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Article

Empirical Evidence for the Potential Climate Benefits of Decarbonizing Light Vehicle Transport in the U.S. with Bioenergy from Purpose-Grown Biomass with and without BECCS

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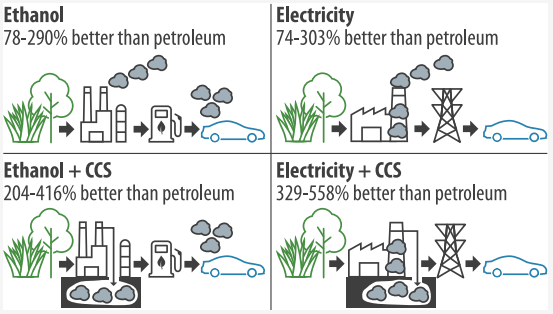
ABSTRACT: Climate mitigation scenarios limiting global temperature increases to 1.5 °C rely on decarbonizing vehicle transport with bioenergy production plus carbon capture and storage (BECCS), but climate impacts for producing different bioenergy feedstocks have not been directly compared experimentally or for ethanol vs electric light-duty vehicles. A field experiment at two Midwest U.S. sites on contrasting soils revealed that feedstock yields of seven potential bioenergy cropping systems varied substantially within sites but little between. Bioenergy produced per hectare reflected yields: miscanthus > poplar > switchgrass > native grasses ≈ maize stover (residue) > restored prairie ≈ early successional. Greenhouse gas emission intensities for ethanol vehicles ranged from 20 to −179 g CO_2e MJ^{-1}: maize stover ≫ miscanthus ≈ switchgrass ≈ native grasses ≈ poplar > early successional ≥ restored prairie; direct climate benefits ranged from ∼80% (stover) to 290% (restored prairie) reductions in CO_2e compared to petroleum and were similar for electric vehicles. With carbon capture and storage (CCS), reductions in emission intensities ranged from 204% (stover) to 416% (restored prairie) for ethanol vehicles and from 329 to 558% for electric vehicles, declining 27 and 15%, respectively, once soil carbon equilibrates within several decades of establishment. Extrapolation based on expected U.S. transportation energy use suggests that, once CCS potential is maximized with CO_2 pipeline infrastructure, negative emissions from bioenergy with CCS for light-duty electric vehicles could capture >900 Tg CO_2e $year^{-1}$ in the U.S. In the future, as other renewable electricity sources become more important, electricity production from biomass would offset less fossil fuel electricity, and the advantage of electric over ethanol vehicles would decrease proportionately.



INTRODUCTION

Dedicated bioenergy crops combined with carbon capture and storage (BECCS) are a feature of almost all Intergovernmental Panel on Climate Change (IPCC) mitigation scenarios that constrain the global temperature increase to 1.5 °C by 2100.[1,2] In these scenarios, biomass is used initially to offset transport sector fossil fuel use by substituting biomass-based fuels for the petroleum (gasoline and diesel) now used to power light-duty vehicles.[3] Later, once carbon capture and storage (CCS) technologies become available, captured biomass carbon (C) is subsequently transferred as CO_2 to long-term geologic storage (BECCS). The energy so released is converted either into liquid fuels to offset hard-to-replace petroleum products (e.g., aviation and long-haul transport fuels) or into electricity for diverse end uses including electric vehicles (EVs).

Thus, future liquid fuel projections are based on a diminishing use for transportation to a point where most but not all transportation needs are met by electricity from a variety of sources in addition to biomass, including wind, solar, nuclear, and hydropower facilities. That said, (a) in the U.S., it will be decades before electric infrastructure is sufficient to deliver electricity to a substantial fraction of the entire U.S. light-duty vehicle fleet[3,4] and (b) once the fleet is converted to electric, the continued need for CO_2 drawdown (negative C emissions) will depend at least in part on biomass to provide CO_2 for geologic sequestration (BECCS).[5] While there are serious land availability limitations for different BECCS scenarios,[6,7] at least in the U.S., a substantial fraction of future biomass needs can come from herbaceous crops grown on former agricultural lands.[8−11] Although this is not necessarily the case elsewhere, requiring our analysis to be extrapolated with care, there is, notwithstanding, an important need globally to understand the climate implications of biomass-derived fuels for light-duty vehicles both during the expected transition to electric vehicles post-mid-century[12] and following maximum

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vehicle electrification, when biomass may be used for CCS while producing electricity.

All components of the measured climate impacts of energy production from mature stands of cellulosic bioenergy crops have yet to be compared experimentally,[9,13−16] and thus direct mitigation estimates remain largely uninformed by direct empirical evidence. When considering liquid transportation fuel as the sole end product,[17−20] life cycle models estimate emission intensities that range from C negative (i.e., net CO_2e uptake) to C positive (i.e., net CO_2e release): from −396 to 61 g CO_2e MJ^{-1} for switchgrass,[21,22] −139 to 13 for miscanthus,[23,24] and −150 to 164 for the maize stover.[22,25,26] The U.S. Environmental Protection Agency[27] estimates an overall range of −10 to 27 g CO_2e MJ^{-1}. This same uncertainty extends to estimates of mitigation potentials for powering electric vehicles and for either ethanol- or electric-powered vehicles used with carbon capture and storage (CCS).[28]

Here, we report the first empirical assessment comparing direct climate impacts of multiple cellulosic bioenergy crops for different bioenergy end uses. Earlier studies, e.g.,[15,25,29−34] while crucially important for informing specific life cycle assessment (LCA) attributes in aggregate, have either not compared feedstocks side by side, or on different soils, or have not used field-based measurements of all important CO_2e flows.[8] Rather, most if not all LCA comparisons to date have drawn on aggregated estimates from multiple studies and/or simulated values from biogeochemical models.[35−37] The absence of comprehensive field-based studies adds to already large uncertainty due to potential indirect effects of land-use change resulting from biomass production on lands now used for food production.[38−41] Thus, current debates on the climate impacts of large-scale implementation of biomass-based renewable fuels suffer from a lack of empirical knowledge of soil greenhouse gas (GHG) emissions, soil organic C (SOC) dynamics, and spatial variability in yields of bioenergy crops.

The current LCA models based on average numbers and specific assumptions are producing multiple more or less plausible scenarios, while failing to predict verifiable real-world effects.[42] And only a handful of studies to date have provided comprehensive measurements of all major components contributing to net CO_2e balances of biomass production.[9,13,43,44] Moreover, we are aware of no empirical studies that have considered alternative end uses—ethanol vs electric, both with and without CCS—of potential value for integrated assessment and earth system models that include a variety of alternative transportation and fuel switching strategies,[2] including ethanol- and electric-powered vehicles.

In this study, we directly compare climate benefits and tradeoffs for a range of anticipated bioenergy feedstocks on two contrasting soils in a climate region that supports rain-fed crop production. We grew seven feedstocks at two former agricultural sites in the United States Midwest, one on soils of average fertility and another on soils of high fertility (Figure S1a).[45] Feedstocks included the maize stover (25−50% of the residue remaining from a continuous no-till maize crop); perennial monocultures of switchgrass, miscanthus, and poplar trees; and perennial polycultures of native grasses, early successional vegetation, and restored prairie. From planting in 2008[46] through 2016, we measured components of each system's direct global warming impact (GWI or net greenhouse gas balance) including aboveground biomass production, soil nitrous oxide (N_2O) and methane (CH_4) fluxes, soil C accumulation, and farming inputs. We combine these results to estimate the fossil fuel offsets associated with four different scenarios for powering light-duty vehicles: first is conversion of cellulosic biomass to ethanol (or its equivalent); second is conversion of the biomass to electricity; and third and fourth are inclusion of CCS for each of the first two scenarios.

MATERIALS AND METHODS

Site Description. Field research was conducted at low- and high-fertility sites that comprise the Biofuel Cropping System Experiment at the U.S. Department of Energy's Great Lakes Bioenergy Research Center (GLBRC). The low-fertility site is located at the W.K. Kellogg Biological Station's Long-Term Ecological Research site in southwest Michigan (KBS, 42°23′47″N, 85°22′26″W, 288 m elevation), and the high-fertility site is located at the Arlington Agricultural Research Station in south-central Wisconsin (ARL, 43°17′45″N, 89°22′48″W, 315 m elevation). Soils at the Michigan site are in the Alfisol soil order[45] (hereafter called the "Alfisol" site) composed of co-mingled Kalamazoo and Oshtemo soil series, both classified as well-drained Mesic Typic Hapludalf loams >1 m depth formed under deciduous forest on parent materials derived from glacial outwash sand intermixed with loess.[47,48] The mean annual air temperature at this site is 10.1 °C (from 1981 to 2010), ranging from a monthly mean of −3.8 °C in January to 22.9 °C in July. Precipitation averages 1005 mm $year^{-1}$ (1981−2010), evenly distributed seasonally.[49] Soils at the Wisconsin site are in the Mollisol soil order (hereafter called the "Mollisol" site),[45] where the predominant soil is in the Plano series, classified as a well-drained mesic Typic Argiudoll silt-loam of >1 m depth formed under tallgrass prairie. The mean annual temperature at the site (1981−2010) is 6.8 °C, ranging from a daily average of −7.7 °C in January to 21.0 °C in July. Precipitation averages 869 mm $year^{-1}$ (1981−2010), evenly distributed seasonally.[46] At both sites, potential evapotranspiration exceeds precipitation for about 4 months of the year. At the time of planting (2008), soil organic C contents were 1.08 and 1.99% for the Alfisol and Mollisol sites, respectively.[46,50]

At both locations, seven candidate bioenergy cropping systems were established in 2008 in a randomized complete block design. Each of five replicate blocks contained seven 27 × 43 m (0.12 ha) treatment (cropping system) plots with at least 12 m between adjacent plots in any direction. The cropping systems comprised a gradient of plant diversity (single to multiple species) and chemical inputs (high to low) and included (1) maize (*Zea mays* L.) in a continuous (maize−maize) rotation, with 50% of its stover (the crop residue that remains after grain harvest) removed as bioenergy feedstock; (2) switchgrass (*Panicum virgatum*), a prairie grass native to North America; (3) giant miscanthus (*Miscanthus* × *giganteus* Greef & Deuter ex Hodkinson & Renvoize), a sterile hybrid cross between the Asian grasses *Miscanthus sinensis* and *Miscanthus sacchariflorus*; (4) poplar trees, a hybrid cross between non-native poplar species *Populus nigra* and *Populus maximowiczii* (variety A. Henry "NM6"); (5) a mixture of six native prairie grasses, including switchgrass; (6) early successional vegetation that grows spontaneously following agricultural abandonment; and (7) restored native prairie. All cropping systems were harvested annually except poplar, which was on a 6 year harvest cycle. Plant species compositions of the native grasses, restored native prairie, and early successional vegetation are provided in Tables S1 and S2.

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Agronomic Management. Planting densities, plant hybrid selection, fertilizer management, and herbicide applications followed local best-management practices.[46] The perennial grass systems, including switchgrass, native grasses, and restored prairie, were planted in June 2008 using a standard drop spreader (Truax Company, Inc.) with two cultipack rollers to smooth the planting bed. Miscanthus rhizomes with one to two active growing points were hand-planted at a depth of 10 cm (76 × 76 cm spacing) in late May 2008. Poplar cuttings were planted by hand in early May 2008 (1.5 m between plants in-row and 2.4 m between rows); cuttings averaged 1.3 cm diameter × 25 cm length with a minimum of two active buds and were planted to expose ∼5 cm above the soil surface. All crop planting densities were based on university extension best-management practices with the purpose of maximizing yields at a reasonable cost to a producer.

Miscanthus at the Mollisol site died of exposure to below-freezing soil temperatures in its first year and was replanted in spring 2010. At the Alfisol site, switchgrass, native grasses, and restored prairie were re-seeded in 2009 due to intense storms in midsummer 2008 that redistributed not-yet-germinated seeds. Poplar at the Mollisol site failed in 2010 due to infection by the leaf spot fungus *Marssonina* spp., which killed most trees;[46] findings for poplar at the Mollisol site are thus not further considered in the present analysis.

Nitrogen fertilizer was added to the maize system based on spring soil tests and averaged 167 kg N ha^{-1} $year^{-1}$ for both locations over the 7 year period. Nitrogen was also applied annually in the early summer at a rate of 56 kg ha^{-1} to the early successional vegetation beginning in 2009 and to switchgrass, miscanthus, and native grasses beginning in 2010, delayed to avoid promoting weed growth during the first 1−2 years. Hybrid poplar at the Alfisol site received a single N application in 2010 at a rate of 155 kg N ha^{-1}. The restored prairie system was not fertilized during the study. No fertilizers other than N were required, based on soil tests.

Carbon Stocks, Pools, and CO_2 Removal from the Atmosphere. There are three main pools and fluxes of C in these systems that may influence atmospheric CO_2 balances: aboveground or harvestable biomass, standing stock belowground (root) biomass, and soil organic C (SOC, which by convention does not include root C). These three C pools differ by C residence times and pathways of C accrual and loss. Harvestable biomass, when converted into fuel or electricity for powering vehicles directly, offsets fossil fuel use and therefore reduces the flow of CO_2 from the fossil pool (i.e., from burning petroleum or other fossil fuels) to the atmosphere; conceptually the CO_2 released during bioenergy combustion is balanced by CO_2 assimilation by vegetation within the past several years, and thus it does not represent an addition of new C to the atmosphere over this time period. For forest biomass, this is not the case, insofar as slow-growing trees can take a century or more to recover sufficient biomass to replace the C added to the atmosphere upon combustion.[51]

Where annual cropland is converted to perennial cropland (as in this study), the increase of root biomass during the first years of production represents the new accumulation of organic C that will help offset CO_2 emissions, which briefly increase during the establishment phase.[36] The converse will occur if land with perennial vegetation is converted to annual crops: in this case, the eventual oxidation of established root and detrital C pools will generate CO_2 and contribute to establishment C debt.[13,43] The belowground biomass pool thus will have a temporary (2−3 years) effect on the overall GHG balance of a perennial bioenergy system, depending on the balance between C lost from decomposing roots of the former vegetation and the C sequestered in new root systems for the duration of feedstock production.

Harvested Biomass. Crops were harvested after natural senescence in the autumn except for poplar, which was harvested at the end of its 6 year harvest cycle in early winter. We used harvest yields as reported[46] for feedstock calculations.[47] Yields are typically lower than aboveground net primary productivity (ANPP) due to retranslocation of C and nutrients to roots during senescence in perennial plants, to leaf loss and decomposition prior to harvest, and to residue remaining after harvest. We harvested our herbaceous perennial systems ∼15 cm above the soil surface. For the maize stover, we used for biomass values the average harvestable stover biomass (± standard error of the mean, SEM; Table S3) of 3.7 ± 0.1 and 6.0 ± 0.1 Mg ha^{-1} $year^{-1}$ for the Alfisol and Mollisol sites, respectively. This represents ∼27 and ∼52% of stover ANPP for Alfisol and Mollisol sites, respectively,[46] percentages intended to allow long-term maintenance of SOC in these no-till systems.[52]

Standing Root Biomass. In the herbaceous systems, we measured root standing stocks annually to determine the time to maximum root biomass accumulation and equilibration and used literature values of root/shoot ratios for established perennial grasses to constrain estimates of changes in root biomass. We measured standing root biomass each year between 2008 and 2013 after the senescence of aboveground vegetation during November or December. At each of three locations in each plot, we sampled root biomass from the plant center, an area adjacent to the plant, and midway to the next plant. We summed all recovered roots from the cores to determine the time course of root biomass accumulation and equilibration after conversion of the cropping system (Figure S1b,c). Four root biomass sampling cores (7.5 cm in diameter × 1 m depth) were taken with a Giddings probe (Giddings Machinery Co, Windsor, Colorado) at the Mollisol site or with a Geoprobe (model 540MT, Geoprobe Systems; Salina, Kansas) at the Alfisol site. Cores were sectioned by depth, and roots were washed, dried, weighed, and analyzed for C content by combustion and gas chromatography [EA 1112 CN automatic elemental analyzer (Thermo Finnigan, Milan, Italy) at Mollisol and Carlo Erba NA1500 series II CN analyzer (Carlo Erba Instruments, Milan, Italy) at Alfisol sites]. Method details are at https://data.sustainability.glbrc.org/protocols/157. The average root C content of surface roots (0−25 cm depth) for switchgrass was 44.1 ± 0.4%; for miscanthus 40.9 ± 0.5%; for native grasses 41.8 ± 0.7%, for early successional vegetation 40.1 ± 0.6%; and for restored prairie 40.8 ± 1.0%.

Root biomass stocks increased in the perennial cropping systems during the first 4 years of the experiment (i.e., 2008−2011) and then stabilized (Figure S1b,c). We thus consider root stocks to have accumulated only until 2011, after which we detected no net long-term change. For established (>3 years post-planting) field-grown switchgrass and other native perennial grasses, the average root/shoot ratio reported in the literature is 1.2 with a range between 0.3 and 9.9 (n = 39).[53−65] Based on a review of studies, the root/shoot ratio of miscanthus has been reported to be ∼1 at peak biomass.[66] We thus calculated the post-establishment root standing stock C pool for our herbaceous systems as average aboveground

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peak biomass for years 2011−2014 multiplied by the root/shoot ratio and then by the root C content. For the poplar system, we obtained an average root standing stock value from the literature[63−65] of 1.17 ± 0.40 kg dry mass m^{-2} and calculated a root standing stock value using reported shoot:root ratios of 3.2 ± 0.3 and 7.2 ± 0.4 and our total harvested aboveground biomass of 55.4 ± 2.9 Mg ha^{-1}. Thus, we averaged three independent estimations from the literature: one based on measurements of root stocks and two based on root/shoot ratios multiplied by our measured total shoot biomass. A root C content of 39.7 ± 1.5% for poplar roots was assumed,[63] and we estimated that the observed root biomass stock equivalent to 1503 ± 390 g CO_2 m^{-2} at year 4 represented the average post-establishment standing stock.

Soil Organic Carbon Pool. As for other sites in the U.S. Midwest,[67] changes in total detrital SOC cannot be reliably detected in fewer than 10 years at either site[68,69] although increases in the surface-horizon active fraction were documented after 5 years.[70] We thus used expected soil C equilibrium values based on observations in adjacent long-term herbaceous perennial vegetation at each site to provide likely future equilibrium values for our perennial bioenergy cropping systems, divided by an expected 50 year pre-equilibration period to provide a conservative estimate of annual SOC accumulation. The estimate is conservative because a 50 year pre-equilibration period possibly overestimates the time it will take for these soils to reach new equilibrium SOC contents, and therefore will underestimate the near-term rate of annual SOC accumulation. Near our Alfisol site, for example, a formerly row-cropped field in the U.S. Department of Agriculture (USDA) Conservation Reserve Program under brome grass (*Bromus inermis*) for 22 years had an SOC content of 20.9 g C kg^{-1} soil,[44] approaching the expected equilibrium concentration of 24.0 ± 3.4 g C kg^{-1} soil, as noted above.

For the Alfisol soils supporting perennial herbaceous vegetation, we used an SOC equilibrium value of 23.0 ± 0.9 g C kg^{-1} soil, which is the SOC concentration reported in Senthilkumar et al.[71] for 0−20 cm depth at a nearby never tilled grassland field converted from forest in 1960. Syswerda et al.[68] reported an SOC content of 24.0 ± 3.4 g C kg^{-1} soil for three mature (two never cut) deciduous forests nearby (A horizon, 17 cm mean depth). The pre-establishment SOC concentration at the Alfisol site was 10.8 g C kg^{-1} soil.[50] To convert SOC concentration measures to an areal basis, we used two values of observed soil bulk density, 1.12 g cm^{-3} at equilibrium and 1.66 g cm^{-3} currently.[49,50] For the Mollisol soils, we used for an SOC equilibrium value the median of reported SOC values[72−79] for remnant prairie sites in south-central Wisconsin (range of 19.9−59.5 g C kg^{-1} soil) of 39.7 g C kg^{-1} soil for 0−20 cm depth with a bulk density of 1.30 g cm^{-3}. The pre-establishment SOC concentration at our Mollisol site[46] was 19.9 g C kg^{-1} soil with a bulk density of 1.36 g cm^{-3}.

We calculated the CO_2-equivalent (g CO_2e m^{-2} $year^{-1}$) SOC change to 20 cm depth using the equation

$$CO_2e\ (SOC) = \frac{(x_1 - x_2)\ kg\ C}{m^2 \times x_3\ year} \times \frac{44\ kg\ CO_2}{12\ kg\ C} \times \frac{10^3\ g\ CO_2}{1\ kg\ CO_2} \quad (1)$$

where x_1 is the estimated SOC in the perennial cellulosic cropping system at equilibrium, corrected for bulk density, x_2 is the SOC in 2008 at the time of cropping system establishment corrected for bulk density, and x_3 is the period of SOC accumulation to reach equilibrium (50 year). Following convention, SOC does not include root biomass C.

The expected longevity of C sequestered into SOC is similar to that for belowground biomass C, i.e., for the duration of feedstock production. Unlike root biomass C, however, SOC will continue accumulating for decades after conversion to perennial vegetation, and thus have a long-term on-going climate benefit even upon cessation of feedstock production, as long as subsequent land use, such as afforestation, does not re-disturb soil. West and Six[80] estimate that, on average, the duration of soil C accretion after management change is about 50 years, after which SOC is at a new equilibrium with little additional net sequestration. If management changes back to annual cropping, however, some or all of the SOC accumulated will be lost back to the atmosphere, depending on tillage intensity.[43] In that case, the long-term mean C stocks would still be higher, however, representing net C sequestration over the bioenergy crop cycle. Our analysis assumes no future soil disturbance that results in the release of stored soil C to the atmosphere.[81]

N_2O and CH_4 Fluxes. Methods for measurement of soil−atmosphere fluxes of N_2O and CH_4 were previously described[82] and are here expressed as positive emissions when there was net release to the atmosphere and negative emissions for net uptake. The CO_2-equivalents (g CO_2e m^{-2} $year^{-1}$) for N_2O and CH_4 emissions were calculated using standard IPCC 100 year horizon factors (298 for N_2O and 25 for CH_4)[83,84]

$$CO_2e\ (N_2O) = \frac{x_1\ g\ N_2O{-}N}{ha \times year} \times \frac{44\ g\ N_2O}{28\ g\ N_2O{-}N} \times \frac{1\ ha}{10^4\ m^2} \times \frac{298\ g\ CO_2}{1\ g\ N_2O} \quad (2)$$

$$CO_2e\ (CH_4) = \frac{x_2\ g\ CH_4{-}C}{ha \times year} \times \frac{16\ g\ CH_4}{12\ g\ CH_4{-}C} \times \frac{1\ ha}{10^4\ m^2} \times \frac{25\ g\ CO_2}{1\ g\ CH_4} \quad (3)$$

where x_1 is the mean annual $N_2O{-}N$ emission rate (g N ha^{-1} $year^{-1}$) and x_2 is the mean annual $CH_4{-}C$ emission rate (g C ha^{-1} $year^{-1}$). While newer global warming potentials for N_2O and CH_4 are available,[85] we use those for national greenhouse gas inventory reporting, consistent with recent IPCC reports.[1] Mean annual soil fluxes of $CH_4{-}C$ and $N_2O{-}N$ for all studied ecosystems are provided in Table S4.

Farming Inputs. Total GHG emissions in CO_2-equivalents associated with farming activities were calculated as the sum of CO_2e emissions from the production of fertilizers and herbicides and from the fuel use by farm machinery (Tables S5 and S6). Calculations are based on actual field practices at the study sites, with average fuel use and production costs.[46] Diesel fuel ($C_{16}H_{34}$) is assumed to be oxidized 100% to CO_2[86]

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Table 1. Emission Intensities (g CO_2e MJ^{-1}) of Biomass Used to Produce Ethanol vs Electricity to Power Light-Duty Vehicles, Prior to SOC Equilibration, with and without Carbon Capture and Storage (CCS)[a]

	biomass for ethanol[b]				biomass for electricity[b]			
	without CCS		with CCS[c]		without CCS		with CCS[c]	
cropping system	Alfisol site	Mollisol site	Alfisol site	Mollisol site	Alfisol site	Mollisol site	Alfisol site	Mollisol site
	g CO_2e MJ^{-1}							
maize stover	20.3 (0.7)	19.2 (0.4)	−98.1 (5.0)	−99.2 (2.8)	22.3 (1.1)	21.2 (0.6)	−217.5 (11.1)	−218.6 (6.2)
switchgrass	−5.2 (0.7)	−44.4 (3.6)	−123.6 (23.1)	−162.8 (16.4)	−5.2 (1.0)	−47.5 (4.8)	−245.0 (45.8)	−287.3 (29.0)
miscanthus	−4.1 (0.7)	−32.4 (6.0)	−122.5 (26.8)	−150.8 (31.6)	−4.1 (0.9)	−34.6 (7.2)	−243.9 (53.4)	−274.4 (57.5)
poplar	−13.2 (0.6)	–[d]	−160.0 (8.4)	–[d]	−11.2 (0.6)	–[d]	−251.0 (13.2)	–[d]
native grasses	−0.2 (0.1)	−82.8 (11.7)	−118.6 (41.5)	−201.2 (35.3)	0.2 (0.1)	−89.1 (15.6)	−239.6 (83.9)	−328.9 (57.7)
early successional	−16.5 (3.9)	−150.3 (32.7)	−134.9 (40.7)	−268.7 (80.4)	−17.4 (5.3)	−162.0 (48.5)	−257.2 (77.7)	−401.8 (120.2)
restored prairie	−53.5 (13.6)	−179.0 (35.7)	−171.8 (57.6)	−297.4 (78.2)	−57.3 (19.2)	−192.9 (50.7)	−297.1 (99.6)	−432.7 (113.8)

[a]Standard error in parentheses (n = 5 replicate blocks). [b]Including SOC accumulation and CO_2 emissions/offsets associated with ethanol production at biorefinery. [c]Assumes 48% CCS efficiency. [d]Failed crop due to *Marssonina* fungal outbreak.

$$\mathrm{CO_2e\ (diesel)} = \frac{x_1\ \mathrm{L\ C_{16}H_{34}}}{\mathrm{ha \times year}} \times \frac{832\ \mathrm{g\ C_{16}H_{34}}}{1\ \mathrm{L\ C_{16}H_{34}}} \times \frac{192\ \mathrm{g\ C}}{226\ \mathrm{g\ C_{16}H_{34}}} \times \frac{44\ \mathrm{g\ CO_2}}{12\ \mathrm{g\ C}} \times \frac{1\ \mathrm{ha}}{10^4\ \mathrm{m^2}} \quad (4)$$

where x_1 is the average annual diesel use for field operations (L ha^{-1} $year^{-1}$).

For the maize stover, only those farming activities associated with stover removal were included as farming CO_2e costs. These activities included the fuel costs of collecting the stover and the need for additional fertilizer inputs due to stover removal. To estimate the additional fertilizer required to replace N removed in the harvested stover, we measured stover N content (1.3 ± 0.1%) at our sites and assumed that all N removed with the stover harvest needed to be reapplied as the urea ammonium nitrate (UAN) fertilizer. The additional fertilizer need was estimated as 76.0 ± 1.5 and 47.8 ± 1.7 kg N ha^{-1} $year^{-1}$ for the Mollisol and Alfisol sites, respectively. To account for additional N_2O emissions due to this additional fertilizer, we used the empirical emission factor for the UAN fertilizer from Shcherbak et al.[87]

$$\mathrm{N_2O\ emission\ (g\ N\ ha^{-1}\ year^{-1})} = 1525 + (8.03 + 0.0067 \times N) \times N \quad (5)$$

where N is the additional fertilizer in kg N ha^{-1} $year^{-1}$.

End Use Scenarios. Subsequent calculations of emission intensities, GWIs, fossil fuel offsets, and BECCS impacts are based on four end use scenarios:

1. Ethanol: cellulosic ethanol (or its equivalent) replaces petroleum;
2. Ethanol + CCS: cellulosic ethanol replaces petroleum with 48% of the CO_2 released from biomass C captured at the biorefinery[2,88] and 100% of captured C transported and stored underground;[2,89]
3. Electric: cellulosic electricity is used to propel electric vehicles;
4. Electric + CCS: cellulosic electricity propels electric vehicles with 90% of the CO_2 released from biomass C captured at the power plant[2,88] and 100% of captured C transported and stored underground.[2,89]

GHG Emission Intensities of Bioenergy Production. Biofuel GHG emission intensity (g CO_2e MJ^{-1}), defined here as the net CO_2e balance per unit of ethanol energy produced by the system, was calculated as

$$\mathrm{GHG\ emission\ intensity_{biofuel}} = \mathrm{net\ CO_2e\ balance} \times \mathrm{biofuel\ energy\ content^{-1}} + \mathrm{biorefinery} \quad (6)$$

where net CO_2e balance is as defined below, and biofuel energy content is the net ethanol energy yield of the system in MJ m^{-2} $year^{-1}$. Biorefinery represents the net emission intensity of the operation of the biorefinery, including CO_2 costs of biomass transport[38] and a GHG credit for the fossil fuel offset by electricity production from biorefinery residues (Table S7). There is no biorefinery emission intensity for the electricity scenarios.[37,90] Net CO_2e balance (g CO_2e m^{-2} $year^{-1}$) is

$$\mathrm{net\ CO_2e\ balance} = \sum \mathrm{CO_2e\ (soil\ GHG,\ farming)} \quad (7)$$

where soil GHG represents the balance of CO_2e (N_2O) + CO_2e (CH_4) + CO_2e (SOC) and farming represents the emissions associated with farming operations and agrochemicals. The net CO_2e balance thus represents the direct CO_2e impact of producing a specific feedstock. As discussed later, the CO_2e cost of indirect land-use change (ILUC) is not included in this calculation, nor is our calculation of the fossil fuel offset affected by future fuel switching as when, for example, coal is phased out completely or large-scale production of bioenergy-based fuels greatly reduces petroleum use. For the BECCS scenarios, we included biomass CO_2e emitted during fuel production and stored with CCS technology (see below). We provide estimates of emission intensities both prior to and after SOC equilibration to evaluate long-term trends.

Net Global Warming Impact Calculation. The direct global warming impact (GWI; g CO_2e m^{-2} $year^{-1}$) is defined here as the difference in GHG emissions between vehicles powered within our two biofuel scenarios compared to the equivalent power provided by petroleum (i.e., the fossil fuel offset of the biofuel)

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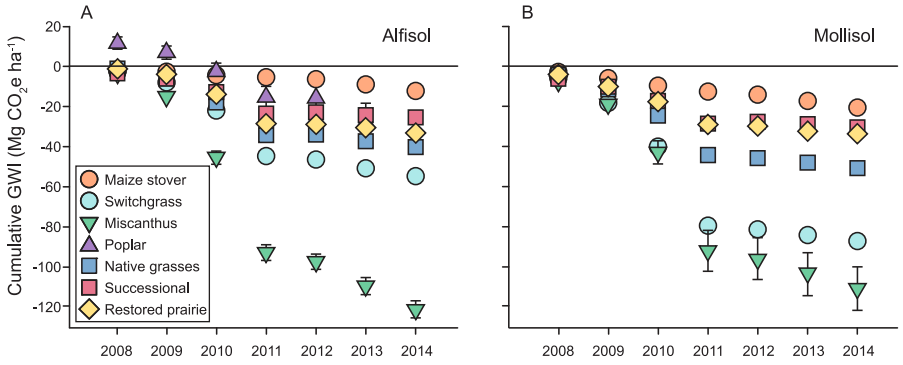

Figure 1. Cumulative global warming impact (GWI) of alternative cellulosic cropping systems at the low-fertility Alfisol (A) and high-fertility Mollisol (B) sites, assuming ethanol to power vehicles as the end use. GWI is expressed in CO_2 equivalent (CO_2e); values <0 indicate a net climate benefit. Error bars represent standard errors (n = 4−5 replicate blocks) and in most cases are smaller than symbols. No CCS assumed. GWI calculations assume a 100 year time horizon.

$$\begin{aligned}&\text{GWI (g CO}_2\text{e m}^{-2}\text{ year}^{-1})\\&= (\text{GHG emission intensity}_{\text{biofuel}}\\&\quad - \text{GHG emission intensity}_{\text{fossil}})\\&\quad \times \text{biofuel energy content}_{\text{system}}\end{aligned} \tag{8}$$

where GHG emission intensity$_{biofuel}$ is calculated by eq 6 (numbers are provided in Table 1), GHG emission intensity$_{fossil}$ is 94 g CO_2e MJ^{-1} (Table S7), and biofuel energy content$_{system}$ is harvestable yield for a given cropping system (Table S4) multiplied by the ethanol production potential for a particular crop (L kg^{-1} feedstock under standard biorefinery operating conditions) from Table S7 and ethanol's lower heating value (LHV) energy content of 21.1 MJ L^{-1}. For the two electric vehicle scenarios, we used an LHV biomass energy content of 17.3 MJ kg^{-1}, which represents the maximum potential energy available to produce electricity, and calculated two GHG emission intensities, one for the electricity produced using cellulosic biomass instead of the current electricity sources for the study region (below) with and without CCS and another for the electricity used to propel the vehicle. We calculated net GWI by summing the two. We provide estimates of GWI both before and after SOC equilibration to evaluate long-term capacities. Unlike GHG emission intensities, which consider climate impacts per MJ of feedstock mass (g CO_2e MJ^{-1}), GWI also includes the land required to produce biomass (g CO_2e m^{-2} $year^{-1}$).

Fossil Fuel CO_2 Offset by Electricity for Vehicles. For the estimation of the fossil fuel offset for electric vehicle use, we assumed that electric vehicles (EVs), for which published LCAs are available,[91] will run entirely with electricity produced using cellulosic biomass, both without and with CCS.[92] We considered the entire life cycle costs of EVs for this analysis, including the substantial CO_2 costs of battery production. The assumptions underlying published LCAs differ significantly,[91] so we detail our calculations here and discuss the implications of these assumptions later. In particular, we calculated the total energy that could be delivered by a battery during the vehicle's lifetime assuming that Li-ion battery production has a life cycle C expense of 5.1 kg CO_2e kg^{-1} battery weight and 48% of battery materials can be recycled.[93] We assumed an average battery weight of 197 kg[94] and an electricity storage capacity of 16.5 kWh or 59.4 MJ.[91] Further, we assumed that the vehicle will have a useful life of 14 years[95] and will be driven ~70 km per day[95] for 260 days $year^{-1}$ (average number of working days in the United States). On average, then, the vehicle battery will deliver 15,444 MJ $year^{-1}$ and have an emission intensity of 2.5 g CO_2e MJ^{-1}. Given that the emission intensity of the gasoline made from petroleum is 94 g CO_2e MJ^{-1}, the battery would offset 91.5 g CO_2e MJ^{-1}. Other than for batteries, we assumed that the CO_2 costs of manufacturing electric and gasoline vehicles are similar.

The use of cellulosic biomass for electricity generation can deliver ~35% of the chemical energy initially stored in the biomass (lower heating value of 17.3 MJ kg^{-1}).[96] For estimation of CO_2e emissions associated with electricity production, we used average emission intensities of electricity production in Michigan and Wisconsin for 2016: 139 and 175 g CO_2e MJ^{-1}, respectively.[97] We assumed further that 20% of the chemical energy initially stored in the cellulosic biomass is available for vehicle propulsion and that the rest is lost during energy conversion.[96] In this way, we calculated fossil fuel offsets associated with the use of electricity in place of petroleum for vehicles. We assumed that existing power plants would serve this purpose without extensive modification and additional CO_2 costs.

Negative Emissions from BECCS. Our estimate of BECCS begins with our measured average biomass production (Table S3) and measured average C content of the biomass (44% of the biomass dry weight), converted to CO_2e. The potential for CCS of the biomass C assumes that (a) 48% of the embedded carbon that ends up as CO_2 could be captured at an ethanol biorefinery[2,88] (the remaining 52% is assumed to escape to the atmosphere, mostly from vehicle tailpipes); (b) electricity produced from the cellulosic biomass is used for electric vehicles; (c) 90% of CO_2 emitted during electricity production is captured and thus available for storage;[2,89] and (d) 100% of CO_2 captured at biorefineries or biomass electricity plants can be transported to and stored in future underground reservoirs (although existing U.S. pipeline infrastructure is inadequate[89,98]).

Data Availability. Data used in this study are available at https://doi.org/10.5061/dryad.44j0zpc8r.

RESULTS AND DISCUSSION

Carbon Debt Payback. Based on the conversion of cellulosic biomass to ethanol for liquid transportation fuel and assuming a 100 year time horizon for all GWI calculations,[81] most of the perennial cropping systems were C neutral after

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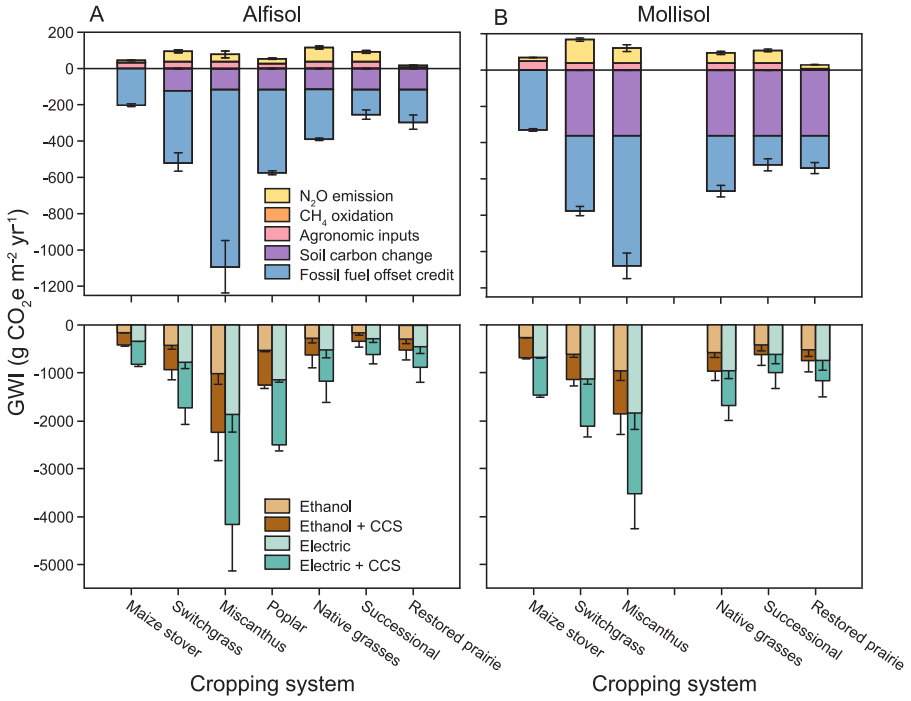

Figure 2. Component (top) and net (bottom) global warming impacts (GWIs) for alternative cellulosic cropping systems at the Alfisol (A) and Mollisol (B) sites. GWI values <0 indicate a net climate benefit. Top: stacked bars represent different components of the GWI balance for a given system including fossil fuel offset credits for ethanol without CCS. Fluxes of methane are too small to be visible in the figure. Bottom: net GWI for each system (means for Alfisol and Mollisol sites) by different end uses: ethanol, ethanol plus carbon capture and storage (CCS; 48% capture efficiency), electricity, or electricity plus CCS (90% capture efficiency) to power vehicles, before SOC equilibration to higher levels. Error bars represent standard errors (n = 4−5 replicate blocks).

year 1 (Figure 1; Tables S10 and S11) and all but poplar achieved a negative GWI by the end of year 2, indicating rapid payback of the C debt incurred at planting.[13,99] Soil organic carbon (SOC) stocks at both sites had been depleted by prior farming, so the initial C debt consisted mainly of farming inputs and N_2O emissions. For poplar, the CO_2 cost of propagules (stem cuttings) together with high initial nitrogen fertilizer inputs (Table S10) and a slow initial growth rate delayed C debt payback an additional year such that the poplar system still had a positive GWI in year 2 (Figure 1; Alfisol site only[46]). For the maize stover, C debt was nil because residue-based feedstocks have no planting costs, which are borne by the cash crop, and thus, the cumulative GWI of the maize stover became negative immediately and incurred only the annual C-equivalent costs of the stover harvest and the fertilizer added to replace nutrients removed in the harvested stover.

Even though some LCA frameworks allow for inclusion of root stocks, e.g.,[100] belowground biomass is rarely included in published LCAs, which instead focus belowground on non-root SOC stocks. Yet, we found that early root accumulation in perennial crops (Table S12) provided a strong initial C credit at both sites (Figure S1b,c). For the first 3 years of establishment, this benefit was even greater than SOC accumulation. By year 4, standing root biomass had stabilized in all perennial systems and no longer represented an increasing climate benefit, though remained an important contributor to cumulative GWI (Figures 1 and S1; Tables S10 and S11).

After the root biomass stabilized, negative GWI sources included only aboveground biomass production (harvestable yield; Table S4), which should persist indefinitely as a potential fossil fuel offset credit, and SOC accumulation, which should continue for 30−50 years until reaching a new equilibrium.[101] As noted earlier, we conservatively forecast annual SOC accumulation based on equilibrium SOC stocks in adjacent undisturbed native soils: mature forest soils at the Alfisol site and native prairie soils at the Mollisol site, corroborated by SOC accumulation rates in USDA Conservation Reserve Program (CRP) lands on the same soils nearby (see the Materials and Methods section). In all cases (Figure 2), SOC accumulation more than offset the GWI costs of farming during and after crop establishment as well as soil N_2O emissions (Table S4), which themselves comprised 30−90% of post-establishment costs (Table S11). Were these sites unfertilized, as could be the case for some biomass crops such as switchgrass,[8,102,103] the farming and N_2O costs would be lower.

Comparative Energy Production Potentials. The maximum energy production potential (bioenergy produced per hectare) of each cropping system varied with yield[46] and decreased in the order miscanthus > poplar > switchgrass > native grasses ≈ maize stover > early successional vegetation ≈ restored prairie (Figure 3A). With the exception of the maize stover, we found surprisingly little variation in feedstock production between the two sites despite differences in soil fertility. Maize productivity was somewhat higher at the more fertile Mollisol site, whereas the productivities of miscanthus and native grasses were similar or even higher at the less fertile Alfisol site.[46] Maize stover differences are primarily due to the greater proportion of the stover removed from the Mollisol (52%) vs the Alfisol (27%) sites. Both sites were subject to a similar climate variability over the study period, including a growing season drought in 2012, and no pest or pathogen

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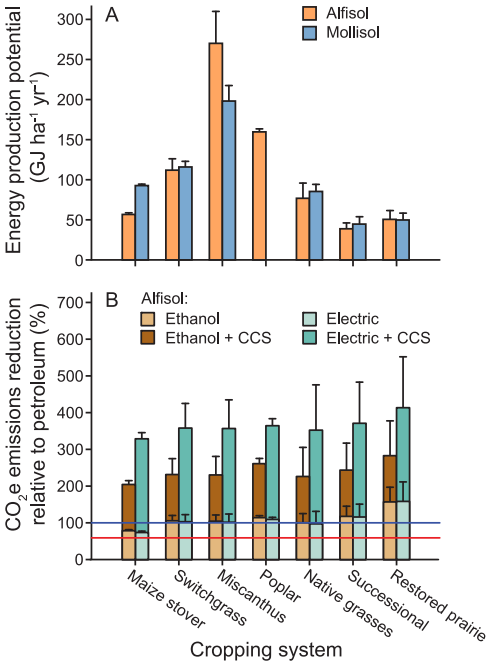

Figure 3. Maximum energy production potential (based on yields and the lower heating value of harvestable biomass) for the Alfisol and Mollisol sites (A) and the percent reduction in emission intensities as CO_2 equivalents (CO_2e) per MJ relative to petroleum averaged for both sites (B) for alternative cellulosic bioenergy cropping systems used for ethanol or electric vehicles. Scenarios include with and without carbon capture and storage (CCS) and are for before SOC equilibration to higher levels (Alfisol site; for Mollisol site see Figure S3). The red horizontal line in (B) represents the 60% reduction currently required for U.S. designation as an advanced biofuel.[105] The blue horizontal line represents the 100% reduction threshold for negative CO_2e emissions. A reduction in emission intensity greater than 100% implies negative emissions (i.e., net CO_2e removal from the atmosphere). Error bars represent standard errors (n = 4−5 replicate blocks). Values appear in Table S17.

stresses were observed other than a fatal pathogenic fungus outbreak in the Mollisol poplar stands, which we excluded from this analysis. While it is not possible to generalize on the basis of only two contrasting sites, similar rates of perennial crop production between these sites corroborate other evidence that perennial herbaceous systems have the potential to thrive in a wide range of soil fertilities.[104]

Emission Intensities. Emission intensities (CO_2e emitted per unit of energy produced) differed substantially from energy production potentials. Even without CCS, all ethanol-producing systems but the maize stover had negative emission intensities (Table 1), reflecting absolute net climate benefits. At the Mollisol site, the average of ethanol emission intensities for restored prairie and early successional vegetation (−165 ± 34 g CO_2e MJ^{-1}, Table 1) was 2 times more negative than that for native grasses (−83 ± 12 g CO_2e MJ^{-1}), which in turn was 2 times more negative than the average for miscanthus and switchgrass (−38 ± 5 g CO_2e MJ^{-1}), the net result of differences in SOC accumulation and energy production potentials. If the GWI of SOC was not included in the total GWI calculation (which will be the case after equilibration in 30−50 years), all studied systems at the Mollisol site would have positive emission intensities post-establishment (from 12.2 ± 2.4 to 68 ± 15 g CO_2e MJ^{-1}; Table S13), similar to that of the maize stover (20 ± 0.4 g CO_2e MJ^{-1}) but still 25−87% lower than the emission intensity of petroleum (Figure S2). At the Alfisol site, emission intensities were not as negative but the net climate benefit before SOC accumulation followed a similar order, with the restored prairie and early successional vegetation showing the most negative intensities in all scenarios (Table 1).

Emission intensities for biomass-based electric vehicles were similar to those for ethanol-fueled vehicles (Table 1); in both cases, most (~65%) of the energy embedded in biomass is lost as waste heat during combustion (for electricity generation) or biorefining (for ethanol production). Whereas the emission intensities for ethanol-fueled vehicles before SOC equilibration and without CCS range from 19 to −179 g CO_2e MJ^{-1}, intensities for electric vehicles without CCS range from 21 to −193 g CO_2e MJ^{-1} (Table 1). After SOC equilibration, intensities for ethanol-fueled vehicles without CCS range from 7 to 63 g CO_2e MJ^{-1} (Table S13).

Although we found similar emission intensities for ethanol and electric vehicles (Table 1), GWIs are very different. The use of biomass to power electric vehicles had substantially more negative GWIs than ethanol whether with or without CCS (Figure 2) mainly because biomass-derived electricity partly displaces coal, which has a much higher emission intensity[98] than the petroleum displaced by ethanol. This difference will diminish in the coming decades as electricity from biomass displaces future electricity sources that are increasingly composed of renewable energy sources with their much lower emission intensities.

All of the cropping systems we examined, including the maize stover, had a substantial climate benefit relative to petroleum (Figures 3B, S2, and S3), especially for the 30−50 year period over which SOC accumulates to its new equilibrium. Even the maize stover, with the lowest benefit, had a 74−80% emission reduction relative to petroleum whether the end use was ethanol or electricity without CCS, above the 60% threshold legislated in the United States for advanced bioenergy feedstocks.[105] For the remaining feedstocks, emission reductions without CCS ranged between 97 and 303% (Table S17), reflecting net uptake of CO_2 from the atmosphere by SOC accumulation (Figures 3 and S3).

Worth noting is that without CCS, SOC accumulation is crucial to the absolute climate benefit of these systems; once SOC equilibrates in 30−50 years, without CCS the climate benefit exists only as an offset relative to fossil fuels (Figure S2). It is also worth noting that although the more biodiverse systems exhibited more negative emission intensities, they were also less productive and thus would require a greater total land area to produce as much energy as more productive crops such as switchgrass or miscanthus (Figures 2 and 3). This is not necessarily an undesirable tradeoff because the conservation benefits of bioenergy cropping systems with greater plant diversity are well recognized.[106−109] Nevertheless, choosing the most productive crop, monoculture miscanthus, would mean giving up the biodiversity benefits of switchgrass, a grass native to North America, which in turn offers fewer conservation benefits than restored prairie (which includes switchgrass as one of many species). In some cases, the choice of non-native species could be environmentally destructive; newly available fertile varieties of our miscanthus, for example, are likely to be highly invasive[110] with potential weed control costs that could be enormous.[111] Thus, a tradeoff exists between the provision of maximum climate mitigation on the one hand and either the maximum provision of other ecosystem services such as those

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Table 2. Net Global Warming Impacts (Mg CO_2e ha^{-1} $year^{-1}$; also Known as Net Greenhouse Gas Balance) for Cellulosic Biomass Used To Generate Electricity for Electric Vehicles Coupled with Carbon Capture and Storage (Scenario 4, See the Materials and Methods Section) before and after Soil Organic Carbon (SOC) Equilibration[a]

	before SOC equilibration		after SOC equilibration	
cropping system	Alfisol site	Mollisol site	Alfisol site	Mollisol site
	Mg CO_2e ha^{-1} $year^{-1}$			
maize stover	−8.1 (0.3)	−14.5 (0.4)	−8.1 (0.4)	−13.3 (0.4)
switchgrass	−17.0 (2.7)	−20.9 (2.2)	−15.9 (3.1)	−15.8 (1.6)
miscanthus	−41.1 (9.6)	−34.8 (7.2)	−39.9 (9.3)	−28.7 (5.8)
poplar	−24.7 (0.1)	–[b]	−23.5 (1.2)	–[b]
native grasses	−11.6 (4.4)	−16.6 (3.1)	−10.4 (3.9)	−11.9 (2.1)
early successional	−6.1 (1.9)	−9.8 (3.3)	−4.9 (1.5)	5.7 (1.8)
restored prairie	−8.6 (3.1)	−11.5 (3.3)	−7.5 (2.6)	−7.3 (2.0)

[a]Values for the electric vehicle scenario without CCS appear in Table S8 and values for ethanol vehicles appear in Table S9. Standard error in parentheses (n = 5 replicate blocks); assuming 90% CCS. [b]Failed crop due to *Marssonina* outbreak.

related to biodiversity,[8,9] which must currently be evaluated separately from carbon benefits,[112] or the avoidance of significant risks[113] such as the spread and substantial economic cost of invasive species.

Bioenergy with Carbon Capture and Storage (BECCS). CCS allows even the maize stover to produce an absolute net climate benefit (Table 2). Assuming that 90% of the cellulosic biomass C used to generate electricity could be captured and stored,[2,88] emission intensities for electric vehicles would range from −217 to −433 g CO_2e MJ^{-1} (Table 1). For ethanol end use, only 48% of biomass C released in the biorefinery could be captured and stored,[2,89] but this would still allow significantly greater climate benefit for cellulosic biomass-derived ethanol, from −98 to −297 g CO_2e MJ^{-1} (Table 1).

Prior to SOC equilibration, then, vehicles powered by renewable biomass-based electricity with CCS would provide negative emissions between −608 and −4110 g CO_2e m^{-2} $year^{-1}$; once SOC equilibrates negative emissions total between −494 and −3995 g CO_2e m^{-2} $year^{-1}$ (Table S9). CCS thus magnifies the climate benefit of electric vehicles by 2- to 3-fold.

Given the existing pipeline network, only ∼30% of centrally captured CO_2 in the United States appears transportable to potential geologic storage locations.[89] Detailed cost analyses,[114] however, suggest that a \$60/tCO_2 sequestration credit could incentivize the construction of necessary infrastructure, including 6900 km of new CO_2 pipelines, to achieve 30 Mt of C storage capacity. Thus, realizing the full potential of CCS requires substantial investment in new CO_2 transport infrastructure. We do not account for the CO_2 costs of building this new infrastructure.

Limitations of This Analysis. Several limitations to this analysis are important to keep in mind. First, we do not include the CO_2e costs of indirect land-use change (ILUC). We assume here that future crops will be established on abandoned cropland[8] and thereby not impact food production to result in land conversion elsewhere. Were this not the case, our CO_2e costs (eq 7) would need to be incremented, although by how much is debatable. ILUC costs are currently highly uncertain and affected by model assumptions.[40,115−117] For maize grain ethanol, estimates of ILUC-associated GHG emissions range from <25 to >210 g CO_2e MJ^{-1},[17,118,119] with most estimates between 4 and 10 g CO_2e MJ^{-1}.[120] For cellulosic feedstocks, estimates are equally uncertain but generally lower: the U.S. Renewable Fuel Standard[119] estimates ILUC GHG emissions between −10 for the maize stover and 12 g CO_2e MJ^{-1} for switchgrass. The GREET model,[17] widely used in the United States, assumes values of −0.5 for maize stover, 7.1 for switchgrass, and ∼2.2 g CO_2e MJ^{-1} for miscanthus grown on grain ethanol lands. Finally, Dunn et al.[120] estimate ILUC emissions of 2.7−19 g CO_2e MJ^{-1} for switchgrass, −10.0 to −2.1 for miscanthus (assumes substitution of maize now grown for grain-based ethanol), and ∼−1.0 for the maize stover. If we were to include ILUC in our total balance, a conservative (median) estimate of ILUC costs would be ∼0.85 g CO_2e MJ^{-1} for the stover and ∼12.0 g CO_2e MJ^{-1} for perennial grasses (ignoring substitution for corn grain ethanol). As noted earlier, however, our expectation is conversion of abandoned U.S. cropland to cellulosic bioenergy, where ILUC effects would be nil and the carbon benefits index[112] would be quite high because with abandoned lands there would be no need for food crop production to shift elsewhere.

A second limitation rests with LCA assumptions regarding current vs future energy sources and energy use efficiency. The current electric vehicle calculations do not consider future technology advances that may reduce the CO_2e costs of vehicles, including improved mining efficiencies, battery chemistries, and manufacturing processes.[91] On the other hand, future changes in the mixture of energy sources, likely toward generating a greater proportion of future electricity from sources with lower C intensities (such as wind and solar) than currently, will lead to cellulosic biomass' offsetting less fossil fuel CO_2 in the future.[91] The same future offset effects will be true for ethanol-powered vehicles, and as well fuel switching can be expected to lower the CO_2e costs associated with farming, as more of the energy costs of inputs and operations are borne by renewable fuels, including bioenergy. As noted earlier, eventually the value of offsets will equilibrate to a fraction of today's needs, although relatively more biomass will be needed to meet future needs due to fuel switching, while the value of CCS for providing negative emissions will increase.[2]

Third, in our sites, initial carbon debt was small because prior land use—row crop agriculture—had already depleted SOC and there were no perennial live biomass C stocks either above or belowground. A prior herbaceous perennial crop would have had root C stores that might have delayed the C debt payback time by 2−3 years, as was the case in a nearby former grassland site.[13] Had aboveground C stores been in forest trees, however, the C debt created by harvested wood

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would likely not be repayable for more than 50−100 years due to slow forest regrowth.[51,99] Furthermore, conversion of forest to a bioenergy cropping system would likely not result in net SOC gain and may result in SOC loss.

Overall Mitigation Potentials. Meeting current expectations for the contribution of bioenergy to the midcentury United States transportation needs will require 33−40 Mha of arable land[118] or, to avoid the impact of ILUC,[40] ∼55 Mha of abandoned cropland with its more modest yields.[15,121] Simple extrapolation of the CO_2e savings at our less fertile Alfisol site for switchgrass, which has yields intermediate to our other cellulosic crops (Table S3) and similar, on average, to switchgrass yields elsewhere,[15,104] to 55 Mha suggests a potential to avoid with ethanol-powered vehicles as much as 231 ± 43 Tg CO_2e year^{-1} for the 30−50 year period before SOC equilibrates (Table S15). The same extrapolation for electric-powered vehicles (Table S16) yields a total near-term mitigation capacity of 422 ± 72 Tg CO_2e year^{-1} with today's electric power generation mix. These mitigation potentials would decline 27% for ethanol and 15% for electric vehicles once SOC equilibrates. Were CCS available, mitigation potentials would approximately double, to 507 ± 114 Tg CO_2e year^{-1} for ethanol and to 939 ± 188 Tg CO_2e year^{-1} for electric vehicles (Tables S15 and S16). In the future, however, as other renewable electricity sources become more important, electricity production from biomass would offset less fossil fuel electricity, and the advantage of electric over ethanol vehicles would decrease proportionately.

These mitigation potentials are substantial and exceed recent U.S. reforestation estimates of 307 Tg CO_2 year^{-1} for 63 Mha,[122] which is, together with natural forest management in that analysis (267 Tg CO_2 year^{-1}), the largest by far of 21 different natural climate solutions considered for the United States; bioenergy for transportation (whether with or without CCS) was not considered.[122] While growing perennial herbaceous crops on abandoned agricultural lands would compete directly with reforestation, substituting bioenergy production for reforestation on most of these lands would increase the overall mitigation potential of natural climate solutions (1200 Tg CO_2 year^{-1}) by a factor of 1.1−2.7 depending on the proportion of substitution, the mix of electric and ethanol vehicles, and the availability of CCS. Moreover, lands used for bioenergy production could, in the future, be reforested with little if any carbon debt, providing another 307 Tg CO_2 year^{-1} of negative emissions for the following 90 years without the need for CCS. In any case, a mix of potential strategies might be attractive to more landowners, some of whom may prefer one option over the other.

Significantly greater mitigation via bioenergy could be achieved by replacing switchgrass with miscanthus, a high-yielding but non-native grass with significant invasive potential.[110] Using a native grass such as switchgrass, either alone or with other native grasses as also evaluated here, provides lower but still substantial mitigation while preserving many biodiversity functions.[106] Rapid advances in switchgrass breeding, e.g., to delay flowering and improve overwinter survivorship, will likely narrow this yield gap within coming decades.[123]

Together with other land management mitigation strategies,[124] decarbonizing U.S. light-duty vehicle transport with bioenergy production coupled with CCS could meet a significant fraction of the negative emissions needed in the United States[125,126] to help ameliorate further global temperature increases.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.9b07019.

Experiment location and root biomass of cropping systems, anticipated CO_2e emissions reduction, CO_2e emissions reduction (Figures S1−S3); species composition and seeding rates, dominant species by percent cover, post-establishment harvestable biomass, post-establishment soil N_2O and CH_4 emissions, estimates of CO_2-equivalent emissions, estimates of ethanol production potentials and greenhouse gas emissions, net global warming impact for cellulosic biomass, greenhouse gas sources, post-establishment greenhouse gas sources and sinks, post-establishment belowground biomass, emission intensities for cellulosic biomass, comparison assumptions for selected inputs of GREET, national mitigation potentials for ethanol powered vehicles, total mitigation potentials for electric vehicles (Tables S1−S17) (PDF)

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Notes

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SUPPORTING INFORMATION

Empirical Evidence for the Potential Climate Benefits of Decarbonizing Light Vehicle Transport in the U.S. with Bioenergy from Purpose-Grown Biomass with and without BECCS

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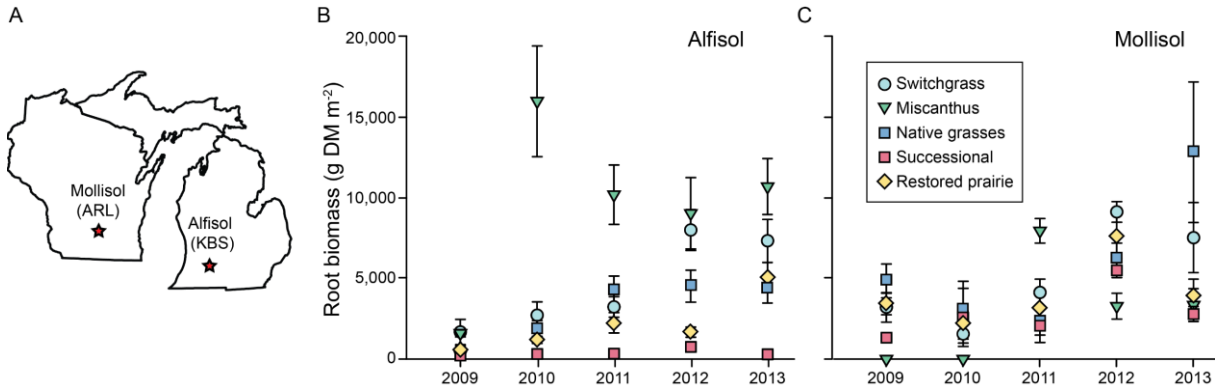

Figure S1. Experiment location and root biomass of cropping systems. (a) Location of the GLBRC Biofuel Cropping System Experiment: Alfisol soils at the Kellogg Biological Station (KBS), Michigan, and Mollisol soils at the Arlington Agricultural Research Station (ARL), Wisconsin. (b and c) Standing root biomass measured in the cellulosic biofuel cropping systems as sum of multiple cores sampling. Values are means ± standard errors (n=4 Alfisol or 5 Mollisol replicate blocks).

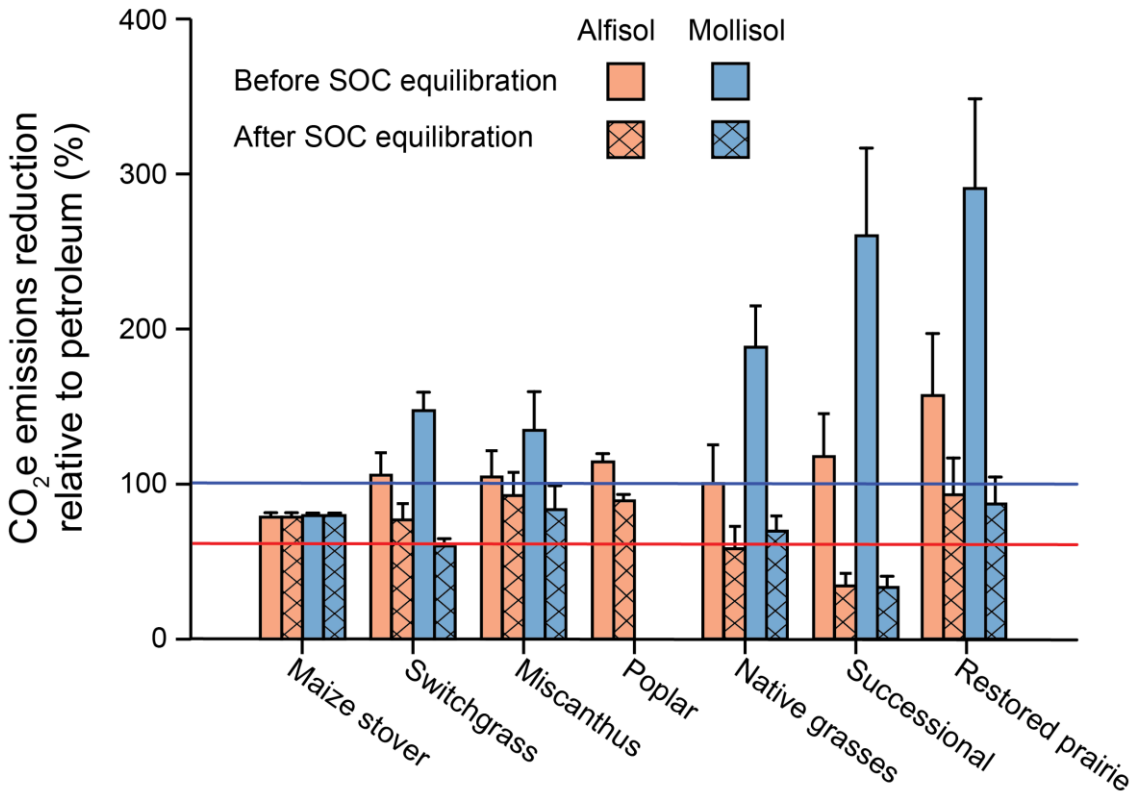

Figure S2. Anticipated CO_2e emission reduction relative to petroleum before and after long-term SOC equilibration to higher levels, assuming ethanol production to power vehicles (scenario 1 – see Materials and Methods). The left-hand bar of each Mollisol and Alfisol pair is the same as in Fig. 3a. The cross-hatched right hand bar in each pair represents CO_2e emission reduction after long-term SOC equilibration, which is likely to take 30-50 years assuming no change in primary productivity (energy production potential). The red horizontal line represents the 60% reduction currently required for U.S. designation as an advanced biofuel. The blue horizontal line represents the 100% reduction threshold for negative CO_2e emissions. Values are means ± standard errors (n=4 Alfisol or 5 Mollisol replicate blocks).

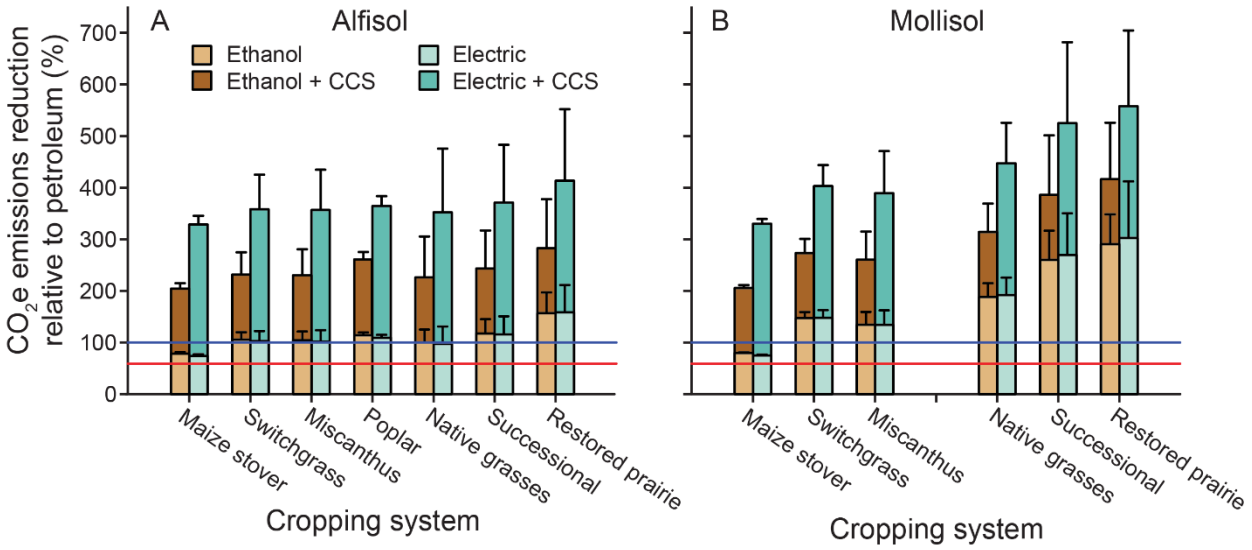

Figure S3. CO_2e emission reduction for alternative cellulosic biofuel cropping systems used for ethanol or electricity with and without carbon capture and storage (CCS) to power vehicles, before SOC equilibration to higher levels. The red horizontal line represents the 60% reduction currently required for U.S. designation as an advanced biofuel. The blue horizontal line represents the 100% reduction threshold for negative CO_2e emissions. Values are means ± standard errors (n=4 Alfisol or 5 Mollisol replicate blocks).

Table S1. Species composition and seeding rates for native grasses and restored prairie systems

	Seeding rate (kg seeds ha^{-1})	
Species	Native grasses	Restored prairie
Grasses		
Big bluestem (*Andropogon gerardii*)	2.4	1.2
Canada wild rye (*Elymus canadensis*)	1.6	1.2
Indiangrass (*Sorghastrum nutans* (L.) Nash ex Small)	2.4	1.2
Junegrass (*Koeleria cristata*)	3.2	0.8
Little bluestem (*Schizacyrium scoparium* (Michx.) Nash)	1.6	1.2
Switchgrass (*Panicum virgatum* var. Southlow)	2.4	0.8
Leguminous forbs		
Round-headed bushclover (*Lespedeza capitata*)		0.4
Showy tick-trefoil (*Desmodium canadense*)		0.4
White wild indigo (*Baptisia lactea* var. lactea (Raf.) Thieret)		0.4
Non-leguminous forbs		
Black-eyed Susan (*Rudbeckia hirta* L.)		0.4
Butterfly weed (*Asclepias tuberosa* L.)		0.4
Canada anemone (*Anemone canadensis* L.)		0.4
Cup plant (*Silphium perfoliatum* L.)		0.4
New England aster (*Aster novae-angliae* L.)		0.4
Pinnate prairie coneflower (*Ratibida pinnata* (Vent.) Barnh.)		0.4
Showy goldenrod (*Solidago speciosa* L.)		0.4
Stiff goldenrod (*S. rigida* L.)		0.4
Wild bergamot (*Monarda fistulosa* L.)		0.4

Table S2. Dominant species by percent cover for the early successional system

Site and year	Species				
	Replicate 1	Replicate 2	Replicate 3	Replicate 4	Replicate 5
Alfisol site					
2011	*Conyza canadensis* (49.0%)	*C. canadensis* (92.9%)	*Echinochloa crus-galli* (57.2%)	*C. canadensis* (82.1%)	*C. canadensis* (44.6%)
2012	*Setaria faberi* (30.9%)	*Lactuca serriola* (63.8%)	*C. canadensis* (50.3%)	*C. canadensis* (34.3%)	*C. canadensis* (32.0%)
2013	*S. faberi* (51.1%)	*S. viridis* (29.0%)	*C. canadensis* (37.8%)	*S. faberi* (39.9%)	*Solidago canadensis* (63.0%)
Mollisol site					
2011	*L. serriola* (29.9%)	*L. serriola* (77.9%)	*Carduus acanthoides* (49.3%)	*Elymus repens* (49.4%)	*L. serriola* (47.5%)
2012	*Elymus canadensis* (48.7%)	*Cerium ravens* (33.3%)	*E. canadensis* (54.4%)	*E. canadensis* (27.0%)	*E. canadensis* (61.2%)
2013	*E. canadensis* (66.4%)	*E. canadensis* (43.0%)	*Ambrosia trifida* (58.0%)	*E. canadensis* (40.6%)	*E. canadensis* (75.8%)

As determined by percent cover (given in parentheses) between years 2011 and 2013. Full species composition by year available at https://data.sustainability.glbrc.org/datatables/543.

Table S3. Post-establishment harvestable biomass* for alternative cropping systems. Values are means (± standard errors, SE; n=5 replicate blocks).

	Harvestable biomass (Mg ha^{-1} yr^{-1})	
Cropping system	Alfisol	Mollisol
Maize stover	3.7 (0.1)	6.0 (0.1)
Switchgrass	7.3 (0.9)	7.5 (0.5)
Miscanthus	17.6 (2.6)	12.9 (1.3)
Poplar	9.2 (0.2)	-
Native grasses	5.0 (1.2)	5.6 (0.6)
Successional	2.5 (0.5)	2.9 (0.6)
Prairie	3.3 (0.7)	3.2 (0.6)

*See Sanford et al.[1] for detailed methods and discussion. Average harvestable yield (dry biomass) determined between years 2011 and 2014 except for maize stover. For the Mollisol site, maize stover yield is an average between years 2008 and 2014, for the Alfisol site it is the average between years 2009 and 2014. For calculation of cellulosic ethanol and electricity production, biomass loss of 11.1% due to transportation and storage was assumed.[2]

Table S4. Post-establishment soil N_2O and CH_4 emissions of alternative cropping systems. Values are means (± SE, n=4 Alfisol or 5 Mollisol replicate blocks).

Cropping system	N_2O emissions* (kg N_2O–N ha^{-1} yr^{-1})	CH_4 emissions† (kg CH_4–C ha^{-1} yr^{-1})
Alfisol site		
Maize stover	0.40 (0.02)	-
Switchgrass	1.22 (0.11)	-0.20 (0.08)
Miscanthus	0.85 (0.11)	-0.49 (0.09)
Poplar	0.58 (0.05)	-0.37 (0.03)
Native grasses	1.66 (0.07)	-0.17 (0.05)
Successional	1.15 (0.27)	-0.24 (0.03)
Restored prairie	0.23 (0.04)	-0.30 (0.04)
Mollisol site		
Maize stover	0.65 (0.01)	-
Switchgrass	2.77 (0.19)	-0.26 (0.03)
Miscanthus	1.76 (0.40)	-0.30 (0.05)
Native grasses	1.21 (0.19)	-0.29 (0.05)
Successional	1.49 (0.16)	-0.38 (0.08)
Restored prairie	0.45 (0.05)	-0.25 (0.01)

Negative emissions indicate net uptake. Soil GHG emissions were calculated using linear interpolation between the sampling dates[3]. Average GHG emissions were determined for years 2011–2014 except for maize stover (2008–2014) and Alfisol poplar (2008–2013); the latter included the full rotation and fertilization during the planting year.

*See Oates et al.[4] for detailed methods. Emissions associated with Maize stover were estimated as described in Methods.

†Negative emissions represent soil CH_4 oxidation.

Table S5. Estimates of CO_2-equivalent (CO_2e) emissions from fuel used for field operations. All fuel is assumed to be diesel with an energy content of 36.4 MJ L^{-1} and with a carbon content of 85%. After initial planting, perennial ecosystems require the following annual field operations: fertilization, mowing, forage raking, and baling except the restored prairie, which is not fertilized, and the poplar system, which is fertilized and harvested only once every 6 years.[1] Differences between GREET and values observed in this study appear in Table S14.

Field operation	Fuel consumption* (L ha^{-1})	CO_2e emissions (g CO_2e m^{-2} yr^{-1})
Herbaceous crops		
Planting‡	7.0	1.8
Fertilizer application	4.3	1.1
Soil finishing	7.0	1.8
Herbicide application	0.6	0.2
Harvest		
Baling (round)	4.3	1.1
Mowing	7.1	1.9
Forage raking	1.9	0.5
Maize stover	14.4	3.7
Poplar trees§		
Planting	2.3	0.4
Harvest (40)	5.5	0.6

*Fuel consumption for field operations were estimated for the following tractors: John Deere 5320 for herbicide application with 13.7-meter application boom; John Deere 7420 for fertilization and planting 4.6-meter wide; John Deere 7230 for the grain, stover, and grasses harvest with John Deere 385 round baler attachment 4.6 meter wide. For miscanthus we assumed fuel use of potato planter, which is similar to a no-till maize planter. Fuel consumption was calculated per number of passes needed to cover one hectare of the field.

†Fuel consumption by individual pieces of equipment was estimated according to University of Nebraska Tractor Test Laboratory (http://tractortestlab.unl.edu/) for maximum fuel consumption during maximum power take-off consumption (PTO) and personal communications with W.K. Kellogg Biological Station farm manager Brook Wilke (wilkebro@msu.edu) who advised on the actual speed

of tractors during different field activities. We assumed the following farming speeds: planting (no-till drilling) – 8 km hr^{-1}; fertilization (no-till injection) – 13 km hr^{-1}; herbicide application (top sprayer) – 16 km hr^{-1}; soil preparation: disking – 10 km hr^{-1} and finishing – 8 km hr^{-1}; harvest: grain – 6 km hr^{-1} and hay mowing – 16 km hr^{-1}; baling/chopping of hay – 8 km hr^{-1}; hay raking – 14 km hr^{-1}.

‡Miscanthus planting was assumed to use same fuel as potato planter. http://www.spriggerschoice.com/index_files/Notillsprigrhizomeplanter.htm

§Fuel use for poplar planting, fertilization, and harvest annualized per rotational basis for six years of rotation based on [9].

Table S6. Estimates of CO_2-equivalent (CO_2e) emissions associated with farming inputs other than fuel.

Input	CO_2e emissions (kg CO_2e kg^{-1})	Reference
Fertilizer		
Urea-Ammonium-Nitrate (UAN)†	6.04	[5]
P	0.22	[6]
K	0.56	[5, 7]
Herbicides*	4.70	[8]
Propagules		
Poplar cuttings†	0.02	[9]
Native grasses‡	5.34	[10]
Miscanthus rhizome†	0.08	[11, 12]

For detailed information on major inputs see Sanford et al. [1]. Differences between GREET and values observed in this study appear in Table S12.

†The value is for the N fraction in the fertilizer, UAN-N.[5]

*At the Mollisol site the following herbicides were used (in parentheses are links to labels): Lexar, Roundup, 2,4-D ester, Callisto, Dual II Magnum, Mirage plus (www.cdms.net/ldat/mp7HH005.pdf), Laudis (www.agrian.com/pdfs/Laudis_CA__NY_Herbicide_Label2.pdf), Prozap + Zinc Phosphide (www.neogen.com/animalsafety/pdf/MSDS/Rodenticides/Rodenticides_SpecLabels/United_States/OR-050009.pdf), Force G3 (www.amvac-chemical.com/products/documents/Force%203G%20Specimen%20Label%2011811-4.pdf), Methylated seed oil, Banvel (www.cdms.net/ldat/ld279006.pdf), and FS transform plus.

At the Alfisol site the following herbicides were used: Lexar, Roundup, and 2,4-D ester.

†Per one cutting/rhizome, planting density of poplars was 2778 tree ha^{-1} and planting density of miscanthus was 17,200 rhizomes ha^{-1} (See Sanford et al.[1] for details). We assume a miscanthus single rhizome weight of 50 g.

‡CO_2 cost of grass seed production was assumed to be equal to that of switchgrass seed.

Table S7. Estimates of ethanol production potential and greenhouse gas emission intensities as compared to gasoline derived from petroleum.

Feedstock	Ethanol production potential (L kg^{-1} feedstock)[†]	GHG emission intensity (g CO_2e MJ^{-1})	Reference
Petroleum	-	94.0	[13]
Maize stover	0.31	-0.38	[13, 14]
Miscanthus	0.31	-0.38	[13,14]
Switchgrass	0.31	-0.38	[13,14]
Poplar	0.25	-0.38	[14, 15]

Based on means for the Alfisol and Mollisol sites. For cellulosic feedstocks, GHG emission intensity includes electricity offset in addition to cellulosic biorefinery operations.[16]

[†] Under standard biorefinery operating conditions.

Table S8. Net global warming impact for cellulosic biomass used for ethanol powered vehicles with and without carbon capture and storage (CCS) technology (scenarios 1 and 2 – see Materials and Methods) and before and after soil organic carbon (SOC) equilibration to higher levels. Values are means (± SE, n=4 Alfisol or 5 Mollisol replicate blocks).

Cropping system	Global warming impact ($gCO_2e\ m^{-2}\ yr^{-1}$)							
	Before SOC/without CCS		Before SOC/with CCS*		After SOC/without CCS		After SOC/with CCS*	
	Alfisol	Mollisol	Alfisol	Mollisol	Alfisol	Mollisol	Alfisol	Mollisol
Maize stover	-158 (8)	-262 (7)	-412 (26)	-677 (23)	-158 (8)	-262 (7)	-412 (26)	-677 (23)
Switchgrass	-420 (79)	-606 (61)	-921 (208)	- 1125 (133)	-305 (57)	-246 (25)	-806 (182)	-765 (90)
Miscanthus	-1002 (219)	-947 (198)	-2211 (584)	-1834 (424)	-887 (194)	-587 (123)	-2096 (553)	-1474 (341)
Poplar	-522 (27)		-1237 (71)		-407 (21)		-1122 (65)	
Native grasses	-274 (96)	-571 (100)	-618 (265)	-953 (194)	-159 (56)	-211 (37)	-503 (215)	-593 (121)
Successional	-162 (49)	-413 (123)	-336 (120)	-613 (222)	-47 (14)	-53 (16)	-221 (79)	-253 (92)
Restored prairie	-282 (95)	-514 (135)	-508 (203)	-737 (231)	-167 (56)	-154 (41)	-393 (157)	-377 (118)

*at 48% carbon capture and storage efficiency

Table S9. Net global warming impact for cellulosic biomass used for electric vehicles with and without carbon capture and storage (CCS) technology (scenarios 3 and 4 – see Materials and Methods) and before and after soil organic carbon (SOC) equilibration. Values are means (± SE, n=4 Alfisol or 5 Mollisol replicate blocks).

Cropping system	Global warming impact ($gCO_2e\ m^{-2}\ yr^{-1}$)							
	Before SOC/without CCS		Before SOC/with CCS*		After SOC/without CCS		After SOC/with CCS*	
	Alfisol	Mollisol	Alfisol	Mollisol	Alfisol	Mollisol	Alfisol	Mollisol
Maize stover	-335 (15)	-669 (18)	-810 (44)	-1447 (44)	-335 (12)	-669 (15)	-810 (44)	-1329 (40)
Switchgrass	-768 (130)	-1115 (107)	-1708 (341)	-2087 (221)	-653 (88)	-755 (56)	-1593 (315)	-1580 (162)
Miscanthus	-1843 (364)	-1816 (338)	-4110 (957)	-3479 (721)	-1728 (280)	-1456 (232)	-3995 (927)	-2867 (580)
Poplar	-1129 (49)	-	-2469 (126)	-	-1014 (39)	-	-2355 (119)	-
Native grasses	-513 (163)	-945 (162)	-1158 (437)	-1662 (307)	-398 (97)	-585 (77)	-1043 (388)	-1193 (212)
Successional	-277 (65)	-609 (192)	-608 (192)	-983 (328)	-168 (32)	-249 (53)	-494 (151)	-566 (177)
Restored prairie	-439 (140)	-733 (202)	-864 (309)	-1151 (333)	-324 (80)	-373 (76)	-749 (263)	-727 (200)

*at 90% carbon capture and storage efficiency

Table S10. Greenhouse gas sources (g CO_2e m^{-2}) in year 1 (2008) for alternative cropping systems. Values are means (± SE, when available; n=4 Alfisol or 5 Mollisol replicate blocks).

	Farming inputs (g CO_2e m^{-2})				Non-CO_2 greenhouse gases (g CO_2e m^{-2})	
			Agrichemicals			
Cropping system	Propagules	Fuel	N	Herbicide	N_2O	CH_4
Alfisol site						
Maize stover	-	3.5	28.9 (1.0)	-	11.9 (0.5)	-
Switchgrass	4.1	10.9	-	0.03	68.2 (9.4)	-1.1 (0.5)
Miscanthus	71.2	6.9	-	0.02	70.9 (1.5)	-0.8 (0.5)
Poplar	41.7	3.4	94.8	0.9	69.7 (8.6)	0.2 (0.3)
Native grasses	4.7	10.9	-	-	80.3 (15.1)	-0.5 (0.2)
Successional	-	1.8	-	-	34.0 (4.6)	-0.3 (0.2)
Restored prairie	4.5	10.9	-	-	67.4 (5.7)	-0.3 (0.1)
Mollisol site						
Maize stover	-	3.5	45.5 (0.9)	-	19.4 (0.4)	-
Switchgrass*	7.6	7.8	-	0.2	45.5 (13.5)	-0.9 (0.1)
Miscanthus*	142.4†	10.8	-	0.5	50.4 (9.1)	-1.2 (0.2)
Native grasses*	8.9	7.3	-	0.1	53.3 (13.2)	-1.4 (0.2)
Successional‡	-	7.1	-	-	21.9 (4.7)	-0.9 (0.2)
Restored prairie‡	8.4	4.8	-	-	21.9 (4.7)	-0.9 (0.2)

GWI is expressed in CO_2 equivalents (CO_2e); values >0 indicate a net climate detriment. Propagules refer to cuttings for poplar, rhizomes for miscanthus, and seeds for all others. CO_2 costs per unit input appear in Tables S9 and S10. Differences between the GREET model and values observed in this study appear in Table S12.

*Greenhouse gas emissions during 2008 for all systems were assumed to be equal to 2009.

†Due to poor establishment almost 100% of the stand was replaced during years 2008 and 2009. For details see Sanford et al.[1]

‡ Greenhouse gas emissions during 2008 were unmeasured and assumed to be average emissions for restored prairie between 2009 and 2014.

Table S11. Post-establishment greenhouse gas sources and sinks (g CO_2e m^{-2}) for alternative cropping systems. Values are means (± SE, when available; n=4 Alfisol or 5 Mollisol replicate blocks).

Cropping system	Post establish-ment period (years)	Farming inputs (g CO_2e m^{-2} yr^{-1})		Non-CO_2 greenhouse gases (g CO_2e m^{-2} yr^{-1})		SOC change (g CO_2e m^{-2} yr^{-1})
		Fuel	N fertilizer	N_2O	CH_4	
Alfisol site						
Maize stover	2008-2014	3.5	28.9	11.9 (0.5)	-	-
Switchgrass	2011-2014	4.6	33.8	56.9 (5.0)	-0.9 (0.3)	-115
Miscanthus	2011-2014	4.6	33.8	40.0 (5.1)	-1.8 (0.4)	-115
Poplar	2008-2013	0.6*	-	27.1 (2.1)	-1.2 (0.1)	-115
Native grasses	2011-2014	4.6	33.8	77.6 (3.2)	-0.6 (0.2)	-115
Successional	2011-2014	4.6	33.8	53.7 (12.8)	-0.8 (0.1)	-115
Restored prairie	2011-2014	3.5		10.9 (1.8)	-1.0 (0.1)	-115
Mollisol site						
Maize stover	2008-2014	3.5	45.9	19.4 (0.4)	-	-
Switchgrass	2011-2014	4.6	33.8	129.8 (8.8)	-1.0 (0.1)	-360
Miscanthus	2011-2014	4.6	33.8	82.6 (18.9)	-1.0 (0.2)	-360
Native grasses	2011-2014	4.6	33.8	56.5 (9.1)	-1.0 (0.2)	-360
Successional	2011-2014	4.6	33.8	69.6 (7.7)	-1.3 (0.3)	-360
Restored prairie	2011-2014	3.5	-	21.0 (2.4)	-0.8 (0.0)	-360

GWI is expressed in CO_2 equivalents (CO_2e); values <0 indicate a net climate benefit. SE in parentheses when available (n=4 replicate blocks for Alfisol and 5 for Mollisol). Post-establishment refers to the period with stable root C stocks and net negative GWI. CO_2 costs per unit input appear in Tables S9 and S10. Differences between GREET model estimates and values observed in this study appear in Table S12.

*After planting and fertilization in 2008, the only fuel needed in the poplar treatment was for harvest in 2013.

Table S12. Post-establishment belowground biomass (root stocks, expressed as $CO_2e\ m^{-2}$) for alternative cropping systems. Values are means (± SE, n=5 replicate blocks).

	Belowground biomass ($g\ CO_2e\ m^{-2}$)	
Cropping system	Alfisol	Mollisol
Switchgrass	2102 (91)	3667 (40)
Miscanthus	3965 (234)	4442 (928)
Poplar	1503 (390)	-
Native grasses	1626 (119)	1801 (138)
Successional	1024 (66)	1083 (89)
Restored prairie	1271 (96)	954 (47)

Table S13. Emission intensities for cellulosic biomass used to produce ethanol or equivalent liquid transportation fuel (scenario 1 – see Materials and Methods) and cellulosic electricity is used to propel electric vehicles (scenario 3 – see Materials and Methods) after long-term soil organic carbon equilibration (without CCS). Values are means (± SE, n=4 Alfisol or 5 Mollisol replicate blocks).

Cropping system	Emission intensity ($gCO_2e\ MJ^{-1}$)			
	Ethanol scenario		Electricity scenario	
	Alfisol	Mollisol	Alfisol	Mollisol
Maize stover	20.3 (0.7)	19.2 (0.4)	22.3 (0.8)	21.2 (0.4)
Switchgrass	21.9 (3.0)	37.8 (3.0)	24.1 (3.3)	41.2 (3.3)
Miscanthus	7.1 (1.2)	15.6 (2.9)	8.1 (1.3)	17.3 (3.2)
Poplar	10.4 (0.5)	-	9.4 (0.4)	-
Native grasses	39.3 (9.8)	28.7 (4.1)	42.9 (10.7)	31.4 (4.5)
Successional	61.9 (14.6)	62.8 (13.7)	67.2 (15.8)	68.3 (14.9)
Restored prairie	6.7 (1.7)	12.2 (2.4)	7.6 (1.9)	13.6 (2.7)

Table S14. Comparison assumptions for selected inputs of GREET (2015) model and current work.

Biofuel feedstock	Ethanol yield* ($L\ kg^{-1}$)		Soil N_2O emission ($g\ CO_2e\ MJ^{-1}$)			Fertilizer† ($g\ CO_2e\ MJ^{-1}$)			Farming‡ ($g\ CO_2e\ MJ^{-1}$)		
	GREET	Current study	GREET	Mollisol	Alfisol	GREET	Mollisol	Alfisol	GREET	Mollisol	Alfisol
Maize stover	0.33	0.31	1.54	5.48	5.51	5.94	12.97	13.34	3.29	0.99	1.62
Switchgrass	0.33	0.31	8.62	30.03	13.63	6.19	8.10	8.10	3.16	0.84	0.84
Miscanthus	0.33	0.31	4.47	11.17	3.97	2.98	3.35	3.35	2.34	0.35	0.35
Poplar	0.33	0.25	2.71	n/a§	5.63	2.05	n/a	3.28	3.22	n/a	0.39

Values calculated for post-establishment years (see Fig. 1). For maize stover, average values between years 2008 and 2013 were used; for the poplar system, average values for full rotation were used. Ethanol energy content assumed to be 21.1 MJ L^{-1}; not including indirect land-use change nor changes in soil organic carbon or belowground biomass carbon stocks (i.e., roots).

*Dry mill.

†CO_2 cost of applied fertilizer divided by energy production.

‡Includes CO_2 cost of maize seeds and poplar cuttings, but not of switchgrass seeds and miscanthus rhizomes.

§Poplar at Mollisol location not included in the analysis (see Materials and Methods).

Table S15. National mitigation potentials for ethanol powered vehicles if cellulosic crops were grown on 55 Mha of abandoned U.S. cropland, and if maize stover were harvested from 50% of 33 Mha of available maize cropland, with and without CCS (scenarios 1 and 2 – see Materials and Methods) and before and after SOC equilibration, based on values in Table S8. Values are means (± SE, n=4 Alfisol or 5 Mollisol replicate blocks).

Cropping system	Global warming impact ($Tg\ CO_2e\ yr^{-1}$)							
	Before SOC/without CCS		Before SOC/with CCS*		After SOC/without CCS		After SOC/with CCS*	
	Alfisol	Mollisol	Alfisol	Mollisol	Alfisol	Mollisol	Alfisol	Mollisol
Maize stover	26 (1.3)	43 (1.2)	68 (4.3)	112 (3.8)	26 (1.3)	43 (1.2)	68 (4.3)	112 (3.8)
Switchgrass	231 (43)	333 (34)	507 (114)	619 (73)	168 (31)	135 (14)	443 (100)	421 (50)
Miscanthus	551 (120)	516 (109)	1216 (321)	1009 (233)	488 (107)	323 (68)	1153 (304)	811 (188)
Poplar	287 (15)	-	680 (39)	-	224 (12)	-	617 (36)	-
Native grasses	151 (53)	314 (55)	340 (146)	524 (107)	87 (31)	116 (20)	277 (118)	326 (67)
Successional	89 (27)	227 (68)	185 (66)	337 (122)	26 (7.7)	29 (8.8)	122 (43)	139 (51)
Restored prairie	155 (52)	283 (74)	279 (112)	405 (127)	92 (31)	85 (23)	216 (86)	207 (65)

*at 48% carbon capture and storage efficiency

Table S16. Total mitigation potentials for electric vehicles if cellulosic crops were grown on 55 Mha of abandoned U.S. cropland, and if maize stover were harvested from 50% of 33 Mha of available maize cropland[17], with and without CCS (scenarios 3 and 4 – see Materials and Methods) and before and after SOC equilibration, based on values in Table S9. Values are means (± SE, n=4 Alfisol or 5 Mollisol replicate blocks).

Cropping system	Global warming impact ($Tg\ CO_2e\ yr^{-1}$)							
	Before SOC/without CCS		Before SOC/with CCS		After SOC/without CCS		After SOC/with CCS	
	Alfisol	Mollisol	Alfisol	Mollisol	Alfisol	Mollisol	Alfisol	Mollisol
Maize stover	54 (2.5)	110 (3.0)	134 (7.3)	239 (7.3)	55 (2.0)	110 (2.5)	134 (7.3)	219 (6.6)
Switchgrass	422 (72)	613 (59)	939 (188)	1148 (122)	359 (48)	415 (31)	876 (173)	869 (89)
Miscanthus	1014 (200)	999 (186)	2261 (526)	1913 (397)	950 (154)	801 (128)	2197 (510)	1577 (319)
Poplar	621 (27)	-	1358 (69)	-	558 (21)	-	1295 (65)	-
Native grasses	282 (90)	520 (89)	637 (240)	914 (169)	219 (53)	322 (42)	574 (213)	656 (117)
Successional	152 (36)	335 (106)	334 (106)	541 (180)	92 (18)	137 (29)	272 (83)	311 (97)
Restored prairie	241 (77)	403 (111)	475 (170)	633 (183)	178 (44)	205 (42)	412 (145)	400 (110)

*at 90% carbon capture and storage efficiency

Table S17. Maximum energy production potential (lower heating value of harvestable biomass converted to useful energy) and the reduction in emissions of CO_2e relative to petroleum for alternative cellulosic biofuel cropping systems used for ethanol or electric vehicles with and without carbon capture and storage (CCS) before SOC equilibration to higher levels. Values are means (± SE, n=4 Alfisol or 5 Mollisol replicate blocks).

		Energy production potential ($GJ\ ha^{-1}\ y^{r-1}$)		CO_2 emission reduction relative to petroleum (%)			
Site	Cropping system	Ethanol	Electric	Ethanol	Ethanol + CCS	Electric	Electric + CCS
Alfisol	Maize stover	21 (0.1)	57 (2)	78 (3)	204 (10)	74 (4)	329 (17)
	Switchgrass	42 (5)	112 (14)	106 (15)	231 (43)	103 (19)	358 (67)
	Miscanthus	102 (5)	270 (40)	104 (17)	230 (50)	102 (22)	357 (78)
	Poplar	49 (1)	160 (4)	114 (5)	270 (14)	109 (6)	364 (19)
	Native grasses	29 (7)	77 (19)	100 (25)	226 (79)	97 (34)	352 (123)
	Successional	15 (3)	39 (7)	118 (28)	243 (74)	116 (35)	371 (112)
	Restored prairie	19 (4)	51 (11)	157 (40)	283 (95)	158 (53)	413 (139)
Mollisol	Maize stover	35 (0.7)	93 (2)	80 (2)	205 (6)	75 (2)	330 (9)
	Switchgrass	44 (3)	116 (7)	147 (12)	273 (28)	148 (15)	403 (41)
	Miscanthus	75 (7)	198 (19)	134 (25)	260 (55)	134 (28)	389 (82)
	Poplar						
	Native grasses	32 (3)	85 (9)	188 (27)	314 (55)	192 (34)	447 (78)
	Successional	17 (3)	45 (9)	260 (57)	386 (115)	270 (81)	525 (157)
	Restored prairie	19 (3)	50 (8)	290 (58)	416 (110)	303 (80)	558 (147)

See also Figure 3.

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