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Long-Term Energy and Climate Implications of Carbon Capture and Storage Deployment Strategies in the US Coal-Fired Electricity Fleet

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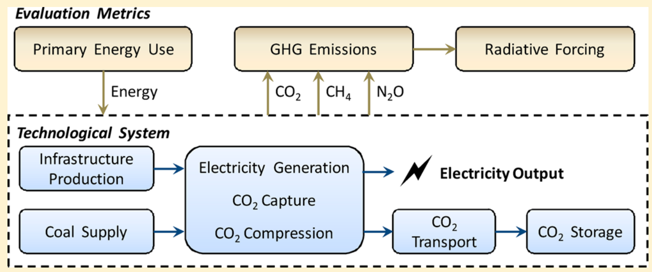
Supporting Information

ABSTRACT: To understand the long-term energy and climate implications of different implementation strategies for carbon capture and storage (CCS) in the US coal-fired electricity fleet, we integrate three analytical elements: scenario projection of energy supply systems, temporally explicit life cycle modeling, and time-dependent calculation of radiative forcing. Assuming continued large-scale use of coal for electricity generation, we find that aggressive implementation of CCS could reduce cumulative greenhouse gas emissions (CO_2, CH_4, and N_2O) from the US coal-fired power fleet through 2100 by 37−58%. Cumulative radiative forcing through 2100 would be reduced by only 24−46%, due to the front-loaded time profile of the emissions and the long atmospheric residence time of CO_2. The efficiency of energy conversion and carbon capture technologies strongly affects the amount of primary energy used but has little effect on greenhouse gas emissions or radiative forcing. Delaying implementation of CCS deployment significantly increases long-term radiative forcing. This study highlights the time-dynamic nature of potential climate benefits and energy costs of different CCS deployment pathways and identifies opportunities and constraints of successful CCS implementation.



INTRODUCTION

Despite significant increases in electricity generation using renewable energy, coal remains the largest single energy source for electricity production in the world. In 2009, coal fuel was used to produce 40% of global electricity[1] and 45% of electricity in the United States (US).[2] Because of the large geologic reserves of coal in many countries, the well established technologies for using coal for electricity generation, and the growing global demand for electricity, it is likely that coal will continue to be used for many years into the future. This raises concerns about climate destabilization due to high carbon dioxide (CO_2) emissions from coal combustion.

At least two means exist for reducing CO_2 emissions per unit of coal-fired electricity: increasing the efficiency of converting coal to electrical energy, and capturing and storing the CO_2 produced by coal combustion. The average conversion efficiency of the US coal-fired electricity fleet in 2010 was 32.8% (HHV basis; calculated by authors based on ref 2). Various technologies exist that could increase the efficiency of coal-fired power plants, such as supercritical or ultrasupercritical steam conditions in Rankine cycle plants, or coal gasification used in combined cycle plants.[3] These technologies would result in less coal being combusted, and correspondingly less CO_2 produced, to generate the same amount of electricity. Another option that is increasingly discussed is carbon capture and storage (CCS), which entails separating CO_2 from other gases and injecting the compressed CO_2 into underground geologic formations.[4] CCS requires energy; thus, more coal would be required to produce a kilowatt-hour (kWh) of electricity, but the net CO_2 emissions per kWh would be reduced.

Analyzing the effectiveness of greenhouse gas (GHG) emission reduction measures applied to the US coal-fired power fleet is challenging due to the dynamic nature of the energy system, which includes supply and demand technologies that evolve and expand over time. Analyses of individual power plants provide important technical information on elements of the supply side but fail to put that information in the overall context of the changing energy system.[5] By considering larger spatial and temporal scales, sectoral-level scenario analysis can provide important context for technological deployment. To date, however, scenario analyses of the coal-fired power sector have focused on near-term time horizons, e.g., through 2030[6,7] or through 2050.[8−11]

As the time horizon extends further, the analytical uncertainties grow but so do the opportunities to overcome barriers imposed by infrastructural and social inertia, thus increasing the range of potential options. Important temporal aspects of the coal-fired power fleet include the retirement dynamics of the existing fleet, the replacement and expansion of the fleet with new plants, changes in coal fuel supply,

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advancements in power plant and carbon capture technologies, and the adoption rate of CCS installations. For example, the expected life span of large coal-fired power plants in the US is on the order of 75 years. Most of the coal-fired plants in the existing US fleet were constructed in the 1960s, 1970s, and 1980s and will reach retirement age around the middle of the 21st century.[12] Results of CCS deployment scenarios through, for example, 2030 or 2050, therefore, are based on a limited range of options tied largely to existing power plants. A greater range of options exists over a longer time horizon, e.g., through 2100, during which time the existing power plants will be retired and possibly replaced by other technologies.

Another important reason to consider system dynamics over a longer time horizon is the time-dependent climate effects of GHGs. The temporal pattern of GHG emissions affects the resulting radiative forcing and hence the climate impact. A common method of analyzing mitigation options is the GHG balance approach, where all emissions that occur during a given time period are simply summed regardless of when they occur. A system with lower cumulative emissions at the end of the time period is considered to have less climate impact than a system with higher emissions. Non-CO_2 GHG emissions (e.g., CH_4 and N_2O) are typically converted to "CO_2 equivalents" (CO_2e) based on the global warming potentials (GWP) of the gases, which express the relative climate impact of the gases compared to an equal mass of CO_2 over a fixed time horizon of 20, 100, or 500 years.[13] This approach introduces inaccuracies, however, because the assumed GWP is static and does not fully take into account the temporal patterns of the GHG emissions and the resulting dynamics of radiative forcing. Within any finite time period, the climate impact depends not only on *how much* GHGs are emitted but *when* they are emitted. Cumulative radiative forcing (also called integrated radiative forcing or absolute global warming potential) is a metric that more accurately estimates the time-dependent climate impacts of dynamic systems.[14] The significance of GHG emission timing is receiving increasing attention in analyses of the mitigation effectiveness of biofuels (e.g., refs 15 and 16) but has heretofore not been addressed in coal-fired fleet deployment analyses.

Large-scale deployment of CCS will depend on many factors, of which some are CCS-specific such as cost and efficiency of current and future CCS technologies, and others are non-CCS-specific such as climate mitigation policy implementation and costs of other low-carbon energy technologies. The present analysis describes several plausible pathways for CCS deployment, acknowledging that long-term scenario projections are inherently uncertain and should not be considered as predictions of future events. Nevertheless, if key assumptions are made transparent and the significance of such assumptions are explored, long-term scenario analysis can contribute to informed policymaking by showing a range of possible futures and their drivers. In this study we create and analyze deployment scenarios for the US coal-fired power fleet extending to the year 2100. This time period includes the full life span of the existing fleet and demonstrates the climate significance of various turnover options. As metrics to compare the scenario options, we calculate the time profiles of primary energy use, emissions of CO_2, CH_4, and N_2O, and radiative forcing. We then calculate cumulative primary energy use, cumulative emissions of the three GHGs, and cumulative radiative forcing. On the basis of these metrics for each scenario option, we discuss appropriate long-term management strategies for the US coal-fired power fleet.

■ METHODS

To determine the energy and climate implications of various fleet deployment pathways, we integrate three analytical elements: scenario projection of energy supply systems, temporally explicit life cycle modeling, and time-dependent calculations of radiative forcing.

Scenario Projection. Key factors determining the future development trajectory of the US coal-fired power sector include potential improvements in generating technology, changes in coal quality, rate and scale of CCS deployment, efficiency of capture technologies, and construction of new power plants vs retrofitting existing plants for carbon capture. We quantitatively model these factors using three scenarios, each describing different deployment pathways of four successive generations of coal-fired power plants (see Table 1). The four generations of power plants (P1 to P4) are defined by their construction and retirement dates. Scenario 1 (No CCS) does not include carbon capture. Scenario 2 (New CCS) includes deployment of carbon capture technology in all new power plants built after 2020. Scenario 3 (retrofit + new CCS) includes carbon capture technology in new power plants built after 2020, plus gradual retrofitting of 75% of P2 generation power plants. Graphic representation of the US coal-fired fleet composition through 2100 under the three deployment scenarios is shown in Figure S1, Supporting Information.

Table 1. Summary Characteristics of Four Generations of Modeled Power Plants (P1−P4) and Their CO_2 Capture Ability in Three CCS Deployment Scenarios

	P1	P2	P3	P4
year of construction of power plants	before 2010	before 2010	2010−2040	after 2040
year of retirement of power plants	before 2040	after 2040	after 2040	after 2040
scenario characteristics				
scenario 1 (no CCS)	no CCS	no CCS	no CCS	no CCS
scenario 2 (new CCS)	no CCS	no CCS	CCS after 2020	CCS
scenario 3 (retrofit + new CCS)	no CCS	75% CCS	CCS[a]	CCS

[a]Carbon capture equipment is installed on all P3 plants built after 2020 and is retrofitted on plants built from 2010 to 2020.

To ensure a constant functional unit that allows objective comparison of the impacts across the three scenarios, the annual net generation of coal-fired electricity for each year is the same in all scenarios, rising from current levels to 2365 TWh/year in 2050, and then remaining constant through 2100. Scenario projections are inherently uncertain, and we do not consider the modeled generation profile to be a definitive forecast. Rather, we view it as one plausible future profile of coal-fired generation which allows us to compare various fleet deployment options for achieving a giving level of electricity generation. The derivation of the functional unit and the uncertainties associated with it are described in the Supporting Information.

Efficiencies of existing plants (P1 and P2 plants without CO_2 capture) are average values based on ref 12. To describe uncertainties in the level of future technological improvements, the efficiency of future power plant (P1 and P2 plants retrofitted with CO_2 capture, and all P3 and P4 plants) is considered under three assumptions of technological improve-

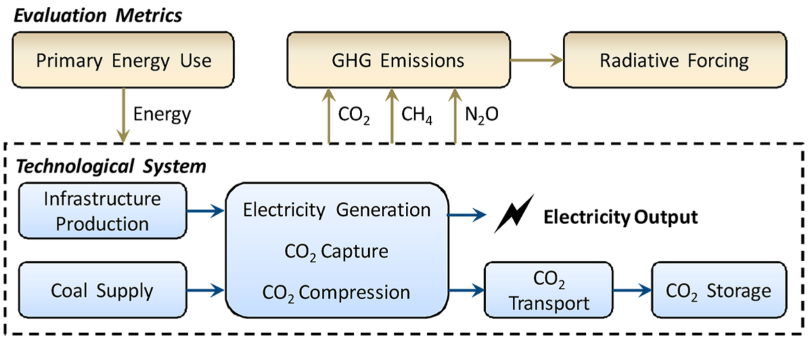

Figure 1. Schematic diagram of the modeled system.

ment: *low* which reflects modest continuing technology improvements, *medium* which we assume in our base case analysis, and *high* resulting from significant advances over current technological performance. Ranges of efficiency values for future plants are the authors' estimates based on a review of relevant literature,[8,9,17−27] with more commonly cited efficiencies becoming our medium values and more extreme values becoming our low and high values. The power plant energy conversion efficiency (HHV basis) without CO_2 capture ranges from 31% to 50%, the efficiency with CO_2 capture ranges from 16% to 44%, and the energy penalty of CO_2 capture (defined here as the percent decrease in electricity output per unit of fuel input) ranges from 12% to 48%. Full details on conversion efficiencies and energy penalties of each plant type are listed in Table S1, Supporting Information.

The power plant fleet dynamics through 2100 are based on the turnover rate of existing plants and the construction of new plants. Using data from ref 12, we created a database of all currently operating coal-fired power plants in the US with nameplate capacity greater than or equal to 15 MW. CO_2 capture is less economically feasible at plants smaller than this size, which make up about 0.1% of total installed capacity. Annual electricity generation of each plant is based on 2010 operation, which we assume remains constant over time. We grouped the existing fleet into 5-year cohorts based on the projected retirement date of each plant.[12] Summary statistics of each 5-year cohort are in Table S2 and Figure S2 of Supporting Information. We assume all new plants in P3 and P4 have a service life of 75 years. In scenarios with CCS, we assume carbon capture technology can be applied beginning in 2020, consistent with aspirational timetables described by IEA[21] and NETL.[26] Depending on the scenario, carbon capture equipment is installed when new plants are constructed or is installed during retrofitting of existing plants. We assume that newer plants are retrofitted before older plants, and that the plant service life is unchanged by retrofitting. We assume that P3 plants are built "retrofit-ready"[28] and have a total service life of 75 years including pre- and post-retrofit phases.

In a sensitivity analysis we vary four parameters. First, we vary the start date for CCS deployment from 2020 to 2030, to explore the impact of potential delays in technology development or policy implementation needed for large-scale CCS deployment. Second, we extend the service life of retrofitted plants an additional 20 years over their projected lifespans, which may occur due to general plant renovation done concurrently with retrofitting. Third, we include retrofitting of 100% of P1 and P2 plants (instead of no P1 plants and 75% of P2 plants), to explore the impact of aggressive retrofitting that manages to overcome limitations of space, water supply, flue gas cleaning capability, and other concerns that may constrain retrofitting.[19] Because the time of retrofitting coincides with the projected retirement time of P1 and older P2 plants, we also include a 20-year service life extension for all retrofitted plants. Fourth, we vary the retirement schedule of nonretrofitted P1 and P2 plants, from the Ventyx[12] projection to an accelerated retirement of all existing plants by 2050, to explore the impact of early retirement of nonretrofitted plants and faster turnover to more efficient plants with or without carbon capture. Fleet composition corresponding to these sensitivity analysis variations is shown in Figure S3, Supporting Information.

From 1969 through 2010 there was a significant shift in US coal supply from predominantly bituminous coal to a mix of bituminous and sub-bituminous coals.[2] AEO projections suggest that this trend will continue through 2035.[29] After 2035 we assume coal supply will stabilize at 33% bituminous and 67% sub-bituminous coal, based on the location, recoverability, and type of coal in the US demonstrated reserve base.[2] The historical and projected future breakdown of US coal supply by coal type is shown in Figure S4, Supporting Information. We include lignite, which comprises less than 7% of current US coal production,[2] in the sub-bituminous group. We include anthracite, which comprises less than 1% of current US coal production,[2] in the bituminous group. We neglect net import and export of coal (which averaged less that 3% of US consumption between 2000 and 2010[2]) and assume that US coal production equals US coal consumption.

Life Cycle Energy and GHG Emissions Modeling. We conduct temporally explicit modeling of primary energy use and GHG emissions of each scenario over a 90-year time horizon from 2010 to 2100, using 5-year time steps. We account for energy use and emissions from the construction of power plants and capture units, the mining and transport of coal fuel, the operation of power plants and CO_2 capture units, and the construction and operation of CO_2 transport and storage infrastructure. We account for emissions of three major GHGs: CO_2, CH_4, and N_2O. The modeling scope is depicted schematically in Figure 1, and the methods and assumptions associated with each modeling element are described below.

Energy use and GHG emissions from coal mining varies depending on whether surface or underground mining is used. We assume the current breakdown of surface and underground mining[30] continues through 2100 (see Table S3, Supporting Information). We estimate energy use and energy-related GHG emissions for surface and underground mining based on fuel use data from Jaramillo et al.,[31] updated with 2008 average emission factors for US electricity production and using default GHG emission factors from IPCC.[32] Specific energy use and GHG emission factors for coal mining from surface and underground mines are listed in Table S4, Supporting Information. To account for future increases in specific energy use as current mines become depleted and coal deposits that are deeper and more remote are exploited, we roughly assume the specific energy and emissions for coal mining will increase linearly from current levels to double current levels by 2100. Coal mine methane emissions vary widely by location and mine type (see Table S5, Supporting Information). We use mine methane data from Jaramillo et al.[31] because they represent national averages for US coal mines.

Projecting forward current trends,[33] we assume 85% of coal is transported 1600 km by train, 10% of coal is transported 480 km by barge, and 5% of coal is transported 25 km by truck, covering the total transport distance from mines to power plants. Mode-specific energy intensity factors for cargo transport[34] are used to estimate final energy use, and GHG emissions are estimated using default vehicle emission factors from IPCC.[32] Specific energy use and GHG emission factors for coal transport are listed in Table S3. Considering that transport efficiencies may improve in the future, but transport distances may increase as more remote mines are exploited, we roughly assume that specific energy and emissions for coal transport will remain constant at current levels.

Quantities of emitted and captured CO_2 from plant operation are calculated based on the conversion efficiency of the plants (Table S1), the carbon intensity of the coal (Table S3), and a 90% capture rate for flue gas CO_2 in plants equipped with carbon capture technology. Emissions of CH_4 and N_2O from plant operations are based on IPCC[32] default emission factors per unit of coal used for stationary combustion in the energy industries, with upper default values applied to the P1 generation, middle values applied to the P2 generation, and lower values applied to the P3 and P4 generations. We assume CO_2 capture equipment does not remove CH_4 and N_2O from the flue gas stream.

Life cycle modeling of the CO_2 capture process is based on current amine technology, acknowledging that other capture technologies may be used in the future. Emissions from the production of capture infrastructure and nonfuel consumables (including capture solvent) are minor compared to emissions from power plant stacks and from coal supply (Table S6, Supporting Information), suggesting that this uncertainty will have little impact on the results. Consumption rates of monoethanolamine (MEA) solvent for CO_2 capture, powdered limestone for flue gas desulfurization, and ammonia for selective catalytic reduction are based generally on IECM[35] then scaled proportional to coal consumption of different plants. Energy use and emissions from the production of MEA, limestone, and ammonia are based on GaBi 4 database.[36] Estimated emissions from power plant construction and retrofitting are based on NETL.[23,25] We disregard energy use and emissions from decommissioning of infrastructure. Primary energy use associated with petroleum and natural gas use is roughly estimated based on the higher heating value (HHV) of the end-use fuel plus 5% fuel cycle inputs.[37]

Transportation of CO_2 from the power plants to the sequestration sites is assumed to be by pipelines, which is more economical for large-scale CO_2 transport than alternatives such as truck, train, and ship. We model a simplified network of feeder pipelines delivering compressed CO_2 from the power plants to a series of trunk pipelines, which transport the CO_2 to the injection sites. We assume feeder pipelines are each 50 km long and trunk pipelines are each 200 km long. Diameters of pipelines vary depending on the volume of CO_2 transported by each pipeline, based on Kuby et al.[38] We assume a wall thickness of 18 mm for all pipelines.[39] Energy use and emissions from steel production are based on Worrell et al.[40] and from pipeline installation on NETL.[23] Power plant efficiencies shown in Table S1 account for the energy used for initial CO_2 compression. CO_2 pressure drop during transport is estimated based on Göttlicher,[17] and recompression to 15 MPa is carried out before sequestration. Energy use for recompression is based on Koornneef et al.,[41] and we calculate energy-related emissions based on current average US electricity production. Leakage of CO_2 from pipelines is based on IPCC[32] default emission factors for pipeline transport of CO_2.

Geological sequestration of CO_2 is assumed to occur via injection into saline aquifers at an average depth of 1200 m. Quantities of fuel, steel, and cement for injection well construction and operation are based on Singh et al.[42] Energy use and emissions from steel and cement production are based on Worrell et al.[40] In our main analysis we assume negligible leakage of CO_2 from geologic reservoirs, though in a sensitivity analysis we consider leakage at rates of 0.1%/year and 1.0%/year based on van der Zwaan and Smekens [43] (see Supporting Information for discussion of methods and assumptions of leakage estimates). In all scenarios we assume that available space for long-term storage of CO_2 in geologic formations will not constrain CCS system deployment.

Radiative Forcing Calculation. On the basis of emissions profiles of CO_2, N_2O, and CH_4 from 2010 to 2100, we model the atmospheric dynamics of the emitted GHGs including atmospheric decay and radiative forcing patterns. We convert emissions occurring during each 5-year time step of life-cycle modeling to annual emissions using linear interpolation, and we treat each annual emission as a pulse emission. The atmospheric decay pattern of each pulse emission is then estimated using eqs S1, S2, and S3 in the Supporting Information.[44,45,13] The total atmospheric mass of each GHG for each year of the study period is then determined by summing the emissions occurring during that year plus the emissions of all previous years minus their decay during the intervening years.

The change in atmospheric mass of each GHG is then converted to change in atmospheric concentration, based on the molecular mass of each GHG, the molecular mass of air, and the total mass of the atmosphere estimated at 5.148×10^{21} g.[46] Annual changes in instantaneous radiative forcing due to the GHG concentration changes are estimated using eqs S4, S5, and S6 in the Supporting Information.[44,45,13] We then estimate the cumulative radiative forcing occurring each year in units of $W{\cdot}s/m^2$, by multiplying the instantaneous radiative forcing of each year by the number of seconds in a year. This operation converts the energy flow per unit of time of the radiative imbalance caused by GHGs into units of energy accumulated in the earth system per year. We use cumulative radiative forcing of each scenario as a proxy for surface temperature change and resulting disruption to physical, ecological, and social systems.[14] Uncertainties associated with the calculation of radiative forcing are discussed in the Supporting Information.

To allow comparison between the calculated values of radiative forcing and the commonly used indicator of cumulative GHG emissions, we also calculate cumulative emissions in units of CO_2 equivalent (CO_2e) using simplified GWP metrics to estimate the climate impact of CH_4 and N_2O

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emissions relative to CO_2 emissions. We use GWPs of 25 for CH_4 and 298 for N_2O, corresponding to a time horizon of 100 years.[13]

■ RESULTS

Annual primary energy use varies significantly over time, between scenarios, and because of differing efficiency improvements (Figure S5, Supporting Information). Cumulative primary energy use of the three deployment scenarios is shown in Figure 2, assuming low, medium (base case), or high efficiency improvements. Without CCS (scenario 1), a total of 2180 EJ of primary energy is used during the 90-year period under base case efficiency improvements. With CCS employed only on new power plants (scenario 2), 17% more primary energy is used. With CCS retrofitted to existing power plants and installed on new power plants (scenario 3), 28% more primary energy is used during the 90-year period. These estimates underscore a critical trade-off associated with CCS: as our ability to sequester CO_2 improves, our dependencies on energy and fuels to produce the same amount of electricity in a low-carbon fashion increase significantly. Compared to cumulative primary energy use in 2100 under base case efficiency improvements, low/high efficiency improvements lead to plus/minus 4% primary energy use for scenario 1, and plus/minus 10−16% primary energy use for scenarios 2 and 3. The larger variation in scenarios 2 and 3 is due to potential changes in efficiency in CO_2 capture technology, in addition to the efficiency changes in electrical generation technologies found in scenario 1.

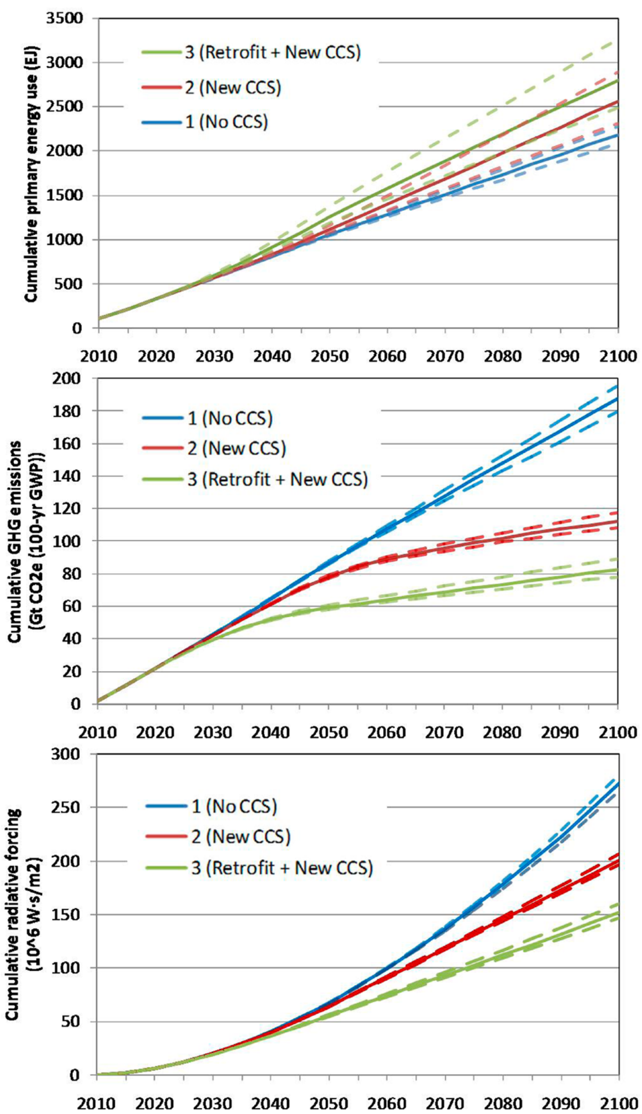

Figure 2. Cumulative primary energy use (EJ), cumulative GHG emissions (Gt CO_2e), and cumulative radiative forcing (10^6 W•s/m^2) of three deployment scenarios. The solid lines represent base case (medium) efficiency improvements, the upper dashed lines represent low efficiency improvements, and the lower dashed lines represent high efficiency improvements. Cumulative GHG emissions include CO_2, CH_4, and N_2O converted to units of CO_2 equivalent based on 100-year GWP.

Cumulative GHG emissions of three deployment scenarios are shown in Figure 2, assuming low, medium (base case), or high efficiency improvements. The GHGs include CO_2, CH_4, and N_2O and are converted to units of CO_2 equivalent based on 100-year GWP. Without CCS (scenario 1), cumulative emissions increase almost linearly from 2010 to 2100. With CCS applied to new power plants (scenario 2), the rate of increase slows in the 2040s and by 2100 the cumulative emissions are 40% less than without CCS. With CCS applied to retrofitted and new power plants (scenario 3), the rate of increase diverges earlier (in the 2030s) and by 2100 the cumulative emissions are 56% less than without CCS. Low/high efficiency improvements lead to about plus/minus 5% cumulative emissions for all three deployment scenarios. For scenario 1, this emissions variation corresponds closely to the variation in primary energy described above, because the emissions and energy use from coal combustion are directly related. For scenarios 2 and 3, the level of efficiency improvement is less significant to cumulative GHG emissions than to cumulative primary energy use because a large fraction of the coal emissions are captured and sequestered, thus energy use and emissions are not directly related. A breakdown of annual GHG emissions by source for the three scenarios is given in Table S6.

Cumulative radiative forcing of three deployment scenarios are shown in Figure 2, assuming low, medium (base case), or high efficiency improvements. In contrast to the linear or decreasing rates of emissions accumulation, the accumulation of radiative forcing begins slowly and proceeds at an increasing rate. This is due to the residence time of GHGs in the atmosphere, leading to their continued radiative forcing for many years after their emission. The differences in cumulative radiative forcing between scenarios with and without CCS are smaller than the differences in cumulative emissions: by 2100, the cumulative radiative forcings of scenarios 2 and 3 are 26% less and 44% less, respectively, than that of scenario 1. Efficiency improvements have little effect on cumulative radiative forcing through 2100, and changing from base case efficiency improvements to low/high efficiency improvements results in only plus/minus 3% cumulative radiative forcing in 2100 for all three deployment scenarios.

The different time dynamics of emissions and radiative forcing have relevance to climate change mitigation policy, which generally seeks to reduce climate impacts within a finite time period. Through 2050, scenario 3 (retrofit + new CCS) gives a 32% reduction in cumulative emissions and 18% reduction in cumulative radiative forcing compared to scenario 1, while scenario 2 (New CCS) gives only 10% and 5% reductions. By 2100, scenario 3 gives a 56% reduction in cumulative emissions and 44% reduction in cumulative radiative forcing, while scenario 2 gives 40% and 26% reductions (see Figure S6, Supporting Information). The emissions rates of scenarios 2 and 3 are greatest during the early part of the study period and decrease over time as CCS is implemented. The emission rate of scenario 1 (no CCS) is roughly constant over time due to continued emissions of GHGs. Radiative forcing, on the other hand, begins to rise at a very low rate corresponding to the atmospheric concentration of GHGs emitted after 2010 but rises at an increasing rate over time as the amount of GHGs in the atmosphere grows. The trajectories over time of cumulative emissions and cumulative radiative forcing are compared graphically in Figure S7, Supporting Information. These results show the importance of early and sustained reductions in GHG emissions for long-term control of radiative forcing.

CO_2 is the dominant contributor to radiative forcing, producing about 94% to 97% of the total radiative forcing. CH_4 contributes 3% to 5% of the total, and N_2O contributes about 1%. As more CO_2 is captured (scenarios 2 and 3), the relative contribution of CO_2 decreases and that of CH_4 and N_2O increases. This is due not only to the reduced emissions of CO_2 because it is captured and sequestered but is also due to increased emissions of CH_4 and N_2O from the mining and transport of greater quantities of coal to fuel the CCS systems. Nevertheless, CO_2 remains the predominant GHG in all scenarios. Details on the contributions of CO_2, CH_4, and N_2O to cumulative radiative forcing in 2100 are shown in Table S7, Supporting Information.

The estimated energy costs of emissions reduction and radiative forcing reduction, defined as the additional cumulative primary energy needed system-wide to reduce a unit of cumulative GHG emissions and cumulative radiative forcing through 2100, are shown in Table 2. Per unit of reduced emissions, more energy is needed in scenario 3 (retrofit + new CCS) than in scenario 2 (new CCS). This is due to the lower efficiencies of older, retrofitted plants relative to newer plants, requiring more energy to capture a ton of CO_2. However, per unit of reduced radiative forcing, scenario 3 uses less energy than scenario 2. This is because the avoided emissions from retrofitting CCS occur earlier than the avoided emissions from new CCS and thus contribute more toward reducing cumulative radiative forcing through 2100. As efficiencies of energy conversion and carbon capture increase from low to medium to high, the energy costs decrease significantly for both new and retrofitted CCS.

Table 2. Amount of Additional Primary Energy (EJ) Needed to Reduce GHG Emissions (Gt CO_2e) and Radiative Forcing (10^6 W·s/m^2) by Implementing CCS in New Plants and Retrofitted Plants through 2100, Assuming Low, Medium (base case), or High Efficiency Improvements

scenario	energy cost of emission reduction (EJ/Gt CO_2e)	energy cost of radiative forcing reduction (EJ/MW·s/m^2)
Low Efficiency Improvements		
2: new CCS	7.9	8.3
3: retrofit + new CCS	9.3	8.1
Medium Efficiency Improvements (base case)		
2: new CCS	5.0	5.2
3: retrofit + new CCS	5.9	5.1
High Efficiency Improvements		
2: new CCS	3.2	3.3
3: retrofit + new CCS	4.0	3.4

A sensitivity analysis of parameters describing fleet dynamics results in complex patterns, because changes made early in the time period continue to affect the system later because of changes in timing and type of replacement technologies (see Table 3). If CCS implementation begins in 2030 instead of 2020, the cumulative emissions of scenarios 2 and 3 increase by 8% and 12%, respectively, while cumulative radiative forcing increase by 5% and 13%. For scenario 2, the main effect of delayed CCS implementation is a cohort of P3 plants that go through their service life without capturing CO_2. For scenario 3, the effect of delaying CCS is more significant because plants of the large P2 generation continue to emit CO_2 for 10 additional years. Extending the service life of retrofitted plants an additional 20 years over their projected lifespans results in minor increases in energy use, emissions, and radiative forcing, due to slower turnover from retrofitted P2 plants to more efficient P4 plants. If it were possible to overcome constraints and retrofit 100% of P1 and P2 plants with 20-year extensions to their service lives, both emissions and radiative forcing would increase slightly. The early emission reductions gained by complete retrofitting are compensated by extended service lives and slower turnover to more efficient P3 and P4 plants. Early retirement of existing plants in scenario 1 (No CCS) gives minor reductions in energy use, emissions, and radiative forcing, due to quicker turnover to more efficient plants. Early retirement of existing plants in scenario 2 (new CCS) reduces emissions by 7% and radiative forcing by 11%, due to the earlier turnover from P2 plants to CCS-equipped P3 plants. Geologic CO_2 leakage significantly increases emissions and radiative forcing at leakage rates of 1% per year, but much less significantly at 0.1% per year, supporting the results of van der Zwaan and Smekens.[43]

■ DISCUSSION

While future events cannot be predicted, we can gain insight into the future by considering possible scenarios and analyzing their drivers and potential consequences. In this study we have integrated scenario projections, life cycle modeling, and radiative forcing calculation to better understand the implications of future CCS deployment options within the US coal-fired power fleet as it meets a predefined electricity demand trajectory. We find that widespread implementation of CCS can significantly reduce GHG emissions and radiative forcing but would require substantial amounts of additional primary energy.

The degree of future efficiency advances in energy conversion and carbon capture technologies is uncertain; thus, we have analyzed deployment scenarios under low, medium (base case), and high levels of efficiency improvements. Changes in efficiency lead to substantial variation in annual and cumulative primary energy use. For example, cumulative energy use of scenario 2 (new CCS) under low efficiency improvements is greater than that of scenario 3 (retrofit + new CCS) under high efficiency improvements. However, changes in efficiency have much less impact on cumulative emissions and cumulative radiative forcing. This suggests that widespread and timely deployment of CCS technology is a primary prerequisite of climate change mitigation, while the efficiency of the technology is of

Table 3. Sensitivity Analysis of Key Parameters[a]

	primary energy (EJ)			emissions (Gt CO_2e)			radiative forcing (10^6 W·s/m^2)		
	scenario 1	scenario 2	scenario 3	scenario 1	scenario 2	scenario 3	scenario 1	scenario 2	scenario 3
base case	2177	2557	2797	187.5	112.1	82.7	273.2	200.9	152.0
CCS start 2030[b]	–	2504	2709	–	120.4	92.7	–	211.3	172.0
percent change	–	−2.1%	−3.1%	–	+7.5%	+12.1%	–	+5.2%	+13.1%
retrofit life extension[c]	–	–	2845	–	–	83.4	–	–	153.1
percent change	–	–	+1.7%	–	–	+0.9%	–	–	+0.7%
retrofit all plants[d]	–	–	2930	–	–	83.8	–	–	152.4
percent change	–	–	+4.8%	–	–	+1.4%	–	–	+0.3%
early retirement[e]	2171	2648	–	186.6	104.7	N/A	267.8	178.7	–
percent change	−0.3%	+3.6%	–	−0.5%	−6.6%	N/A	−2.0%	−11.1%	–
leakage 0.1%/year[f]	–	2557	2797	–	115.2	88.4	–	203.2	157.0
percent change	–	0.0%	0.0%	–	+2.8%	+6.9%	–	+1.2%	+3.3%
leakage 1.0%/year[f]	–	2557	2797	–	138.0	128.5	–	221.1	193.8
percent change	–	0.0%	0.0%	–	+23.1%	+55.5%	–	+10.1%	+27.5%

[a]The table shows values of cumulative primary energy use, cumulative GHG emissions, and cumulative radiative forcing for the year 2100. [b]CCS deployment begins in 2030 instead of 2020. [c]Service life of retrofitted plants is extended by 20 years beyond originally projected service life. [d]All P1 and P2 plants are retrofitted (instead of no P1 and 75% P2 plants), plus 20-year service life extension for retrofitted plants. [e]Nonretrofitted P1 and P2 plants are retired at an accelerated rate (see Figure S3). [f]Leakage of CO_2 from geologic storage; percent of total current stock per year.

secondary importance to climate mitigation goals. This confirms and extends the results of Bistline and Rai,[7] who found that the start time and diffusion rate of CCS deployment had greater impact on emissions through 2030 than did the heat rate and type of capture technology. Importantly, however, higher efficiency may improve the economic performance of the technology, which may better facilitate its widespread deployment.

This modeling exercise shows the value of a long-term perspective (e.g., through 2100) using appropriate metrics (i.e., radiative forcing) when considering the climate effects of changes to the energy system. Previous scenario studies of CCS implementation have extended to 2050, but Figure 2 shows that limiting the time horizon to 2050 considerably reduces the insight generated. A longer-term perspective is important to understand the slow rate of change due to the inertia of the physical infrastructure. Furthermore, while divergence of cumulative emissions between the three scenarios is evident by 2050, the cumulative radiative forcing shows little divergence between scenarios by 2050. By 2100, scenario 1 (No CCS) has clearly higher cumulative emissions than the two scenarios with CCS, while there is less differentiation in cumulative radiative forcing between the three scenarios. This is due to the long atmospheric residence time of CO_2 that gives long-term significance to emissions occurring early in the time horizon. This analysis could, of course, be extended further in time. A longer analysis, for example, to 2200, would further emphasize the divergence between scenarios but would introduce additional uncertainties about events in the more distant future. As the time horizon extends to infinity, differences between cumulative emissions and cumulative radiative forcing diminish.

Numerous factors may constrain eventual scale-up of CCS, which we acknowledge but do not explicitly model. These include economic costs, public acceptance, water availability, and suitable policy framework. Furthermore, large-scale CCS as modeled here will require a continuing supply of affordable coal fuel throughout the 21st century, which has conventionally been assumed to exist (e.g., see ref 47). Through 2100, scenario 1 (no CCS) requires an estimated 94 billion Mg of coal, or 40% of the estimated recoverable US coal reserves of 237 billion Mg.[2] Scenarios 2 (new CCS) and 3 (retrofit + new CCS) require 108 and 118 billion Mg of coal, respectively, or 46% and 50% of estimated recoverable US coal reserves. However, recent research has suggested that the rate of global coal production may be constrained earlier than previously expected (e.g., see ref 48), which could potentially limit the scope of CCS-equipped coal-fired power. Additional research on coal reserves and resources may reduce this uncertainty and provide a stronger basis for decision making regarding long-term shifts in energy supply systems.

This analysis shows that CCS is not a panacea for climate change concerns but if deployed early can reduce climate impacts compared to an alternative of continued large-scale coal use without CO_2 capture. This study also highlights the slow turnover rate of energy infrastructure and the long time period required to shift from widespread use of one technology type to another (e.g., the transition from P1 to P4 power plant generations that we model). The physical implementation of the scenarios developed here may reinforce a path dependency on coal-fired electricity that extends long after the end of these scenarios in 2100. CCS is considered attractive because it may allow society to continue to fulfill its demand for energy services while reducing the rate of CO_2 emissions and the potential for climate destabilization. However, long-term energy security depends on numerous factors besides fuel availability and climate concerns, such as efficiency, affordability, dependence, and sustainability.[49] As we contemplate and implement changes to today's energy system, it is important to consider long-term dynamics to ensure that actions made now will continue to benefit society well into the future.

ASSOCIATED CONTENT

Supporting Information

Seven tables, eight figures, and additional explanatory text provide further information on this analysis. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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Supporting Information

Environmental Science & Technology

Long-term Energy and Climate Implications of Carbon Capture and Storage Deployment in the US Coal-fired Electricity Fleet

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Table S1. Power plant energy conversion efficiencies (percent; HHV basis) of four generations of power plants (P1-P4), with and without carbon capture, fired with bituminous and sub-bituminous coal, under three assumptions of technological improvement. The values in bold type (P1 and P2 plants without carbon capture) are average efficiencies of existing plants. The remaining values are the authors' estimates based on a review of relevant literature (see main text). The energy penalties are calculated as the percent decrease in electricity output per unit of fuel input.

	Bituminous coal				Sub-bituminous coal			
	P1	P2	P3	P4	P1	P2	P3	P4
Low efficiency improvement								
No carbon capture	**32**	**35**	38	41	**31**	**34**	36	38
With carbon capture	17	20	24	30	16	19	22	27
Energy penalty of carbon capture	47	43	37	27	48	44	39	29
Medium efficiency improvement (base case)								
No carbon capture	**32**	**35**	39	44	**31**	**34**	38	42
With carbon capture	20	23	29	36	19	22	27	33
Energy penalty of carbon capture	38	34	26	18	39	35	29	21
High efficiency improvement								
No carbon capture	**32**	**35**	41	50	**31**	**34**	39	46
With carbon capture	22	25	33	44	21	24	31	40
Energy penalty of carbon capture	31	29	20	12	32	29	21	13

Table S2. Number of plants, and average plant efficiency, of 5-year cohorts of US coal-fired power plants projected to retire between 2010 and 2070+. Based on data from [12].

Year of retirement	Number of plants	Average plant efficiency (HHV basis)
2010-2014	64	25.9%
2015-2019	66	30.7%
2020-2024	40	29.5%
2025-2029	106	33.0%
2030-2034	150	34.0%
2035-2039	97	34.1%
2040-2044	111	34.6%
2045-2049	129	34.9%
2050-2054	145	34.3%
2055-2059	138	33.9%
2060-2064	43	34.1%
2065-2069	21	35.9%
2070+	51	35.1%

Table S3. Energy content, carbon intensity, and source of bituminous and sub-bituminous coals.

Type of coal	Higher Heating Value	Carbon intensity	Surface mined	Underground mined
	GJ/Mg coal	Mg CO_2/Mg coal	Percent	Percent
Bituminous	27.1	2.37	40%	60%
Sub-bituminous	19.4	1.77	98%	2%

Table S4. Specific energy use and GHG emission factors for coal mining from surface and underground mines, and coal transport by rail, barge, and truck.

Activity	Energy use	GHG emissions		
		CO_2	CH_4	N_2O
Coal mining [a]	Primary energy use (GJ/Mg coal)	Emissions (kg/Mg coal)		
Surface mining	0.85	40.0	0.00030	0.0019
Underground mining	0.22	10.2	0.000048	0.00030
Coal transport by mode	End-use energy use (kJ/Mg·km)	Emissions (g/Mg·km)		
Rail	240	17.2	0.00096	0.0066
Water	320	22.5	0.0021	0.00061
Truck	2310	163	0.0089	0.036
Total coal transport [b]	Primary energy use (GJ/Mg coal)	Emissions (kg/Mg coal)		
	0.37	24.7	0.0014	0.0091

[a] We assume the energy and emissions for coal mining will increase linearly from the levels shown here in 2010 to double these levels in 2100, to account for future increases in specific energy use as current mines become depleted and coal deposits that are deeper and more remote are exploited.

[b] Assuming 85% of coal is transported 1600 km by train, 10% of coal is transported 480 km by barge, and 5% of coal is transported 25 km by truck.

Table S5. Coal mine methane emissions estimated by previous studies.

Reference	Methane emission (kg CH_4 per Mg coal)	Description
[S1]	1.9	Illinois surface mine, 1996
	4.2	Illinois underground mine, 1996
[S2]	0.16	Minimum (typical emission from US surface mine)
	14	Maximum (western European mine)
	0.23	European surface lignite mine
[31][a]	0.72	Average of US surface mines, 1997
	6.0	Average of US underground mines, 1997
[S3][b]	1.4	Minimum (US underground mines)
	130	Maximum (US underground mines)
[23][b]	6.9	Illinois underground mine, 2002-2006

[a] Reported by Jaramillo et al. [31] in units of tons of CO_2 equivalents. Converted by authors to kg CH_4 based on 100-year GWP of methane.

[b] Reported by EPA [S3] and NETL [23] in units of cubic feet of methane. Converted by authors to kg CH_4 based on density of methane.

Table S6. Annual GHG emissions (million t CO_2e per year) from different sources from 2010 to 2100 under three deployment scenarios, with medium (base-case) efficiency improvements. Emission of CH_4 and N_2O are converted to CO_2e using 100-year GWP values.

Year	Plant stack emissions	Coal mining and transport	Plant infrastructure production	Non-fuel consumables production [a]	CO_2 transport and storage	Total emissions
Scenario 1 (No CCS)						
2010	1834	100	0.0	6	0	1940
2015	1889	104	0.6	6	0	2000
2020	1928	108	0.5	6	0	2042
2025	1968	112	0.4	6	0	2087
2030	2009	116	0.7	6	0	2132
2035	2062	121	1.4	7	0	2191
2040	2070	124	0.9	7	0	2201
2045	2074	126	1.8	7	0	2208
2050	2045	126	3.1	6	0	2180
2055	1978	124	2.5	6	0	2111
2060	1908	122	2.6	6	0	2038
2065	1881	122	1.0	6	0	2010
2070	1872	123	0.3	6	0	2002
2075	1867	125	0.2	6	0	1998
2080	1866	127	0.1	6	0	1998
2085	1857	128	0.3	6	0	1991
2090	1846	129	0.7	6	0	1982
2095	1840	130	0.5	6	0	1977
2100	1835	132	0.4	6	0	1973
Scenario 2 (New CCS)						
2010	1834	100	0.0	6	0	1940
2015	1889	104	0.6	6	0	2000
2020	1928	108	0.5	6	0	2042
2025	1924	113	0.9	7	3	2048
2030	1889	119	1.4	8	3	2021
2035	1793	128	2.8	11	5	1940
2040	1707	133	1.8	12	6	1860
2045	1540	139	3.5	14	8	1705
2050	1223	145	6.0	18	11	1403
2055	920	147	4.9	21	13	1107
2060	603	150	5.1	24	14	797
2065	483	152	2.0	25	15	677
2070	441	155	0.7	26	13	634
2075	419	157	0.3	26	12	615
2080	413	159	0.1	26	12	610
2085	372	162	0.7	27	13	574
2090	295	165	1.4	27	14	502
2095	240	168	1.0	28	14	450
2100	239	169	0.8	28	13	449
Scenario 3 (Retrofit + New CCS)						
2010	1834	100	0.0	6	0	1940
2015	1889	104	0.6	6	0	2000
2020	1928	108	0.5	6	0	2042
2025	1615	121	5.4	12	14	1768
2030	1387	133	3.8	16	11	1551
2035	1098	148	5.2	22	15	1289
2040	818	160	4.1	26	18	1027
2045	456	171	5.9	32	23	688

2050	288	173	5.1	33	16	515
2055	275	169	3.2	32	15	493
2060	261	164	3.3	30	14	473
2065	256	163	1.3	30	14	464
2070	254	165	0.4	30	14	463
2075	253	167	0.2	30	14	464
2080	253	169	0.1	30	14	465
2085	251	171	0.4	29	13	465
2090	249	171	1.3	29	13	464
2095	247	173	1.0	29	13	463
2100	244	173	2.0	29	13	461

[a] Non-fuel consumables include MEA, limestone, and ammonia.

Table S7. Percent contribution of CO_2, CH_4, and N_2O to cumulative radiative forcing in 2100, under three deployment scenarios with low, medium (base case), or high efficiency improvements.

	CO_2	CH_4	N_20
1: No CCS			
Low efficiency improvements	96.8%	2.6%	0.6%
Medium efficiency improvements	96.9%	2.6%	0.6%
High efficiency improvements	96.9%	2.5%	0.6%
2: New CCS			
Low efficiency improvements	94.8%	4.3%	0.9%
Medium efficiency improvements	95.2%	4.0%	0.8%
High efficiency improvements	95.4%	3.7%	0.8%
3: Retrofit + New CCS			
Low efficiency improvements	93.5%	5.4%	1.0%
Medium efficiency improvements	94.0%	5.0%	1.0%
High efficiency improvements	94.4%	4.6%	1.0%

Figure S1. Composition of US coal-fired generating fleet through 2100 under three deployment scenarios.

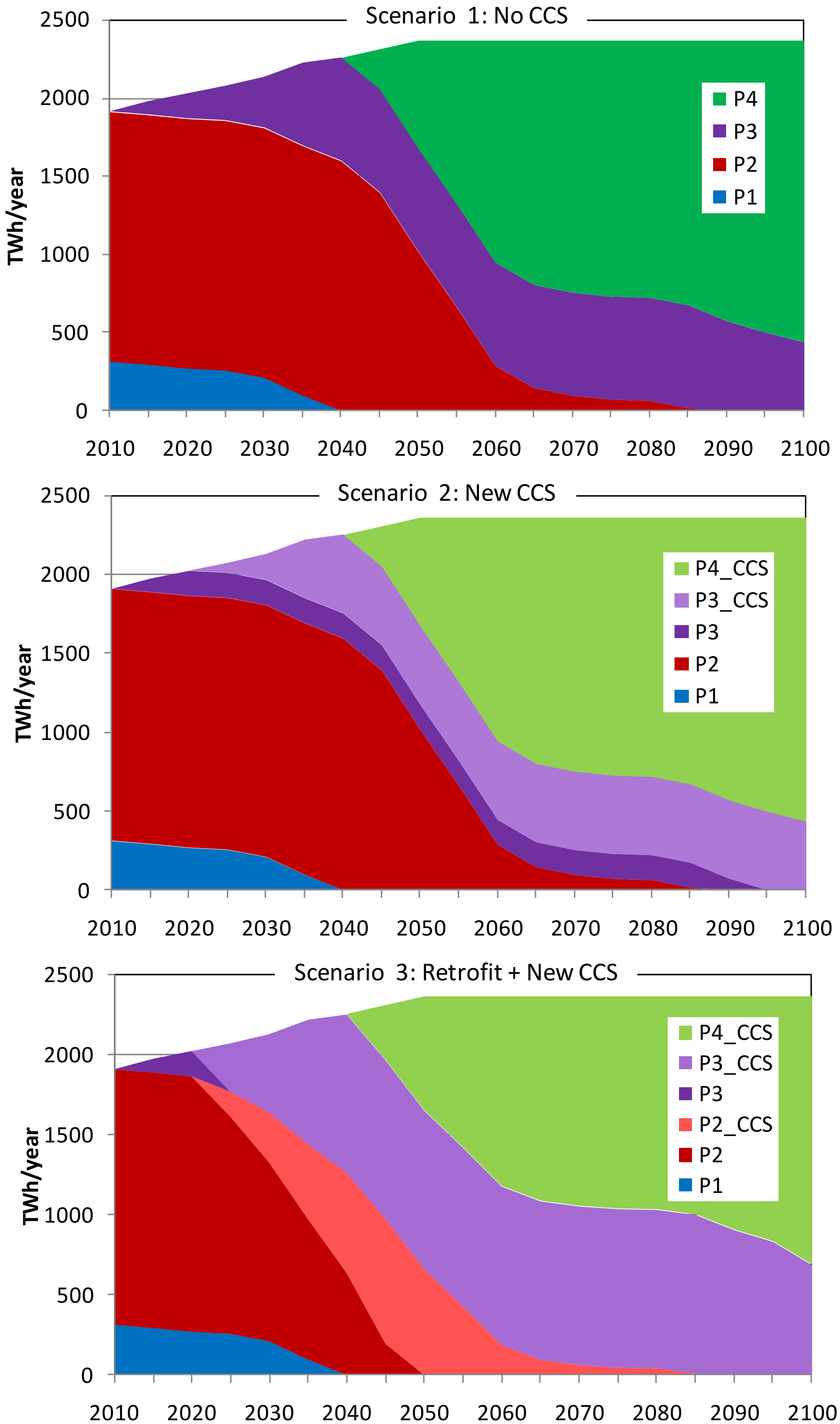


Figure S2. Annual electricity generation in 2009 of 5-year cohorts of US coal-fired power plants projected to retire between 2010 and 2070+, broken down by type of coal fuel. Based on data from [12].

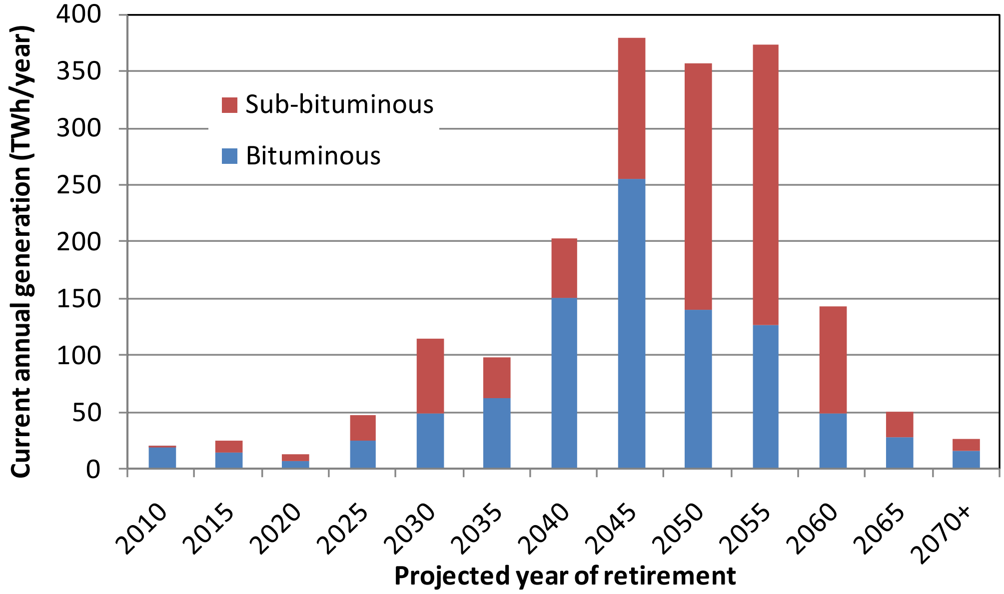


Figure S3. Composition of US coal-fired generating fleet through 2100 under sensitivity analysis conditions.

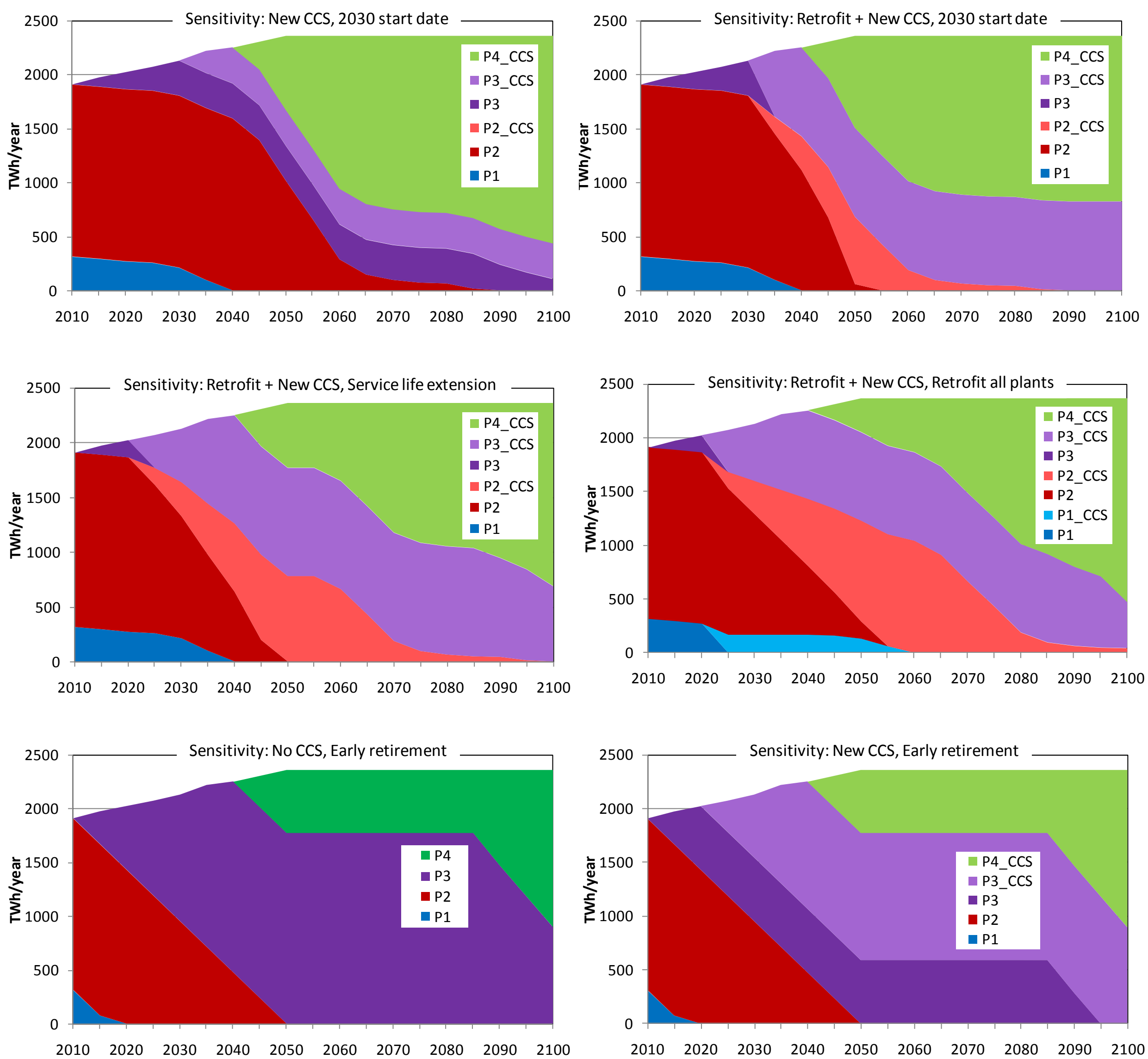


Figure S4. Historical and projected breakdown of US coal supply (by weight) into sub-bituminous rank (including lignite) and bituminous rank (including anthracite).

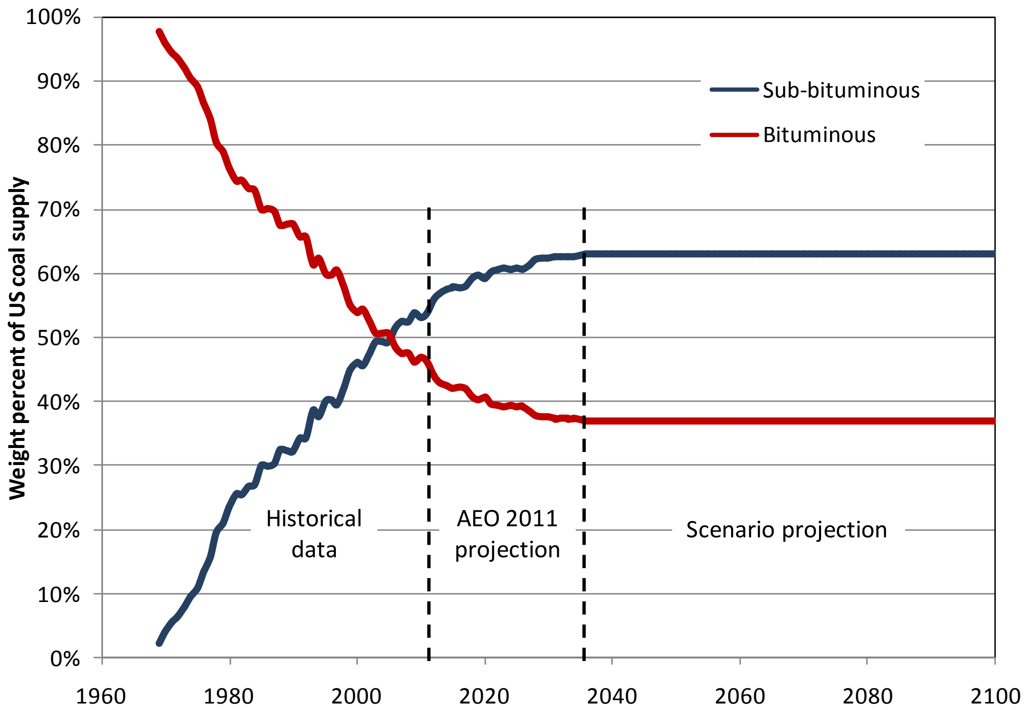
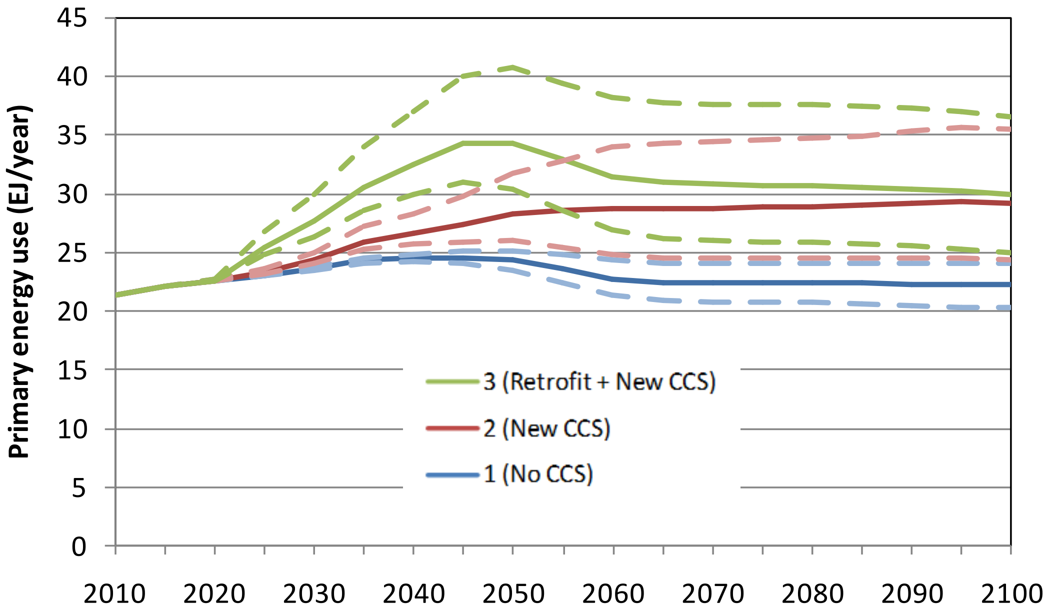


Figure S5. Annual primary energy use (EJ/yr) of three deployment scenarios. The solid lines represent base case (medium) efficiency improvements, the upper dashed lines represent low efficiency improvements, and the lower dashed lines represent high efficiency improvements.



Primary energy use in scenario 1 (No CCS) remains fairly constant, ranging between 22 and 25 EJ/yr under base case efficiency improvements. The increasing conversion efficiency of successive generations of power plants compensates for the increased electricity output through 2050, resulting in relatively stable primary energy use. In scenario 2 (New CCS), primary energy use increases to 28 EJ/yr by 2050 under base case efficiency improvements and then remains fairly stable through 2100. The increase in primary energy use is due largely to the energy penalty of CCS operations in newly-built plants. In scenario 3 (Retrofit + New CCS) the primary energy use increases rapidly and exceeds 34 EJ/yr by 2050 under base case efficiency improvements, then gradually decreases. The rapid increase is due to the energy penalty associated with carbon capture installations in older, inefficient power plants. As those plants are retired later in the century and replaced by more efficient new plants equipped with carbon capture, the primary energy use decreases. The variation between low and high efficiency improvements reaches about plus/minus 5 EJ/yr for scenarios 2 and 3. The variation for scenario 1 is limited to plus/minus 2EJ/yr because it includes only variation in energy conversion efficiency and not in carbon capture efficiency.

Figure S6. Percent reductions in cumulative GHG emissions (CE) and cumulative radiative forcing (CRF) of Scenarios 2 (New CCS) and 3 (Retrofit + New CCS), relative to Scenario 1 (No CCS).

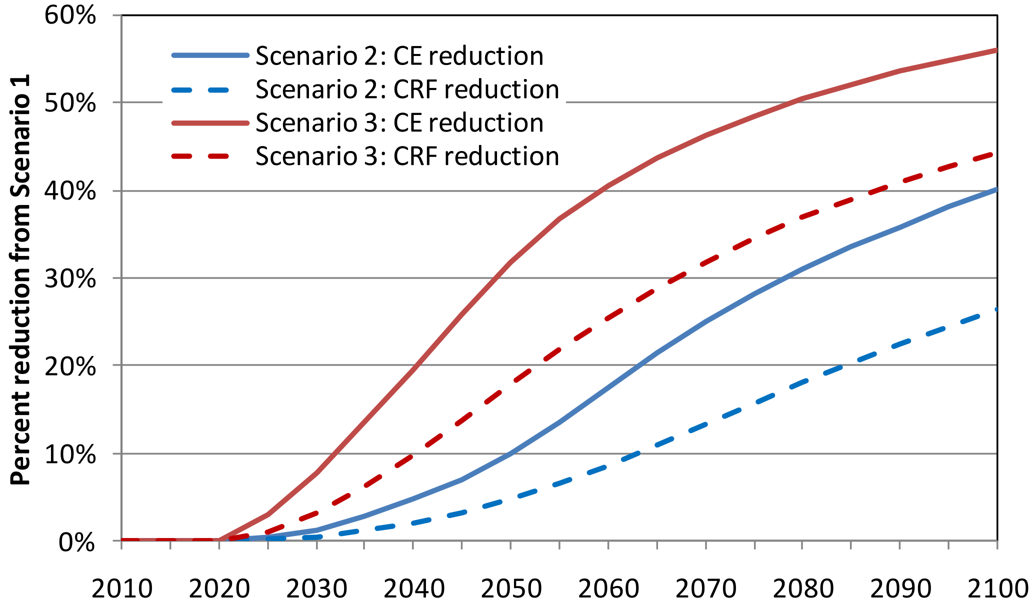
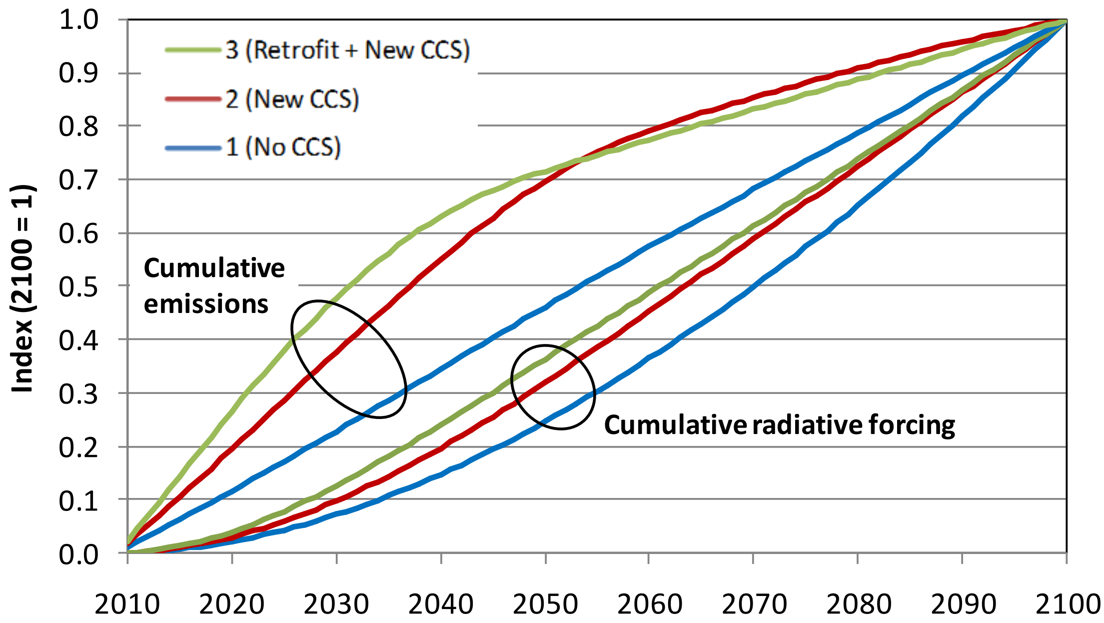


Figure S7. Comparison of trajectories of cumulative emissions and cumulative radiative forcing of three main deployment scenarios, with base case (medium) efficiency improvements. Each line shows the value over time as a proportion of the 2100 value.



The trajectories through 2100 of cumulative emissions are convex up, while those of cumulative radiative forcing are convex down. The emissions rates of scenarios 2 and 3 are greatest during the early part of the study period and decrease over time as CCS is implemented. The emission rate of scenario 1 is roughly constant over time due to continued emissions of GHGs. Radiative forcing, on the other hand, begins to rise at a low rate corresponding to the atmospheric concentration of GHGs emitted after 2010, but rises at an increasing rate over time as the amount of GHGs in the atmosphere grows.

Discussion of Functional Unit

A comparative analysis requires the definition of a reference entity or "functional unit" to allow objective comparison of the alternatives. A functional unit is a measure of the required properties of the system, providing a reference to which input and output flows can be related. To ensure a constant functional unit that allows objective comparison of the primary energy use, emissions, and radiative forcing across the three scenarios, the annual net generation of coal-fired electricity for each year is the same in all scenarios. Through 2035, this generation follows the reference-case projections of the 2010 Annual Energy Outlook [29], selected because the AEO reference-case is an authoritative and commonly-used projection for future development of the US power sector. The AEO reference case is then extrapolated linearly until 2050, and then assumed to remain constant at 2365 TWh/year through 2100.

Long-term energy supply forecasts are inherently uncertain, as future events will depend on demographic, technological, and behavioral factors and their complex interactions [S4]. In particular, there is significant uncertainty regarding the future of coal use, vis-à-vis the introduction of GHG mitigation policies and the diffusion of low-carbon energy technologies. An important uncertainty associated with the selected functional unit is that an incentive to install CCS (e.g., a carbon price) may just as well incentivize a shift away from coal and towards gas, nuclear, renewables, and conservation. Integrated modeling of dynamic interactions between the coal-fired power sector and other power sectors would provide additional information on other means of ensuring that supply of low-carbon electricity meets demand. Nevertheless, our assumption of large-scale deployment of CCS is not implausible, and is similar to deployment scenarios made in previous analyses. For example, EPRI [8] assumed US coal-fired power with CCS to reach about 2400 TWh in 2050 (compared to our 2365 TWh), although the updated EPRI [9] reduced that amount to about 1900 TWh. Bistline & Rai [7] counted their CCS emissions reductions off of a baseline of the 2006 AEO reference case, thus implicitly assuming that total coal-fired power would not decrease due to a GHG price. IEA [11] assumed that emission reductions due to CCS will be large and growing by 2050, suggesting that a GHG price will not necessarily cause other primary energy sources to displace coal-fired power.

Calculation of Radiative Forcing

The atmospheric decay of each annual pulse emission is estimated using the following equations [43,44,13]:

$$(CO_2)_t = (CO_2)_0 \times \left[0.217 + 0.259e^{\frac{-t}{172.9}} + 0.338e^{\frac{-t}{18.51}} + 0.186e^{\frac{-t}{1.186}} \right]$$

(Equation S1)

$$(N_2O)_t = (N_2O)_0 \times \left[e^{\frac{-t}{114}} \right]$$

(Equation S2)

$$(CH_4)_t = (CH_4)_0 \times \left[e^{\frac{-t}{12}} \right]$$

(Equation S3)

where t is the number of years since the pulse emission, $(GHG)_0$ is the mass of GHG emitted as a pulse emission at year 0, and $(GHG)_t$ is the mass of GHG remaining in the atmosphere at year t.

Annual changes in instantaneous radiative forcing due to the GHG concentration changes are estimated using the following equations [43,44,13]:

$$F_{CO_2} = \frac{3.7}{\ln(2)} \times \ln\left\{1 + \frac{\Delta CO_2}{CO_{2ref}}\right\}$$

(Equation S4)

$$F_{N_2O} = 0.12 \times \left(\sqrt{\Delta N_2O + N_2O_{ref}} - \sqrt{N_2O_{ref}}\right) - f(M,N)$$

(Equation S5)

$$F_{CH_4} = 0.036 \times \left(\sqrt{\Delta CH_4 + CH_{4ref}} - \sqrt{CH_{4ref}}\right) - f(M,N)$$

(Equation S6)

where F_{GHG} is instantaneous radiative forcing in W/m^2 for each GHG, ΔGHG is the change in atmospheric concentration of the GHG (in units of ppmv for CO_2, and ppbv for N_2O and CH_4), CO_{2ref} = 383ppmv, N_2O_{ref} = 319ppbv, CH_{4ref} = 1774ppbv, and *f(M,N)* is a function to compensate for the spectral absorption overlap between N_2O and CH_4 (IPCC 1997, 2001, 2007).

In our calculations of radiative forcing we have assumed relatively minor marginal changes in atmospheric GHG concentrations, such that radiative efficiencies and atmospheric decay patterns of the gases remain constant. However, significant increases in the atmospheric concentration of CO_2 can be expected during this century. As atmospheric CO_2 concentration increases, the marginal increase in radiative efficiency of CO_2 will become smaller, but the rate of atmospheric CO_2 decay will also become smaller. These will have opposite and therefore offsetting effects on radiative forcing [S5], thus we expect the significance of this uncertainty to be minor. We have estimated cumulative radiative forcing by integrating instantaneous radiative forcing over time. This is a simplification, as we ignore the feedback effect that the accumulated energy will have on future outgoing radiation. Radiative forcing is a measure of the radiative imbalance given that atmospheric temperatures remain unchanged. In fact, the radiative forcing calculated here will increase the heat energy accumulated in the earth system, leading to more outgoing longwave radiation and an eventual restoration of radiative balance. Since instantaneous radiative forcing does not account for this feedback effect, our results may therefore slightly overestimate the amount of cumulative radiative forcing.

Estimation of CO_2 Leakage Effects

Long-term storage of CO_2 in geologic formations is required for sustained climate benefit of CCS systems. IPCC suggests that CO_2 leakage from appropriately selected and managed geological reservoirs is likely to be less than 1% over 1,000 years, due to a combination of physical and geochemical trapping mechanisms that become more secure over longer time periods [4]. If these mechanisms are inadequate in some future storage reservoir, however, CO_2 leakage may occur which would fully or partially negate the atmospheric concentration reduction gained from sequestration. At worst, a complete leakage of CCS-derived CO_2 would result in greater overall emissions, because the energy penalty of the initial capture and compression of the CO_2 requires that more fuel be burned and more CO_2 produced than if CCS were not implemented. These emissions may occur far in the future, raising questions of trade-offs between generations.

A limitation of current LCA methodology is its poor suitability for extended time horizons or for low probability, high impact events, which may be needed for robust analysis of CCS systems [5]. The permanence of CO_2 storage in geologic deposits is typically taken for granted in life cycle studies of CCS systems. A review [S6] of numerous LCA studies of CCS systems found only one study [S7] to have considered sensitivity to CO_2 leakage. Viebahn et al. [S7] distinguished between long-term (>10,000 years) CO_2 emissions and middle-term (0 to 10,000 years) CO_2 emissions. They noted the challenge, still not solved, of balancing inter-generational impacts and benefits.

A variety of mathematical functions can be used to represent natural process patterns including leakage rates. Following the methodology of Viebahn et al. [S7] and van der Zwaan & Smekens [43], we use a simple exponential decay function where a fixed proportion of the then-current stock of CO_2 in geologic storage is leaked to the atmosphere each year. We consider two leakage rates, 0.1% per year and 1.0% per year. These two threshold values were suggested by van der Zwaan & Smekens [43] as being acceptable (0.1% per year) and unacceptable (1.0% per year) leakage rates. We calculate iteratively the time profiles of stored and leaked quantities of injected CO_2. Figure S8 shows the quantities of CO_2 remaining in geologic storage and the quantities leaked to the atmosphere for scenario 3 (new + retrofit CCS) through 2100. Table 3 in the main article shows that geologic CO_2 leakage of 0.1% per year would modestly increase overall emissions through 2100 by up to 7% and radiative forcing by up to 3%. At leakage rates of 1.0% per year, however, emissions through 2100 increase significantly by up to 56% and radiative forcing by up to 28%. This appears to support the observation by van der Zwaan and Smekens [43] that these two leakage rates bracket the region of acceptability of leakage, from the perspective of mass flow management.

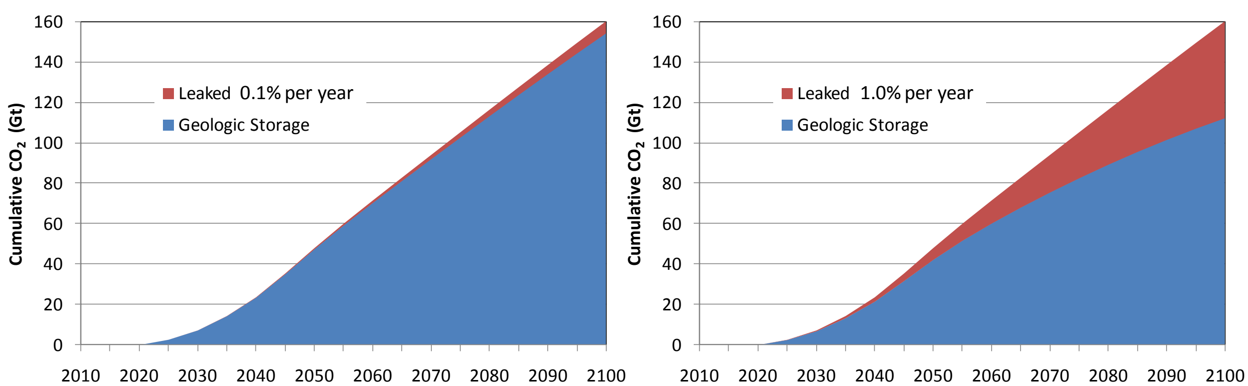


Figure S8. Amounts of CO_2 stored in geologic formations and leaked from storage at rates of 0.1% per year (left) and 1.0% per year (right), through 2100.

Over longer time horizons, however, even slow leakage rates result in significantly increased emissions of CO_2 to the atmosphere. Figure S9 shows the quantities of CO_2 remaining in geologic storage and leaked to the atmosphere through the year 2400, assuming injection of CO_2 continues through 2100 and then stops. With a leakage rate of 0.1% per year, 4.2% of the total injected CO_2 will have leaked by 2100, and

29% will have leaked by 2400. With a leakage rate of 1.0% per year, 33% of the total injected CO_2 will have leaked by 2100, and an overwhelming 97% will have leaked by 2400. Thus, depending on the time horizon and the perspective of various stakeholders, a CO_2 leakage rate of zero may be the only acceptable option [S8].

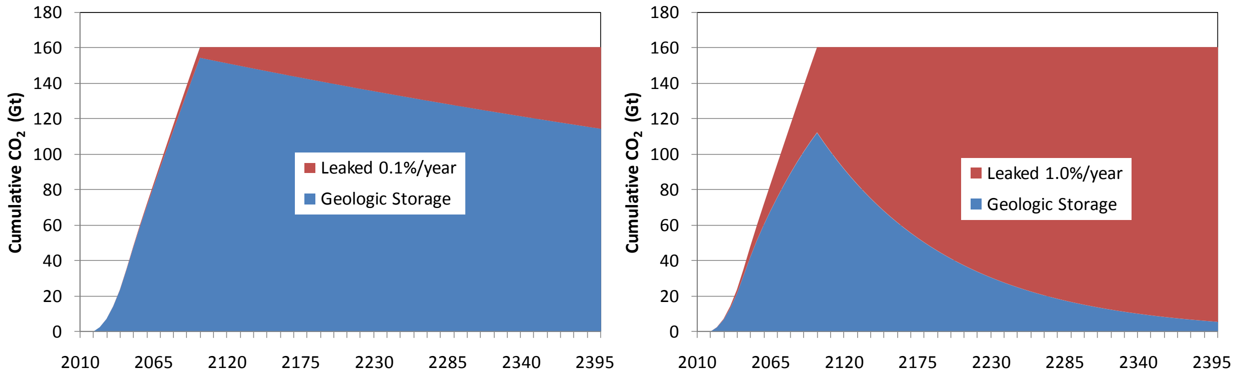


Figure S9. Amounts of CO_2 stored in geologic formations and leaked from storage at rates of 0.1% per year (left) and 1.0% per year (right), through 2400.

Here we have considered slow, steady leakage over long time periods, not short-term catastrophic blow-outs. While the overall mass flow effects of most blow-outs may be negligible in terms of total quantities of CO_2 emitted, the potential for serious accidental injury exists. While we have used an exponential decay function where a fixed proportion of the CO_2 in geologic storage is leaked each year, we note that a function involving a progressively decreasing leakage rate may be more appropriate over long time periods, due to the increasing security of geochemical trapping mechanisms over time. In addition to leakage, other potential impacts from underground CO_2 storage may be of concern, such as groundwater contamination and induced seismicity. These issues merit further study.

Additional references

References that are cited only in the Supporting Information are listed below. All other references are listed in the References section of the main article.

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