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Reassessing the Efficiency Penalty from Carbon Capture in Coal-Fired Power Plants

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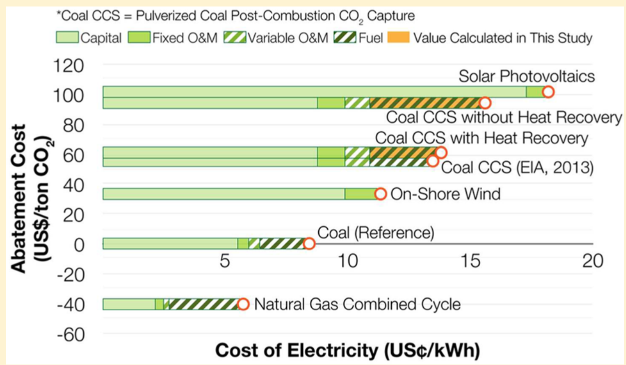
Supporting Information

ABSTRACT: This paper examines thermal efficiency penalties and greenhouse gas as well as other pollutant emissions associated with pulverized coal (PC) power plants equipped with postcombustion CO_2 capture for carbon sequestration. We find that, depending on the source of heat used to meet the steam requirements in the capture unit, retrofitting a PC power plant that maintains its gross power output (compared to a PC power plant without a capture unit) can cause a drop in plant thermal efficiency of 11.3−22.9%-points. This estimate for efficiency penalty is significantly higher than literature values and corresponds to an increase of about 5.3−7.7 US¢/kWh in the levelized cost of electricity (COE) over the 8.4 US¢/kWh COE value for PC plants without CO_2 capture. The results follow from the inclusion of mass and energy feedbacks in PC power plants with CO_2 capture into previous analyses, as well as including potential quality considerations for safe and reliable transportation and sequestration of CO_2. We conclude that PC power plants with CO_2 capture are likely to remain less competitive than natural gas combined cycle (without CO_2 capture) and on-shore wind power plants, both from a levelized and marginal COE point of view.



INTRODUCTION

Fossil fuel combustion in stationary boilers for generation of electricity and heat generation accounts for more than half of the world's annual carbon dioxide (CO_2) emissions.[1] More than 40% of these boilers are fired by coal,[2] which is the most carbon-intensive of all fossil fuels per unit of useful heat delivered, and over 65% of these coal-fired units are less than 30 years old.[3] With much service life left in them, these coal-fired units are poised to emit at least 263−351 gigatons of CO_2 by 2050 (see Supporting Information (SI) for calculation). Coal-fired boilers are likely to meet a growing demand for electricity in developing countries such as China and India. As a result, every major study on technology pathways for climate change mitigation[4−10] has recommended the retrofit of existing coal-fired plants with carbon capture and sequestration (CCS) units as a necessary option in curbing carbon dioxide (CO_2) emissions, with estimates for total installed coal CCS capacity as high as 800 GW by the year 2050.

Three main capture processes have been explored for CCS: postcombustion, precombustion, and oxyfuel CCS. Postcombustion capture involves recovery of CO_2 from flue gases arising from combustion of primary fuel in the presence of air. Precombustion capture involves recovery of CO_2 produced during the synthesis of syngas from the primary fuel using oxygen and steam. Oxyfuel capture involves recovery of CO_2 from flue gases from combustion of primary fuel in the presence of pure oxygen. Of these three main capture processes, precombustion and oxyfuel capture are still at the early technology development stage. As such, postcombustion capture remains the "most common and commercially mature" CCS technology option,[11] particularly given its suitability for retrofitting the existing global coal plant fleet, and its lower energy requirements relative to precombustion and oxyfuel CCS at present levels of technology advancement. Postcombustion capture is therefore the focus of this article.

The additional electricity and heat needed to operate the CCS unit in a power plant either reduces the rated power output of the plant or increases the amount of fuel consumed to produce the same electricity output as a plant without a CCS unit. This creates what is commonly termed as an "energy penalty" for the power plant, which is defined in the literature as the relative increase in energy input or the relative decrease in electric power output of a CCS-equipped power plant compared to a power plant without a CCS unit. Another commonly expressed form of energy penalty that captures potential changes to both the heat input and the power output from the addition of a CCS unit is the "efficiency penalty." Defined as the drop in the thermal efficiency (ratio of net electric energy output to total heat input) from the addition of a CCS unit, the efficiency penalty is used to represent energy penalty in this work.

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Table 1. Efficiency Penalties and CO_2 Capture Energy Requirements Reported in the Literature on Energy and Emissions Analyses of Pulverized Coal (PC) Plants with CCS

literature study	heat (kJ/kg CO_2)	electricity (kJ/kg CO_2)	efficiency penalty	source(s) of CCS energy use data
Booras and Smelser (1991)[13a]			12%-points[d]	pilot-scale plant
Suda et al. (1992)[14]	3768[d]	432[d]		pilot-scale plant
Sander and Mariz (1992)[15a]			12%-points[e]	Booras and Smelser (1991)[13]
Göttlicher and Pruschek (1997)[16b]	1224[e]	432[d]	8−11%-points for heat, 3.2−5.1%-points for compression[e]	about 300 studies surveyed including Booras and Smelser (1991),[13] not all sources cited
Rao and Rubin (2002)[17]	3775−4220[e]	329[e]	9.7%-points[d]	Suda et al. (1992),[14] Desideri and Paolucci (1999)[18]
IEA GHG (2004)[19]	3,456[d]		9.2%-points[d]	report submitted by Fluor Corporation
IPCC (2005)[5]	2700−3300[e]	460−510[e]	9.2%-points[e]	IEA GHG (2004)[19]
Khoo and Tan (2006)[20b]	1188−1224[e]	432[e]		Göttlicher and Pruschek (1997)[16]
Koornneef et al. (2008)[21]	4000−4320[e]	400[d]	11%-points[d]	Chapel et al. (1999),[22] Rao and Rubin (2002),[17] Alie et al. (2005),[23] Abu-Zahra et al. (2007)[24]
Odeh and Cockerill (2008)[25a]			9.6%-points[d]	Sander and Mariz (1992)[15]
Schreiber et al. (2009)[26a]			10.5%-points[e]	efficiency drop assumed based on "existing coal power plants or experts' expectations"
Pehnt and Henkel (2009)[27abc]	1368[e]	402[e]	18.2%-points[d]	Idrissova (2004)[28]
Singh et al. (2011)[29a]			10.2%-points[e]	IPCC (2005)[5]

[a]Specific values for capture heat and electricity requirements not reported. [b]Heat requirement expressed as loss in turbine power output per kg CO_2. [c]Energy input includes average grid mix electricity use for recompression and storage of CO_2. [d]Reported value calculated in study. [e]Reported value assumed from literature.

In addition to increased fuel use per unit of electric output from a CCS-equipped power plant, emission of chemical solvents such as monoethanolamine (MEA) used for separation of CO_2 from the flue gas creates concerns with increased toxicity, and the degradation products of these solvents such as ammonia and aldehydes also amount to increased acidification and smog formation, respectively. It is thus important to account for all significant energy and material flows in CCS-coupled power plants to evaluate their environmental and economic benefits and trade-offs.

To this end, several studies in the literature have performed energy and/or emissions analyses of coal power plants with CCS. Estimates for the efficiency penalty in these studies, which are summarized in Table 1, typically range between 8−16%-points for pulverized coal (PC) plants with postcombustion capture units. These estimates, which are summarized in Table 1, account for the energy required for separation of CO_2 from flue gases using chemical absorption with MEA, stripping of the absorbed CO_2, and compression of CO_2 to roughly 100−130 bar pressure. The additional cost of installing, operating, and maintaining the capture unit for a pulverized coal (PC) plant retrofit is estimated to add 2.6−5.1 US¢/kWh[12] of electricity generated based on a range of estimates and assumptions for efficiency penalty values and financial calculation parameters.

Further Investigation of Previously Reported Efficiency Penalty Estimates. A deeper investigation of the underlying data and assumptions of studies shown in Table 1 as well as several other reports[7,30] reveals that the efficiency penalty values are obtained from seminal studies from the early 1990s, which discuss the chemistry and thermodynamics of the MEA-based Econamine FG^{SM} postcombustion capture process developed by Fluor Corporation (Irving, TX, U.S.A.). The Econamine process studies themselves use data reported in an article by Booras and Smelser[13] on a pilot-scale Econamine process-based coal plant retrofit project commissioned by the Electric Power Research Institute (EPRI) and the International Energy Agency (IEA).

Although the pilot-scale power plant examined in the study by Booras and Smelser[13] maintained its gross power output of 554 MW, its net capacity was reduced (derated) from 513 to 336 MW after the CO_2 capture unit retrofit. The heat rate (ratio of heat input in Btu to power output in kWh) of the power plant was reported to increase from 9800 Btu/kWh to 15 000 Btu/kWh, which corresponded to an efficiency penalty of 12%-points. The pilot CCS plant did not use any recovered heat from the power plant or capture unit. Their study also noted that turbines designed for thermal power plants without capture were likely to meet only part of the total steam demand of power plants with capture units while operating within safe limits for turbine stresses. The pilot CCS power plant overcame this challenge by swapping out the old turbines with new ones with larger capacity. Finally, they mention that the drop in power output due to the addition of a capture unit will need to be compensated by a "replacement power source" to continue meeting consumer demand for electricity.

These outcomes hold significant implications for the studies listed in Table 1 that use the efficiency penalty value calculated by Booras and Smelser.[13] CCS retrofits where turbine replacement may be economically or operationally infeasible, a separate low-pressure boiler will be required to fully or partially meet the total steam demand of the CO_2 capture unit. The excess fuel combusted in the boiler(s) to meet the additional steam demand will lead to generation of more CO_2. If the power plant compensates for the lost power output by installing another turbine driven by steam from existing or additional coal-fired boiler(s), then this will also lead to generation of additional CO_2. Capture of this additional CO_2 generated will create a feedback in the mass and energy balance of the power plant. If the additional CO_2 is instead released into the atmosphere, then this should be allocated to the power plant itself, thus increasing the CO_2 emission intensity per unit

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of power output and decreasing the effective carbon capture efficiency of the power plant to a value less than the capture efficiency of the CO_2 separation and recovery process itself. When considering the effect of mass and energy feedbacks and the allocation of emissions from the replacement power source, the drop in plant thermal efficiency from the addition of a capture unit will be larger than the estimated values in the literature listed in Table 1. This paper quantifies the additional drop in thermal efficiency.

CCS and CO_2 Quality Concerns. Another factor that can increase the efficiency penalty for CCS-coupled power plants is the additional purification steps that may be needed to reduce the concentration of impurities in the recovered CO_2 to prevent accelerated corrosion of CO_2 transport infrastructure, to avoid human health and environmental risks in the event of an accidental release, and/or to ensure the long-term integrity of the sequestration sites. Raw (unpurified) CO_2 in postcombustion flue gases contains various amounts of water, air gases, carbon monoxide, sulfides, mercaptans, sulfur oxides, mercury, amines, nitrogen oxides, and volatile organic compounds (VOCs) such as xylene and benzene.[31,32] These impurities can cause problems ranging from toxicity to increased corrosion risk in pipelines. Table S2 lists some of the concerns that impurities present in CO_2 recovered from CCS plants pose. Although safe levels of impurities in CO_2 for CCS are undetermined at this point, it is likely that further treatment of recovered CO_2 beyond amine absorption and separation will be required for CCS. For instance, pretreatment of flue gases may be necessary for adequate removal rates of SO_2 beyond those achieved by installed emission control equipment if any, so as to minimize formation of heat-stable salts that lead to excess MEA consumption in the absorber, and to avoid excessive acidification of the brine present in the environment within which the CO_2 will be sequestered.[33] Additional purification steps such as desiccant drying for removal of moisture, and activated carbon treatment for removal of sulfur compounds and VOCs may also be required.

Much of the knowledge to date on allowable levels of impurities in recovered CO_2 for CCS is derived from practices used for enhanced oil recovery (EOR) and from laboratory and field studies on pipeline corrosion in the gas industry. On the basis of these studies, the European Enhanced Capture of CO_2 (ENCAP) project[34] and the U.S. National Energy Technology Laboratory (NETL)[35] have outlined general guidelines for maximum allowable levels of impurities in CO_2 recovered for sequestration projects. These values are also listed in Table S2.

The values for efficiency penalties listed in Table 1 do not take into account the influence of potential CO_2 quality requirements for pipeline and ship transportation of the captured CO_2, as noted by Zapp et al.[36] in their review of LCA studies on PC and NGCC plants with CCS. Further purification of CO_2 before transportation can significantly increase the energy input to the capture plant due to the energy requirements of unit operations and yield losses from additional treatment steps.[37]

In this work, we evaluate the efficiency penalty for pulverized coal (PC) plants coupled with postcombustion CCS units while incorporating the mass and energy feedbacks, and CO_2 quality factors, into the analysis. We begin by describing the system boundaries, and the mass- and energy-balance framework used for the analysis. Next we describe the separation and purification processes used for CO_2 recovery from PC plants and compare the environmental impacts of CCS-coupled power plants with their non-CCS counterparts. Quality-related factors are accounted for by building on well-understood processes used to recover high purity CO_2 from sources such as natural gas processing and ethanol plants for use in established markets such as EOR and the food and beverage industries. Finally, we discuss the results of the efficiency penalty and emissions analysis in the context of environmental trade-offs, CO_2 abatement costs for CCS systems at the current technology frontier, and design targets for future CO_2 separation technologies to make CCS competitive with other carbon abatement technology options.

■ MATERIALS AND METHODS

System Boundaries. Mass and energy balances are calculated assuming that the CCS-coupled plant continues to generate the same amount of electricity for sale as the plant without a CCS unit, and thus 1 MJ of electricity output is treated as the functional unit for comparing the efficiency penalty and emission values for a PC−CCS power plant relative to a PC power plant without CCS. Figure 1 shows the system boundaries and mass and energy balances used for calculation of the CO_2 recovery process emissions and energy use. The analysis focuses on air emissions from the combustion of bituminous coal for electric power generation. Nonfuel material and energy inputs to the power plant, as well as energy and emissions embodied in the power plant's construction and the capital equipment contained within it fall outside the scope of the analysis.

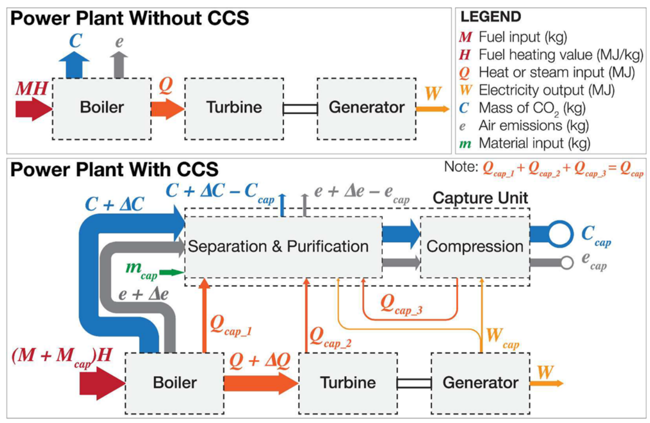

Figure 1. System boundaries, mass and energy balance used for the efficiency penalty and emissions analysis. The subscript "Cap" indicates association of quantity with capture unit, and Δ indicates the additional amount of the respective quantity required as a result of addition of the capture unit.

The recovery process begins with separation of CO_2 from flue gases using a chemical absorption/stripping processes enabled by an aqueous solution of MEA. It is assumed that an advanced flue gas desulfurization (FGD) unit and a selective catalytic reduction (SCR) unit are installed upstream of the CO_2 absorber. These unit operations are essential to prevent excessive consumption of the expensive MEA through reactions with SO_2 and NO_2 to form heat-stable salts during the chemical absorption process.[38] The expected heat rate for PC plants without CCS units was obtained from U.S. EPA's estimates[39,40] for new plants. These estimates include parasitic energy consumption from the operation of emission control equipment. The degradation of MEA into ammonia and aldehydes is

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Table 2. Scenarios for Process Conditions Examined for Their Influence on Energy Use and Emissions

scenario	description	$\zeta_{recovery}$	$\zeta_{turbine}$	ζ_{boiler}	α
Representative #1 (applicable to retrofits, and new plants)	turbine steam can partially supply capture heat; compressor heat recovery; nominal values for process parameters reported in literature	0.3[53]	0.4[13]	0.6[13]	0.30[52]
Representative #2 (applicable to retrofits)	steam extraction from turbine not feasible; boiler supplies steam for capture heat; no heat recovery; higher end of reported values for flue gas contaminant concentrations, MEA makeup, carbon and desiccant bed regeneration steam temperature and flow rate	0[13]	0[13]	1.0[13]	0.30[52]
Future (applicable to new plants in future)	heat recovery and advanced solvents significantly reduce capture heat requirements	0.37[54]	0.4[13]	0.6[13]	0.22[52]

modeled based on a detailed characterization of the process by Veltman et al.[41]

Raw CO_2 recovered from natural wells as well as other high purity sources such as natural gas processing and ethanol plants has significantly lower initial levels of impurities (>95% CO_2 v/v) than postcombustion sources (7−15% CO_2 v/v). Postcombustion CO_2 contains additional impurities such as HCl, HF, NO_x, halocarbons, vinyl chloride, and amines, which may not necessarily be present in CO_2 from other sources.[32] Without extensive studies analyzing the corrosion effects of each impurity and its interaction with other impurities at various pressures and temperatures, it is difficult to determine their safe levels in CO_2 for pipeline and ship transportation as well as permanent storage. This consideration is the motivation behind the recently initiated IMPACTS[42] research project, which proposes to study the impacts of potential CO_2 impurities on the fluid properties and chemical reactions on CCS infrastructure and storage sites.

Here we examine two purification configurations to assess the effect of CO_2 quality on overall energy use and emissions. Configuration A assumes the CO_2 purification process to comprise only of separation using MEA-based chemical absorption/stripping, while Configuration B, modeled based on commercial CO_2 purification literature, assumes further purification after the chemical absorption/stripping process with activated carbon filtration and desiccant drying. Final compression of the recovered CO_2 to 110 bar is achieved using a six-stage intercooled compressor. Table S3 provides details of the processes included in the two purification train configurations. Pollutant removal efficiencies of various emission control equipment for flue gas pretreatment and postrecovery purification are obtained from the literature on gas treatment and purification.[25,43−51] Figure S1 shows the block flow diagram of the CO_2 recovery process.

Mass and Energy Balance. Assuming H as the heating value of the fuel, M as the fuel input to the power plant without capture, and W as the total electricity output, the thermal efficiency of the power plant without CCS based on Figure 1 can be expressed as $\eta_{plant}^{nocap} = W/MH$. If M_{cap} is the additional fuel burnt to run the capture unit, then the thermal efficiency of the power plant with CCS can be expressed as $\eta_{plant}^{withcap} = W/(M + M_{cap})H$, since both plants are assumed to produce identical electric power for sale. The relative drop in power plant efficiency (ε_{CCS}) due the addition of a capture unit is then calculated using eq 1.

$$\varepsilon_{CCS} = \frac{(\eta_{plant}^{nocap} - \eta_{plant}^{withcap})}{\eta_{plant}^{nocap}} = \hat{c}\cdot\eta_{cap}\left\{\left(\frac{(1-\zeta_{recovery})\cdot\zeta_{boiler}}{\eta_{boiler}}\right)\cdot q_{cap} + \frac{(\alpha\cdot(1-\zeta_{recovery})\cdot(1-\zeta_{boiler}))\cdot q_{cap} + w_{cap}}{\eta_{plant}^{nocap}}\right\} \quad (1)$$

Here, $\hat{c}$ is the amount of CO_2 generated per unit of energy input to the power plant (kg CO_2/MJ input), and $q_{cap} = Q_{cap}/C_{cap}$ and w_{cap}/C_{cap} are the amount of heat and electricity inputs respectively to the capture unit per unit of CO_2 recovered (MJ/kg recovered CO_2). η_{cap} represents the plant's overall CO_2 capture efficiency, which is assumed as 90%.[13] $\zeta_{recovery}$ represents the fraction of the *total* heat requirement in the capture plant met by steam from heat recovery, and ζ_{boiler} represents the fraction of the *balance* heat requirement not met by heat recovery that is met by steam from the boiler. The remaining heat requirement ($\zeta_{turbine} = 1 - \zeta_{boiler}$) is met by extracting steam from the low-pressure (LP) turbine exit. The rate at which power output from the turbine drops per unit of heat withdrawn from the turbine for the capture plant is called the power equivalent factor, denoted by α, and its value for the different steam temperatures considered in the analysis is obtained from the literature.[52] η_{boiler} is the boiler efficiency, which is assumed as 90%.

Scenarios and Process Configurations. Three scenarios with different sets of process conditions were tested for each purification configuration to obtain a range of emissions and efficiency penalty values, and to assess their sensitivity to key process parameters and CO_2 quality. Table 2 lists the key characteristics of these scenarios and purification configurations, and Table S3 in the SI provides detailed values, assumptions, and sources used for the process parameters. All scenarios assume that an additional boiler will be needed to meet the steam demand not met by the turbine or recovered heat. The representative scenarios are particularly applicable to CCS retrofits.

While many CCS retrofit projects would likely utilize waste heat recovered from the interstage coolers of the multistage compressor, the upfront capital costs associated with heat recovery units and modification of piping may preclude waste heat utilization in some CCS retrofits. Heat recovery in the Representative Scenario #1 is modeled based on the detailed

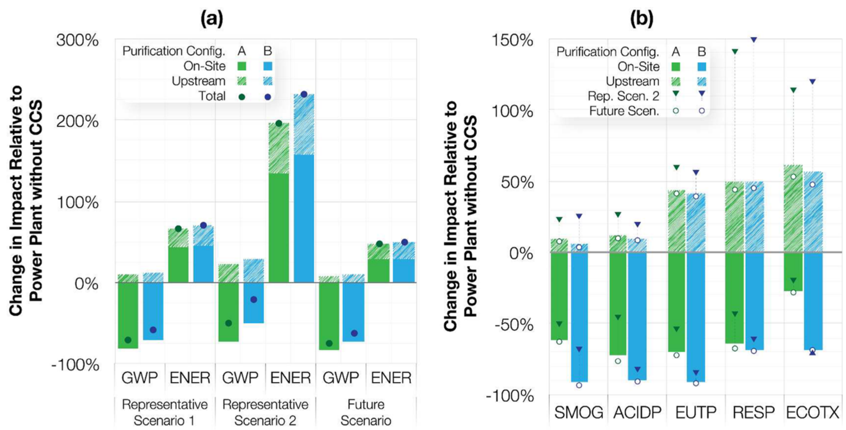

Figure 2. Environmental impacts of pulverized coal (PC) power plants with CCS unit expressed as the relative change in their value over PC power plants without CCS.

thermodynamic modeling of multistage pumps for CCS power plants presented by Alabdulkarem et al.,[53] and includes any parasitic energy consumption and losses in the ammonia refrigerant loop needed for recovering the heat. The resulting contribution of recovered heat to the total heat requirement of the CCS plant is 30%. Representative Scenario #2 assumes no heat recovery. The Future Scenario assumes both heat recovery and solvent technology advancements, which can reduce regeneration heat requirements to as low as 2600 kJ/kg CO_2.[54] This technology improvement for the Future Scenario is modeled as a 37% reduction in the total heat requirement of the CCS unit.

Emission inventories are characterized using the U.S. EPA's TRACI 2.0 method.[55] Impact categories chosen for analysis are global warming (GWP), smog formation (SMOG), acidification (ACIDP), eutrophication (EUTP), respiratory effects (RESP), and energy use (ENER). ENER includes energy content of the nonrenewable fossil fuel used in the power plant as well as embodied energy of upstream processes associated with fuel and other material supply chains that are within the scope of the analysis. Characterization factors for each pollutant examined are obtained from the Ecoinvent database.[56] The emissions inventory includes emissions from upstream processes such as extraction, purification and transportation of the fuels, as well as emissions from on-site processes such as combustion and capture.

RESULTS AND DISCUSSION

Efficiency Penalty and CO_2 Purity. Using the mass and energy balance illustrated in Figure 1, and the process flow and process operating conditions described in Figure S1 and Table S3, the emissions and energy use for producing 1 MJ of electric power output from a PC power plant with and without a CCS unit are presented in Figure 2. The range of efficiency penalty values for Representative Scenario # 1 and Representative Scenario # 2 was estimated as 11.3−21.4%-points for a PC−CCS plant operating with a Configuration A purification train, and 11.8−22.9%-points for operation of the PC−CCS plant with a Configuration B purification train. For the Future Scenario, the efficiency penalty value for purification Configurations A and B was estimated to be 9.3 and 9.6%-points, respectively.

Comparing the efficiency penalty of 21.4%-points in Representative Scenario #2 and Configuration A purification train to the literature values summarized in Table 1, we observe the significant influence of mass and energy feedbacks in CCS power plants. The amount of recovered heat used to meet the CO_2 stripper steam demand, or any corresponding reduction in the heat requirement of the process itself through advancements in technology is found to significantly reduce the efficiency penalty. In addition to reducing steam draw from the turbine, use of recovered heat in the capture plant leads to a compounding and nonlinear effect on reducing efficiency penalty by also reducing the amount of CO_2 generated for capture from combustion of additional fuel.

It is possible to evaluate the effect of CO_2 quality on energy use by comparing the range of efficiency penalty values for purification configurations A and B. The concentration of critical impurities from a pipeline corrosion and accidental exposure safety point of view will thus be closer to the lower bound of the range of recommended values in Table S2 with configuration B, while configuration A will likely yield impurity concentrations closer to the upper bound. With this approach, we find that the efficiency penalty increases by 0.5−1.5%-points for the higher purity configuration. The increase is due to two factors, the first of which is the additional heat and electricity requirements for desiccant and carbon bed regeneration and running additional pumps for scrubbing water. The second factor is yield losses in the purification train, which increase in the energy input and emissions per unit of final CO_2 recovered from processes upstream of them. For instance, about 8% of dry CO_2 product is vented to the atmosphere during desiccant bed regeneration.

The purification train considered in the Configuration B does not remove trace impurities such as O_2, N_2, Ar, H_2, and CH_4. These impurities have lower critical temperatures and pressures than CO_2, which lowers the density of the dense-phase CO_2, thereby increasing the risk of two-phase flows in pipelines and thus requiring higher pipeline operating pressures.[54] To limit their concentration to less than 4% v/v as advised by studies in the literature on CO_2 purification for CCS,[34,54,57] CO_2 would have to be selectively condensed from the mixture using a distillation step based on Supekar and Skerlos.[37] If this process is included as the final step of purification before CO_2 transportation, then the yield loss from refluxing pure CO_2 product into the distillation column can further increase the efficiency penalty in Representative Scenario #1 from 11.8%-points to 12.9%-points.

Comparing efficiency penalties for Configurations A and B across Representative Scenario #1, Representative Scenario #2,

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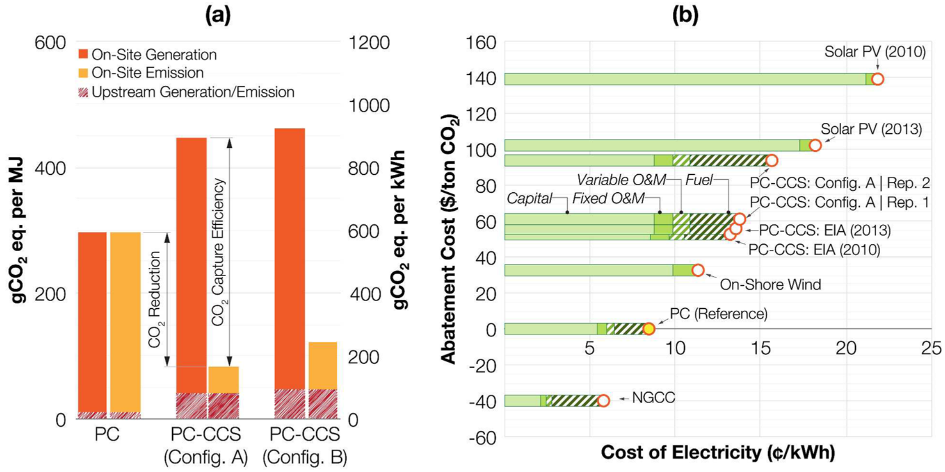

Figure 3. (a) Comparison of upstream and on-site greenhouse gas *generation* and *emission* from pulverized coal (PC) plants with and without CCS. (b) Cost of electricity for pulverized coal plants with CCS and other comparable energy options. Hatched bars represent marginal cost of electricity. Abatement costs are calculated using PC plants without CCS as reference, and do not consider carbon price or credits from SO_x and NO_x reduction.

and Future Scenario, we observe that not only does reduction in the total heat requirement of the capture unit have a strong and nonlinear effect on reducing the efficiency penalty, but it also greatly reduces the magnitude of additional penalty introduced by additional purification steps. Implementation of efficiency measures such as recovered heat usage in the capture plant can thus not only help plants reduce their fuel costs, but also largely insulate them from the incremental operational costs of meeting stricter CO_2 quality regulations that could be introduced down the line.

Another general point about CCS efficiency penalties that can be readily observed by examining eq 1 is that under identical heat, electricity, and nonfuel material inputs to a capture unit, plants using fuels such as natural gas that give a higher overall thermal efficiency ($\eta_{\text{plant}}^{\text{nocap}}$) and have a lower carbon intensity per unit heat input ($\hat{c}$) will have a lower efficiency penalty than plants using fuels such as coal. This also means that increase in plant thermal efficiency through heat recovery and other measures within the power plant itself can help reduce the efficiency penalty due to CCS. For instance, advanced ultrasupercritical PC plants today can have thermal efficiencies of up to 49%. Compared to the reference PC power plant with 37.2% thermal efficiency, the addition of a capture unit to a PC power plant with 49% thermal efficiency under process conditions from Representative Scenario #1 will lead to about a 13% lower value for the relative drop in efficiency given by eq 1.

Pollutant Control and Upstream Emissions. Increased fuel use affects upstream processes associated with fuel extraction, refining, and transportation. With efficiency penalty estimates higher than those previously used in the literature listed in Table 1, corresponding upstream emissions from increased fuel use will also be higher. When accounting for upstream emissions, we find that the net reduction in GHG emissions, assuming no accidental release of CO_2 during transportation and storage is about 51–72%. Purifying CO_2 to a higher quality using Configuration B train causes the net GHG reduction from capture to be about 20–59%. Comparing these estimates to reported values in the literature, we find that our highest estimate of 72% net reduction in CO_2 emissions is comparable to the lowest value of 70%[36] reported in the literature. Life cycle CO_2 reduction in the future scenario is estimated at 62–74% due to technology advancements and higher recovered heat utilization. Figure 3(a) shows the breakdown of CO_2 emission intensity of PC and PC–CCS plants, contrasting CO_2 generation and emission. It also shows that the capture efficiency of a CCS plant is considerably different from the total reduction in life cycle CO_2 emissions.

An often discussed potential benefit of postcombustion CCS plants is the added removal of particulate matter, sulfur oxides, and nitrogen oxides emitted to the atmosphere during the various recovery and purification steps in the capture plant. The resulting reduction in the on-site release of these pollutants can reduce local smog, acidification, eutrophication, and harmful respiratory effects. Given the spatial and temporal difference between on-site and upstream emission of these pollutants, the aggregation of on-site and upstream emissions does not lend itself to a meaningful interpretation. As such, we present these values separately as shown in Figure 2(b), and caution against the blind interpretation of combined on-site and upstream impacts.

We find that the addition of a capture unit can result in about 36–61% reduction in on-site smog emissions and 59–72% reduction in on-site emissions causing acidification. Ammonia emissions from the degradation of MEA and fugitive losses in the chiller unit offset some of the reduction in on-site eutrophication effects from the nearly 100% removal of NO_x. Similarly, any gains in ecotoxicity from removal of organic compounds, sulfides, and mercury during activated carbon treatment in Configuration B purification train are offset by the release of MEA (which has comparable toxicity to cyanide) and its degradation products. Atmospheric release of MEA remains a significant environmental trade-off with CCS in addition to increased nonrenewable energy use, causing a potential increase in excess of 150% in ecotoxicity effects depending on CO_2 quality and extent of turbine steam extraction for the capture plant.

To compare our estimates for impacts other than GWP to aggregated values for on-site and upstream emissions reported in the CCS LCA literature (compiled in Zapp et al.[36]), we present aggregated values for these impact categories in Table S7. It should be noted that existing CCS LCA studies do not consider heat recovery in their efficiency penalty calculations. The scenario to be used for comparison should thus be

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Representative Scenario #2. We observe a net reduction in smog and acidification effects irrespective of the purification train configuration in contrast to literature values that report a 25−100% increase in acidification and 5−100% increase in smog potential. The likely reason behind this is the higher removal rates for SO_x and NO_x assumed in this study to minimize loss of MEA. Aggregated eutrophication potential is found to increase, although the increase is lower than estimated values of 100−190% in most literature studies. We note that emerging solvents such as MDEA may have different acidification and other life cycle impacts, which should be considered in future studies.

Levelized Cost of Electricity and Carbon Abatement Cost. The pronounced effect of mass and energy feedback and CO_2 quality effects on efficiency penalty and fuel consumption has a significant effect on the levelized cost of electricity (COE) in USD/kWh and CO_2 abatement cost in USD/ton of CO_2 avoided, which are metrics used to compare the economics of various energy technologies. Numerous cost estimates for PC−CCS have been provided in the literature, and there is a fair amount of variability in these estimates based on their calculation method and assumptions for financial parameters and reference cases.[58] It should be noted that these cost estimates do not account for the feedback and quality issues presented in this work. Using capital and nonfuel operation and maintenance (O&M) cost estimates provided by the U.S. Energy Information Administration (EIA),[59] and efficiency penalties obtained in this work for fuel costs, the annual levelized COE for PC−CCS plants was calculated. Carbon abatement costs were calculated based on an equivalent annual generation basis using PC power plant without CCS as a reference. Figure 3(b) shows these costs, and Table S8 provides a detailed description of the assumptions for financial parameters and capacity factors used for the analysis. Costs and emissions associated with CO_2 transportation and storage were excluded to obtain a lower bound estimate for both COE and abatement cost.

The levelized COE for a PC−CCS plant is estimated to increase to about 13.8 US¢/kWh from about 8.4 US¢/kWh for its non-CCS counterpart. This is an increase of about 64%, which is similar to the 60% value reported by EIA in 2013,[59] and the 62% value reported by Finkenrath[12] in his revised estimates of CCS plant costs from 14 different studies published between 2007 and 2010. Without heat recovery, the levelized COE is estimated to be 15.6 US¢/kWh. Projections for capital costs of CCS units indicate a drop of 9−17% as technologies and their supply chains mature. Fuel costs on the other hand are unlikely to decrease based on EIA's projections.[60] As a result, fuel costs and factors such as efficiency penalty that influence them can have a significant effect over decisions to build or retrofit coal plants with CCS, particularly given that electricity dispatch is heavily influenced by the marginal cost of production. Marginal COE is estimated to increase from 2.5 US¢/kWh to 3.9−5.8 US¢/kWh.

Comparing COE values for PC−CCS with NGCC plants, the economic case for building new NGCC plants over PC−CCS plants is obvious. However, it is interesting to note that the on-shore wind power has a lower COE than PC−CCS on an equivalent generation basis. Anticipated reduction in the capital costs of renewables is likely to push this difference further in favor of wind power and make solar photovoltaics (PV) more competitive relative to CCS (see Figure 3(b)). The economic competitiveness of new and retrofit PC−CCS plants as a carbon abatement option thus heavily depends on whether advancements in separation technologies and efficiency measures can reduce the capital costs and energy requirements by a factor of 2 or more, and whether the price of natural gas returns to its preshale gas boom period.

Retrofit PC−CCS plants can also be operated at their derated capacity, as assumed by a few studies in the literature.[21,30] This would lead to a different mass and energy balance, which is beyond the scope of this work and should be part of future work on this subject. Coal plants constitute much of the base load electricity generation in most parts of the world, and thus large scale derating could lead to substantial electricity shortages unless it is supplemented by other energy sources capable of meeting base load generation. This would have a significant impact on the cost per MJ of energy delivered.

The use of nuclear power as a replacement energy source will require a huge and unprecedented expansion of this energy resource. Concerns surrounding operational safety in the wake of nuclear accidents, issues with safe disposal of spent fuel, high regulatory costs, and construction costs and lead times have precluded the expansion of nuclear plant fleets for several decades, with recent instances of countries such as Germany making policy changes to decommission their nuclear fleet. Using more coal to compensate for the derating will lead to significant efficiency penalties and costs as demonstrated in this work. Another alternative is to use natural gas given its higher thermal efficiency, lower carbon intensity per unit energy input, and lower prices, which will all contribute to lower efficiency penalties and operating costs. Replacement of retired base load coal-fired power plants with NGCC plants is already occurring in the United States. Countries such as India and China are likely to rely on coal as the replacement power source since their coal reserves are abundant, natural gas prices are high, and energy consumption rates are projected to grow significantly. The use of any fossil fuel to compensate for derating of fossil fuel-based plants will inevitably lead to generation of additional CO_2 per unit of energy output. Regardless of what the replacement energy source is, the issue of derating creates an interesting and important question about whether or not the marginal emissions from replacement sources should be allocated to the derated coal plant or the replacement source. Further research on this allocation question is warranted to inform policy measures related to CCS.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b03052.

> Detailed descriptions of the CO_2 recovery process, operating conditions, mass and energy balance expressions, and electricity and carbon abatement cost calculations (PDF)

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Notes

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