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Technoeconomic Analysis of Negative Emissions Bioenergy with Carbon Capture and Storage through Pyrolysis and Bioenergy District Heating Infrastructure

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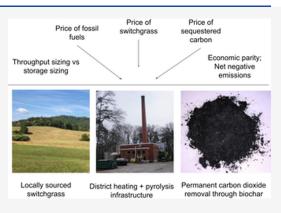
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ABSTRACT: Bioenergy with carbon capture and storage (BECCS) has been identified as a cost-effective negative emission technology that will be necessary to limit global warming to 1.5 °C targets. However, the study of BECCS deployment has mainly focused on large-scale, centralized facilities and geologic sequestration. In this study, we perform technoeconomic analysis of BECCS through pyrolysis technology within a district heating system using locally grown switchgrass. The analysis is based on a unique case study of an existing switchgrass-fueled district heating system in the rural southeastern United States and combines empirical daily energy data with a retrospective analysis of add-on pyrolysis technology with biochar storage. We show that at current heating oil and switchgrass prices, pyrolysis-bioenergy (PyBE) and pyrolysis BECCS (PyBECCS) can each reach economic parity with a fossil fuel-based system when the prices of carbon is \$116/Mg CO₂-eq and \$51/Mg CO₂-eq, respectively. In addition, each can reach parity with a direct



combustion bioenergy (BE) system when the prices of carbon is $$264/Mg\ CO_2$ -eq and $$212/Mg\ CO_2$ -eq, respectively. However, PyBECCS cannot reach economic parity with BE without revenue from carbon sequestration, while PyBE can, and in some cases, PyBECCS could counterintuitively require more reliance on fossil fuels than both the PyBE case and BE.

KEYWORDS: decarbonization, carbon dioxide removal (CDR), lifecycle analysis (LCA),

bioenergy and carbon capture and storage (BECCS), district heating, switchgrass, biochar, energy infrastructure transition

1. INTRODUCTION

With each year that climate change mitigation action is delayed, pressure increases to not only reduce CO_2 emissions but also to remove CO_2 that has accumulated in the atmosphere. Since 2014, Negative emissions technologies (NETs), also known as carbon dioxide removal (CDR), have already been incorporated into the majority of integrated assessment model scenarios that meet global warming limit targets. Of NETs, one technology in particular, bioenergy and carbon capture and storage (BECCS), has emerged as one of the most promising. Unlike other alternatives, such as direct air capture, all of the technologies required to implement BECCS are already developed and are either already commercially viable or in prototyping stages and may be economically viable now. 2

The dominant technological configuration imagined for BECCS infrastructure is the large, centralized bioenergy production facility that captures carbon dioxide waste streams of either a fermentation or thermochemical conversion process and permanently stores it in underground geologic formations. Currently, there is only one large-scale (sequestering ~ 1 Mt CO₂ eq yr⁻¹) BECCS facility in the world, located in Decatur, IL. This facility processes corn into ethanol for use as a

gasoline additive and injects the CO₂ emitted from the fermentation process into a sandstone geologic formation near the bioethanol plant.³ This kind of carbon capture and storage (CCS) infrastructure is not only geographically limited (based on suitability of geologic formations for CO₂ storage) it is also understood to require both scale and payment for the carbon removal in order to be economically feasible.

An alternative pathway for scaling BECCS implementation is through smaller-scale, distributed facilities, ranging from single institutions to small district energy systems (<100 MW capacity). Because conventional CCS infrastructures are impractical for this scale, carbon sequestration is typically achieved through the production and land application of biochar, a byproduct of pyrolysis. Several studies have begun to explore the greenhouse gas (GHG) emissions' mitigation

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potential and economic implications and requirements for such a technology configuration. To our knowledge however, no study addresses how the daily operational decisions of a pyrolysis-bioenergy plant interact with infrastructure sizing decisions and variability in fuel and carbon prices. Our study aims to address this gap through the following research questions:

- At this scale, how are bioenergy and biochar operation modes distinct and what implications are there for reaching economic parity and carbon sequestration goals?
- What prices (of fossil and biomass fuels, and of carbon offsets or credits) will be required to justify the addition of pyrolysis infrastructure (to produce biochar for sequestration, enabling the attainment of net negative emissions) to a biomass heating system?
- How will economic factors impact the feasibility of distributed, small-scale BECCS?

This research aims to make two contributions. First, our case study presents two base case scenarios: one in which the pyrolysis add-on technologies are compared to a fossil fuel (heating oil)-based system, and one in which the pyrolysis addon technologies are compared to the direct combustion of switchgrass for bioenergy (BE). We consider three pyrolysis operations scenarios: (1) pyrolysis for CCS only (PyCCS), (2) pyrolysis for bioenergy (PyBE), and (3) pyrolysis-bioenergy CCS (PyBECCS). Our BE base case scenario uses daily empirical data from an operational, district heating system that is currently fueled with locally sourced switchgrass and heating oil and more accurately reflects the operational challenges of direct combustion of switchgrass in solid fuel boilers. Second, our study explores tradeo-ffs in stock and flow dynamics during the daily operation of the pyrolysis-bioenergy plant. The consideration of operational modes is important given the competing goals of biochar for sequestration and affordable and consistent energy production. More details regarding the distinctions between our operational scenarios are presented in Section 3.4.

2. BACKGROUND

2.1. Gaps and Challenges with Scaling BECCS. For more information on the state of the art of research for negative emissions technologies and how BECCS fits into the greater NETs literature, the reader is referred to a series of articles in *Environmental Research Letters.* ^{4–6} In most studies, BECCS integration into energy systems reflects the dominant technological imaginary of how BECCS infrastructure will scale: from bench-scale laboratory experiments to pilot facilities, and to large-scale, centralized energy facilities.²

However, there are major practical concerns with large-scale, centralized BECCS facilities, often due to the relatively low energy density of bioenergy feedstocks. Global supply chains similar to those currently used for fossil fuels and agricultural commodity crops would require a 10-fold increase in global container volume if applied to the amount of biofuel required by some Intergovernmental Panel on Climate Change (IPCC) scenarios. Other studies have shown how both net carbon sequestration and economic feasibility will be impacted by the sourcing radius for biomass feedstock, bringing into question trade-offs between economies of scale that could be reached with large centralized facilities and smaller, more distributed plants or pre-processing facilities. Depending on the

sources of energy for the transportation sector, the embodied carbon in long-distance supply chains can impede a project from reaching net negative emissions.

Few studies address how the transition to bioenergy crops by local growers will happen (notable exceptions being refs 16–18), which relates problems in market penetration and technological diffusion. ^{19,20} In addition, large-scale, centralized, and financialized global flows of carbon capture and sequestration may also induce short-term or accelerated increases in carbon emissions, which could result in missing opportunities to prevent irreversible consequences of climate change. ^{21,22}

One unique context in which the above concerns could be addressed is by district-scale heating. ^{23,24} Local sourcing and vertical integration of agricultural biomass for district heating systems result in synergies that create economically competitive biomass supply chains with reduced greenhouse gas emissions. ²⁵ In the US, converting boilers to accept biomass feedstocks is feasible in a diverse range of locations and has been shown to result in many economic, social, and environmental benefits for local communities. ^{26,27} This motivates consideration of local scale technological configurations as potential transition pathways for greater BECCS adoption.

2.2. District-Scale BECCS and Pyrolysis. Integration of carbon sequestration into district biomass heating systems will likely be different from the sequestration techniques used in large, centralized facilities. Studies of smaller-scale BECCS have different technological configurations that allow them to be economically viable. Instead of carbon capture through chemical scrubbing and sequestration through high-pressure injection into geologic formations, which would be cost prohibitive at small scales, CCS at small scales may be achieved through the production of biochar in a process called "pyrolysis". ^{28,29}

In pyrolysis, biomass is heated in the absence of oxygen, resulting in three products—a solid (biochar), liquid (bio-oil), and gas (syngas)—the proportions of which depend primarily on the temperature of the reaction. Biochar is a stable form of carbon that is resistant to oxidation on the order of 100s or even 1000s of years, meaning that biomass in this form is effectively "sequestered". Biochar can be directly applied to the land surface and has been shown to increase crop yields, improve soil water holding capacity, and decrease erosion. 30,31 The production of biochar has been estimated to potentially sequester 1.8 Gt CO₂ yr⁻¹ without negatively impacting food production or biodiversity, globally.²⁹ Sequestration of bio-oils and syngas products is considered more challenging technically, economically, and socially to implement, and is estimated to be possible in the 10-20 year range, and +20 year range, respectively.²⁸

Each product, biochar, bio-oil, and syngas, can be completely combusted for energy generation; therefore, the pyrolysis process can be adjusted for optimal production of either bioenergy or the pyrolysis products. ^{32–34} Several studies have completed lifecycle analyses (LCA) of pyrolysis-bioenergy (heat and/or power) systems to better understand this technology configuration for reaching net negative emissions. A range of scales have been considered, expressed in terms of pyrolysis throughput (200 kg/h in Yang et al. ³⁵ and ~10 Mg/h in Roberts et al. ³⁶), thermal output (66 MW_{th} in Pröll and Zerobin), ³² or power output (1.5 MW in Yang et al., ³⁵ and ~50 MW in Woolf et al. ³³ and Azzi et al. ³⁷).

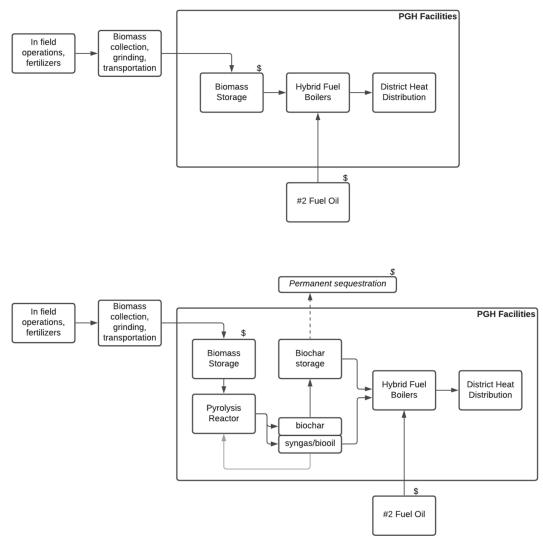


Figure 1. System boundary diagrams for PGH's current district heating plant operations (top) and heating plant with the pyrolysis facility (bottom).

However, in our review of the literature, studies that carried out LCA of pyrolysis and bioenergy technology configurations usually only calculated economic parity with respect to a fossilfuel or economic payback in relation to the capital cost of infrastructure investment and not to the operations of a bioenergy-only base case. Also, the majority of LCAs for carbon dioxide removal technologies for reaching net negative emissions rely on the "system expansion" approach that includes counting of substituted conventional fossil fuels, but may not actually result in more carbon dioxide removed from the atmosphere than produced, which is the goal of NETs. In addition, none of the reviewed studies conducted modeling at the daily scale.

We hypothesize that addressing these areas is important to understanding technological transition due to the following: (1) if a particular rural heating facility was to consider incorporating pyrolysis of a dedicated biomass fuel into their heating system, then likely they would also be considering direct combustion of the dedicated biomass, perhaps as an intermediary infrastructure transition, and (2) in climates with large fluctuations in heating demand, stockpiling biochar produced during warm weather months could offer additional savings while building capacity toward future CCS operations.

In the following section, we describe our case study and methods for exploring the integration of pyrolysis into a rural district heating system and implications for operations, economics, and carbon sequestration.

3. METHODS

3.1. Case Study and System Descriptions. Piedmont Geriatric Hospital (PGH) is a state-run hospital for seniors who require inpatient treatment located in Southside Virginia, USA. A district heating plant provides steam for space heating and hot water to the patient care and staff buildings of the PGH campus. More detail on the facilities and district energy system is provided in the Supporting Information.

In the late 90s, PGH entered into a 10-year fixed contract with a supplier to provide ground, dried, locally harvested switchgrass to the facility for \$191/Mg of delivered biomass with under 15% moisture content. The facility still uses heating oil as a back-up fuel when the biomass system is not functioning as intended; heating oil is also needed because the low energy density of switchgrass needs a higher energy density fuel to handle fluctuations (ramp up) in the system's heating demand. Between 2015 and 2019, 7 to 24% of the annual total steam produced was from the combustion of oil.

Table 1. Scenarios Considered

	Heating Oil Base Case	Bioenergy (BE) Base Case	Pyrolysis + Bioenergy (PyBE)	Pyrolysis + Bioenergy and Carbon Capture and Storage (PyBECCS)	Pyrolysis Carbon Capture and Storage (PyCCS)
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Goal	Base case assuming all heating demand met with heating oil	Base case (current operation at PGH)	Eliminate residual heating oil demand	Reach net carbon removal from the atmosphere	Benefit from future positive price of carbon sequestration
Infrastructure	Hybrid solid fuel (unused)/heating oil boilers	Hybrid solid fuel/heating oil boilers	+ pyrolysis reactor and interseasonal biochar storage systems	+ pyrolysis reactor with excess throughput capacity and one day's throughput biochar storage	Pyrolysis reactor with one week's biochar storage
Operations	Hypothetical fossil fuel only case	Existing PGH case	Stockpile biochar when heating demand is low; use storage biochar when demand is high. Empty biochar in storage at end of heating season and land apply for permanent sequestration	Use only syngas and biooil products to meet heating demand. Send all biochar to permanent sequestration. Empty all biochar storage daily.	Minimal stockpiling of biochar on site; produce biochar and land apply for permanent sequestration.
Supplemental Fuel	NA	Heating oil	Heating oil	Heating oil	NA
Basis of Economic Parity/Payback	NA	NA	Parity through offsetting remaining heating oil demand	Parity through receiving credits for carbon sequestration	Payback for infrastructure investment through receiving credits for carbon sequestration

We obtained daily records from PGH's facilities manager for steam production and amounts of heating oil and grass used in direct combustion in each of the boilers for 2015–2019. This data was used to build models for a 20-year period from January 1, 2000—December 31, 2019 for the heating demand, fuels required to meet this demand given the current system's function, and the fuels required to meet this demand given the addition of pyrolysis and biochar storage infrastructure. We then calculated the 20 year lifecycle GHG emissions from various scenarios of pyrolysis infrastructure sizing and net present values of the pyrolysis infrastructure investment compared to the direct combustion case. Figure 1 shows a conceptual diagram of our study's system boundaries.

3.2. Estimation of the Heating Demand and the Direct Combustion Bioenergy (BE) Model. Empirical data on the daily heating demand and proportions of the heating demand met with fuel oil and grass were used to fit a predictive model for a 20-year time period of daily grass and oil demands assuming direct combustion of switchgrass. We estimated these demands in a two-step statistical model, which is described in more detail in the Supporting Information.

3.3. Pyrolysis Model. We modeled slow pyrolysis occurring at 500 °C using equations presented by Schmidt et al. for yields. Although other sources 34,41,42 provide models for the yield of biomass pyrolysis, we found the Schmidt et al. model to more closely approximate the pyrolysis conditions we propose for PGH. Schmidt provides temperature-dependent equations for the yields of slow pyrolysis products (chars, liquids, and gas) and provides tabulated results for the elemental makeup of biochar and energy content of pyrolysis products. Using these results, we calculated the yield of pyrolysis products and their energy content per Mg of switchgrass as shown in Table S1. We also calculated the energy in the incoming switchgrass by assuming an 8% moisture content. Switchgrass is presumed to be delivered dried to 10% moisture content before arriving on the site, and therefore drying to 10% moisture content was excluded from the system boundary. Switchgrass is then dried to 8% moisture before entering the pyrolysis reactor using residual heat. In subsequent sections, we will refer to syngas and bio-oil as separate products of the pyrolysis reaction; however, in reality, bio-oils are assumed to be directly combusted with the

pyrolytic gases and are never condensed into the liquid product at any time in the process. Therefore, the energy content of neither the syngas nor the bio-oil can be stored to meet the later demand. The subsequent analyses are based on feasibility and the function of an indirect heated recirculating pyrogas convection-type moving bed reactor.

3.4. Daily Heating Oil, BE, PyBE, PyBECCS, and PyCCS **Operation Models.** The pyrolysis model described above was applied to the modeled daily heating demands for the 20-year study period. For each day of the PyBE scenario, the pyrolysis reactor is presumed to operate at 100% of its maximum daily throughput capacity unless no biochar storage capacity remains, in which case the pyrolysis reactor is not operated and heating demands are met by combusting the stockpiled biochar. For each day that the pyrolysis reactor does operate, syngas and bio-oil pyrolysis products (from the pyrolysis reaction that day) are first combusted to meet the daily heating demand. If syngas and bio-oil production exceeds the heating demand, the extra syngas and bio-oil products are assumed to be combusted or "flared". To minimize complications associated with storage, no liquid products will be condensed or stored from the process and the storage and waste disposal is not accounted for in our cost models. After the combustion of syngas and bio-oil, biochar (produced that day or stored) is combusted to fulfill any unmet heating demand; otherwise, the biochar product is stored in the biochar storage volume. Finally, for the BE, PyBE, and PyBECCS scenarios, heating oil is used to meet any remaining demand.

The amount of heating oil that can be offset compared to the BE base case is therefore a function of how the throughput of the pyrolysis reactor and the biochar storage volume capacity work together to meet seasonally variable heating demands. Because the heating demand is lower during the summer, the pyrolysis reactor can be used to top-off storage, which can then be used to supplement the throughput capacity during winter months when the heating demand is much higher. When biochars from the pyrolysis reactions are used to store energy to meet the seasonally fluctuating thermal demands more efficiently, we call this the PyBE (pyrolysis bioenergy) operation model: storage and throughput are chosen in order to minimize heating oil needed to supplement the system.

In contrast, in the PyBECCS (pyrolysis bioenergy and carbon capture and storage) scenario, the production of biochar for sequestration is prioritized and it is not stockpiled to meet the winter heating demand. In this scenario, storage capacity can be downsized since biochar is presumed to be removed from the site every day. Only the syngas and bio-oil products are combusted to meet the site's heating demand. When the heating demand cannot be met, heating oil is used to meet the remaining demand.

We also used the pyrolysis model to estimate the amount of biochar that could be produced from a Mg of switchgrass. We called this operational model PyCCS (Pyrolysis Carbon Capture and Storage) since it is not linked to any heating demand; it merely converts biogenic carbon in the switchgrass to biochar, the sequesterable form. Table 1 summarizes the scenarios considered.

3.5. Emissions, Costs, and Net Present Value Calculations. 3.5.1. Greenhouse Gas Emissions. One objective of the pyrolysis facility heating system is to decrease net greenhouse gas emissions of PGH and assess how the system can be designed to achieve net negative emissions. To

evaluate these goals, we calculated lifecycle emissions for switchgrass production and transport to the PGH facility. The switchgrass currently used at PGH is grown on fallow agricultural land with the inputs shown in Table S3. Because the grass is grown on fallow agricultural land, we assume that direct land use change emissions are negligible and that there are no indirect land use change emissions.

For transportation of switchgrass, we assumed that each truck has a maximum wet payload of 20 tons and fuel efficiency of 6.5 mpg of diesel fuel.⁴³ Using emission factors from GREET (Argonne National Lab, 2018) and ecoinvent,⁴⁴ we calculated GHG emissions for switchgrass production as shown in Table S4, which contains an example of calculated emissions for four scenarios over 5 years of operation from 2015 to 2019.

3.5.2. Capital Costs. Capital costs of the pyrolysis reactor equipment included the system components for the pyrolysis reactor and equipment for the biochar storage infrastructure. Construction costs associated with the pyrolysis reactor and equipment included the following: pyrolysis equipment (infeed, reactor, heat exchanger, cooling, exhaust, syngas generation, and condensing), biochar equipment (processing, cooling, densification, and storage), and controls. Details regarding cost models applied can be found in the Supporting Information.

3.5.3. Annual Costs and Net Present Value (NPV). NPV was calculated using the difference in annual cash flow for each scenario compared to each base case for the 20 year period. PyBE and PyBECCS NPVs are calculated with respect to the BE or the fossil fuel base case, meaning that only additional capital costs associated with the pyrolysis system are counted, and all annual operational costs or savings are relative to the annual costs of each respective base case, with savings reached over time through the offset of heating oil demand (we refer to this as reaching "economic parity" with the base case). In contrast, the PyCCS NPV is calculated by presuming that biochar can be sold at a profit to offset the capital cost of the pyrolysis equipment over time (we refer to this as reaching "payback" of the initial infrastructure investment capital cost associated with the pyrolysis process). The following were considered in the annual cash flows: costs of fuels (fuel oil, biomass), annual maintenance costs (calculated as 5% of the capital expense each year), labor expenses associated with each operation scenario, and a "price of carbon" (see the Supporting Information Section S4 for more details on the "price of carbon)".

A discount rate of 5% was applied to the cash flow to calculate the NPV. This reflects the lower range of debt service interest rates that are currently being offered in the US to support infrastructure development capital costs. Because the NPV is calculated with respect to a base case scenario, a negative NPV denotes that the scenario costs more in the long term than the base case (does not reach economic parity). For PyBE and PyBECCS, a positive NPV denotes that the scenario costs less in the long term than the base case (reaches economic parity). For PyCCS, a positive NPV denotes that the scenario is able to recover the capital and operation costs of the pyrolysis infrastructure over the 20 year period, by the revenue generated through the sale of carbon offsets. The different base cases for PyBE/PyBECCS and PyCCS mean that PyBE and PyBECCS could potentially reach positive NPVs even without a price on sequestered carbon, while PyCCS requires a price of carbon to reach a positive NPV.

3.6. Scenario Testing, Sensitivity Analysis, and Uncertainty Analysis. The BE, pyrolysis, emissions, cost, and NPV models were all programmed using Python. This allowed us to test a range of scenarios with different combinations of input parameters, including ranges in prices for heating oil (\$2-\$5.5/gal), delivered switchgrass (\$87.5-\$200/Mg), and sequestration of CO₂-eqs (\$0-\$300/Mg CO₂eq); ranges of throughputs for the pyrolysis reactor (16-24 Mg/d); and ranges of the maximum biochar storage volume (0-500 Mg). Scenarios of various combinations of input parameters were tested in order to illustrate broad trends exhibited by the various operations scenarios. In addition, we completed a sensitivity analysis to determine which parameters had the largest effects on the model outputs. The most sensitive parameters (the price of fuel oil and the price of delivered grass) were used in uncertainty analysis, in which 1000 random values of the parameters were drawn from probability distributions to understand the uncertainty around estimates for the price of carbon (\$/Mg CO₂-eq) necessary to reach economic parity with the heating oil and direct combustion BE base cases. The price of fuel oil was drawn from an empirical distribution of heating oil prices in the United States. The price of delivered grass was drawn from a uniform distribution ranging from 20% lower to 20% higher than the price of delivered grass to the PGH site.

4. RESULTS

4.1. Pyrolysis Reactor Throughput and Biochar Storage Sizing Configurations. We first found that given the seasonal fluctuations in daily heating demand over the 20year simulated period, each pyrolysis reactor daily throughput rate could be associated with a maximum biochar storage capacity above which additional capacity would not result in additional carbon sequestration (Table S4). The interactions between interseasonal storage volumes and daily throughput result in variable utilization rates of the pyrolysis infrastructure. We more closely examine the 16 Mg/day throughput pyrolysis reactor size for the PyBE scenario since the utilization rate was found to be the most effective in meeting the case's existing heating demand. In the PyBE scenario, this throughput was optimally coupled with a biochar storage capacity of 250 Mg. This configuration resulted in a net removal of 791 Mg of CO₂-eq over the 20 year simulation period.

In the PyBECCS case, an equally sized reactor (16 Mg/d), but with only one day's storage volume for biochar, with all biochar removed daily for land application and permanent carbon sequestration resulted in a net removal of 36,222 Mg/CO $_2$ -eq over the 20 year simulation period.

4.2. Price Conditions Necessary for PyBE to Meet Economic Parity with the Fuel Oil and BE Base Cases. Holding the price of delivered switchgrass fixed at \$191/dry Mg for switchgrass, we solved for the price per Mg CO₂-eq necessary to reach economic parity with each base case (heating oil and direct combustion of switchgrass (BE)), given a range of $P_{\rm oil}$ (ranging from \$1.00–\$5.50/gal). For the 2010–2020 10 year mean $P_{\rm oil}$ of \$2.36/gal, the price of carbon at which PyBE reaches economic parity reaches parity with the heating oil base case is \$51.13/Mg CO₂-eq; however, when compared to the BE base case, the price of carbon needed to reach economic parity is \$265.29/Mg CO₂-eq (Figure S1).

There are also interactions between pyrolysis throughput and biochar storage to consider for both economic parity and negative emissions potential (Figure S3). When prices of

heating oil are low (\$2/gallon), no amount of storage of biochar can result in economic parity of the PyBE system with the BE base case over a 20 year period. When the price of heating oil is raised to \$3.75/gallon, and the price of switchgrass is \$87.5/Mg, the 16 Mg/day pyrolysis reactor can reach economic parity with BE but only when 250–300 Mg of biochar storage is built. Also, while the 16 Mg/day pyrolysis system with various biochar storage scenarios can reach economic parity with BE when the price of heating oil is high and the price of switchgrass is low, not all scenarios result in negative emissions. Under these conditions, a 20 Mg/d throughput pyrolysis reactor would reach negative emissions and economic parity with the BE base case with any amount of biochar storage.

4.3. Comparison of PyBE, PyBECCS, and PyCCS Scenarios. The PyBECCS operational scenario with 16 Mg/d throughput and 16 Mg of storage, under current (10year mean) heating oil prices and a \$191/Mg switchgrass price, reached economic parity with the fuel oil base case when the price of carbon is \$116.17/Mg CO₂-eq (compared to \$51.13 for PyBE). Under the same conditions but compared to the BE base case, the PyBECCS scenario reached economic parity with the BE base case when the price of carbon is \$211.87/Mg CO₂-eq (compared to \$265.29 for PyBE). The economic payback of PyCCS decreases as the throughput of the pyrolysis increases (Figure S4). At 16 Mg/d throughput, PyCCS reaches economic payback at \$41/Mg CO₂-eq and is not affected by the choice of base case. This is because payback is reached solely with respect to pyrolysis and biochar storage infrastructure and labor investments, rather than through offsetting heating oil demands. This economic comparison is quite different from the parity conditions for PyBE and PyBECCS. Table 2 summarizes the operational scenarios' emissions performance and required carbon prices to reach economic

Table 2. Comparison of PyCCS, PyBECCS, and PyBE Operational Scenarios^a

Operational scenario	Total negative emissions (20 years)	Required $P_{\rm C}$ (assuming a 10 year mean price of fuel oil: \$2.36/gal)	Required $P_{\rm C}$ (assuming a high price of fuel oil: \$5.5/gal)	Required P_{oil} (if P_{C} = 0)				
PyCCS ^b (112 Mg storage)	57,620 Mg	\$41/Mg CO ₂ -eq	\$41/Mg CO ₂ -eq ^c	NA				
Base case = fuel oil								
PyBECCS (16 M storage)	g 36,222 Mg	\$116.17/Mg CO ₂ -eq	\$38.71/Mg CO ₂ -eq	\$7.07/ gal				
PyBE (250 Mg storage)	791 Mg	$$51.13/Mg$ CO_2 -eq	\$0/Mg CO ₂ - eq	\$2.90/ gal				
Base case = direct combustion of switchgrass								
PyBECCS (16 M storage)	g 36,222 Mg	\$211.87/Mg CO2-eq	\$268.46/Mg CO ₂ -eq	NA				
PyBE (250 Mg storage)	791 Mg	\$265.29/Mg CO2-eq	\$0/Mg CO ₂ - eq	\$5.43				

"All comparisons assume the current price of delivered switchgrass (\$191/Mg). ^bPyCCS scenario does not provide any energy to the district heating plant; energy demands are met through the BE base case (direct combustion of switchgrass and unmet demands met with heating oil). ^cEconomic case is based on payback against pyrolysis infrastructure and labor costs only, therefore not dependent on the price of heating oil; if prices of heating oil were to rise to \$5.5/gal from the 10 year mean of 2.36/gal, the district energy plant's heating oil costs over a 20 year period would be \$29.6 M higher.

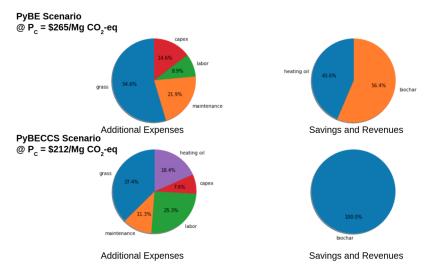


Figure 2. Comparison of sources of additional expenses and savings and revenues (compared to BE) of PyBE and PyBECCS, shown at the price of carbon for each scenario that results in both economic parity with BE, and net removal of carbon dioxide from the atmosphere (\$248 and \$208/Mg, respectively). Both PyBE and PyBECCS scenarios incorporate a pyrolysis reactor with 16 Mg/day throughput. Note that "heating oil" appears as a source of savings for PyBE, while it appears as an additional expense for PyBECCS.

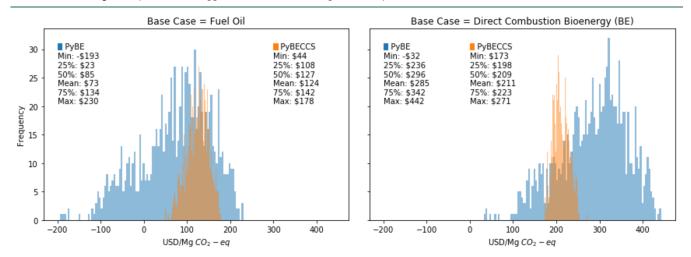


Figure 3. Uncertainty around the predicted price of carbon ($\$/Mg\ CO_2$ -eq) needed to reach economic parity with each base case. The estimates for P_C necessary given the PGH's cost of delivered switchgrass (\$191/Mg) and the 10 year mean fuel oil price (\$2.36/gallon) for PyBE and PyBECCS compared to a fuel oil base case are \$51 and \$116, respectively, and compared to a BE base case are \$265 and \$212, respectively. These values are near the central tendencies of each distribution. However, there is a much wider range of variability around the PyBE operational scenario than the PyBECCS scenario.

parity with BE (PyBECCS and PyBE) or economic payback for pyrolysis infrastructure (PyCCS).

Figure 2 shows a comparison of sources of additional expenses and savings and revenues between PyBE and PyBECCS at the prices of carbon at which each reaches economic parity with the BE base case. PyBECCS does not achieve any savings through offsetting demand for heating oil, and in fact, has a higher heating oil demand than the BE case. Instead, both economic parity and net negative emissions are achieved through the production of biochar.

4.4. Sensitivity and Uncertainty Analysis. The input parameters that were found to be the most sensitive in each of the comparisons were the price of fuel oil and the price of delivered grass (Table S5). Figure 3 shows the results of the uncertainty analysis of the model's predicted price of carbon necessary to reach economic parity, given 1000 random draws of the two most sensitive input parameters.

DISCUSSION: IMPLICATIONS FOR INFRASTRUCTURE TRANSITION

The above results have several implications for BECCS implementation in district heating applications. First, considerations of combinations of pyrolysis reactor throughput and storage capacity to meet daily and seasonally fluctuating heating demands are necessary. When compared to a BE-only system, pyrolysis makes the combustion of the solid fuel more efficient through the increased energy density of biochar compared to switchgrass. In other studies, switchgrass has been associated with more operating issues than conventional fuels, which causes maintenance issues and inefficiency. 45,46 The low energy density of switchgrass in the PGH's experience has also necessitated continued reliance on supplemental heating oil to ramp up steam production when the heating demand increases during daily low temperatures and with changing seasons. Modeling remaining interseasonal storage capacity in the PyBE case makes it possible to explore stock and flow dynamics.

Second, we demonstrate more detailed economic considerations of PyCCS, PyBECCS, and PyBE. While PyCCS requires the lowest carbon prices given current heating oil and switchgrass prices, the way it achieves economic payback means that it will not shield the district energy plant from high heating oil prices. In addition, the attractiveness of PyBECCS compared to PyBE depends on the choice of base case. When the base case being considered is a fossil fuel-only facility (heating oil), PyBE is able to achieve economic parity when the price of carbon is \$51/Mg CO₂-eq, while PyBECCS requires a price of \$116. When the base case is the direct combustion of switchgrass, then PyBECCS requires a lower price of carbon (\$212/Mg CO₂-eq) than PyBE (\$265/Mg CO₂-eq). However, given high heating oil prices, PyBE has the potential to reach net negative carbon dioxide emissions and economic parity with BE, without any price on sequestered carbon, while PyBECCS does can never reach economic parity if there is no price on sequestered carbon. This is because nearly half of the source of economic parity is achieved through offsetting the remaining heating oil demand in the PyBE case, while the PyBECCS case relies on selling carbon credits and actually requires additional heating oil to supplement syngas and bio-oil to meet winter-month heating demands.

The prices for carbon that enable our PyBE and PyBECCS scenarios to reach economic parity are much higher than other studies that estimate prices of carbon dioxide equivalents at which it is economically feasible to reach negative emissions from \$7³³ to over \$50/Mg CO₂-eq.^{34–36} This is primarily for two reasons. First, in our models, we compare PyBE and PyBECSS to a direct combustion BE case, which may be a more realistic infrastructure transition pathway for district-scale BECCS infrastructure, for which connections between buyers and sellers of a dedicated energy crop like switchgrass need to be established first. However, even in our comparisons to a fossil fuel-only base case, our estimated required price of carbon is often higher than others reported in the literature. Another explanation for this might be the higher price of delivered switchgrass compared to other studies, which assume low prices for commodities and agricultural or forestry residues 32,33,37,47-49 (see the Supporting Information Section S6 for more discussion).

Operations around the variation in the seasonal heating demand in the PyBE case result in net negative emissions over the 20 year period without intentionally changing operations to prioritize reaching negative emissions. This outcome is likely dependent on the heating profile (daily/interseasonal) of our case site. Future research should explore how different heating profiles result in different outcomes. As has been found by others, direct land application of biochar (in our case, the PyBECCS scenario) was found to remove much more carbon dioxide equivalents than when biochar was used for fuel, even though our lifecycle analysis did not assume additional reduced emissions associated with soil improvement, and therefore may be a conservative estimate. 31,38,50–53

Most interesting, however, was the counterintuitive finding that PyBECCS operations, in which biochar is not used to meet heating demands but directly applied to land for permanent sequestration, could actually result in increased demands for heating oil compared to a direct combustion of the switchgrass base case. This was found to occur because the daily throughput of the pyrolysis reactor is often less able to meet the daily (and seasonally varying) heating demands of the

site than the BE case. This is not the case for PyBE, which includes additional storage infrastructure to stockpile biochar during the non-heating season to supplement throughput in the heating season.

Lastly, while there are benefits to a case-study-based analysis, including the ability to incorporate empirical data analysis from a functioning switchgrass-fueled heating plant, and more concrete understanding of operational concerns and constraints, there are also drawbacks. Drawbacks of case-studybased analyses include a need to consider limited generalizability. Southside Virginia comprises much marginal and underutilized agricultural land, formerly intensively farmed for tobacco production. The land has been rated as very suitable for conversion to switchgrass production, and there are few competing alternatives for commodity crops. 54,55 District energy systems in the United States are also not as common as in other parts of the world; however, institutions such as hospitals and universities that do rely on district energy facilities have been identified as unique niches for adoption of decarbonization technologies. 56,57 Future research should aim to evaluate the potential of coupled land-infrastructure planning at the district scale to generate negative emissions on a broader scale, such as in an entire state, region, or country.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.1c03478.

(S1) Case study detail, (S2) empirical BE models, (S3) pyrolysis model, (S4) lifecycle analysis model, (S5) cost model, (S6) supplemental results, and (S7) supplemental discussion (PDF)

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Notes

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