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Ecological limits to terrestrial biological carbon dioxide removal

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Abstract Terrestrial biological atmospheric carbon dioxide removal (BCDR) through bioenergy with carbon capture and storage (BECS), afforestation/reforestation, and forest and soil management is a family of proposed climate change mitigation strategies. Very high sequestration potentials for these strategies have been reported, but there has been no systematic analysis of the potential ecological limits to and environmental impacts of implementation at the scale relevant to climate change mitigation. In this analysis, we identified site-specific aspects of land, water, nutrients, and habitat that will affect local project-scale carbon sequestration and ecological impacts. Using this framework, we estimated global-scale land and resource requirements for BCDR, implemented at a rate of 1 Pg C y^{-1}. We estimate that removing 1 Pg C y^{-1} via tropical afforestation would require at least 7×10^6 ha y^{-1} of land, 0.09 Tg y^{-1} of nitrogen, and 0.2 Tg y^{-1} of phosphorous, and would increase evapotranspiration from those lands by almost 50 %. Switchgrass BECS would require at least 2×10^8 ha of land (20 times U.S. area currently under bioethanol production) and 20 Tg y^{-1} of nitrogen (20 % of global fertilizer nitrogen production), consuming 4×10^{12} m^3 y^{-1} of water. While BCDR promises some direct (climate) and ancillary (restoration, habitat protection) benefits, Pg C-scale implementation may be constrained by ecological factors, and may compromise the ultimate goals of climate change mitigation.

1 Introduction

Global fossil fuel use emits roughly 8 Pg carbon (C) y^{-1} to the atmosphere (EIA 2011). Because the oceans and terrestrial biosphere take up only roughly 55 % of these emissions

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(Ballantyne et al. 2012), atmospheric carbon dioxide (CO_2) concentrations are growing at roughly 2 ppm y^{-1} (NOAA 2012). Given this trend, strategies for atmospheric carbon dioxide removal (CDR), are being considered for climate change mitigation. One category of CDR, *terrestrial biological* carbon dioxide removal (BCDR), increases terrestrial reservoirs' carbon uptake and storage by increasing plant productivity and/or carbon residence times, reducing the fraction of emissions that remain airborne, which currently averages 45 %. If BCDR could be implemented on the scale of 1 Pg of carbon (C) per year—the magnitude of stabilization wedges used in Pacala and Socolow (2004)—it could contribute substantially to climate change mitigation.

One BCDR approach is bioenergy with carbon capture and storage (BECS). The next generation of bioenergy technology aims to replace current feedstocks such as corn, sorghum, sugarcane, rapeseed, soy, and oil palm with dedicated cellulosic crops (Kszos et al. 2000; Heaton et al. 2008b), such as woody tree species and the grasses switchgrass (*Panacum vergatum*) and miscanthus (*Miscanthus x giganteus*) (Lewandowski et al. 2000). These fuel crops can produce usable energy with <10 % the energy inputs of corn (McLaughlin and Walsh 1998), with lower water and nutrient requirements (Msangi et al. 2007; Heaton et al. 2008a). In addition to offsetting fossil fuel emissions and sequestering carbon stocks in soils and biomass (Tilman et al. 2006), cellulosic BECS using geologic reservoirs could provide a continual flow of carbon out of the atmosphere.

A second BCDR strategy is forestry-based sequestration, which removes atmospheric carbon and stores it in forest biomass by increasing forest area and/or carbon density. Forestry BCDR projects are grouped into three categories: (1) reforestation (planting trees in deforested areas), (2) afforestation (planting trees in historically treeless areas such as grasslands or shrublands), and (3) forest and harvest management. Afforestation and commercial reforestation projects often use monocultures of fast-growing species such as pine and eucalyptus (Wright et al. 2000).

Third, soil carbon sequestration aims to increase soil carbon stocks through land management practices such as reducing agricultural tillage, planting more deeply-rooted species, and incorporating relatively persistent compounds (e.g., biochar) into soil for long-term storage. Soil carbon sequestration is often conceived of as optimizing inputs and plant cover to restore carbon lost from past land use (West and Marland 2002; Brown et al. 2004; Lal 2004); global soils have lost an estimated 230 Pg C due to land use in the last 10,000 y (Lal 2001), and conventional cultivation decreases soil carbon by 25–50 % after 30–50 years (Johnson 1992; Post and Kwon 2000).

Many studies of carbon sequestration consider potential co-benefits, such as increased soil fertility, erosion control, habitat improvement, and community development (Cook and Beyea 2000; Silver et al. 2000; Sauerbeck 2001; Paul et al. 2002; West and Marland 2002; Lal 2008). Negative ecological effects deserve detailed analyses as well, but have received less attention than co-benefits. The most comprehensive analysis to-date, the IPCC third assessment report (Kauppi et al. 2001), included the sequestration capacity and potential ecosystem effects of various BCDR strategies, but had a relatively brief treatment of potential negative ecological effects.

Ecological constraints have been identified in individual studies and reports (Schaeffer et al. 2006; van Vuuren et al. 2007; Dornburg et al. 2008; Thomson et al. 2009), but not synthesized or reviewed. This paper offers a systematic overview of the likely ecological and biophysical limits, negative impacts, and resource implications of BCDR implemented at the Pg C y^{-1} scale.

2 Ecological considerations

Much of the literature presents an optimistic view of BCDR. Studies estimate that BECS could sequester as much as 5 Pg C annually (Azar et al. 2006) or 771 Pg C cumulatively by 2100 (Lenton and Vaughan 2009). Estimates of afforestation BCDR potential are based on steady-state stocks and land availability over the next 100 years, ranging from a cumulative 1.5 Pg C in California (Brown et al. 2004) to 104 Pg C globally (Nilsson and Schopfhauser 1995). Maximum soil carbon sequestration estimates, based on past soil carbon losses and land availability over the next 50 years, are as high as 30–60 Pg C (Lal 2004)

These optimistic estimates overlook or downplay a number of ecological considerations. BCDR requires productive land, nutrients, and water, all of which are limited in the global biosphere. In light of such limitations, actual BCDR capabilities likely fall well below the literature estimates, and efforts to maximize carbon sequestration may negatively affect biodiversity and compete with other resource demands. This section discusses these ecological realities in relation to BCDR.

2.1 Land and productivity

BCDR depends on increasing and/or utilizing plant productivity, and most strategies require land of at least moderate fertility (except degraded land restoration). The upper bound on large-scale BCDR could be set by available land and its rate of carbon uptake; conversely, large scale BCDR could intensify competition for arable or manageable land.

Large scale expansion of cellulosic crops for BECS may put pressure on food security (Msangi et al. 2007), forest conservation (Danielsen et al. 2009), and other uses of productive land. Such competition is already apparent, and global demands for food are projected to nearly double over the next 50 years (Tilman et al. 2001), increasing the land area needed for both food and biofuel production.

Like bioenergy, forestry and soil-based BCDR face land and productivity constraints and may compete with other land uses. Although some forestry projects provide co-benefits for local communities while sequestering carbon (e.g., agroforestry), others isolate people from ecosystem services, as when commercial plantations prevent local communities from harvesting wood or other forest products (Jindal et al. 2008).

2.2 Nutrient balance and soil fertility

Plants require nitrogen, phosphorous, and other elements to fix carbon. Nitrogen limits productivity in many temperate ecosystems, and phosphorous limits productivity in much of the tropics (i.e., adding the nutrient increases productivity) (Herbert and Fownes 1995; Cleveland et al. 2002), so insufficient nitrogen and phosphorous may limit future forest biomass increases (Hungate et al. 2003; Reich et al. 2006). To overcome these limitations, tree plantations are typically fertilized at the nursery and transplant stages (O'Connell 1994), and long-term increased nutrient demands from afforestation can lead to decreased soil nutrient availability (Jackson et al. 2000; Berthrong et al. 2009), soil acidification, (Jobbágy and Jackson 2004), and decreased capacity to retain added nutrients (Berthrong et al. 2009).

Nutrient limitations pertain to BECS as well. Harvesting bioenergy biomass removes nutrients, particularly potassium and other structural components of aboveground plant tissue. While most cellulosic bioenergy grasses require less fertilization than corn or other ethanol feedstocks (Lewandowski et al. 2000), a series of U.S. studies determined optimal

fertilization rates of 40–120 kg nitrogen ha^{-1} y^{-1}, with yield improvements realized up to 224 kg nitrogen ha^{-1} y^{-1} (McLaughlin and Adams Kszos 2005).

Mobilizing nutrients for fertilization has negative environmental consequences downstream and downwind. Fertilizer runoff to oceans, rivers, and lakes has led to species composition and food web shifts, eutrophication, toxin formation, and other impacts (Mitsch et al. 2001; Camargo and Alonso 2006). For example, 70 % of the nitrate loading responsible for the large hypoxic zone in the Gulf of Mexico is attributed to agriculture (Rabalais et al. 2002). Furthermore, producing and applying fertilizer requires energy from fossil fuels; such activities have embedded carbon costs that counteract the carbon benefits of increased productivity (Schlesinger 2010).

2.3 Water

Plant photosynthesis requires water to fix carbon. Ecosystems consume water, i.e., withdraw water permanently from a catchment, via evaporation and transpiration (evapotranspiration, ET). By altering plant species, density, and productivity, BCDR activities alter ET, seasonal water use patterns, and rooting depth for water use—all of which can affect water quality and flows for other human and ecosystem uses.

Numerous studies have demonstrated that afforestation affects local hydrology (Bosch and Hewlett 1982; Brown et al. 2005; Nosetto et al. 2011). Because replacing grasslands or shrublands with forests increases ET, afforestation projects can lower the water table, reduce streamflow, and decrease watershed water yield (precipitation - ET) (Farley et al. 2005). On average, where studied, afforestation of grasslands and shrublands cuts streamflow by 1/3 to 3/4, often leading to a loss of perennial streamflow in regions where water yield is already low (Farley et al. 2005). In South Africa, a paired catchment experiment saw streams dry up completely 9 and 12 years after eucalyptus and pine afforestation (Lesch and Scott 1997), and similar effects were seen in Argentina (Nosetto et al. 2011). More generally, a 10 % increase in tree cover decreases water yield by 25–40 mm across a range of ecosystems (Sahin and Hall 1996; Jackson et al. 2000).

By changing the water table and soil texture, forestry projects also affect soil and water salt balance in site-specific ways. In Australia, reforestation is used to reduce salinity; tree growth increases water table depth, reducing infiltration of saline groundwater into surface soils (George et al. 1999). In Argentina's humid grasslands, by contrast, afforestation salinizes soils by using up the thin freshwater lens above saline groundwater (Jobbágy and Jackson 2004).

The amount of water consumed by BECS for energy generation and for carbon capture and storage is two orders of magnitude lower than that used for biomass growth, but this industrial usage requires water diversion, conveyance, and treatment with intensive localized effects (Scown et al. 2011). As with nutrient additions, water infrastructure and groundwater pumping require energy as well, reducing net carbon sequestration gains (Schlesinger 2000).

2.4 Biodiversity

Afforestation and bioenergy projects can threaten biodiversity. Recent decades have seen increased conversion of natural forests to pine or eucalyptus monocultures (Zurita et al. 2006), and policy incentives for further land conversion may negatively affect biodiversity (Caparrós and Jacquemont 2003). Bioenergy expansion can directly convert native habitats to cropland or indirectly drive land clearing elsewhere by displacing cropland or rangeland (Cook and Beyea 2000; Lapola et al. 2010).

How a BCDR project affects biodiversity depends on the native ecosystem type, land use history, planted species, and spatial arrangement of remaining native habitats. Existing research is difficult to synthesize (Barlow et al. 2007), but in many cases, BCDR-type land use reduces biodiversity. Primary forests tend to have higher plant and animal diversity than secondary or plantation forests (Lindenmayer and Hobbs 2004; Zurita et al. 2006; Barlow et al. 2007), and even restored grasslands or forests often have lower biodiversity than nearby native ecosystems (Camill et al. 2004).

2.5 Non-CO_2 climate impacts

In addition to CO_2 emissions, other greenhouse gas emissions, albedo, and latent heat fluxes influence regional and global climate (Diffenbaugh 2009; Georgescu et al. 2009). As a result, non-CO_2 climate impacts of BCDR may offset intended climate mitigation.

Fertilizer use in BCDR is an important part of this tradeoff. 1–5 % of global nitrogen fertilizer is converted to nitrous oxide (global warming potential 296 times higher than CO_2) (De Klein et al. 2006; Solomon et al. 2007; Crutzen et al. 2008). Where quantified, adding manure or inorganic fertilizer, planting legumes, or incorporating crop residues all resulted in nitrous oxide emissions offsetting 75–310 % of the sequestered CO_2 (Robertson et al. 2000; Brown et al. 2004; Li et al. 2005).

The energy balance of a given land area is determined primarily by the albedo and ET, associated with plant cover type and productivity (Pielke and Avissar 1990). By altering land cover and land use, BCDR may therefore influence local climate directly, intensifying or counteracting the effects of atmospheric CO_2 reduction in site-specific ways (Gibbard et al. 2005). Increases in forest cover generally cause cooling in the tropics where the ET effect dominates (Claussen et al. 2001) and warming in mid- and high-latitudes where the albedo effect is strong (Betts 2000). In temperate latitudes, where substantial bioenergy cultivation may occur, net biophysical effects are difficult to predict (Bonan 2008).

3 Estimated requirements for 1 Pg C y^{-1} sequestration

The challenge for BCDR as a mitigation measure is implementing it at a meaningful scale. In this section, we estimate resource requirements and ancillary damages associated with 1 Pg C y^{-1} atmospheric carbon removal using either eucalyptus afforestation or switchgrass BECS. Using emissions factors, typical resource inputs, and efficiencies from the literature, we generate order of magnitude estimates of land area requirements, nitrogen (N) and phosphorous (P) applications, water consumption, and nitrous oxide (N_2O) emissions.

3.1 Inputs to the calculation

We calculated resource requirements using literature values for typical carbon accumulation rates, carbon inputs and emissions from agricultural and silvicultural practices, ecosystem-specific evapotranspiration rates, and industrial processing efficiencies (Table 1). Where the literature offers a range of values for a given process, technology, or ecosystem property, we calculate low and high estimates (equations in Electronic Supplementary Material).

Switchgrass production requires fossil fuel inputs for machinery used in establishment (soil preparation and seed sowing), cultivation, harvest, and transportation to the processing plant (Qin et al. 2006). Fertilizer, pesticides, and herbicides applied during switchgrass production carry additional carbon costs, from fossil fuels used in their manufacture

Table 1 Literature-derived values used to calculate resource requirements for 1 Pg C y^{-1} BCDR with tropical eucalyptus afforestation or temperate switchgrass BECS

	Units	Value	Value reference(s)
(a) Afforestation			
C sequestration capacity[a]	t C ha^{-1}	65–195	Winjum et al. 1992
Time to reach sequestration capacity	y	113	Silver et al. 2004
		158	Lugo et al. 1988, Stape et al. 2008
Time to reach sequestration capacity	y	50	Winjum et al. 1992
		61	Silver et al. 2004
Nursery N additions	(kgN) ha^{-1}	0.22	Stape et al. 2001
			Stape et al. 2010
Nursery P additions	(kg P) ha^{-1}	0.13	Stape et al. 2001
			Stape et al. 2010
Planting N additions	(kgN) ha^{-1}	14–47	Stape et al. 2001
			Stape et al. 2010
			de Aguiar Ferreira and Stape 2009
Planting P additions	(kg P) ha^{-1}	30–50	Stape et al. 2001
			Stape et al. 2010
			de Aguiar Ferreira and Stape 2009
ET, forested land with MAP of 1,250–1,500 mm	mm y^{-1}	1050	Zhang et al. 2001
			Farley et al. 2005
ET, grassland with MAP of 1250–1,500 mm	mm y^{-1}	750	Zhang et al. 2001
			Farley et al. 2005
(b) Switchgrass			
Gross photosynthetic uptake	g CO_2 kg^{-1}	1540	Qin et al. 2006
Production CO_2 losses[b]	%	5.0–8.5	Qin et al. 2006
Storage dry biomass losses	%	4	Qin et al. 2006
Gasification/conditioning	%	38	Rhodes and Keith 2005
Carbon capture efficiency	%	89	Rhodes and Keith 2005
Transport/injection losses	%	5.9–12	Weisser 2007
Dry biomass productivity	kgha^{-1} y^{-1}	5600–7000	Thomson et al. 2009
		10000	Heaton et al. 2004
		9100–23000	McLaughlin and Adams Kszos 2005
Typical N additions	kgha^{-1} y^{-1}	80	Kszos et al. 2000
			McLaughlin and Adams Kszos 2005
N converted to N_2O	%	2	De Klein et al. 2006
ET water consumption	Lm^2 y^{-1}	750	Dominguez-Faus et al. 2009
			Hickman et al. 2010
IGCC water use	gal (kg biomass)$^{-1}$	0.59	Mani et al. 2004
			Rhodes and Keith 2005
			Klett et al. 2007

[a] Includes aboveground biomass, belowground biomass, soil carbon, and litter

[b] Embedded emissions include machinery involved in soil preparation and seed sowing; production, application and transport of agricultural chemicals; harvest and baling or pelletizing; and transport to the processing plant

(Schlesinger 2010), transportation, and application. We subtract the CO_2 emissions embedded in these processes from overall carbon sequestration gains. Similarly, carbon lost or embedded via physical biomass losses or energy inputs in the combustion, capture, and storage processes increase the resources required per unit sequestered carbon (Rhodes and Keith 2005; Qin et al. 2006; Weisser 2007).

The switchgrass calculation does not include carbon emissions from direct or indirect land clearing, for our estimate represents the resources required by BECS at steady-state. However, land clearing can seriously affect biodiversity, ecosystem services, and carbon storage over decades or centuries, discussed by Fargione et al. (2008), Searchinger et al. (2008), and Plevin et al. (2010). Additionally, irrigation infrastructure introduces additional carbon emissions. Our calculation also omits carbon emissions reductions from avoided fossil fuel use, addressing BECS with respect to BCDR only, without including its potential value for energy security or as an alternative to fossil fuels to avoid greenhouse gas emissions.

The per-Pg carbon resource demands of afforestation depend on climate, location, tree species, monoculture vs. polyculture, carbon content of the replaced ecosystem (Winjum et al. 1992; Silver et al. 2004), time for the forest carbon to reach steady state, the two ecosystems' relative ET rates (Zhang et al. 2001), and fertilization practices during seedling growth and transplantation (de Aguiar Ferreira and Stape 2009). Due to this high variability, we constrained our estimate to a monoculture eucalyptus plantation in a tropical region with mean annual precipitation of 1,200–1,500 mm y^{-1}.

Whereas the flow of carbon out of the atmosphere using BECS is theoretically continuous through time, afforestation sequesters carbon by maximizing biomass stocks. As forest biomass reaches steady state, the net carbon uptake rate declines, with little additional CDR after 50 years. Because of this saturation effect, achieving an annual flow of 1 Pg C y^{-1} CDR would require planting new forests annually and protecting existing afforested land (Fig. 1).

3.2 Results

For BECS, a field-to-reservoir accounting of carbon flows shows that net sequestration of 1 Pg C requires fixing 2.1 Pg C, considering the CO_2 losses from farm and transport fossil fuel use, pre-capture storage and processing, and CO_2 capture and injection (Fig. 2). We estimate that supporting 2.1 Pg C y^{-1} of switchgrass productivity—an increase equal to roughly 3 % of global terrestrial net primary productivity (Haberl et al. 2007)—would use 218–990 Mha of land, 17–79 Tg N y^{-1} fertilizer, and $1.6–7.4\times10^{12}$ m^3 water y^{-1} from ET and industrial processing (Table 2). Lower fertilization levels should be possible, but no studies of mature switchgrass under repeated harvest management were found to confirm this. This 1 Pg C y^{-1} net sequestration would commandeer ~16–75 % of current global N fertilizer production and 14–65 times the U.S. land area currently under bioethanol production.

To sequester a stock of 1 Pg C in forest biomass, tropical afforestation projects would cover 6.7–15 Mha (equal to 4 % of global plantation forest area in 2000), apply $2.0–7.7\times10^5$ Mg P and $9–73\times10^4$ Mg N as fertilizer, and consume $1.2–2.7\times10^{12}$ m^3 y^{-1} more water than the grasslands they replace, increasing the percentage of mean annual precipitation consumed via ET on those lands from 50 % to 75 %. To a coarse approximation, 50 times this land area must be converted from grass/shrub to silvicultural management to provide an average annual CDR rate of 1 Pg C y^{-1} for 50 years. This ~300–750 Mha land requirement (equal to as much as half the global tropical grassland area) would be accompanied by a cumulative fertilizer demand of $10–15\times10^6$ Mg P and $4.5–15\times10^6$ MgN over 50 years, and a water demand $1.2–2.7\times10^{12}$ m^3 y^{-1} greater than grassland. To continue CDR beyond this

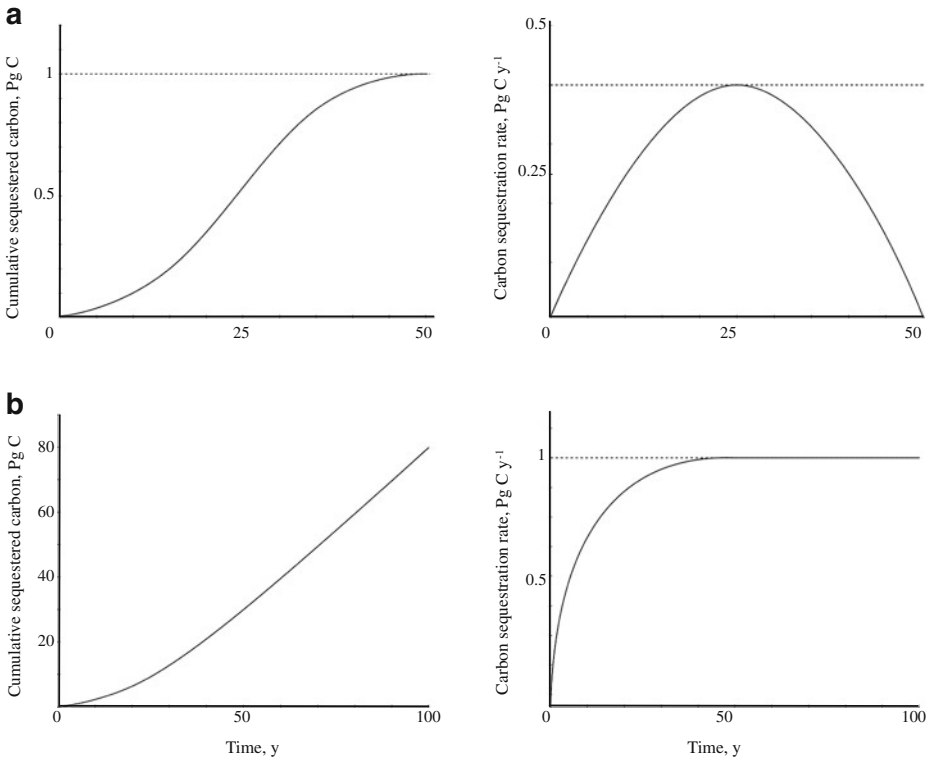

Fig. 1 Afforestation carbon sequestration scenarios. Schematic of carbon sequestration by afforestation under two different land conversion scenarios. **a** Land with steady state sequestration capacity of 1 Pg C is afforested in year 1. The C accumulation rate peaks at an intermediate year and then saturates after 50 years. **b** The same land area is converted each year perpetually. Total carbon accumulation reaches 1 Pg y^{-1} after 50 years, and continues at that rate as further land conversion continues

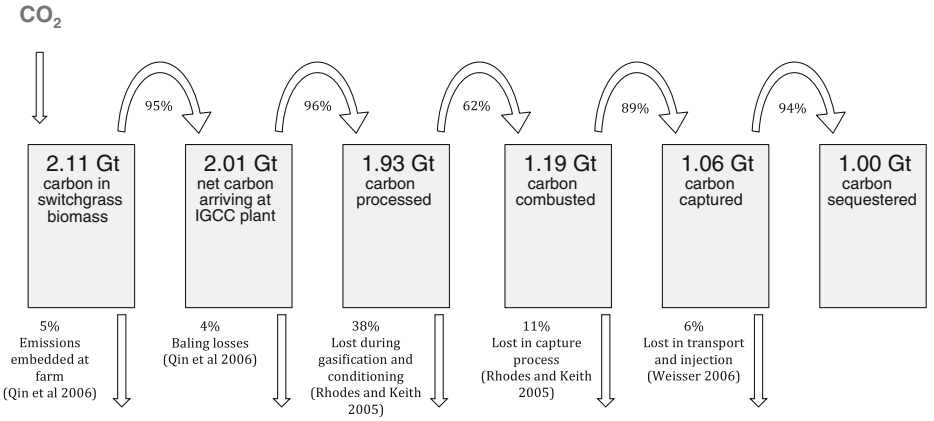

Fig. 2 BECS carbon flow. Schematic of carbon flow, life cycle emissions, and carbon losses during temperate switchgrass production and processing with carbon capture and storage. The percentage values are carbon losses calculated from literature values in Table 1 for an IGCC facility retrofitted to burn biomass only

Table 2 Estimated resource requirements for 1 Pg y^{-1} BCDR using tropical eucalyptus afforestation or temperate switchgrass BECS. All values calculated using the literature inputs, stocks, and flows listed in Table 1. Eucalyptus monoculture plantation is assumed to replace grassland or shrubland, and forest biomass is maintained at steady-state levels. Switchgrass biomass processing is assumed to use integrated gasification combined cycle, retrofitted to burn biomass only. Carbon losses or gains due to land use change are not included in calculation

	Strategy	Mean	Range	Equivalent figure
(a) Land area	Afforestation	8.5×10^{6} ha y^{-1}	$6.7\times10^{6}-1.5\times10^{7}$	4.5 % of the total global plantation forest in 2000 (FAO 2012), or 0.6 % of the tropical grassland area per year (Scurlock and Hall 1998)
	Switchgrass	5.8×10^{8} ha	$2.2\times10^{8}-9.9\times10^{8}$	17 times the land under maize production in the US in 2010 (FAO 2012), or 38 times the land under bioethanol production in the US in 2010 (EIA 2011)
(b) Nitrogen	Afforestation	2.6×10^{8} (kgN) y^{-1}	$9.0\times10^{7}-7.3\times10^{8}$	0.2 % of the global annual N fertilizer production in 2009 (FAO 2012)
	Switchgrass	4.6×10^{10} (kgN) y^{-1}	$1.7\times10^{10}-7.9\times10^{10}$	44 % of the global annual N fertilizer production in 2009 (FAO 2012)
(c) Phosphorous	Afforestation	3.4×10^{8} (kg P) y^{-1}	$2.0\times10^{8}-7.7\times10^{8}$	0.7 % of the global annual P fertilizer production in 2009 (FAO 2012)
(d) Water	Afforestation	4.7×10^{12} m^3 y^{-1} y^{-1}	$3.5\times10^{12}-8.1\times10^{12}$	Local ET increase from 50 % of mean annual precipitation to 75 % of mean annual precipitation (Zhang et al. 2001)
	Switchgrass	4.4×10^{12} m^3 y^{-1}	$1.6\times10^{12}-7.4\times10^{12}$	0.6 m^3 water (160 gal) per kg switchgrass grown (Mani et al. 2004; Rhodes and Keith 2005; Klett et al. 2007)

50-year window, additional land conversion would be required, at a rate of 6.7–15 Mha per year, if land of similar fertility were available.

4 Discussion

BCDR faces ecological constraints at the local project level and at large scale. The land area required for 1 Pg C y^{-1} BCDR ranges broadly in our calculation, from ~200 to ~1,000 Mha, due to literature differences in switchgrass productivity. Average productivities over broad regions are relatively low (Thomson et al. 2009), suggesting that actual land requirements would fall at the high end of our calculated range. A land requirement of as much as 990 Mha for switchgrass or 15 Mha y^{-1} for afforestation implies important opportunity costs and impacts on biodiversity and food, fuel, and fiber production. Of the 100 million km^2 of productive land area on earth, nearly 40 % is used for agriculture or grazing lands, and scarcity of unexploited arable land constrains agricultural production in many areas (De Fraiture et al. 2008). Humans have already appropriated one third of global net primary productivity, using it primarily as cropland (14 %), pasture (8 %), and managed forest (3 %) (Vitousek et al. 1986; Haberl et al. 2007). Much of the remaining terrestrial vegetation supports valuable ecosystem services such as water provision, local climate regulation, erosion and flood control, and genetic resource protection (Millennium Ecosystem Assessment 2005).

At the scale relevant to global climate change mitigation, BCDR would entail significant consumption of the world's fertilizer supply, with attendant downstream consequences. This is particularly true for BECS, for which our estimated 17–79 Tg N y^{-1} applied per sequestered Pg C y^{-1} amounts to as much as 75 % of global annual nitrogen fertilizer production. (As with the land area estimate, the low-end estimate is unrealistic at large scale.) Humans have already more than doubled the natural nitrogen fixation rate to over 250 Tg N per year (Vitousek 1994), and phosphorous fertilizer use increases 1 % annually (FAO 2012).

In some cases, fertilizer availability may constrain large-scale BCDR. Fertilizer phosphorous, for example, is a non-renewable resource whose global use increased 5-fold between 1960 and 2000 (FAO 2012). Prices of mineral fertilizers reflect relative scarcity; phosphorous prices tripled in the last decade and potassium chloride prices quadrupled since 2002 (USDA 2012).

BCDR activities increase water demands in a world where water scarcity constrains plant productivity in many regions (De Fraiture et al. 2008) and a third of humanity lives in water-stressed regions (Vörösmarty et al. 2000). ET water losses from switchgrass production amount to as much as 7×10^{12} m^3 y^{-1} for 1 Pg C y^{-1} sequestration, and replacing grasslands with forest increases ET water losses by as much as 50 %. If BCDR intensifies pressure on water resources, it could increase tapping of groundwater reserves (Postel et al. 1996), reduce access to clean water (Vörösmarty et al. 2000), divert water from ecosystems, and compete with food demands (Falkenmark 1997).

Scale matters for BCDR, but scaling up may disproportionately impact ecosystems if land suitable for BCDR becomes scarce. If financial incentives spur BCDR expansion, ecosystems and vulnerable societies may pay the cost. Current biodiversity losses, for example, are tightly tied to land use. The fossil record indicates that the past 100 years has seen species extinctions at 100–1,000 times the background rate (Millennium Ecosystem Assessment 2005), and among five drivers of global biodiversity loss between now and 2100 (climate change, land use change, atmospheric CO_2 increases, nitrogen deposition, and species introductions), land use change—not climate change—is predicted to be the most important (Sala et al. 2000).

While this paper provides an estimate of resource requirements for BCDR, the ranges and magnitudes of associated impacts are harder to quantify. Local air and water pollution, reduced biodiversity, and competition with food security and local livelihoods are all context-dependent negative effects that depend on the project, location, land use history, and societal needs. Our abilities to predict and monitor these impacts are limited by model uncertainty (Friedlingstein et al. 2003), insufficient ecological knowledge, unaccounted-for greenhouse gas emissions (Searchinger et al. 2008), measurement difficulties (Baker et al. 2007), and political and economic complexity (Murray 2008).

In conclusion, achieving large-scale BCDR would require project siting that depends on regional resource constraints and localized impacts, as well as management of globally traded resources. Such ecological constraints are underrepresented in the literature, but will limit BCDR's role in climate change mitigation. Our calculated land and nutrient requirements suggest that previous estimates of sustained BCDR potential of 3–5 Pg C y^{-1} by BECS and afforestation were unrealistically high. Even at the scale of 1 Pg C y^{-1}, either strategy represents a major perturbation to land, water, nitrogen, and phosphorous stocks and flows. Climate change mitigation must be a global priority, but more detailed and comprehensive analyses are needed before pursuing large-scale BCDR.

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