times VO_{u,p'} + \sum_f (FC_{u,f} + EC_u \times EP_{g=CHP}/PF_{g=CHP} - VO_{u,p'} \times LHV_{p'}) \times EF_f \right) \quad (7)$$

Where $VO_{u,p'}$ is the volume of refinery byproduct p' that can be used as a fuel, such as methane, ethane, and hydrogen in refinery gas and catalyst coke in the CCU regenerator. $EF_{p'}$ and EF_f are the emission factors of byproduct p' and fossil fuel (kg/oil barrel), respectively. $LHV_{p'}$ is the Lower Heating Value (LHV) of refinery byproducts. The second term in Eq. (7) calculates total purchased fossil fuels that are burned to supply heat and power to the refinery.

Because it is very difficult to track the destination of each refinery byproduct within all U.S. refineries, a simplified approach used in this research is to assume that refinery byproduct that can be used as fuels are firstly used by the unit process producing them. This is a reasonable assumption given the fact that refinery byproducts can be used in a variety of burners such as boilers, fired heaters, and furnaces, and it is typically economical to burn byproducts on-site to avoid transportation cost (Maples, 2000; Speight and Ozum, 2001). For some unit processes, refinery byproducts generated can provide heat more than the energy requirement, in those cases, the excess byproducts (e.g., hydrogen, ethane, and methane) are assumed to be used as fuels and supplied to steam systems and SMR. Two exceptions are the CRU and the CCU. The refinery gas produced by the CRU is used as a source of hydrogen instead of fuel due to its high concentration of hydrogen (Aitani, 1996). The heat supplied to the CCU is provided by a catalyst regenerator that burns the catalyst coke formed on the surface of the catalyst. Therefore, the refinery gas produced by the CCU is supplied to utility steam systems and the SMR.

The CO₂ emission factors of refinery fuels used in this study are shown in Table 1. The primary purchased fuel burned in the U.S. refining industry is natural gas (Brueske et al., 2012). Therefore, the

purchased fossil fuel here is assumed to be natural gas whose major component is methane. The LHV of ethane, methane, hydrogen are also provided in the same table (Morrow et al., 2015). All emissions shown in Table 1 are for fired heaters and boilers as described in the previous discussion of energy balance calculation.

2.4. Carbon capture systems

In this section, different CC systems and their cost estimations are discussed. Three CC systems are considered in this study: post-combustion, pre-combustion, and oxyfuel-combustion (see Fig. 2). The differences and scenario design of three CC systems are discussed in the following paragraphs. Then the calculation method of estimating the costs of different scenarios are presented.

In the post-combustion system, CO₂ is captured from the flue gas generated by combustion units (e.g., boilers and furnaces) (IEA, 2016). The most developed and studied technique for post-combustion CC is monoethanolamine (MEA) absorption, which has been used in the natural gas industry for more than half of a century (Dutcher et al., 2015; IEA, 2016; Luis, 2016; Yang et al., 2008). Other more efficient solvents are also available (Meerman et al., 2012). Because those solvents are not commercially viable, they are excluded in this study. In the pre-combustion system, fuels are first converted to syngas, then carbon is removed from the fuel before the combustion (IEA, 2016; Yang et al., 2008). The technology options for pre-combustion CO₂ capture include chemical/physical absorption (Meerman et al., 2012), pressure swing adsorption (PSA), and membrane technology (Yang et al., 2008). In the oxyfuel-combustion system, pure O₂ generated from an air separation unit is sent to a combustion unit to produce flue gas with a high purity of CO₂ that does not require further CO₂ separation or can be easily separated before sequestration (Tan et al., 2016).

Post-combustion systems are the most likely to be applied to fired heaters and boilers in existing refineries because these combustion units have similar operational conditions and CO₂ concentration of flue gas (4–6%) (van Straelen et al., 2010) to their counterparts in power plants that use post-combustion systems. As fired-heaters are associated with most of the unit processes, post-combustion systems would be distributed throughout the refinery, as shown in the industrial case studies conducted by oil companies (van Straelen et al., 2010). Post-combustion CC systems can also be applied to centralized utility steam systems and SMR systems. To explore which unit processes or utility systems have the largest potential for post-combustion CC adoption, a scenario was developed to investigate the CO₂ emission reduction potential and cost of post-combustion systems applied to different unit processes, utility steam systems, and SMR systems.

Another scenario was developed for analyzing centralized pre-combustion systems that decarbonize all fuel gases before they are sent to combustion units. Compared to the point-to-point design in the post-combustion scenario, the centralized design in this scenario may be more suitable for existing refinery with limited empty space. From an economic perspective, this option may be preferable to distributed post-combustion systems for the economies of scale. One exception in this scenario is that oxyfuel-combustion is assumed to be implemented for catalyst regeneration at the CCU for two reasons. First, previous research and studies indicate that oxyfuel-combustion is a feasible option for the CCU (CO₂ Capture Project, 2013). Second, H₂-rich fuel gas generated from pre-combustion systems is not suitable for the catalyst

Table 1
LHV and emissions of major fuels used in the refineries (EIA, 2011; Morrow et al., 2015).

	Hydrogen	Ethane	Methane	Catalyst Coke
LHV (MJ/kg)	120	47.6	50.1	N/A
Emission factor (kg CO ₂ /GJ)	N/A	60.3	50.3	109.5

Fig. 2. Process schematics of three CO₂ capture systems (IEA, 2016).

regeneration, which primarily uses air.

The leveled costs of CC are calculated by the equation below:

$$CC_s = CT_s \times \frac{r(1+r)^Y}{(1+r)^Y - 1} + FC_s + OM_s \quad (8)$$

Where CT_s is the un-leveled capital cost of CO₂ capture system for scenario s , and r is the discount rate used to allocate the capital cost over the lifetime of CC system (Y : years). These two parameters are assumed to be 8% over 25 years (IEAGHG, 2012). FC_s and OM_s are the costs of fuel, and operational and maintenance (O&M) other than fuel. To estimate the cost of CC systems in the two scenarios, typical costs of post-combustion, pre-combustion, and oxyfuel-combustion were collected from the recent literature and harmonized as shown in Table 2. The costs were adjusted to 2014 dollars based on the Chemical Engineering Plant Cost Index (Access Intelligence LLC, 2015; GCCSI, 2011; IEA, 2016; Manzolini et al., 2013). The capital cost was normalized on the basis of annual capacity (\$/t of CO₂ captured annually (\$/tCO₂ PA)). The fuel penalty is the quantity of fuel consumed by CC systems, as listed in Table 2. In addition to fuel cost, other O&M costs are also provided in Table 2. The cost of CO₂ transmission and storage is not included in this research because it requires a location analysis to see if there would be transport infrastructure and storage sites nearby all U.S. refineries, which is beyond the scope of this study. However, the location analysis can be easily integrated into the model when future information on possible infrastructure systems and storage sites is publicly available.

As discussed earlier, the post-combustion system is implemented for distributed unit processes and centralized utility steam and SMR systems. Therefore, the capital cost of the post-combustion system was estimated based on the CO₂ emissions coming from each unit process, the utility steam systems, and the SMR systems, as shown in Eq. (9). The equation allows a consideration of the impact of economies of scale instead of assuming fixed process configuration (e.g., the number of absorbers and strippers).

$$CT_{s=post,u} = (CP_p \times CF_p) \times \left(\frac{CR_u \times CE_u}{CF_p} \right)^{SF} / CR_u \quad (9)$$

Where $CT_{s=post,u}$ is the capital cost of post-combustion system (\$/tCO₂ PA) applied to different systems u , including all unit processes, the utility steam system, and the SMR system. CP_p is the capital cost (\$/t

Table 2

The cost data of CC systems applied to power plants (2014\$).

Literature	Power plant type	CC	CO ₂ captured (Mt CO ₂ PA) ^a	Total Capital (\$/t CO ₂ PA) ^b	Fuel Penalty (GJ/t CO ₂)	Other O&M (\$/t CO ₂) ^c
(DOE, 2011)	Natural Gas	Post	1.3	380	4.7	23.1
(IEAGHG, 2012)	Combined Cycle		2.3	588	3.8	13.8
(Rubin and Zhai, 2012)	(NGCC)		2.2	407	3.2	8.8
			1.1	292	4.5	6.3
(DOE, 2013)			1.3	381	4.5	5.2
(EPRI, 2013)			1.3	539	3.9	N/A
(DOE, 2013)	Pulverized	Post	4.1	244	5.0	20.9
(EPRI, 2011)	Coal (PC)		4.0	438	5.4	16.9
(Léandri, 2011)			5.3	299	2.7	16.7
(IEAGHG, 2014)			5.6	420	3.3	19.3
(GCCSI, 2011)			3.0	382	5.9	15.3
(ZEP, 2011)			3.8	320	2.5	7.0
(IEAGHG, 2012)	Natural Gas	Pre	2.5	476	7.2	16.9
(Manzolini et al., 2013)	Combined Cycle		2.5	260	7.1	23.3
	(NGCC)		1.9	407	8.1	18.5
			3.0	183	4.2	10.7
			1.8	301	7.2	17.4
			1.2	482	12	27.6
			3.0	183	4.2	10.7
			1.9	300	7.2	17.4
(DOE, 2010)	Pulverized	Oxy	4.0	240	3.0	4
	Coal (PC)		4.1	295	3.6	2
(EPRI, 2011)			3.6	492	3.0	16
			3.6	489	3.0	15
			3.9	473	2.9	10
(IEAGHG, 2014)			5.5	420	3.1	38

The costs from different years are harmonized to base year 2014 costs on the Chemical Engineering Plant Cost Index (Access Intelligence LLC, 2015).

^a CO₂ captured = CO₂ generated – CO₂ emitted; this parameter is used as the capacity of CC system.

^b tCO₂ PA is tonne per annum based on the capacity of CC system.

^c Other O&M are operation and maintenance cost.

CO₂ PA) of the CC system implemented in power plant type p including Natural Gas Combined Cycle (NGCC) and Pulverized Coal (PC). CF_p is the CO₂ captured per year (10^6 t CO₂ PA) by the CC system applied to different power plant type p . CR_u is the total CO₂ emitted from the different unit processes, the utility steam system, and the SMR system in the refinery process. CE_u is the CC efficiency (%). $CR_u \times CE_u$ calculates the quantity of CO₂ that can be captured by post-combustion system for each unit process and system in a year, which is regarded as the capacity of the CC system. SF is the scaling factor that is used to scale the capital cost for the CC system with different capacities. The scaling factor is assumed to be 0.8 based on literature (Atsonios et al., 2013; IEAGHG, 2012).

The capital cost and CC capacity of CC systems applied to different types of power plants are listed in Table 2. For CC systems applied to all unit process excluding the CCU, the capital cost is estimated based on the NGCC case ($p = \text{NGCC}$) due to the similar CO₂ concentration (4–6%) of flue gas (van Straelen et al., 2010) coming from fired heaters for refinery unit processes and NGCC power plants. The CC system applied to PC power plants ($p = \text{PC}$) is used as the comparative case for the CCU because the CO₂ concentration of flue gas coming from power plants (approximately 13.5%) is similar to that from the CCU fired-heaters. Similarly, based on the CO₂ concentration of flue gas, the capital cost of CC systems applied to the utility steam system and SMR (Collodi and Wheeler, 2010) is estimated based on NGCC and PC power plants, respectively. The average capacities of the utility steam system and SMR system are based on the average capacity of the entire refinery plant that is equal to the capacity of the first unit process ADU. The total CO₂ emissions from different unit processes are estimated based the average capacity data of each unit process listed in Table 3 (EIA, 2015c; Oil and Gas Journal Research Center, 2014), which are used as inputs to the energy and CO₂ emission analysis discussed in the previous section. The CO₂ concentration of flue gases from different unit processes except CCU is assumed to be 4% (van Straelen et al., 2010), while the CO₂ concentration of the flue gas of CCU is assumed to be 13.5% (de Mello et al., 2013).

The fuel cost in the post-combustion scenario was estimated based on the fuel penalty of CC systems applied to power plants as shown in Table 2 and the fuel price. Similar to the capital cost estimation, fuel penalty data from PC cases were used to estimate the fuel consumption of post-combustion systems applied to the CCU and SMR system; while the fuel consumption of the rest of the unit processes and the utility steam system are estimated based on the NGCC case. The fuel price is assumed to be the U.S. natural gas price in 2014 (\$5.1/GJ) (EIA, 2016b) since natural gas is the most common fuel used in the U.S. refinery process (Brueske et al., 2012). Using a similar philosophy, the other O&

M costs in the post-combustion scenario are estimated based on the corresponding cost of NGCC and PC cases as shown in Table 2.

The costs in the pre-combustion scenario are estimated in a manner similar to the post-combustion scenario. The only difference is that it is unnecessary to estimate the CC cost for each unit process in the pre-combustion scenario, because the CC system is implemented as a centralized process to decarbonize fuels before further use in various unit processes. The capital cost of the pre-combustion system is estimated by using the Eq. (9) based on the pre-combustion cases for NGCC power plants shown in Table 2 and the average quantity of CO₂ emitted from an average refinery excluding the CCU as displayed in Table 3. The capital cost of the CC system applied to the CCU is estimated using Eq. (9) based on an oxyfuel-combustion system applied to PC. Other costs are estimated similarly based on cost data collected for pre-combustion systems of NGCC power plants and oxyfuel-combustion of PC power plants.

After the cost was estimated for the two CC scenarios, the CO₂ mitigation potential (CP_s ; 10^6 t/year) is estimated based on the equation below:

$$CP_s = CR_u \times CE_u \times (1 - FP_s \times EF_{f=NG}) \quad (10)$$

Where FP_s is the fuel penalty calculated within the different scenarios (GJ/t CO₂ captured). In this study, we assumed that the additional energy required by CC system can be met onsite by the capacity expansion of steam systems and CHP plants. The CO₂ generated due to the energy consumption of CC systems are assumed to be emitted instead of being processed by CC systems, which is a standard assumption for CC analyses. Basically, Eq. (10) calculates the difference between the CO₂ captured and the CO₂ emitted due to the energy used by the CC systems, giving the net CO₂ reduction potential.

3. Results

The energy analysis results for the typical U.S. refinery (prior to CC technology adoption) are shown in Fig. 3. Four energy consumption sources are presented for each unit process, including fired heaters (blue bar), utility steam systems (red bar), SMR systems (purple bar), and electricity end-uses (green bar). As mentioned previously, steam systems and SMR systems are centralized facilities that provide steam and hydrogen for each unit process. Therefore, the primary energy consumption of steam systems and SMR systems are allocated to each unit process based on their associated usage of utility steam and hydrogen. To avoid double counting, the primary energy consumption of electricity end-uses shown in Fig. 3 only covers purchased electricity, and the electricity generated on-site by CHP is included in the energy

Table 3
Total CO₂ emissions from different unit processes and utility system in the U.S. refineries.

Unit Process	Average Capacity (M BPCD) (EIA, 2015c; Oil and Gas Journal Research Center, 2014)	Total CO ₂ in Flue Gas (Mt CO ₂ PA)	Comparative Cases
ADU	145.4	0.113	NGCC
VDU	89.4	0.045	NGCC
CKU	39.1	0.058	NGCC
CCU	66.0	0.623	PC
HCU	37.3	0.078	NGCC
CTU	49.3	0.027	NGCC
GTU	24.3	0.022	NGCC
NTU	23.7	0.021	NGCC
KTU	20.7	0.011	NGCC
DTU	20.5	0.011	NGCC
CRU (continuous catalyst regeneration)	29.8	0.101	NGCC
CRU (semi-regenerative catalytic reformer)	29.5	0.100	NGCC
ISU (C4 Isomerization)	8.3	0.000	NGCC
ISU (C5/C6 Isomerization)	15.5	0.008	NGCC
Refining Total for Unit Processes	145.4	0.594	
Steam System	–	0.200	NGCC
SMR System	–	0.256	PC

Fig. 3. Primary energy consumption of unit processes in U.S. refineries (2014 year).

consumption of steam systems.

The on-site GHG emissions results for the typical U.S. refinery (prior to CC technology adoption) are shown in Fig. 4, which is consistent with 2014 CO₂ emissions for the U.S. refining industry reported by the U.S. Environmental Protection Agency (174×10^6 t CO₂/year) (EPA, 2016). As mentioned previously, the CO₂ emissions coming from raw material extraction as well as electricity generation of grid-purchased-electricity are excluded in the CO₂ emission analysis, given that the system boundary of this study includes only onsite refinery operations. Because CC systems can be implemented for centralized steam systems and SMR systems, the CO₂ emissions associated with these two systems are shown as accumulated values, whereas the emissions associated with each unit process in Fig. 4 include only their fired heaters, which combust fossil fuels and fuel products. The top five GHG emissions sources in U.S. refineries are, in descending order: CCU, SMR, steam systems, ADU, and CRU. The CCU generates the most CO₂ emissions due to its high energy consumption and usage of catalyst coke, a fuel with a high CO₂ emission factor.

As discussed in the previous section, two application scenarios were developed in this study. The cost versus CO₂ emission reduction potential of CC technology in the two scenarios are shown in Fig. 5. Because the costs of CC systems applied to power plants have large variances as shown in Table 2, the estimated costs of CC technology applied to refinery processes are shown in high and low cases based on the minimum and maximum cost data collected from the literature. The costs of distributed post-combustion CC systems applied to utility steam systems, SMR systems, and different unit processes are shown explicitly as a black step-wise line in Fig. 5; the dashed green line represents the total average post-combustion CC cost. The red line in Fig. 5 represents the estimated cost of the pre-combustion scenario. As mentioned in the previous section, in the pre-combustion scenario, centralized CC systems are constructed to de-carbonize all fuels used in the refinery unit processes except the CCU that is more suitable for oxyfuel-combustion technology. Therefore, only two costs are shown in the pre-combustion scenario; one is the cost of centralized pre-combustion systems, the other is the cost of oxyfuel-combustion systems applied to the CCU. The gap between the CO₂ reduction potential of two scenarios is highlighted by the dashed blue line.

There are several major findings from these results. First, a large fraction of the current CO₂ emissions from the U.S. refining industry could be reduced by applying CC technology (110 – 126×10^6 t/year,

accounting for 62%–71% of total CO₂ emissions). Second, the pre-combustion scenario has a smaller CO₂ reduction potential compared to the post-combustion scenario, but it is likely to have a lower CC cost (the average CC cost of post-combustion and pre-combustion scenarios are 62–128 \$/t CO₂ captured, and 52–127 \$/t CO₂ captured, respectively).

The large ranges of CC costs are mainly attributed to the large differences in costs and system designs in underlying studies used in this study (shown in Table 2). However, some observations here can still provide valuable indications. First, the slightly lower cost of pre-combustion scenario is primarily attributed to the centralized system design that can take advantage of the economy of scale. Second, the lower CO₂ reduction potential of pre-combustion systems (highlighted as the gap between two scenarios in Fig. 5) is mainly caused by more fuels used by the pre-combustion system (see data shown in Table 2). In this analysis, CO₂ emissions from the energy penalty are not captured, the differences between two scenarios might be smaller if such emissions are captured or the energy penalty of pre-combustion systems is reduced by more advanced technologies (IEA, 2016). Another solution to reducing CO₂ emissions from energy used for CC system is to utilize excess heat in the refinery (Johansson et al., 2013). Both measures could increase the costs and should be analyzed on a case-by-case basis. Despite these

Fig. 4. On-site CO₂ emission breakdown of U.S. refineries by unit process and utility system.

Fig. 5. Cost of CC applied to utility systems and unit processes in U.S. refineries (2014\$).

inherent limitations, our findings provide an improved quantitative understanding of the CO₂ reduction potential of CC technology application in the U.S. refining industry, even though the variations and differences among CC systems in various studies are large. It can be further used by energy/environmental analysts, policymakers, as well as the refining industry, for strategic planning of industry-wide reduction of CO₂ emissions.

Another critical observation is that in the post-combustion scenario, the costs and CO₂ mitigation potential of CC technology applied to different unit processes and utility systems have large variations. Among all unit processes and utility systems, applying CC to the CCU and utility systems (steam systems and SMR) is likely to be the most effective regarding both cost and CO₂ emission reduction potential. On the contrary, installing post-combustion for smaller CO₂ emitting unit processes such as the ISU, KTU, and DTU, might be too expensive given their small CO₂ mitigation potentials. Although the conclusion that large sources will be economical and small sources will be uneconomical is intuitively obvious because CC technology costs need economies of scale to be acceptable, the questions of “how small is too small for CC adoption” or “how large is suitable for a CC adoption” did not have clear answers prior to this study. Therefore, the results shown in this case study might be used by refinery engineers, project managers, and energy and environmental analysts to screen and rank

suitable unit processes for CC adoption, especially to eliminate small emission sources with high CC costs, based on different budgets and goals of capturing CO₂ emissions.

4. Discussion

4.1. Limitations of the study

There are some limitations of this study. First, this study focuses on industrial tested applications, and the integration with emerging technologies (e.g., more advanced solvents) is not taken into consideration. Some studies indicated that integrating with other emerging technology such as selective carbon membrane (a type of carbon membrane with average pore size of about 5 Å) may reduce CC costs (Kuramochi et al., 2012). The results shown in this work can be used as the reference cases for future studies that aim at evaluating the potential of cost reduction by integrating CC technology with other emerging technologies. Second, in this study, we assume that the fuel penalty would be handled by the capacity expansion of refinery steam and CHP plants. If the refinery is unable to size the facilities to handle the energy demand of CC system, refineries might need to purchase energy either steam or electricity from the utility industry, in which case, those emissions would be outside of a refinery's control, and the cost would be different (likely

higher than the estimates in this study as in many cases when producing energy on-site is more economical than purchasing energy from the utility industry, but could be lower depending on utility industrial customer class tariffs). Lastly, we assume that the emissions from energy supply of CC systems are not captured (parasitic CO₂ emission) and the net CO₂ emission reduction would be less than 100%, an assumption common to most electric power sector CC scenarios.

As discussed in the previous section, the costs collected from literature have large variations and it is very difficult to break down the data to a detailed level so that specific sources of uncertainties can be identified (e.g., detailed equipment costs data and retrofitting costs). In this work, the implications of cost uncertainty from literature are analyzed by quantifying possible uncertainty ranges of CC costs and CO₂ reduction potential as shown in Fig. 5. Sensitivity analysis on specific process parameters that will have impacts on costs such as equipment and process design, construction activities, and availability of material supply can be performed as future work when more transparent data and literature of industrial-scale CC applications are publicly available.

Another limitation is that although we have included the uncertainty of all costs based on the data collected, there are some other uncertainties that are hard to be quantified and may have impacts on the analysis. For example, retooling processing units to add additional equipment might not be permissible in some refineries due to the tight spatial arrangement of existing process equipment (Berghout et al., 2013). Additionally, refurbishing process units require downtime posing an additional financial risk due to loss of production time and finished products. Assuming that these barriers are overcome, firing furnaces with oxygen-rich combustion air and/or hydrogen will require furnace overhauls with specific attention to tuning the furnaces to control against potentially higher temperatures than furnace ratings and/or product temperature set-point ranges. While higher temperatures can be controlled with cooling air or recycled flue gases, doing so requires new equipment with added space requirements, process controls, and overheat protocols. Higher temperature combustion could also require new and/or more exotic furnace materials to withstand higher temperatures – leading to potentially significant research and development efforts. In the post-combustion scenario, variability in furnace fuels could require specifically tuned and/or dynamic CO₂ scrubbers to capture CO₂ without fouling the absorption medium. The additional costs associated with those barriers and uncertainties are hard to be quantified in a general way as they may vary by plants; however, such costs should not be neglected when conducting the refinery-by-refinery analysis.

In addition, this study used a representative model for typical operations in U.S. refineries and average capacities of different unit processes were used as input data. The capacity of individual refinery varies and will have impacts on the CC costs due to the economies of scales. When using the results in this work for individual refineries, necessary adjustments should be made using Eq. (9) based on specific capacity and scale data.

4.2. Future directions

Lowering the cost of CC is the key to promote CC application in refineries. Compared with the 2014 carbon prices in different regions in the world (Table 2 in SI), even the CC cost shown in the low estimates is much higher than the majority of carbon prices in existing markets (lower than 35 \$/t) (Interagency Working Group on Social Cost of Carbon, 2013; World Bank Group, 2014). To create commercial markets for CC adoption in the U.S. refining industry, based on our results, a carbon price at least higher than \$50/t CO₂ would be needed, which is very challenging at current status. A possible solution suggested by previous CC studies (Blunt et al., 1993; IEA, 2015; Rubin et al., 2015) is that CO₂ captured can be sold for enhanced oil recovery (EOR). EOR is a technique that injects chemicals and gas into a reservoir to improve the

crude oil production (Lyons and Plisga, 2011). Through long-term geologic storage for EOR, approximately \$15–\$40/t CO₂ net credits can be potentially achieved (negative storage cost when the cost of CO₂ transmission and storage is taken into consideration) (Rubin et al., 2015). If such credits can be realized, it might lower the costs of CC systems in this analysis down to \$10/t CO₂, which is a much more reasonable level from an economic point of view. However, such credits are subject to many factors such as reservoir locations, oil price, and oil companies' willingness to pay. In addition, the transportation and distribution cost of CO₂ may compensate the EOR credits for refineries which do not have CO₂ transmission infrastructure or suitable storage locations nearby.

Other possible avenues have been proposed by previous studies to lower CC costs, such as using more advanced and cost-effective solvents (Wardhaugh and Cousins, 2017), integrating with emerging separation technologies like membrane (Kuramochi et al., 2012), using waste heat through heat integration (Andersson et al., 2013; Berghout et al., 2015), advanced process control (Mechleri et al., 2017), etc. However, most of the proposed measures are analyzed through simulation or lab-scale test, and many of them are designed for power plant applications. Intensive efforts are needed in the future to validate the performances of those measures by pilot tests or industrial demonstration in refineries.

To promote CC application in the refining industry, future research and development should focus on directions such as:

- Develop transparent and validated datasets for different CC systems and process configurations, identify the sources and impacts of the uncertainty of CC costs and techno-economic performance at facility levels
- Evaluate and compare the technical feasibility and economic effectiveness of integrating CC with process improvement measures available to petroleum refineries, such as mass and heat integration, combined heat and power systems, petroleum coke gasification, etc.
- Analyze the potential of CC with feedstock, product slate, and fuel switching in refineries, such as switching to lighter crude oil or bio-based, renewable feedstocks, product slates that require less-severe processing, and low-carbon in-plant fuel.
- Comprehensively assess infrastructure needs of different CC systems and evaluate the potential of using existing infrastructure or strategies of building new infrastructure in a cost-effective way
- Investigate technology developments in the refining sectors (e.g., catalyst development and advanced separation techniques such as PSA), and assess their impacts on CO₂ emissions and CC application.
- Assess space footprints for different CC systems and evaluate various process intensification strategies to reduce space requirement of CC as well as enhance their techno-economic performance.
- Evaluate the potential of using waste or low-grade heat within refineries and petrochemical plants for CC systems.

5. Conclusions

In summary, a bottom-up techno-economic model was developed to analyze the U.S. national CO₂ mitigation potentials and costs of three types of CC technologies applied to U.S. refineries in two different design scenarios – distributed design with post-combustion technology and centralized design with pre-combustion and oxyfuel-combustion technologies. This study provided quantitative answers to the three research questions discussed in the Introduction Section – (1) By how much could CO₂ emissions be reduced by applying CC to the entire U.S. refining industry? (2) How much do different CC systems cost and where are the associated uncertainties? (3) What are the future directions for promoting increased CC application moving forward? The results show that 62%–71% of CO₂ emissions from the U.S. refineries could be reduced by applying CC technology. The CO₂ reduction potential and costs of two CC scenario have large variations due to different system design and cost data collected from the literature.

However, even the lowest CC cost estimated in this work is much higher than the majority of carbon prices in existing systems. More efforts need to be made on research, technology development, infrastructure building, and effective policy support to make CC technology economically feasible to be applied in the refining industry. The results shown in this work provides quantitative and useful information for policymakers, oil companies, energy/environmental analysts to identify and develop suitable strategies for CC technology adoption as well as to reduce CO₂ emissions of the U.S. refining industry. Although this work focuses on the application of CC technology in the U.S. refining industry, the modeling approached presented in this work could be adapted and used to analyze the costs and CO₂ mitigation potentials of CC applications in other industrial sectors or other regions.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <https://doi.org/10.1016/j.ijggc.2018.04.020>.

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