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Distributed biochar and bioenergy coproduction: a regionally specific case study of environmental benefits and economic impacts

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Abstract

Biochar has been advocated as a method of sequestering carbon while simultaneously improving crop yields and agro-ecosystem sustainability. It can be produced from a wide variety of biomass feedstocks using different thermochemical conversion technologies with or without the recovery of energy coproducts, resulting in chars of differing quality and a range of overall system greenhouse gas (GHG) mitigation outcomes. This analysis expands on previous sustainability studies by proposing a mechanistic life cycle GHG and economic operating cost assessment model for the coproduction of biochar and bioenergy from biomass residue feedstocks, with a case study for north-central Colorado presented. Production is modeled as a continuous function of temperature for slow pyrolysis, fast pyrolysis, and gasification systems. Biochar environmental benefits (C sequestration, N₂O suppression, crop yield improvements) are predicted in terms of expected liming value and recalcitrance. System-level net GHG mitigation is computed, and net returns are estimated that reflect the variable economic costs of production, the agronomic value of biochar based on agricultural limestone or fertilizer displacement, and the value of GHG mitigation, with results compared to the alternate use of char for energy production. Case study results indicate that slow pyrolysis systems can mitigate up to 1.4 Mg CO₂eq/Mg feedstock consumed, provided a favorable feedstock is utilized, production air pollutant emissions are mitigated, and energy coproducts are recovered. The model suggests that while financial returns are generally greater when char is consumed for energy (biocoal) than when used as a soil amendment (biochar), chars produced through high-temperature conversion processes will have greater GHG-mitigation value as biochar. The biochar scenario reaches economic parity at carbon prices as low as \$50/Mg CO₂eq for optimal scenarios, despite conservative modeling assumptions. This model is a step toward spatially explicit assessment and optimization of biochar system design across different feedstocks, conversion technologies, and agricultural soils.

Keywords: air pollution, biochar, biocoal, bioenergy, carbon sequestration, economic assessment, life cycle assessment, social cost of carbon

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Introduction

Biochar is the carbon-rich solid coproduct of thermochemical biomass conversion technologies. Its production and application to agricultural soils has been advocated as a greenhouse gas (GHG) mitigation strategy capable of rapid deployment, substantial total annual abatement potential, and significant cobenefits for agricultural system sustainability (Lehmann, 2007b; Molina *et al.*, 2009; Woolf *et al.*, 2010). Biochar is

characterized by stable aromatic C structures, low O and H to C ratios, low bulk density, moderate cation exchange capacity (CEC), and high ash content, pH, and surface area (Lehmann, 2007a). Because the C in biochar is derived from atmospheric CO₂ fixed in biomass via photosynthesis, the stable storage of biochar in soils represents a long-term removal of atmospheric C, i.e., terrestrial C sequestration. In addition, when applied as a soil amendment in agricultural systems, biochar has been shown, in many cases, to suppress N₂O emissions (a byproduct of the microbial metabolism of nitrogen fertilizer that dominates the GHG balance of many modern agricultural systems) (Clough & Condon, 2010;

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Singh *et al.*, 2010; Zheng *et al.*, 2012) and improve crop yields (Atkinson *et al.*, 2010; Jeffery *et al.*, 2011). Like any other organic matter addition to soil, biochar application affects a multitude of soil properties including bulk density, water-holding capacity, drainage, CEC, and pH, resulting in a substantial reengineering of the soil environment (Atkinson *et al.*, 2010) with respect to basic physical (Busscher *et al.*, 2010) and chemical properties (Gaskin *et al.*, 2010), water dynamics (Gaskin *et al.*, 2007), and macro- and microfauna viability (Liesch *et al.*, 2010; Lehmann *et al.*, 2011).

Biochar can be made from a variety of feedstock materials via several different thermochemical conversion pathways (Goyal *et al.*, 2008; Meyer *et al.*, 2011), resulting in chars with different chemical properties (Brewer *et al.*, 2009; Keiluweit *et al.*, 2010) and associated differences in recalcitrance (Spokas, 2010), agronomic performance (Atkinson *et al.*, 2010; Deal *et al.*, 2012), and overall economic value (Lin & Hwang, 2009; Yoder *et al.*, 2011). Thermochemical conversion involves the heating of biomass feedstocks in oxygen-restricted environments, causing the biomass to undergo a series of depolymerization, volatilization, and reorganization processes resulting in a mixture of low-molecular weight gases, high-molecular weight condensable liquid vapors, and solid char (Laird *et al.*, 2009). A range of thermochemical conversion technologies exist, with process conditions such as temperature, heating rate, and atmosphere optimized to favor either solid, liquid, or gas yields (Goyal *et al.*, 2008; Laird *et al.*, 2009; Brown *et al.*, 2011; Meyer *et al.*, 2011). Slow pyrolysis typically involves the low-temperature (300–550 °C) conversion of biomass with long residence times (e.g., hours), favoring yields of char (Gaunt & Lehmann, 2008; Laird *et al.*, 2009; Brown *et al.*, 2011). Fast pyrolysis is characterized by much faster heating rates, shorter residence times (e.g., seconds) and potentially higher temperatures (350–900 °C), and has been explored as a method of generating high yields of stable, energy-dense pyrolysis oils (Wright & Brown, 2007; Coleman *et al.*, 2010). Gasification implies the high temperature (600–1200 °C) intermediate-duration (10 s) auto-thermal conversion of biomass, at less than stoichiometric air–fuel ratios, into a primarily gaseous product rich in H₂, CO, and CH₄ that can be used for power generation (Alauddin *et al.*, 2010; Dasappa *et al.*, 2011; Mai Thao *et al.*, 2011). While each of these thermochemical conversion technologies have been used in the past for the production of fuels or feedstock chemicals, only gasification is typically employed for energy production today, and only in certain niche markets. However, pyrolysis technologies with the potential to coproduce fuels and biochar are currently the subject of intensive research efforts (Bridgwater *et al.*, 2002; Ringer *et al.*, 2006) and numerous commercial ventures (Butler *et al.*,

2011; Kauffman *et al.*, 2011; Solantausta *et al.*, 2012; US Biochar Initiative, 2012).

Although all of these processes will produce a solid, liquid, and gaseous product fraction, the yield and chemical composition of each fraction will vary considerably. As a result, the derived char can display a wide range in properties such as pH and CEC that will affect its function as a soil amendment (Spokas & Reicosky, 2009), and likewise the liquid and gas fractions will exhibit a range of different heating values that will dictate their value as energy products (Tsai *et al.*, 2007). In real-world thermochemical bioenergy systems not all product fractions are necessarily recovered for productive use, particularly in distributed small-scale systems where potential revenues from the smaller fractions are insufficient to justify capital investments in the required separation, filtration, and other cleanup equipment. Many slow and fast pyrolysis systems may in practice lack the capacity for pyrolysis gas recovery (Brick & Lyutse, 2010), while many gasification systems make no provision for generating energy from the pyrolysis oils filtered from the gas stream. Pyrolysis gas management is particularly important for system sustainability, as the CH₄ it contains is a potent GHG. Although not typically included in quantitative sustainability assessment studies, improper management (e.g., venting or incomplete flaring) of these gases has been hypothesized as a potentially significant source of GHGs and other air pollutants (Laird *et al.*, 2009; Brick & Lyutse, 2010).

Several aspects of biochar production system sustainability can be quantified scientifically. Life cycle assessment (LCA) is the systematic study of input and output flows of materials and energy across a given production chain to determine its full cradle-to-grave impact on areas such as anthropogenic GHG emissions, environmental quality, or human health (Finnveden *et al.*, 2009). LCA techniques have been widely applied to biofuel and bioenergy systems (Farrell, 2006; Bai *et al.*, 2010; Wang *et al.*, 2011). Several recent LCA studies and less formalized GHG-mitigation assessments have analyzed the coproduction of biochar and bioenergy from slow pyrolysis of various biomass feedstocks, considering GHG emissions associated with feedstock sourcing, bioenergy coproduction, and agronomic effects of biochar, in addition to the direct C sequestration effect (Gaunt & Lehmann, 2008; Roberts *et al.*, 2010; Woolf *et al.*, 2010; Hammond *et al.*, 2011). These studies conclude that such systems will mitigate 0.7–1.4 Mg CO₂eq per Mg of feedstock consumed. It is also recognized that there exists an inherent trade-off between bioenergy and biochar production (Fowles, 2007). Char produced through the thermochemical conversion of biomass has significant heating value and can be used as a fuel, or alternately the conversion system can be configured for

the complete combustion of the feedstock for maximum energy generation with no char production. These biochar GHG-mitigation studies generally conclude that biochar-producing systems can have greater GHG-mitigation value than systems configured for maximum bioenergy production, although the underlying analysis typically relies on coarse estimates of crop yield increases and N₂O suppression based on extrapolations of small numbers of greenhouse or field trials.

Significant work has also been conducted in the area of economic assessment of biochar systems (Islam & Ani, 2000; Lin & Hwang, 2009; McCarl *et al.*, 2009; Pratt & Moran, 2010; Roberts *et al.*, 2010; Galinato *et al.*, 2011; Shackley *et al.*, 2011; Yoder *et al.*, 2011). These studies typically find that the potential economic profitability of biochar production systems varies depending on the feedstock used (Lin & Hwang, 2009; Roberts *et al.*, 2010), the conversion technology employed (Pratt & Moran, 2010; Brown *et al.*, 2011), or the inclusion of carbon credits reflecting the social value of GHG mitigation (Pratt & Moran, 2010; Roberts *et al.*, 2010; Galinato *et al.*, 2011; Shackley *et al.*, 2011). One study has explored the implications of different production techniques and resulting variations in biochar properties for overall system performance, modeling the trade-off between product yield and product quality as conversion temperature increases (Yoder *et al.*, 2011). Taken together, most of the existing biochar sustainability literature tends to focus on a somewhat narrow and idealized case of dedicated biochar production in modern, efficient slow pyrolysis systems. However, practically speaking, much of the biochar available today or expected to be available soon will be either (i) produced in small-scale carbonization systems that lack the capacity for air pollutant mitigation and energy coproduct recovery; or (ii) a by-product from fast pyrolysis or gasification systems optimized for energy production rather than biochar production (e.g., Brick & Lyutse, 2010; Deal *et al.*, 2012).

The purpose of this study is to construct an integrated life cycle GHG and economic operating cost assessment tool around a detailed thermochemical biomass conversion dataset coupled with a mechanistic model of agronomic responses to assess the GHG mitigation and variable costs of systems that coproduce bioenergy and biochar, and to apply that tool to a biochar production case study. Yields and product qualities are compiled for slow pyrolysis, fast pyrolysis, and gasification across a range of reaction temperatures, with the recovery of individual product fractions adjusted as appropriate for the type of system modeled. Biochar recalcitrance is estimated as a function of production temperature, and agronomic response is modeled based on the biochar liming effect. While biochar addition affects a variety of physical, chemical, and biological soil properties, this

analysis focuses exclusively on the liming effect because (i) pH increases have been observed across a wide variety of biochar trials (Blackwell *et al.*, 2009; Streubel *et al.*, 2011); (ii) liming effects are relatively straightforward to simulate quantitatively (Galinato *et al.*, 2011); and (iii) meta-analyses show agronomic responses to be better correlated with pH changes than other biochar effects (Verheijen *et al.*, 2009; Jeffery *et al.*, 2011). Several aspects of the sustainability of biochar systems will vary regionally, including the availability of different feedstocks, the prices of system inputs and outputs, and the agronomic response to amendment of a specific soil type. A regional case study is presented to ground the analysis for a specific production case with realistic feedstock materials, transportation distances, energy pricing, and agronomic conditions. Following the presentation of the case study, the analysis is generalized to investigate sustainability in systems based on different conversion technologies and feedstocks, and with different transportation distances and native soil qualities, to enrich the analysis and bound the range of scenarios that are likely to achieve positive results.

Overall, this analysis expands on previous LCA and economic assessment methods in the literature to elucidate: (i) how system configuration and production-phase air pollutant management affect net environmental benefits and economic returns; (ii) how the quality, agronomic performance, and associated value of biochar changes across a range of thermochemical conversion conditions; and (iii) under what circumstances system optimization for environmental vs. economic outcomes are in competition.

Materials and methods

An integrated life cycle GHG and economic operating cost assessment tool is developed to examine the feasibility of establishing a biochar and bioenergy coproduction facility in north-central Colorado. This tool considers a variety of feedstocks and thermochemical conversion technologies, and models biochar properties, recalcitrance, and agronomic responses in a continuous, mechanistic manner. An LCA approach is employed, and the assessment is consequential in that it focuses on marginal emissions associated with a specific case study, the principle of system expansion is used to value the GHG impact of energy coproducts, and indirect effects are included where appropriate (specifically, indirect N₂O) (Brander *et al.*, 2009; Kauffman *et al.*, 2011). The functional unit considered is the management of 1 dry Mg of biomass residue. The life cycle inventory is constructed from a variety of sources, with many of the upstream embodied emissions and system expansion factors derived from the Argonne National Laboratory GREET model (Wang, 1999) version 1.8d. The impact assessment considers climate change impact using the metric of global warming potential (GWP).

The model follows the convention of treating CO₂ emissions from harvested biomass as neutral (e.g., assuming rapid biomass regrowth), but includes emissions of non-CO₂ GHGs such as CH₄ from the uncontrolled open burning of those feedstocks or from pyrolysis in scenarios where pyrolysis gas emissions are not captured or flared. Both CO₂ and non-CO₂ GHG emissions associated with life cycle fossil energy use are included where necessary. Capital embodied emissions are assumed to be similar across the different scenarios investigated and small enough relative to other life cycle emissions (Hill *et al.*, 2006) that they could be considered negligible. Likewise, the economic model specifically focuses on operating costs, otherwise known as variable costs, associated with the coproduction of energy and biochar, and does not include capital costs. Capital costs are not negligible for determining the overall feasibility of commercial systems; a recent economic analysis found that they contribute 27–31% to total biochar production costs from forestry residues, depending on system scale (Shackley *et al.*, 2011). However, focusing exclusively on operating costs is a reasonable approach for the first-order estimate of design trade-offs investigated in this study, and a fully developed enterprise budget-based profitability assessment of biochar production systems is outside the scope of the current analysis. An overview of the integrated analysis methods is presented below, while some of the more technical details are given in the online Supporting Information (SI).

Summary of case study scenarios

A case study is conducted for locating a thermochemical biomass conversion facility in Larimer County, Colorado, operating on one of two different locally available biomass residue feedstocks. The case study life cycle inventory includes: (i) operations associated with sourcing feedstock material; (ii) feedstock transport to a centralized conversion facility; (iii) processing and thermochemical conversion into biochar and energy coproducts with associated air pollutant emissions; (iv) transport of the resulting biochar to appropriate agricultural regions; and (v) biochar application to agricultural soils with associated direct sequestration of C, as well as displacement of agricultural inputs and suppression of N₂O emissions due to the liming effect (Fig. 1). Agronomic benefits are evaluated in the context of winter wheat production under two different sets of assumptions: (i) an initially limed system in which biochar application displaces an equivalent amount of agricultural limestone (aglime) application (Galinato *et al.*, 2011) while soil pH, nitrogen fertilizer inputs, and crop yield remain constant; and (ii) an initially nonlimed system in which biochar application increases soil pH, reducing the amount of N fertilizer required to maintain a given crop yield and partially suppressing N₂O emissions (Gaunt & Lehmann, 2008; Roberts *et al.*, 2010; Woolf *et al.*, 2010).

The case study evaluates the use of pine wood and slash sourced from Jackson County, Colorado (Fig. 2), as a waste biomass feedstock material. Forests in Jackson County have been devastated by an outbreak of the Mountain Pine Beetle (US Forest Service, 2012), and dead pine trees in proximity to roads, homes, and recreational areas are being cleared to reduce the

Fig. 1 System schematic illustrating the different life cycle phases and processes included in the assessment.

Fig. 2 Map showing the locations of feedstock sourcing (Jackson County, Colorado for pine and Larimer County, Colorado for spent grains), thermochemical conversion (Larimer County, Colorado) and agricultural soil incorporation (Hall County, Nebraska), and the highway network (US-14, I-25, and I-90) connecting them, for the hypothetical case study assessed.

risks of wildfire and falling trees. The material is typically piled and open-burned for disposal, with significant associated air pollutant emissions. In this analysis, the feedstock is transported via diesel truck to Larimer County, Colorado, where it is then ground prior to thermochemical conversion. The second biomass residue feedstock is spent grains produced at one of the many breweries in Larimer County. Similar to distillers grains with solubles (DGS) derived from corn ethanol production, these spent grains have value as animal feed capable of offsetting corn and soy consumption (Arora *et al.*, 2008). In this case, the spent grains are assumed to be dried in a natural gas-fired drier and consumed in a thermochemical conversion facility colocated with the brewery. Forgone emissions avoidance and revenues associated with alternate management of the biomass residue feedstocks are treated as opportunity costs or credits (Woolf *et al.*, 2010).

The analysis considers the conversion of these biomass feedstocks to char and energy coproducts through traditional charcoal production methods (carbonization), slow pyrolysis, fast pyrolysis, or gasification, as these are the most well-developed thermochemical conversion technologies (Meyer *et al.*, 2011). It is assumed that char is recovered for each technology and consumed locally as a substitute for coal (biocoal) in industrial boilers or power plants, or transported to Hall County, Nebraska and used as a soil amendment in winter wheat farms, applied a single time in the course of normal tillage operations at a rate of 25 Mg biochar per hectare. This particular site is selected as it is one of the closest areas with agriculture on native low-pH soils, specifically a Corzad loam of pH 5.6 and CEC of 15 cmol_c kg⁻¹ (Soil Survey Staff, Natural Resources Conservation Service, United States Department of Agriculture, 2012). In the same manner, other recovered product fractions are assumed to be used for energy generation locally, displacing the use of fossil fuels.

Greenhouse gas accounting

Standard UN Intergovernmental Panel on Climate Change values for the GWP of CH₄ and N₂O on a 100-year analytical time horizon are used for this analysis (Forster *et al.*, 2007), and values for CO, nonmethane hydrocarbons, and particulate matter emissions are taken from Grieshop *et al.* (2011) (Data S1 of the SI). The direct C sequestration value of biochar is estimated as a carbon stability factor (Hammond *et al.*, 2011) in CO₂-equivalent terms according to the equation

$$\text{CO}_{2,\text{sequest}} = 3.66 \left(1 - e^{(t_{1/2} \cdot \ln(0.5)) / \text{TH}} \right) \quad (1)$$

where 3.66 is the ratio of the molecular weight of CO₂ to that of C, $t_{1/2}$ the half-life of biochar in soil, and TH the analytical time horizon, in this case 100 years. The sequestration value of the char varies from zero to 3.66 Mg CO₂eq/Mg biochar-C as recalcitrance increases.

Feedstock sourcing

Harvest costs for beetle-kill pine are estimated from local US Forest Service studies (Lynch & Mackes, 2003; Duda, 2008),

and associated GHG emissions are estimated by applying a general emissions intensity term for the US forestry sector (US Department of Commerce, 2010). The authors have made a conservative assumption of attributing these costs and emissions fully to the biochar life cycle, even though large quantities of beetle-killed pine wood and slash will continue to be collected and piled for disposal via open burning in Jackson County regardless of the existence of a local biochar industry. As the alternate disposal method is uncontrolled open burning with high associated emissions of CH₄, particulate matter, and other products of incomplete production, an emissions credit based on data from McMeeking *et al.* (2009) is applied to the biochar production scenario for the avoidance of those emissions. It is assumed that the material will air dry to a moisture content of 10% after harvest but prior to transport, which is described in the next section. Feedstock handling and grinding costs at the conversion facility are estimated from Hess *et al.* (2009), and associated GHG emissions are computed using the same emissions intensity factor for the US forestry sector employed previously. Modeling of the spent grains feedstock sourcing is described in Data S2 of the SI.

Transport

Diesel fuel consumption and associated emissions are modeled for the transport of pine feedstock 100 miles (160 km) from Jackson County to Larimer County, Colorado, and for the transport of biochar to Hall County, Nebraska (400 mi/645 km, Fig. 2). The analysis models transport in heavy trucks using default payload capacity and fuel economy values from a trucking cost model (Norris, 2009) and from the GREET model. The full life cycle emissions associated with consuming a unit of diesel fuel are calculated by combining tailpipe CO₂ emissions along with an estimate of the embodied emissions associated with the upstream extraction, refining, and distribution of the fuel. Both estimates are derived from the GREET model (detailed in Data S3 of the SI).

Thermochemical conversion

Thermochemical conversion product yields and associated heating values, as well as char C-content, are modeled continuously as a function of temperature based on bench-top scale analyses from the literature specific to pine wood and spent grains feedstocks. A composite dataset simulating the slow pyrolysis of pine is assembled with data from Şensöz & Can (2002), Şensöz (2003), and DeSisto *et al.* (2010) for temperatures from 350 to 500 °C. Likewise, fast pyrolysis of pine is modeled from 400 to 600 °C with data from DeSisto *et al.* (2010), and gasification from 650 to 775 °C with data from Herguido *et al.* (1992) and Brewer *et al.* (2009). Note that the slow pyrolysis and gasification datasets are composites assembled from across multiple studies (the assumptions underlying which are detailed in Data S4 of the SI) and are thus somewhat more speculative than the fast pyrolysis dataset. No scaling factors are applied to bench-top scale results, and there is uncertainty around achieving these exact product distributions in a large-scale system. This is particularly true in the case of fast

pyrolysis, where the ability to match bench-top scale heat transfer rates at commercial scale is the subject of considerable research (Ringer *et al.*, 2006), and as a result this scenario should be viewed more as a bound than an expected value. However, it is not uncommon for assessment studies to assume bench-top scale product distributions directly for large-scale systems (Bridgwater *et al.*, 2002; Ringer *et al.*, 2006). In addition to these continuous process models, point estimates for other technologies and feedstocks are included in the model for comparison. The production of char from wood via carbonization in a more traditional batch charcoal kiln design is modeled with data from Pennise *et al.* (2001) to contrast traditional char production processes with modern ones. Although the authors are unable to identify continuous data or slow pyrolysis data for the spent grains feedstock in the literature, fast pyrolysis of spent grains at a single temperature (500 °C) is modeled for barley DGS as per Mullen *et al.* (2009). Details of the studies underlying these datasets are given in Table 1.

These product fractions are then corrected to reflect (i) the consumption of energy coproducts on-site to drive the endothermic pyrolysis process; or (ii) those that are not typically recovered in a useable form. It is assumed that the energy equivalent of 0.21 kg of pyrolysis gas is required to drive the pyrolysis (fast or slow) of 1 kg of dry biomass (Brown *et al.*, 2011), and that the higher energy requirements of higher temperature pyrolysis is compensated by the increased heating value of the gas produced under these conditions. In production regimes where gas yields are insufficient to meet this requirement, a fraction of one of the other products (pyrolysis oil for slow pyrolysis and char for fast pyrolysis) is consumed to fulfill the requirement. Gasification is auto-thermal and thus no products are consumed externally to drive the process, but it is assumed that the liquid fraction (tar) produced is not recovered for energy generation due to the relatively small yield and low quality. In the traditional charcoal kiln scenario, it is assumed that pyrolysis oil and gas are not recovered for energy production but rather are vented to the atmosphere or flared.

Recovered pyrolysis oils are modeled as being consumed locally to displace heavy fuel oil use on an energy-equivalent basis. Pyrolysis gases are modeled as being converted to electricity to offset grid electricity demand on-site. For biocoal scenarios, the char is assumed to displace local coal consumption

on an energy-equivalent basis. It is assumed that non-CO₂ GHG emissions rates are similar between heavy fuel oil and bio-oil (Solantausta *et al.*, 2012) and between coal and biocoal, so no additional GHG burden is calculated at this step. The details of these calculations are described in Data S4 of the SI.

Biochar amendment to agricultural soils

Biochar recalcitrance to biotic and abiotic mineralization after its application to soil is modeled as per Spokas (2010), which compiles data from a number of studies and maps biochar half-life estimates ranging over several orders of magnitude to char production temperature using char O : C ratio as proxy. A conservative fit of half-life vs. O : C is used here, as detailed in Data S5 of the SI. Biochar half-life estimates are then converted to CO₂-equivalent sequestration terms using Equation 1. The potential for a biochar 'priming effect' leading to changes in native soil organic matter dynamics is ignored in this analysis. Although some previous LCA studies attempt to include such an effect (e.g., Woolf *et al.*, 2010), recent studies suggest that the direction of the effect varies among soils and its magnitude is small (e.g., Stewart *et al.*, 2012).

Several sources in the literature report the liming value of various biochars in terms of calcium carbonate equivalence (CCE), along with their elemental makeup (Van Zwieten *et al.*, 2009, 2010a,b). The authors compile a composite dataset from these sources and supplement the analysis with additional biochar samples (see Table S1 of the SI for additional details). The measured CCE is regressed against biochar elemental composition including C, ash, and base element content using JMP Pro 9 (SAS Institute Inc., Cary, NC, USA). It is found that CCE is well-predicted based on the final base and ash content of the biochar (adjusted $R^2 = 0.78$, $P = 0.0047$) according to the relation

$$\text{CCE}(\%) = 5.378 + 1.582 \cdot B - 0.2136 \cdot A \quad (2)$$

where CCE is the acid-neutralizing capacity of material relative to that of pure CaCO₃, B the percentage of base elements (Ca, Mg, K, and Na) in the biochar, and A the total ash content, all on a mass basis. The base and total ash content of the pine feedstock are estimated from Bramryd & Fransman (1995), and those of spent grains using data on DGS from Spiehs *et al.* (2002). These mineral fractions are not perfectly conserved

Table 1 Thermochemical conversion studies used in model

Conversion scenario	Charcoal kiln (pine)	Slow pyrolysis (pine)	Fast pyrolysis (pine)	Fast pyrolysis (spent grain)	Gasification (pine)
Primary data source(s)	Pennise <i>et al.</i> (2001)	Şensöz and Can (2002), Şensöz (2003)	DeSisto <i>et al.</i> (2010)	Mullen <i>et al.</i> (2009)	Herguido <i>et al.</i> (1992), Brewer <i>et al.</i> (2009)
Temperature range* (°C)	–	350–500	400–600	500	650–775
Feedstock	Eucalyptus	Pine, pine bark	Pine	Barley DGS	Pine chips
Max particle size (mm)	–	10, 0.60	0.4	2	10
Atmosphere	Air	Pyrolysis gas	N ₂	N ₂	Steam/N ₂ , Air/N ₂
Reactor type	Circular Brazilian kiln	Fixed bed	Fluidized bed	Fluidized bed	Fluidized bed

*Temperature range used in this model; may be a subset of the full temperature range reported in the source study.

during thermochemical conversion (Gaskin *et al.*, 2008; Novak *et al.*, 2009) and a uniform recovery factor of 80% is assumed for both base and total ash content of the feedstocks across all scenarios.

The liming effect is then evaluated in the context of a previously limed scenario in which aglime consumption is displaced, and a previously unlimed scenario in which fertilizer is displaced. In the first case, biochar displaces an equivalent amount of aglime of 100% CCE value (pure calcitic limestone). A fraction of the C in aglime is released as CO₂ during the re-acidification of the soil over time (West & McBride, 2005), and this avoided emission is credited to the biochar in addition to the embodied emissions from the manufacture and distribution of the displaced aglime as estimated in GREET. Final soil pH and associated crop yields are assumed constant in this scenario. In the second case, it is assumed that soil is not initially being limed, and any biochar additions will result in an increase in soil pH, computed based on the initial soil CEC (a proxy for soil buffering capacity) according to the Adams–Evans method (Evans & Adams, 1962) and comparable in duration to that encountered with an application of aglime as reported in Lukin & Epplin (2003). In an acid soil, this pH increase can result in an improvement in crop productivity and associated reduction in the amount of nitrogen fertilizer needed to maintain a given yield (assuming a baseline fertilizer rate below that of maximum yield response), as well as a reduction in N₂O emissions based on the decreased fertilizer application rate combined with a pH-mediated reduction in the N-to-N₂O emissions factor (Clough & Condon, 2010; Zheng *et al.*, 2012). The associated calculations are detailed in Data S5 of the SI.

Economic assessment

An economic assessment of system operating costs is performed by applying prices to all life cycle model inputs of material and labor, and all system outputs, as detailed in Data S6 and Table S2 of the SI. In the case of the spent grains feedstock, DGS prices reflect the opportunity cost of using the material as a feedstock rather than as animal feed. Prices for commodities subject to high price volatility (e.g., some fuels and energy-intensive products such as nitrogen fertilizer) are computed based on multiyear averages. The value of biochar is inferred from the cost of aglime or nitrogen fertilizer displaced, depending on the scenario. All prices are adjusted for inflation to 2012 US dollars.

Complex nonmarket valuation models can be conducted for a variety of environmental externalities associated with the energy and agricultural sectors (e.g., air and water pollutant emissions) (Keske, 2011). However, the nonmarket valuation in the current analysis is limited to the pricing of GHG emissions to quantify the social benefit of systems that mitigate GHGs relative to the fossil fuel *status quo*. Marginal damage estimates are taken from the United States Government Interagency Workgroup on Social Cost of Carbon (2010), with a median estimate of the social cost of carbon (SCC) of \$23.09/Mg CO₂eq when adjusted for inflation. Note that while most carbon emissions trading systems focus on a narrow set of GHGs (e.g. CO₂, CH₄, and N₂O as per the Kyoto protocol) and approved mitigation

technologies, CO₂-equivalent forcings from particulate matter emissions and direct sequestration of CO₂ as biochar-C are monetized here as well to reflect the best estimate of total system climate impact. Conversely, the price of carbon necessary to achieve parity of returns between a low-financial return, high GHG-mitigation biochar scenario and a higher return but lower GHG-mitigation biochar scenario can be calculated as follows:

$$P_C = \frac{R_{\text{coal}} - R_{\text{char}}}{\text{GHG}_{\text{char}} - \text{GHG}_{\text{coal}}} \quad (3)$$

where R denotes financial returns (\$/Mg feedstock) in the absence of nonmarket or social costs of carbon, GHG denotes the net GHG mitigation of the scenario (Mg CO₂eq/Mg feedstock), and the subscripts *coal* and *char* correspond to the biochar and biochar scenarios, respectively.

Results

Slow pyrolysis: effects of technology configuration

The analysis suggests substantial GHG mitigation but weak economic performance for the slow pyrolysis case study scenario assessed. The net mitigation of 1.41 Mg CO₂eq (100-year time horizon) and a net revenue of −\$78 (i.e., unprofitable operation) are predicted for every metric ton of dry pine feedstock processed through this system (Fig. 3), assuming slow pyrolysis at 500 °C with pyrolysis oil recovery, biochar application at a rate of 25 Mg ha^{−1} (2% by mass at an incorporation depth of 10 cm), and biochar valuation based on fertilizer displacement (note the sign convention of showing GHG avoidance and revenues as positive, and GHG emissions and costs as negative). The largest positive contributor to GHG mitigation is the direct sequestration of carbon in soil as biochar-C. Under these conversion conditions, the model predicts a char mass yield of 29% with a C concentration of 89% and an O : C ratio of 0.21, corresponding to a conservative soil half-life estimate of 240 years and a resulting C sequestration value of 0.76 Mg CO₂eq/Mg feedstock processed. System economic returns are dominated by pyrolysis oil production at a 21% yield and a higher heating value of 34.1 MJ kg^{−1}. This results in a heavy fuel oil displacement rate of 0.8 : 1 that contributes 0.61 Mg CO₂eq/Mg and \$61/Mg to the system GHG mitigation and financial returns, respectively. Significant costs are incurred to harvest the pine feedstock (−\$109/Mg) and from life cycle energy use associated with transport, handling, and grinding of the feedstock and transport and field incorporation of the derived char (−\$63/Mg), although the GHG burden associated with these operations (−0.03 and −0.05 Mg CO₂eq/Mg) is relatively small. Smaller GHG benefits are accumulated from the avoidance of open burning of the pine residue material (0.10 Mg CO₂eq/Mg) and the

Fig. 3 Detail of the GHG balance and economic returns for the slow pyrolysis of pine feedstock at 500 °C, assuming biochar application at 25 Mg ha⁻¹ to a Corzad loam soil of pH 5.6 and CEC 15 cmol_e kg⁻¹. Valuation of GHG mitigation is based on a \$23/Mg CO₂eq estimate of the social cost of carbon using a 100-year analytical time frame for the calculation of CO₂ equivalence.

displacement of fertilizer and N₂O emissions suppression (0.03 Mg CO₂eq/Mg). Finally, monetizing total system GHG mitigation at a SCC of \$23/Mg CO₂eq contributes a further \$33/Mg of revenue. When all of these terms are combined the system shows a strong GHG-mitigation potential, although production costs

are so high that it would operate at a net loss even before capital costs are considered.

These GHG mitigation and economic return estimates are highly dependent on the configuration of the char production system (Fig. 4). For the same scenario relying on a traditional batch carbonization method with

Fig. 4 Net GHG mitigation and economic returns for different conversion technology configurations, with the resulting char used either as biochar or bio-coal.

uncontrolled air pollutant emissions and no energy coproduct recovery, net system GHG-mitigation value drops to virtually zero; this is primarily the result of losing the mitigation value of pyrolysis oils displacing heavy fuel oil use and the accumulation of an additional GHG burden of $-0.72 \text{ Mg CO}_2\text{eq/Mg feedstock}$ from the uncontrolled release of high-GWP pyrolysis gases. Associated net economic returns drop to a deficit of $-\$171/\text{Mg feedstock}$ processed. System GHG performance is improved somewhat with the addition of pyrolysis gas flaring, although performance is better still for modern slow pyrolysis systems that completely combust excess pyrolysis gases in the course of electricity generation. Net economic returns are particularly poor across all system configurations that do not include recovery of the pyrolysis oil fraction. For both traditional kilns and slow pyrolysis systems, the char produced has relatively high heating value (31 MJ kg^{-1}). As a result, the biocoal scenario leads to better GHG and economic results from the displacement of coal than the biochar scenario does through the direct sequestration of C and displacement of fertilizer and suppression of N_2O in agricultural soils. GHG mitigation and economic returns are reasonably well correlated across all of the scenarios plotted in Fig. 4 (Spearman $\rho = 0.70$, $P = 0.05$), suggesting that optimizing the system configuration to maximize profitability will also tend to maximize GHG performance even in the absence of carbon social cost valuation. Monetizing system GHG mitigation increases net returns, but even the best-performing pine slow pyrolysis scenario still operates at a significant loss.

Fast pyrolysis: effects of feedstock choice

The choice of feedstock can significantly affect system GHG balance and profitability. Pine and spent grains

feedstocks are contrasted directly for a fast pyrolysis process at 500°C in Fig. 5, and spent grains system performance is characterized by lower net GHG-mitigation value (0.97 vs. $1.58 \text{ Mg CO}_2\text{eq/Mg feedstock}$) but higher economic returns ($-\$8$ vs. $-\$45/\text{Mg feedstock}$, again both net losses) than pine. The difference in GHG performance is driven primarily by the emissions associated with sourcing the feedstock itself. While in the case of pine the avoidance of air pollution from open burning more than offsets emissions from feedstock harvest, the spent grain carries a large opportunity emissions burden ($-0.44 \text{ Mg CO}_2\text{eq/Mg}$) associated with diverting the feedstock away from use as an animal feed replacing corn and soy consumption, as well as significant emissions associated with drying the material down to 10% moisture content ($-0.19 \text{ Mg CO}_2\text{eq/Mg}$). The spent grain scenario is also characterized by lower biochar-C concentration (51%) and higher pyrolysis oil heating value (32.9 MJ kg^{-1}) as compared with the pine scenario (76% and 24.7 MJ kg^{-1}). However, the resulting effects of less direct soil C sequestration and greater heavy fuel oil displacement roughly cancel each other out on a GHG basis. Although the spent grain feedstock carries a large opportunity cost ($-\$91/\text{Mg}$), it is still somewhat lower than the harvest cost of pine ($-\$109/\text{Mg}$). This, in addition to larger revenues associated with the greater heavy fuel oil replacement rate, makes the spent grains scenario more profitable than the pine feedstock, although net returns are still slightly negative.

Biochar valuation

This analysis includes the valuation of biochar by two different methods (aglime displacement and fertilizer displacement), the results of which are shown in

Fig. 5 Simplified detail of the GHG balance and economic returns for the fast pyrolysis of pine and spent grains feedstocks at 500°C .

Table 2 Different biochar valuation results calculated over the lifetime of the liming effect

		ΔpH	Displacement rate (kg/Mg char)	GHG mitigation (kg CO ₂ eq/Mg char)	Value (\$/Mg char)*
Case 1	Aglime displacement	0	61	Avoided emissions (embodied & field): 53	0.53
Case 2	N fertilizer displacement	0.13	3.9	Avoided emissions (embodied & field): 28 Additional pH-mediated N ₂ O suppression: 58	1.48

*Does not include valuation of GHG mitigation.

Table 2 for the scenario of modern slow pyrolysis of pine at 500 °C described above. When this biochar is used in place of aglime, it results in the displacement of 61 kg lime per Mg char (based on the predicted CCE value), which is associated with the avoidance of 53 kg CO₂eq/Mg biochar and a value of \$0.53/Mg. In the alternate scenario where the biochar is introduced into a nonlimed system, it is predicted to increase soil pH by 0.13 units, improving fertility by approximately 2% and allowing a reduction in fertilizer application rate of 11 kg ammonia per hectare for the duration of the liming effect. The combined effect of N₂O reduction and nitrogen fertilizer embodied emissions avoidance in this case is 87 kg CO₂eq/Mg biochar, and the fertilizer savings is valued at \$1.48/Mg biochar. For the remainder of the analysis, the fertilizer displacement method is used for biochar valuation.

However, additional emissions and costs are incurred for the transport of biochar from the thermochemical conversion facility to the farm and for soil application, detracting from its overall value. This effect can be bounded in terms of the maximum transport distance possible before transport and application emissions or costs outweigh agricultural GHG-mitigation benefits (not including the direct C sequestration value of the biochar) or revenues with or without carbon social cost valuation, given a farm soil buffering capacity. For scenarios where fertilizer displacement is considered, positive GHG mitigation is the least-binding criteria, and maximum transport distances of 1640 and 520 km can be tolerated for biochar that will be applied to soils with a CEC of 5 and 20 cmol_e kg⁻¹, respectively (assuming an initial pH of 5). Achieving positive revenues is more constraining, with maximum transport distances of 50 and 125 km with and without carbon valuation, respectively, for low buffering capacity soils (CEC of 5 cmol_e kg⁻¹). When aglime displacement is considered, incorporation costs will always outweigh biochar value, even at transport distances of zero. Note that the case study scenario includes a biochar transport distance of 645 km and application to a native soil of CEC of 15

cmol_e kg⁻¹. In this case, the costs associated with biochar transport and incorporation outweigh the agronomic value of the char, even when the nonmarket values of agronomic GHG mitigation are calculated. The associated emissions value is negative when biochar is valued as displacing aglime but positive when displacing fertilizer.

Biochar vs. biocoal across conversion technologies

This analysis finds that financial returns from using char as biocoal are higher than that of using it as biochar across all of the production technologies, conversion temperatures, and agricultural soil properties explored. However, the net GHG-mitigation value of biochar does exceed that of biocoal under certain conditions, as plotted for three different production technologies as a function of conversion temperature and soil CEC in Fig. 6. Regions shaded bright red and purple represent regimes for which biochar GHG mitigation outperforms that of biocoal by up to 0.11 Mg CO₂eq/Mg feedstock consumed. This occurs for fast pyrolysis scenarios at high conversion temperatures and low soil CEC values, and across all gasification scenarios investigated. These conversion conditions are associated with relatively high char C-concentration, recalcitrance, and liming values, and relatively low heating values. As biochar is shown to outperform biocoal on a GHG basis but underperform on a revenue basis, Equation 3 can be applied to compute the price of carbon necessary to make up the revenue deficit (P_C). These results are shown for gasification in Fig. 7, indicating that biochar will be more valuable than biocoal at carbon prices as low as \$49/Mg CO₂eq when produced at high conversion temperatures and used in soils with low buffering capacity, to as high as \$155/Mg CO₂eq under the opposite conditions.

Sensitivity analysis

Sensitivity analysis is performed on the price of carbon for biochar–biocoal parity (P_C), as this parameter

Fig. 6 The relative GHG-mitigation advantage of biochar over biocoal, plotted for char produced from multiple conversion technologies as a function of production temperature and soil CEC. Regions in which biochar outperforms biocoal are plotted in red and purple.

Fig. 7 The price of carbon necessary for biochar revenue to equal biocoal revenue (P_C), for char produced from the gasification of pine, as a function of production temperature and farm soil CEC.

encompasses both the GHG and revenue aspects of the assessment into a single metric. Sensitivity of results is evaluated in response to a standardized perturbation to key input parameters, as is commonly done for speculative assessment scenarios where rigorous bounding of total uncertainties is problematic (e.g., Bridgwater *et al.*, 2002; Bergqvist *et al.*, 2008). The analysis of pine gasification at 700 °C and biochar application in soils of pH 5 and 10 $\text{cmol}_e \text{kg}^{-1}$ CEC (see Fig. 7) is perturbed by increasing or decreasing the value of key model input parameters by 1%, and the resulting response in P_C is plotted in Fig. 8. This analysis shows that results are dri-

Fig. 8 Sensitivity analysis for the carbon price for biochar–biocoal parity (P_C), for char produced from pine gasification at 700 °C and applied to soil of pH 5.0 and CEC of 10 $\text{cmol}_e \text{kg}^{-1}$.

ven primarily by the physical properties of the char; a 1% reduction in biochar C-content increases P_C by 9%, while a 1% increase in char heating value increases P_C by 8%. Input parameters of intermediate influence (0.2–0.9% response to a 1% perturbation) include labor and energy prices and variables describing the duration of the char liming effect and the stability of biochar in soil, as well as the magnitude of the baseline farm soil N_2O emission rate. The char CCE regression coefficient and the price of fertilizer exert minimal influence on overall results (0.1% response to a 1% perturbation). The model is even less sensitive to changes in biochar incorporation costs.

Discussion

The goal of this analysis is to develop an integrated life cycle GHG and economic operating cost assessment tool applicable to biochar production in north-central Colorado. The model captures some of the diversity in biochar production technologies encountered in the real world, models the C sequestration and agronomic value of biochar in a mechanistic manner, and estimates the value of biochar based on its displacement of other agricultural inputs. Overall, the analysis suggests that: (i) slow pyrolysis biochar systems based around modern conversion technologies can mitigate up to 1.4 Mg CO₂eq/Mg pine wood feedstock in Colorado, but this performance depends on air pollutant management and energy coproduct recovery; (ii) the locally available biomass residue feedstocks considered are generally too expensive for system profitability; (iii) the agronomic value of biochar makes only a very small contribution to total system GHG mitigation and profitability; and (iv) that using char as biochar only unambiguously outperforms using it as biocoal when it is produced via high-temperature fast pyrolysis or gasification and when GHG mitigation is valued above \$50/Mg CO₂eq.

The net GHG-mitigation estimate for the biochar-producing slow pyrolysis system assessed in this case study is very comparable to that estimated by other assessment studies in the literature, which typically report mitigation values between 0.6 and 1.4 Mg CO₂eq/Mg depending on the details of the scenario (Gaunt & Lehmann, 2008; Roberts *et al.*, 2010; Woolf *et al.*, 2010; Hammond *et al.*, 2011; Kauffman *et al.*, 2011), as summarized in Data S7 of the SI. The relatively low GHG footprint of sourcing the pine feedstock considered here contributes to the positive overall results. Also, the assumption of pyrolysis oils being used to displace heavy fuel oil locally rather than being combusted directly for electricity generation further improves the GHG balance relative to other studies, as fuel oil always has a high GHG footprint whereas the footprint of electricity generation being displaced can vary regionally. This analysis implicitly assumes that total regional biochar production is sufficiently low that local fuel oil demand will fully consume the associated pyrolysis oil coproduct.

The lack of profitability of the slow pyrolysis scenario modeled in this case study is somewhat unexpected, although not inconsistent with other economic assessment studies that report both positive and negative net returns depending on the feedstock used (Roberts *et al.*, 2010), conversion technology employed (Pratt & Moran, 2010), and price of carbon (Galinato *et al.*, 2011). This result is primarily driven by the cost of feedstock sourcing; spent grains have a significant opportunity cost

stemming from their value as animal feed, and the harvest of beetle-killed pine is very costly. Studies suggest that large quantities of cellulosic biomass will be available at the national level for the future bioenergy industry at prices starting as low as \$25–60/Mg (Jain *et al.*, 2010; Brown *et al.*, 2011; Egbendewe-Mondzozo *et al.*, 2011; U.S. Department of Energy, 2011), a range low enough to move some of the scenarios analyzed here into profitability if located in appropriate regions.

The finding that biocoal will be more profitable across all scenarios analyzed and will mitigate more GHG emissions when char is produced by slow pyrolysis and under most fast pyrolysis conditions also runs counter to the conventional wisdom. While previous studies typically make assumptions about crop yield response and N₂O suppression that are largely independent of specific biochar physical and chemical properties, those authors find that biochar outperforms energy-maximizing scenarios on a GHG basis. There is, however, wide disagreement as to whether that result is sensitive or insensitive to factors such as biochar recalcitrance in soil (Gaunt & Lehmann, 2008; Hammond *et al.*, 2011), displaced electricity GHG footprint (Woolf *et al.*, 2010), or even system analytical boundaries (Roberts *et al.*, 2010). This study is similar in that it reports a nuanced result in which biochar will only outperform an energy-maximizing scenario within certain limits of production temperature and application rate, outside of which the recalcitrance level and liming effect are too low to produce GHG mitigation in excess of what would be achieved from displacing fossil coal emissions. However, both the biochar and biocoal scenarios lead to significant total GHG mitigation, and the difference between these two values is relatively small and thus highly sensitive to certain modeling assumptions, as illustrated in the sensitivity analysis.

The approach of this study to estimate the GHG benefits and economic value of biochar based solely on its liming potential is very conservative, in that it neglects other potential benefits from biochar application in agricultural systems. In addition to liming effects, evidence suggests that biochar can have agronomic value with respect to water dynamics, nutrient retention, and microbial activity. In some cases, the underlying drivers (CEC, surface area) of these effects will show the same positive relationship with production temperature (Lehmann, 2007a) that liming capacity is predicted to have here. It is effects such as these, rather than a transient liming effect, that underlie the long-term improved fertility of the Terra Preta soils that inspired the biochar concept (Glaser *et al.*, 2001; Laird *et al.*, 2009). However, such effects have been neglected here due to an incomplete understanding of their mechanism that makes assessment extremely challenging. While it is debatable

whether the biochar market is sufficiently well developed that current prices accurately reflect the underlying agronomic value of biochar, the fact that there are a diversity of niche markets willing to pay several orders of magnitude more for biochar (Keske & Lohman, 2012) than the valuation estimated here strongly suggests that biochar has additional benefits beyond what can be explained through the liming effect alone. The analysis is also conservative to the extent that no attempt is made to correct for the decreasing value of the biochar alternative as char ash concentration increases with production temperature, resulting in a fuel with greater propensity for slagging (Vamvuka *et al.*, 2010).

Finally, it is also likely that the true GHG-mitigation value of biochar is underestimated with the carbon stability factor approach used here and in most other biochar LCA studies. While this analysis only considers the GHG-mitigation impact of the fraction of biochar-C remaining in the soil at the end of 100-year analytical time horizon, a more dynamic accounting approach would consider the CO₂-equivalent value of the transient sequestration of the char volatile fraction. Dynamic accounting of changes in carbon sinks is becoming more common in bioenergy LCA studies (O'Hare *et al.*, 2009; Cherubini *et al.*, 2011), and it is conceivable that it might significantly raise the GHG-mitigation estimate of biochars, particularly those with relatively low soil half-lives relative to the analytical time horizon.

These limitations notwithstanding, the authors believe this study makes an important step in improving the methodology of sustainability assessment in biochar systems. It expands the scope of analysis beyond slow pyrolysis systems to account for the diversity of thermochemical technologies that are currently producing biochar around the world, and includes effects such as conversion air pollutant emissions that are neglected in other studies. The transition to continuous, mechanistic estimates of thermochemical conversion product yields and biochar recalcitrance and agronomic benefits has the potential to improve assessment accuracy and address the trade-offs inherent in the design of such systems. This is consistent with the increasingly common use of biophysical models of feedstock production and soil management in bioenergy LCA (Zhang *et al.*, 2010) and economic assessment studies (Jain *et al.*, 2010), allowing for the regionally specific assessment of system performance and sustainability that reflects variance in climate, soil type, and land use history. Further development of such mechanistic assessment methods will enable spatially explicit assessments of biochar systems in the context of feedstock availability, energy coproduct demand, and agricultural needs, facilitating the design and siting of biochar production facilities to maximize both profitability and environmental benefits.

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Supporting Information

Additional Supporting Information may be found in the online version of this article:

- Data S1.** Greenhouse gas accounting.
- Data S2.** Feedstock sourcing.
- Data S3.** Transport.
- Data S4.** Thermochemical conversion product yields and qualities.
- Data S5.** Biochar amendment to agricultural soils.
- Data S6.** Economic assessment.
- Data S7.** Results of other biochar assessment studies.