The Global Potential for Carbon Dioxide Removal

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ABSTRACT

The global physical potential of different methods of carbon dioxide removal (CDR) from the atmosphere is reviewed. A new categorisation into plant-based, algal-based and alkalinity-based approaches to CDR is proposed. Within these categories, the key flux-limiting resources for CDR are identified and the potential CO_2 removal flux that each technology could generate is quantitatively assessed – with a focus on the present, 2050 and 2100. This reveals, for example, that use of waste nutrient flows to feed macro-algae for biomass energy with carbon capture and storage (algal BECCS), shows significant CDR potential, without needing the large land areas or freshwater supplies of plant biomass energy crops. Adding up the potentials of different CDR methods, the total CDR potential at present is 1.5–3 PgC yr^{-1} (Petagram of carbon per year), comparable in size to either the natural land or ocean carbon sinks. Already 0.55–0.76 PgC yr^{-1} of this potential has been realised through afforestation and inadvertent ocean fertilisation. The total CDR potential (without including direct air capture) grows such that by mid-century it is 4–9 PgC yr^{-1} and by the end of the century it is 9–26 PgC yr^{-1}, comparable with current total CO_2 emissions of 10 PgC yr^{-1}. The CDR that can be realised under social, economic and engineering constraints is always going to be less than the physical potential. Nevertheless, if combined with reducing CO_2 emissions (conventional mitigation), CDR has the physical potential to help stabilise atmospheric CO_2 by the middle of this century.

1 Introduction

The global carbon cycle is currently out of balance. Human fossil fuel burning and land use change activities are producing a combined source of CO_2 to the atmosphere of around ~ 10 PgC yr^{-1} (petagrams or Pg $= 10^{15}$).[1] This is causing atmospheric CO_2 concentration to rise at ~ 2 ppm yr^{-1} and causing carbon to accumulate in the ocean and in land ecosystems.[1] The rise in atmospheric CO_2 concentration is in turn making the single largest contribution to increasing global temperatures. Thus, in order to minimise the rise of global temperature, the rise of atmospheric CO_2 must be halted (or sunlight reflection methods of geoengineering must be deployed).

In simple terms, stabilizing atmospheric CO_2 concentration demands that carbon sinks (removal fluxes from the atmosphere) match carbon sources (fluxes to the atmosphere). Lowering atmospheric CO_2 concentration demands that sinks exceed sources. The conventional policy framework for achieving stabilization is to reduce CO_2 emissions to match natural (land and ocean) sinks, and then to reduce CO_2 emissions to zero (at least as fast as natural sinks decay). The cumulative carbon emission will then determine the resulting change in global temperature, called the "cumulative warming commitment".[2] This policy approach poses a profound collective challenge to transform the current exponential increase in CO_2 emissions ($\sim 2\%$ yr^{-1} over the past 25 years and $> 3\%$ yr^{-1} at the beginning of the 21st century)[3,1] into a comparable or greater rate of decrease in CO_2 emissions. The required transition of the global energy system must start soon and be completed within decades, if global warming is to be restricted to less than 2 °C above pre-industrial.[2,4,5] Already it demands rates of technological and economic change that may be politically unachievable.[3]

Hence there is growing interest in the potential for deliberate carbon dioxide removal from the atmosphere to augment reductions in CO_2 emissions. Carbon dioxide removal (CDR) – or Negative Emissions Technologies (NETs) – describes a suite of methods that remove CO_2 from the ambient air by biological, chemical or physical means and store the resulting carbon in long lived reservoirs. If both anthropogenic emissions of CO_2 are reduced and CO_2 sinks are created, then the rise of atmospheric CO_2 concentration (and global temperature) can be halted sooner and at a lower level than by reducing emissions alone. Indeed, CDR is already implicit in most scenarios to stay under 2 °C of global warming above pre-industrial, including the IPCC RCP2.6 scenario, where it takes the form of widespread biomass energy with carbon capture and storage (BECCS).[6] CDR effectively reduces the cumulative carbon emission and hence reduces the corresponding global warming commitment.[2] Ultimately, CDR could be used to bring atmospheric CO_2 concentration down to whatever is considered a safe level. CDR may also be used to counter-balance some "essential" or "unavoidable" fossil fuel CO_2 emissions, without increasing the CO_2 concentration.

However, most CDR technologies are more expensive than most conventional emissions reduction options, and hence are unlikely to be used

until after the cheaper mitigation options. Furthermore, because CDR offers the option to reduce atmospheric CO_2 concentrations at some later time, this may reduce the urgency to start cutting emissions now. Indeed, knowing that global temperature change lags changes in radiative forcing, some studies have framed CDR as allowing a temporary overshoot in atmospheric CO_2 concentrations above safe levels,[7] without overshooting a corresponding safe temperature target. This would of course be a risky strategy if the imagined future potential for CDR cannot be realised in practice.

There are thus several reasons to want to know the global potential for CDR from a scientific perspective, before also thinking about the engineering, the costs and the social acceptability of CDR technologies. The most critical factor in determining the global potential of CDR is the flux of CO_2 removal that can be achieved at a given time. The achievable CDR flux, together with the anthropogenic emissions flux and natural sinks fluxes, determines whether CO_2 concentration can be stabilised, reduced, or will continue rising, at a given time. In the longer term, the total storage capacity for removed CO_2, together with the total cumulative CO_2 emission, will determine how much anthropogenic CO_2 remains in the atmosphere–ocean system, and therefore the long-term concentration of CO_2 and the corresponding warming.[2,8–10] Also important in the long term is whether there is leakage of CO_2 from the storage reservoirs back to the atmosphere, and if so, at what rate.

The various methods available for carbon dioxide removal (see Figure 1) have been summarised in previous work.[11,12] They can be categorised into biological, chemical and physical approaches, or land and ocean based approaches. Here I suggest a new categorization into plant-based, algal-based, and alkalinity-based CDR approaches. The various CDR options include permanent afforestation and reforestation, biomass burial, biochar production, biomass energy with carbon capture and storage (BECCS), either from plant or algal material, ocean fertilization with macronutrients (nitrogen, phosphorus), or micronutrients (iron), enhanced weathering, ocean liming, and direct air capture (DAC). Current assessments suggest that land-based methods of CO_2 removal either *via* biological (photosynthesis) or chemical and physical means, have greater potential than ocean-based methods.[11,12] Furthermore, existing economic assessment suggests that land-based biological CDR has a better cost–benefit ratio than direct air capture of CO_2 using chemical and physical means (although direct air capture would take up far less land space).[13]

Before getting into the specifics of the different CDR pathways, let us note some general overarching constraints for the generation of any CDR flux. All CDR fluxes can be viewed as depending upon: (i) a supply of some limiting resource(s) to capture CO_2; (ii) a yield of carbon per unit input of limiting resource; and (iii) a conversion efficiency of that carbon to long-lived storage, including a supply of resource(s) to achieve that capture. For example, biomass energy with carbon capture and storage (BECCS) using terrestrial plants depends upon a supply of land area, along with nutrients and freshwater, a yield of carbon per unit land area (which will depend on
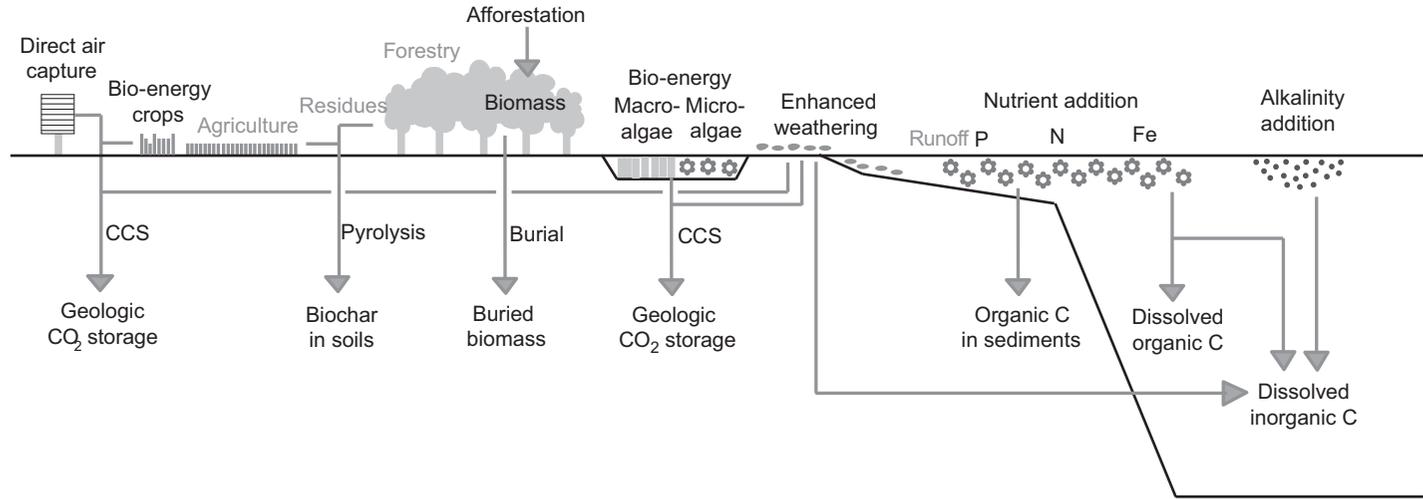
Figure 1 Methods of carbon dioxide removal (CDR).

nutrient and water availability and the chosen biomass crop), and a conversion efficiency to liquid CO_2 stored underground, which takes into account the energy penalty for the conversion and storage process. For each CDR proposal, it may be possible to identify a rate-limiting step in this sequence that limits the CDR flux that can be generated.

The aim in this chapter is to provide an up-to-date review of the global, physical CDR flux potential of leading candidate technologies up to the century timescale. I do not dwell on CDR proposals where existing assessments indicate they have minimal potential,[11] such as enhancing ocean upwelling or down-welling. I assess the flux potential of plant-based CDR, algal-based CDR and alkalinity-based CDR and then the combined total CDR potential, seeking to avoid double counting. Having quantified the physical potential for CDR, I then briefly discuss the implications for climate policy, the caveats, and some directions for future research.

2   Plant-based CDR

"Plant-based CDR" refers to all pathways where CO_2 is removed from the atmosphere by terrestrial plants and some of the resulting biomass flows are converted to stored carbon (see Figure 1). The simplest plant-based CDR pathway is to accumulate carbon in woody biomass through permanent afforestation, perhaps augmenting the sink by harvesting some of the biomass as wood products and thus maintaining the corresponding forestry plantations in a high growth phase.[14,15] Alternative suggestions are to deliberately bury wood in soils,[16] or crop residues in ocean sediments,[17,18] to store carbon. None of these pathways make use of the chemical energy in biomass. Alternatively, if energy is extracted from biomass, some of the associated carbon can in principle be captured and stored as biochar (from pyrolysis of biomass),[19] or as CO_2 (from fermentation, gasification or combustion processes),[20,21] either as liquid CO_2 in geological storage or in seawater when combined with a balanced source of alkalinity. The feedstocks for these bioenergy CDR pathways could include deliberately grown energy crops, forestry wood that is surplus to other uses, and residues (*i.e.* waste products) from agriculture, energy crops and forestry. The different end storage forms for carbon can be derived from the same land and/or the same biomass feedstock, so in estimating global CDR flux potential one must be careful to avoid double counting. Hence I first consider potential supplies of "new" land and the afforestation or bioenergy crops that could be grown on it. Then I consider additional biomass flows from forestry and agriculture.

2.1   Resource Supplies

For plant-based CDR, a key limiting resource supply is land area, especially in a world where demand for food is increasing rapidly and there is a desire to preserve natural ecosystems, including forests. Abandoned agricultural land is a prime target for plant-based CDR as it is generally the most

productive land. It could either be afforested or devoted to biomass energy crops. Historically, since the early 1960s there has been little net change in land area under cultivation despite a doubling of population,[22] but some agricultural land has been abandoned, whilst new land went under cultivation elsewhere. Looking ahead, most scenarios project a continuing supply of abandoned cropland,[23] of 0.6–1.3 Gha in 2050 and about double this in 2100.[24] The corresponding supply rate of abandoned agricultural land fluctuates over 0–17 Mha yr^{-1} across the scenarios.[25] However, if gains in agricultural efficiency of around 1% per year cannot be sustained in the face of growing food demand, global land area under cultivation will have to increase – leaving little high-productivity land for plant-based CDR.[26] Supplies of nutrients and freshwater could also limit plant-based CDR potential – especially when trying to achieve high productivity and yields with dedicated biomass energy crops.[27]

Low-productivity land (including grazed grassland) is projected to dwindle in area, and to have negligible potential for deliberate biomass growth.[24] However, one ambitious proposal suggests that large areas of very low-productivity desert in Australia and the Sahara could be irrigated by desalination of seawater, and forests grown there.[28] In that case, the key limiting resources will be the supply of energy to desalinate seawater and pipe it, and the corresponding nutrients needed to establish forest soils.

2.2 Afforestation and Reforestation

The conversion of unforested land to permanent forest creates a net carbon sink and a store of carbon in the biomass of the trees and in the soil, although there can be transient (and even net) loss of carbon from soil depending on location. The afforestation CDR flux grows both as planted trees approach their peak rates of carbon accumulation and as progressively more land is subject to planting. Once a forest reaches maturity, the sink declines to zero with respiratory carbon losses matching photosynthetic carbon uptake, although recent studies point to a persistent carbon sink in old growth forests.[29] By harvesting carbon in the form of wood products and replanting, forestry plantations can be maintained in a higher average yield state, thus increasing the CDR flux.[15] Yields of carbon for permanent afforestation are of the order 1 MgC ha^{-1} yr^{-1},[15] with average values of 0.8–1.6 MgC ha^{-1} yr^{-1} used in global projections.[14,15]

Large afforestation programs have already been undertaken, with an estimated 264 Mha afforested in 2010.[30] In China alone, the corresponding CDR flux is estimated to have been 0.19 PgC yr^{-1} over 1988–2001.[31] If the 264 Mha of existing plantations are accumulating carbon at an average rate of 0.8–1.6 MgC ha^{-1} yr^{-1},[14,15] then the corresponding CDR is already 0.21–0.42 PgC yr^{-1}. Conceivably this is an under-estimate as yield can be considerably greater in the tropics.

Afforestation area increased at $\sim$5 Mha yr^{-1} over 2005–2010,[30] which simply extrapolating forward could produce an additional 200 Mha afforested

by 2050. On the resulting total area of 464 Mha, the afforestation CDR flux in 2050 could be 0.37–0.74 PgC yr^{-1} (assuming 0.8–1.6 MgC ha^{-1} yr^{-1}). This is in the range of projections for 2050[32] with more detailed models of 0.2–1.5 PgC yr^{-1},[15,25,33,34] which is constrained by the supply of abandoned cropland.

In 2100, afforestation CDR projections from detailed models range over 0.3–3.3 PgC yr^{-1},[15,25] again constrained by the supply of abandoned cropland. Much larger technical potentials have been suggested on shorter time horizons (e.g. ~10 PgC yr^{-1} in 2035)[35,36] but these imply major conflicts with food production and/or the preservation of natural ecosystems and hence are ignored here.[32]

2.3 Bioenergy Crop Supplies

Woody bioenergy crops (e.g. Pinus, Eucalyptus – globally the two main species), have higher achieved yields than afforestation of ~1.5–7 MgC ha^{-1} yr^{-1} (assuming ~0.5 gC g^{-1} average carbon content of wood).[37,38] Even higher global average yield levels are assumed in some projections, ranging over 1.5–15 MgC ha^{-1} yr^{-1} or 8–10.5 MgC ha^{-1} yr^{-1},[37,39] which seem ambitiously high.[14] Other energy crops generally have yields less than or equal to woody crops.

Current bioenergy supplies meet about 6% of global energy demand but only a small fraction of this is in the form of modern biomass energy crops, amenable to large-scale capture and storage technology.

In 2050, it has been estimated that bioenergy crops could supply 40–330 EJ yr^{-1} (exajoules or EJ = 10^{18}),[37,40] which assumes that the plantation area will range over 390–750 Mha, with yields typically 4–7.5 MgC ha^{-1} yr^{-1} and global production of around 2.5–3 PgC yr^{-1} (assuming ~0.5 gC g^{-1}).[37] Alternative estimates are that if 1% yr^{-1} gains in agricultural efficiency can be sustained to 2050, with the expected trend toward higher meat diets, 1.7 PgC yr^{-1} of energy crop could be produced in 2050.[26] If in addition there were a reversion to lower meat diets, bioenergy crop supplies could increase to 3.4 PgC yr^{-1} in 2050.[26] However, this would demand massive nutrient inputs. For each ~1 PgC yr^{-1}, roughly 20% of present global fertiliser nitrogen would be required, based on switchgrass as the crop.[27] Expressed another way this is a ratio of nitrogen input to carbon output of C : N ~58, which with typical grass N : P ~15–20, corresponds to C : P ~1000, and is consistent with around 20% of global fertiliser phosphate inputs (of 0.39–0.45×10^{12} mol P yr^{-1}) also being required for each ~1 PgC yr^{-1}.

By 2100, integrated assessments of bioenergy potential constrained by the supply of suitable land area tend to be roughly double what they are in 2050.[24,37] The greatest potential is on abandoned agricultural land, with one study giving a range of 240–850 EJ yr^{-1} in 2100 for woody energy crops,[24] corresponding to 6–21 PgC yr^{-1} (assuming 20 GJ Mg^{-1} and 0.5 gC g^{-1}). However, the required nitrogen and phosphorus inputs to sustain this would be roughly 100–400% of present fertiliser production, unless efficient nutrient recycling systems can be implemented.

2.4 Additional Biomass Supplies

With large areas of the land surface already managed, wood supplies from forests and waste biomass flows from agriculture and forestry can provide major sources of carbon for conversion to long-lived forms.

Based on year 2000 figures,[41] global wood removals from forests were ~ 1 PgC yr^{-1}, roughly half of which went to wood fuel and half to industrial uses, but it is unclear whether any of the ~ 0.5 PgC yr^{-1} used as wood fuel is available for capture and storage. Global flows of carbon in unused residues from cropland were ~ 0.6 PgC yr^{-1} (assuming 0.4 gC g^{-1}).[41] Alternative estimates of maximum current potential agricultural residues are 0.28 PgC yr^{-1} from rice, 0.18 PgC yr^{-1} from other cereals, 0.13 PgC yr^{-1} from sugar cane waste bagasse and field trash, and 0.19 PgC yr^{-1} from manures, giving 0.78 PgC yr^{-1} in total.[42] In addition, year 2000 felling losses from forestry were ~ 0.33 PgC yr^{-1} (assuming 0.5 gC g^{-1}).[41] Thus combined total residues are in the range 0.9–1.1 PgC yr^{-1} which, with the addition of all wood fuel, gives ~ 1.5 PgC yr^{-1}.

In 2050, forecast bioenergy supply from surplus forest biomass is 60–100 EJ yr^{-1},[40] which corresponds to 1.5–2.5 PgC yr^{-1} (assuming 20 GJ Mg^{-1} and 0.5 gC g^{-1}), and would represent a significant increase over current global wood removals of ~ 1 PgC yr^{-1}.[41] In 2050, the supply of agricultural and forest residues is estimated at 30–180 (mean 100) EJ yr^{-1},[40] which corresponds to 0.8–4.8 (mean 2.7) PgC yr^{-1} (assuming 15 GJ Mg^{-1} and 0.4 gC g^{-1} for residues). The lower end of this range is less than unused crop residues and felling losses at present,[41] but the upper estimate looks unrealistic. Alternative estimates for 2050, including manure and food waste as potential feedstocks, give total biomass "waste" streams of 2.2–2.7 PgC yr^{-1} (across four scenarios) as a more reasonable upper limit.[26]

2.5 Conversion Routes and Efficiencies

Having established the supply of carbon in biomass the next key question is; how much can be converted to long-term storage?

If one leaves biomass in permanent forests and their soils (where previously the land stored less carbon) the conversion efficiency is often treated as 100%, although natural disturbances such as pests and fire that reduce carbon storage cannot be completely prevented.[43] Biomass burial has a conversion efficiency $> 97\%$ based on data for solid wood in landfill sites, where $< 3\%$ of the carbon is converted to CH_4 and CO_2 in a roughly 1 : 1 ratio.[44] Burial of biomass in the deep ocean is also assumed to be near 100% efficient.

BECCS technologies generally lend themselves to relatively uniform feedstock such as dedicated bioenergy crops. There are several pathways for converting biomass carbon to captured CO_2 including: (i) biomass combustion with flue gas CO_2 capture (which can also use mixed feedstock); (ii) biomass gasification then CO_2 capture (with an optional CO shift) before combustion or conversion to fuel; (iii) air separation of pure O_2 for biomass combustion with CO_2 capture; (iv) biomass fermentation to biofuel

(sometimes preceded by saccharification) with CO_2 capture; and (v) biomass conversion to biofuel *via* the Fischer–Tropsch process with CO_2 capture.[21]

CO_2 capture potential varies considerably across these technologies (as do the offsets of fossil fuel burning). Carbon capture yields of 90% (and possibly higher), with a corresponding $\sim$30% energy yield as electricity, are claimed for (ii) a biomass integrated gasification combined cycle (BIGCC) with CCS.[45,46] However, other authors estimate only a 55% carbon yield with 25% energy yield as electricity for the same type of system.[47] Higher energy yields are estimated in the form of hydrogen production (55%) or heat production (80%).[45] Lowest carbon capture yields are for converting sugar cane to ethanol (iv), which leaves 67% of the carbon in the ethanol and releases 33% as CO_2. However, future biofuel is likely to be dominated by lignocellulosic crops, for which processing by saccharification and fermentation (iv) or Fischer–Tropsch (v) leaves around half of the carbon in the fuel, with carbon yields of (iv) $\sim$13% or (v) $\sim$41% as high purity CO_2.[48]

Biochar production lends itself to biomass residues from agriculture and forestry and other mixed feedstocks like food waste and manure, providing a convenient recycling mechanism for organic wastes. Charcoal is typically produced by pyrolysis of biomass although thermo-catalytic depolymerisation has also been demonstrated. The carbon and energy yields of biochar production vary greatly with the temperature of pyrolysis. In systems optimised for biochar yield,[49] up to 63% carbon capture is possible *via* pressurised flash pyrolysis, with an energy yield of around 35% in gas (59% of the energy is left in the char and 6% lost). A more conservative figure is $\sim$50% carbon capture with a similar energy yield.[42] When returned to soil as biochar a significant (but debated) fraction of the carbon in charcoal, *e.g.* 85%, is long-term resistant to biological decay.[42] Thus, the maximum overall conversion efficiency for biochar is probably around 50%.

2.6 Combined CDR Potential

The combined potential for plant-CDR (see Table 1) depends on the choice of land use and the choice of conversion pathways for biomass supplies. To maximise CDR potential, abandoned agricultural land should be devoted to dedicated bioenergy crops rather than afforestation. Then burial of all biomass could probably maximise CDR, but runs the risk of damaging ecosystems by withdrawing carbon and nutrients and, with no energy benefit, is unlikely to be favoured. Instead, uniform feedstock including dedicated bioenergy crops and surplus wood from forestry could be directed to BIGCC with CCS as it offers the potential to fix up to 90% of the carbon from the feedstock, whilst also yielding energy. For heterogeneous feedstock, including various biomass "waste" streams, pyrolysis and return of biochar to the soil has the potential to sequester around 50% of the carbon content of the feedstock, whilst also returning some associated nutrients to the soil.

The maximum combined plant-CDR potential at present is around 0.75–1.5 PgC yr^{-1} (see Table 1). Current afforestation CDR is conservatively 0.21–0.42 PgC yr^{-1}. The present potential for bioenergy CDR from

Table 1 Estimates of global plant-CDR flux potential by pathway for present, 2050 and 2100.

Carbon source	Carbon store	CDR flux $(PgC\ yr^{-1})$	Key conditions / assumptions	Reference
Present				
Afforestation	Biomass	0.21–0.42	264 Mha×0.8–1.6 MgC ha^{-1} yr^{-1}	30,32
Energy crops	BECCS	0.19–0.23	Sugarcane, ethanol and pulp mills	21
	Biochar	0.18	All biomass energy by pyrolysis	19
Forestry residues	Burial	0.33	0.65 Pg yr^{-1} of felling losses	32,41
	Biochar	0.16	All felling losses from forestry	32,41
Crop residues	Burial	0.18–0.6	1.5–5 Pg yr^{-1}, 30% removed	18,32,41
	Biochar	0.18	50% of unused crop residues	50
All residues	Biochar	0.16–0.34	original-revised estimates	19,32,41
Shifting cultivation	Biochar	0.21–0.35	All shifting cultivation fires	19,32,51
Charcoal	Biochar	0.01	All waste from charcoal making	19
All	Various	**0.75–1.5**	Total	
2050				
Afforestation	Biomass	0.2–1.5	All abandoned cropland	15,25,33,34
	Biomass	0.2–0.4	No expansion of afforested area	30,32
Energy crops	BECCS	1.25–1.5	390–750 Mha, 8–15 Mg ha^{-1} yr^{-1}	32,37
	BECCS	1.5–3.0	1.7–3.4 PgC yr^{-1} across 4 scenarios	26
Surplus wood	Biochar	0.75–1.25	60–100 EJ yr^{-1}, 20 GJ Mg^{-1}, 0.5 gC g^{-1}	32,40
All residues	Biochar	1.35	∼100 EJ yr^{-1}, 15 GJ Mg^{-1}, 0.4 gC g^{-1}	32,40
	Biochar	1.1–1.35	2.2–2.7 PgC yr^{-1} across 4 scenarios	26
All	Various	**2.3–5.75**	Total (avoiding double counting)	
2100				
Afforestation	Biomass	0.3–3.3	SRES A2, B1, B2, A1b range	15,25
Energy crops	BECCS	5.4–19	240–850 EJ yr^{-1}, 20 GJ Mg^{-1}, 0.5 gC g^{-1}	24,32
	Biochar	5.5–9.5	180–310 EJ yr^{-1} all pyrolysed	19
Surplus wood	Biochar	0.75–1.25	2050 estimates (as above)	32,40
All residues	Biochar	1.1–1.35	2050 estimates (as above)	26
All	Various	**∼5–20**	Total (avoiding double counting)	

sugarcane-based ethanol production and chemical pulp mills has been estimated at 0.19–0.23 PgC yr^{-1}.[21] Alternatively, if all "modern" biomass energy were converted to biochar by pyrolysis, the estimated CDR flux is 0.18 PgC yr^{-1}.[19] In addition, if global flows of crop residues from agriculture were all converted to biochar,[41] 0.3 PgC yr^{-1} could be removed,[50] or if 30% of these residues were buried in the deep ocean, 0.18 PgC yr^{-1} could be removed. In addition, the ∼0.33 PgC yr^{-1} of felling losses from forestry,[41] could produce a CDR flux of 0.32 PgC yr^{-1} if all buried in soils

or 0.16 PgC yr^{-1} as biochar. For the upper estimate, an additional 0.21–0.35 PgC yr^{-1} as biochar is included assuming "slash-and-char" shifting cultivation was adopted in place of current slash-and-burn practices.[19,32,51] Finally, there is 0.01 PgC yr^{-1} from wastes of charcoal production.[19] However, it is assumed that none of the wood currently being used as fuel is available for alternative conversion pathways (*e.g.* pyrolysis to biochar).

The maximum combined plant-CDR potential in 2050 could range over 2.3–5.75 PgC yr^{-1} (see Table 1). The lower estimate comes from a scenario in which gains in agricultural efficiency cannot be sustained and agricultural land has to expand in area, with current bioenergy crop land having to be reclaimed for food production. In that case, residues are maximised and their conversion to biochar can produce 1.35 PgC yr^{-1}. Present afforested area (with no further expansion) creates a minimum additional CDR flux of $\sim$0.2 PgC yr^{-1}, and wood removal from all managed forests could produce an additional 0.75 PgC yr^{-1} if converted to biochar. The upper estimate comes from a world in which agricultural efficiency continues to increase, the global trend towards eating more meat is reversed, and all abandoned agricultural land is devoted to bioenergy crops, which support a CDR flux of 3.0 PgC yr^{-1}. Wood removal is maximised giving 1.25 PgC yr^{-1} as biochar. Residues are somewhat less (2.2 PgC yr^{-1}) due to increases in agricultural efficiency, but produce a biochar CDR flux of 1.1 PgC yr^{-1}. Even though afforestation has not expanded, the current area supports an additional CDR flux $\sim$0.4 PgC yr^{-1}. Integrated assessment modelling of realised CDR potential in 2050 gives either 0–2.7 PgC yr^{-1} by BECCS or 0–1.1 PgC yr^{-1} by afforestation,[52] which, as would be expected, is less than the maximum potential estimates here. Furthermore, the freshwater and nutrient demands of removing several PgC yr^{-1} by BECCS and/ or afforestation may be "unrealistically high".[27]

The maximum combined plant-CDR potential in 2100 is particularly uncertain but could range over *circa* 5–20 PgCyr^{-1} (see Table 1). The lower limit comes from a maximum afforestation scenario (3.3 PgC yr^{-1}) with the addition of 2050 values for wood removal (0.75 PgC yr^{-1}) and forest and crop residue (1.1 PgC yr^{-1}) conversion to biochar. If, instead, one takes a lower limit for woody bioenergy crop production and converts it by BECCS (5.4 PgC yr^{-1}), plus the wood removal and residue biochar production fluxes, the total CDR flux is 7.25 PgC yr^{-1}. For an upper limit, the maximum woody bioenergy crop production with conversion by BECCS could yield up to $\sim$19 PgC yr^{-1} to which can be added wood removal and residues generated fluxes. The overall range spans earlier estimates of 5.5–9.5 PgC yr^{-1} potential biochar CDR flux (from 180–310 EJ yr^{-1} biomass energy supply in 2100).[19,37] Of course the realised potential would be expected to be lower, with integrated assessment modelling giving a BECCS potential of 0–5.4 PgC yr^{-1} in 2100.[52]

3   Algal-based CDR

"Algal-based CDR" refers here to all pathways in which CO_2 is removed from the atmosphere by algae (including cyanobacteria and macro-algae) and

some of the resulting biomass flows are converted to stored carbon (see
Figure 1). This categorisation groups ocean fertilization methods and algal
bioenergy production with carbon capture and storage. The latter could be
conducted in freshwater or salt water.

3.1   Resource Supplies

For algal-based CDR the input of nutrient(s) (and their subsequent recycling)
is the key limiting resource, especially for cyanobacteria and micro-algae
which have relatively low carbon-to-nutrient ratios compared with terrestrial
plants. If algal-based CDR is attempted on land, the supply of suitable area
for water containers also needs to be considered, whereas for ocean fertil-
ization the supply of ships is important.

3.2   Algal BECCS

Micro-algae are an attractive source of bioenergy because their photo-
synthetic efficiency (10–20%) far exceeds that of land plants (1–2%) and they
can achieve very high growth rates, doubling their biomass within a few
hours under ideal conditions. Also many taxa grow in seawater, eliminating
the need for freshwater inputs. Countering this is the problem that the
typical carbon-to-nutrient ratio of micro-algae is much lower than that of
land plants – especially woody plants – therefore the total nutrient demand
to fix a given flux of carbon is much higher. Macro-algae have the advantage
of higher carbon-to-nutrient stoichiometry than micro-algae and also have
high productivity. Micro-algae can be cultivated in flow-through open
"raceway" ponds or in closed "photobioreactors", whereas macro-algae can
be cultivated in tidal-flat farms, nearshore farms, rope system farms, open-
ocean farms or as floating seaweed.[53] However, it can take up to 20% of the
energy captured to harvest algal biomass (greater than for terrestrial plants).
To maximise photosynthetic carbon fixation in seawater, CO_2 needs to be
continually resupplied by replenishing algae with CO_2-rich water, especially
when grown in closed systems. Hence linking micro-algal biofuel production
to high CO_2 concentration flue gases from power stations has been con-
sidered – but this only amounts to a CDR technology if the CO_2 is derived
from biomass combustion, in which case algae can just be viewed as part of
the capture technology. Instead, true algal-BECCS should be based on algae
in water exchanging CO_2 with the free air. The subsequent conversion of
biomass can involve biofuel production with CCS, anaerobic digesters pro-
ducing methane (as well as CO_2) with subsequent combustion and CCS, and/
or biomass gasification (BIGCC) with CCS.

Micro-algae in raceway ponds can capture up to 36.5 MgC ha^{-1} yr^{-1}
(10 gC m^{-2} d^{-1}), which is well above woody biomass energy crops. With the
harvesting penalty (80% efficiency of energy conversion) and maximum cap-
ture efficiency (90% for BIGCC with CCS) this could translate to a CDR flux
~25 MgC ha^{-1} yr^{-1}. Thus with an area of 0.1 Gha (10^6 km^2) one might

imagine capturing 2.5 PgC yr^{-1}, but the corresponding nutrient demand would be huge; with classic "Redfield" ratio (C : P = 106 : 1) and no recycling, it would require ~2.9×10^{12} mol P yr^{-1}, which is nearly an order of magnitude more than the global fertiliser inputs to the land of 0.39–0.45×10^{12} mol P yr^{-1}.

To get a more reasonable upper limit on algal-BECCS CDR potential, consider the global flux of phosphorus in treated or untreated sewage of 0.048×10^{12} mol P yr^{-1} (1.5×10^{12} gP yr^{-1}). If we imagine that this could all be linked through wastewater processing plants to fuel micro-algal productivity, with the above conversion efficiencies and no nutrient recycling, the resulting CDR flux would be 0.044 PgC yr^{-1} on a relatively modest area of 1.7 Mha. Experience with anaerobic digestion of algal biomass however suggests a nutrient recycling efficiency of 60–80% can be achieved (for nitrogen) at an energy conversion efficiency of 75%,[54] in which case the CDR flux might be increased to 0.1–0.2 PgC yr^{-1}. Still the CDR potential remains modest because of the low C : P stoichiometry of micro-algae and the assumed constraint on nutrient input. The genetic-engineering of micro-algae that excrete carbon rich (and nutrient poor) compounds such as long-chain hydrocarbon fuels may improve the forecast, but such liquid fuels are likely to be used in the transport sector where it is difficult to link them to carbon capture and storage technology.

Macro-algae cultivation has achieved productivity of 10–34 MgC ha^{-1} yr^{-1}, with much higher C : N : P stoichiometry than micro-algae.[53] Taking the "Atkinson" ratio of C : P = 550 : 1 and the global sewage flux of phosphorus,[55] with the same energy conversion and carbon capture efficiencies, a CDR flux of 0.23 PgC yr^{-1} could be generated without any nutrient recycling, although this would require a larger area than micro-algae of ~9–32 Mha (depending on productivity). Existing anaerobic digestion systems have shown a nutrient recycling efficiency of 60–80% can be achieved with 75% efficient energy conversion,[54,56] thus the CDR flux could conceivably be increased to 0.53–1.07 PgC yr^{-1} (on the same area). Such algaculture systems would likely be operated in estuarine (total global area ~100 Mha) or coastal shelf sea settings, so the required area could pose a constraint on scaling up of this technology.

Looking ahead, if the human sewage flux of phosphorus scales with total projected waste phosphorus fluxes we can expect a ~50% increase in 2050 and a ~100% increase in 2100. Thus the corresponding algal BECCS CDR flux could be ~0.15–0.3 PgC yr^{-1} for the micro-algal route and ~0.75–1.5 PgC yr^{-1} for the macro-algal route in 2050, and ~0.2–0.4 PgC yr^{-1} or ~1–2 PgC yr^{-1} in 2100. Of course if one assumed the same nutrient inputs as woody biomass energy crops grown on land then these CDR fluxes could be considerably increased.

3.3   Ocean Fertilisation

The phenomenal area of the ocean offers obvious algal-CDR potential, if carbon in biomass can be transferred to long-lived reservoirs, but once again

the required nutrient inputs will be massive, and without a bioenergy gain the economics are more prohibitive. Sequestration of carbon can occur in two main reservoirs: marine sediments or the deep ocean itself. Ocean fertilisation proposals focus on adding new nutrient to the surface ocean (or increasing nutrient supply from depth), as this ultimately controls the sequestration flux of carbon. Candidate fertilisers are the macro-nutrients nitrogen (N) and phosphorus (P), and the micro-nutrient iron (Fe), noting that relieving a deficit of one nutrient is likely to lead to limitation by another. In addition to N, P and Fe limitation, silicate (Si) limitation is a possibility, especially for diatoms.

In the surface ocean, carbon fixed by photosynthesising organisms comes from dissolved inorganic carbon (DIC) in the water, creating a deficit of DIC that in turn drives an air–sea flux of CO_2. Much of the resulting organic carbon is recycled back to DIC by heterotrophic organisms in the upper ocean, nullifying the effect on atmospheric CO_2. However, a modest fraction of the carbon fixed by marine phytoplankton escapes respiration and sinks to greater depth. In coastal and shelf seas it soon hits the bottom sediments, whereas in the open ocean it can keep sinking for kilometres. However, as it does so, the sinking flux of carbon decays away due to remineralisation, following a power law function with depth,[57] with the power known to vary significantly with surface community structure.[58] The ''export flux'' refers to the flux of carbon sinking out of the sunlit photic zone. This has been estimated at 17 PgC yr^{-1} globally at 75 m,[59] but the bottom of the photic zone is more typically taken to be 100 m where 11 PgC yr^{-1} has been estimated.[60,61] What is critical for long-term carbon storage in the deep ocean is the ''sequestration flux'' below the depth of winter wind-driven mixing, which ranges over 200–1000 m, depending on location.[62] Any organic carbon remineralised to DIC above the depth of winter mixing will not create a DIC deficit on the annual or longer timescale and hence will not drive a CO_2 sink. Taking 500 m as a reference depth, there the global sequestration flux is estimated to be in the range 2.3–5.5 PgC yr^{-1} (*i.e.* 20–50% of the export flux at 100 m),[58] with a long-favoured formula giving 2.8 PgC yr^{-1} (*i.e.* 25% of the export flux).[57]

Some algal-CDR is already (inadvertently) occurring in the ocean, because humans mine and add phosphorus to the land surface 0.39–0.45×10^{12} mol P yr^{-1} and much of this leaks to the ocean. The riverine flux of biologically-available (dissolved and particulate) phosphorus to coastal seas (including sewage and detergent) has increased by 0.31×10^{12} mol P yr^{-1}, suggesting a 70–80% transfer efficiency.[63] Some of this P input is converted to organic carbon in coastal and shelf seas and buried in their sediments with a typical burial ratio under oxic (where oxygen is present) bottom waters of C : P ~ 250.[64] In the North Sea this sediment burial flux of P is around 20% of the estimated anthropogenic P input.[65] Extrapolating these figures to global coastal regions gives an estimated 0.18 PgC yr^{-1} being sequestered in their sediments at present. If the remaining 80% of anthropogenic P inputs are transferred to the open ocean and go to fuel export production there, assuming (as an upper limit) half of this flux contributes

to long-term sequestration below 500 m, then a further sink of 0.16 PgC yr^{-1} is being generated at present, giving a total of 0.34 PgC yr^{-1}. Alternatively, if all anthropogenic P loading ends up in the open ocean (*i.e.* none is buried in coastal and shelf sea sediments) then the estimated sequestration flux is 0.20 PgC yr^{-1} at present.

It has been suggested that mined phosphate could be directly added to the surface ocean,[62] despite this finite resource being essential for food production. More likely we can expect inadvertent phosphorus addition to the ocean to increase in future. One projection gives a linear increase to 0.42×10^{12} mol P yr^{-1} in 2035,[63] which could drive a sink of 0.27–0.47 PgC yr^{-1}. Extrapolating the linear trend forwards gives 0.47×10^{12} mol P yr^{-1} added to the ocean in 2050 driving 0.30–0.53 PgC yr^{-1}, and 0.64×10^{12} mol P yr^{-1} added to the ocean in 2100 driving 0.41–0.71 PgC yr^{-1}. This assumes that nitrogen fixation will cause nitrogen availability in the ocean to track increased phosphorus availability,[64,66] and that micro-nutrients (*e.g.* iron) do not limit new production in the (currently coastal) regions to which phosphate is added. On the millennial timescale, the total reservoir of mineable phosphate of 323–645×10^{12} mol P could readily be drained, which of course would precipitate a food crisis. Instead societies are going to be compelled to develop efficient recycling systems for phosphorus on land, which in turn will reduce algal-CDR potential in the ocean.

Nitrogen fertilisation of the ocean is a slightly more sustainable proposition in that nitrogen can be fixed from the atmosphere, but it comes at great energy expense. Potentially, the deficit of available nitrogen relative to phosphorus in the world ocean could be alleviated. This deficit averages 2.7 μmol kg^{-1},[67] compared to an average deep ocean nitrate concentration of 30.9 μmol kg^{-1}. Thus, removing the nitrogen deficit would result in a ∼9% (2.7/30.9) increase in the export flux, corresponding to about 1 PgC yr^{-1} at 100 m depth. However, it is the sequestration flux below ∼500 m depth which is critical and an upper limit for the increase in that is 0.5 PgC yr^{-1}. Assuming this could be achieved in 2100 on a linear growth trajectory, the 2050 CDR flux might be ∼0.2 PgC yr^{-1}.

Iron is the favoured nutrient for ocean fertilisation because often the C : Fe >10000 in algal biomass, so the required nutrient inputs are much less massive than for N or P. However, iron is only limiting in remote high-nutrient low-chlorophyll (HNLC) areas of the world ocean,[68–70] notably the Southern Ocean. A number of model studies have assessed the potential carbon sink that could be generated by iron fertilisation. The maximum potential is indicated by simulations that remove iron limitation globally for 100 years.[68] Global export production across 100 m is increased initially by 3.5 PgC yr^{-1}, decaying after 100 years to 1.8 PgC yr^{-1}, and totalling 226 PgC. Diatoms are predicted to make a greater contribution to export production, creating fast sinking particles that should maximise the sequestration flux. Taking the predicted time mean increase in export flux across 100 m of 2.26 PgC yr^{-1}, and likening the remineralisation with depth to that at a station where diatoms dominate (K2 in the Northwest Pacific),[58] then the estimated increase in sequestration flux across 500 m is 1 PgC yr^{-1}. Assuming a linear

growth trajectory, ~ 0.4 PgC yr^{-1} might be achieved in 2050 and 1 PgC yr^{-1} in 2100.

3.4 Combined CDR Potential

The combined potential of algal-based CDR (see Table 2) is clearly set by nutrient supply fluxes, and in the case of phosphorus and nitrogen these

Table 2 Estimates of global algal-CDR flux potential by pathway for present, 2050 and 2100.

Carbon source	Carbon store	CDR flux (PgC yr^{-1})	Key conditions / assumptions	Reference
Present				
Micro-algae	BECCS	0.044	0.048×10^{12} mol P yr^{-1} sewage	This study
	BECCS	0.1–0.2	60–80% efficient P recycling	This study
Macro-algae	BECCS	0.23	0.048×10^{12} mol P yr^{-1} sewage	This study
	BECCS	0.53–1.07	60–80% efficient P recycling	This study
River excess P flux	Sediment	0.18	$C/P = 250$, 0.31×10^{12} mol P yr^{-1}	32
	DIC	0.16	80% of excess P to open ocean	32
All	Various	**0.9–1.4**	Total potential	
2050				
Micro-algae	BECCS	0.15–0.3	50% increase in sewage P, recycling	This study
Macro-algae	BECCS	0.75–1.5	50% increase in sewage P, recycling	This study
River excess P flux	Sed.+DIC	0.30–0.53	0.47×10^{12} mol P yr^{-1}	32
N fertilisation	DIC	0.2	Remove 40% of global N deficit	32
Fe fertilisation	DIC	0.4	Remove 40% of global Fe deficit	32,68
All	Various	**1.6–2.6**	Total potential	
2100				
Micro-algae	BECCS	0.2–0.4	100% increase sewage P, recycling	This study
Macro-algae	BECCS	1.0–2.0	100% increase sewage P, recycling	This study
River excess P flux	Sed.+DIC	0.41–0.71	0.64×10^{12} mol P yr^{-1}	32
N fertilisation	DIC	0.5	Remove 100% global N deficit	32
Fe fertilisation	DIC	1	Remove 100% of global Fe deficit	32,68
All	Various	**2.9–4.2**	Total potential	

are the same nutrients that we need to support intensive agriculture – a function that is obviously of greater direct value to us. Thus, the potential for N and P based algal-CDR is likely to be built on the waste products of intensive agricultural systems – and will be determined by future agricultural trends. The combined, current potential of wastewater P based algal BECCS and inadvertent ocean P fertilisation appears to be significant at 0.9–1.4 PgC yr^{-1}. Conceivably this could rise to 1–2 PgC yr^{-1} in 2050, and 1.4–2.7 PgC yr^{-1} in 2100. To this might be added ocean nitrogen and iron fertilisation, to give a total algal CDR flux potential of 1.6–2.6 PgC yr^{-1} in 2050, and 2.9–4.2 PgC yr^{-1} in 2100. The present total potential is broadly comparable with plant-based CDR, and although the future potential appears smaller, this is because the woody biomass energy crop estimates implicitly include much larger nutrient inputs than allowed here for macroalgal production.

4   Alkalinity-based CDR

"Alkalinity-based CDR" refers here to all pathways in which CO_2 is removed from the atmosphere using a source of alkalinity (see Figure 1) and stored either as charge-balanced solution in seawater, or as liquid CO_2 in geologic formations (with regeneration of the alkali). It thus groups together methods of engineered direct air capture (DAC), enhanced weathering on land, and addition of alkalinity to the oceans. For methods of enhancing weathering and direct addition of alkalinity to the ocean, the key limiting resource is the supply flux of alkaline material, *i.e.* basic minerals, and the associated energy required to mine, crush and transport it. The CDR flux in turn may be limited by the kinetics of reaction of the alkali source with carbonic acid. For direct air capture (DAC) the key limiting resources are the energy required to capture CO_2 and the supply of engineered DAC devices, because the chemical sorbent system itself can be regenerated.

4.1   *Enhanced Weathering – Land*

Enhanced weathering refers to a suite of CDR options whereby a new source of alkalinity, from the dissolution of either carbonate or silicate minerals, is added to the land surface or to the oceans. Both silicate and carbonate weathering act to transfer excess CO_2 from the atmosphere to the ocean, and silicate weathering (followed by carbonate deposition) also ultimately acts to remove CO_2 to the Earth's crust. The central issue with all of these methods is that a greater mass of rock is needed than the mass of carbon removed from the atmosphere, thus the scale of the extractive industry needed to produce a useful CDR flux has to be comparable to the fossil fuel industry causing the problem in the first place.

Original suggestions for enhancing silicate weathering on land involve adding crushed olivine to soils,[71,72] particularly in the moist tropics where

there are olivine sources and the reaction kinetics will be most favourable.[72,73] However, even saturating the Amazon and Congo basins with 2.2 Pg olivine yr^{-1} would only produce a CDR flux of 0.6 PgC yr^{-1}, and the efficiency penalty of crushing and milling rock is estimated at 17% making the net flux $\sim$0.5 PgC yr^{-1}.[74] This requires loadings everywhere of $\sim$3 kg m^{-2} yr^{-1} which may be unachievable in remote tropical locations, thus the stated upper limit potential of $\sim$1 PgC yr^{-1} should be treated with caution. Also, some studies are internally inconsistent in that they show the impact on atmospheric CO_2 of larger olivine weathering CDR fluxes than they calculate to be feasible.[73,75]

4.2 Enhanced Weathering – Ocean

The weathering process can be short-circuited by adding alkalinity directly to the ocean. Proposals include adding crushed olivine directly to coastal regions[74] or to the open ocean.[76] The problem in both cases is that olivine dissolution proceeds slower under the relatively high pH of seawater and therefore smaller particle sizes are required to achieve a given kinetic dissolution rate. The idea behind coastal addition of olivine is to use the energy of waves to break down particles to smaller sizes, thus increasing the surface area for dissolution. However, the available barge fleet probably limits the CDR flux potential to an estimated 0.09–0.16 PgC yr^{-1}, which with an energy penalty of crushing and milling of $\sim$17% would be reduced to 0.08–0.14 PgC yr^{-1}.[74] Open ocean olivine addition requires even smaller particles of $\sim$1 μm or they will sink before dissolution is complete, increasing the energy penalty to $\sim$30%.[76] With a dedicated fleet of ships or addition to the ballast water of commercial ships, the resulting CDR potential[76] is 0.18–0.20 PgC yr^{-1}.

An alternative proposal – sometimes termed ''ocean liming'' – is to increase the alkalinity of the ocean by adding either calcium bicarbonate[77] or calcium hydroxide,[78] thus increasing CO_2 uptake. A detailed account has been presented based on using a flotilla of ships to sprinkle finely ground limestone ($CaCO_3$) on areas of the surface ocean where the depth of the saturation horizon is shallow (250–500 m) and the upwelling velocity is large (30–300 m yr^{-1}).[79] A CDR flux of 0.27 PgC yr^{-1} has been calculated after a century of linearly ramping up activity.

4.3 Direct Air Capture (DAC)

Direct air capture (DAC) refers to chemical (and physical) methods of removing CO_2 directly from the atmosphere, followed by regeneration and CO_2 storage. There are two main methods of direct air capture being developed – using a solid sorbent system (*e.g.* solid amines) or an alkaline aqueous solution (*e.g.* sodium hydroxide). The capture step is generally more expensive with solid sorbents, whereas the regeneration step is more

expensive with alkaline aqueous solutions. Storage of CO_2 could be in liquid form in geological reservoirs, where ultimately the amount of CO_2 sequestered may be limited by the size of these reservoirs.[80,81] Alternatively, DAC could be combined with enhanced weathering to produce a charge neutral solution to add to the ocean, but with the additional energy cost of mining, crushing and milling the required alkaline rock material.

In principle, DAC could generate whatever size of carbon sink societies were willing to pay for, as it is unlikely to be limited by available substrates or land surface area.[13] However, the rate of development and production of DAC devices could place a serious constraint on the CDR potential in the short-medium term. One study suggests $\sim$3 PgC yr^{-1} might be achievable on a 2030–2050 timeframe.[82] However, the crucial constraint on DAC is probably its high cost, which will mean other cheaper methods of CDR will be deployed before it. When considered in integrated assessment models, DAC is generally not be deployed until later this century, with one model predicting a rise from 0 PgC yr^{-1} DAC in 2065 to $\sim$10 PgC yr^{-1} DAC by 2100.[83]

4.4 Combined CDR Potential

There is a shortage of studies from which to attempt alkalinity-based CDR flux estimates on different time horizons, so just their ultimate potential (assumed realisable by 2100) is considered here (see Table 3). A combination of land and ocean enhanced weathering methods might achieve 1–1.6 PgC yr^{-1}, which is comparable to the current potential of either plant-based or algal-based CDR, but much less than their ultimate potential. Direct air capture, in contrast, is the wildcard CDR technology in that its physical potential could be >10 PgC yr^{-1} and the real constraints on its use will be economic, social and technical.

Table 3 Estimates of global alkalinity-CDR flux potential by pathway.

Process	Carbon store	CDR flux (PgC yr^{-1})	Key conditions / assumptions	Reference
Enhanced weathering				
Terrestrial olivine	DIC	0.5–1	Amazon+Congo, global	73
Coastal olivine	DIC	0.08–0.14	Constrained by barge fleet	74
Open ocean olivine	DIC	0.18–0.20	Dedicated fleet or commercial ships	76
Ocean liming	DIC	0.27	Dedicated ship fleet	79
All	DIC	**1–1.6**	Total potential	

5   Overall CDR Flux Potential

In estimating the overall physical flux potential for CDR, care must be taken to avoid double counting, when the same resource (*e.g.* land or nutrient) is required by different technologies. Competition for land (or biomass) among plant-based CDR methods has been considered above. I have also tried to avoid double-counting phosphorus supplies by only allowing algal-based CDR to access waste flows of phosphorus, either in the form of sewage through wastewater treatment, or washing off the land directly into the ocean. However, a broader competition for phosphorus between food production and plant- or algal-based CDR also ought to be considered, especially in a world where the price of rock phosphate has increased roughly 4-fold over 2006–2013 (with a peak increase of more than 8-fold in 2008). Nitrogen supplies are usually assumed to be unconstrained, because nitrogen can be fixed from the atmosphere, but there is a high energetic cost to doing that. Competition for freshwater supplies between food production and plant-based CDR also deserve more consideration.

Overall achievable CDR fluxes might also conceivably be constrained, on the century timescale, by the storage capacity for carbon in different forms. Various methods suggest that permanent forest plantations could store up to 150 PgC by 2100,[84,85] and up to $\sim$300 PgC in the longer term,[32] thus more than reversing historical cumulative carbon emissions from deforestation of $\sim$150 PgC.[86] Global storage capacity for biochar in cropland, grassland and abandoned land soils is estimated at $\sim$500 PgC (representing a $\sim$25% increase in the carbon content of the world's soils),[32] which would be difficult to saturate this century. Estimates of geologic storage capacity for liquid CO_2 range upwards from $\sim$500 PgC to $\sim$3000 PgC.[87] However, if global geologic storage is at the low end of this range and both CDR methods and conventional CCS on fossil fuel combustion are competing for this capacity, it could be filled up within this century. This would then limit BECCS and DAC fluxes ending in liquid CO_2 geological storage (see Figure 1). However, there is the alternative option to marry BECCS or DAC with enhanced weathering (at high CO_2 concentration of flue gases) and thus switch the form of carbon storage to charge neutral solution in seawater. In that case, the carbon storage capacity depends on the total mineable reserves of alkaline minerals such as olivine, limestone, and chalk, which are vast. The ocean is already the largest reservoir of carbon in the surface Earth system at $\sim$38000 PgC and as long as added DIC is balanced by added alkalinity, the ocean carbon store can continue to grow.

The situation is more subtle when nutrient fertilising the ocean and increasing the efficiency of the "biological carbon pump" – which transfers carbon from the equilibrated surface ocean and atmosphere to the deep ocean or the sediments as a mixture of particulate organic carbon, dissolved organic carbon, and DIC – but essentially this can increase the ocean and sediment carbon store by an amount that will be determined by the total

Table 4 Total physical CDR flux potential (PgC yr^{-1}) on different time horizons.

	'Present'	2050	2100
Plant-based CDR	0.75–1.5	2.3–5.75	5–20
Algal-based CDR	0.9–1.4	1.6–2.6	2.9–4.2
Alkalinity-based CDR	0	0.5–0.8 (+DAC?)	1–1.6 (+DAC)
Total CDR	1.65–2.9	4–9 (+DAC?)	9–26 (+DAC)

reservoirs of nutrient that can be added and the stoichiometry of the organic material produced. For phosphorus, the total mineable reservoir of phosphate of 323–645×10^{12} mol P, if 70–80% ends up in the ocean, could sequester ~ 600 PgC.

To summarise, overall CDR is unlikely to be limited by storage constraints on the century timescale, but particular methods of CDR that end in geologic storage of CO_2 might be constrained.

Accepting these caveats, a summation of the estimated plant-based, algal-based and alkalinity-based CDR contributions (see Table 4) produces some indicative upper limit figures for the total physical flux potential for CDR. The total "present" potential of ~ 1.5–3 PgC yr^{-1} is based on known afforestation, known (mostly waste) flows of biomass and know "waste" flows of phosphorus. Part of this present CDR flux potential is already being realised; afforestation plus inadvertent phosphorus fertilisation of the ocean are generating CDR of 0.55–0.76 PgC yr^{-1}. The 2050 total CDR potential of ~ 4–9 PgC yr^{-1} is roughly three times the present potential. It includes (in the upper limit) a contribution from enhanced weathering of roughly half its ultimate potential. However, it does not include direct air capture, which is treated a potential additional flux in 2050 and in 2100. The 2100 total CDR potential of ~ 9–26 PgC yr^{-1} carries the largest uncertainties, but is again roughly three times the 2050 potential.

6   Discussion

The literature review and "ball-park figure" estimates herein suggest there is considerable physical potential for carbon dioxide removal at a global scale. The already realised CDR flux (albeit somewhat inadvertent) of 0.55–0.76 PgC yr^{-1} is offsetting at least half of land-use change CO_2 emissions of 1.1 PgC yr^{-1}. The current physical CDR potential of ~ 1.5–3 PgC yr^{-1} is of a comparable size to the natural land or ocean carbon sinks and thus if realised could make a very valuable contribution to slowing the rate of rise of atmospheric CO_2.

The 2050 CDR potential of ~ 4–9 PgC yr^{-1}, if combined with substantive efforts to mitigate CO_2 emissions below present levels of ~ 10 PgC yr^{-1}, could together stabilise atmospheric CO_2 concentration. Although it is tempting to argue that stabilisation would happen even sooner because of

the existence of natural CO_2 sinks, in fact just as CO_2 emission create these natural sinks, CO_2 removal will create a counterbalancing natural CO_2 source. Hence one must consider the net flux of CO_2 to or from the atmosphere, and when that net flux is zero, natural net CO_2 exchange fluxes will also tend towards zero.

By the end of the century, CDR potential of $\sim$9–26 PgC yr^{-1} (without including direct air capture) could offer substantial leverage on atmospheric CO_2 concentration. However, despite the large physical potential of CDR, it would not make economic sense to imagine that CDR could be ramped up to counterbalance growing CO_2 emissions, which following "business-as-usual" type scenarios could reach $\sim$30 PgC yr^{-1} in 2100. Instead economics would dictate that cheaper emissions reductions options would be taken before deploying generally more expensive CDR.

Indeed the actual CDR flux that can be realised for any method is always going to be less than the physical potential thanks to social, economic and engineering constraints. Integrated assessment models provide a framework for considering these constraints, especially cost. From the estimates herein, those models seem broadly right to bank on considerable future CDR potential. However, there are considerable uncertainties around the future estimates of CDR potential.

Starting with land plant-based CDR, the largest CDR potential is in BECCS pathways. However, land for woody biomass energy crops may simply not be available before mid-century. At the same time some integrated assessment models suggest that BECCS will only become affordable mid-century, producing a fortuitous correspondence in time. However, nutrient and freshwater demands for global scale biomass energy cropping are large and might end up being the true constraining factor.

Considering area, water and nutrient demands leads to the tentative conclusion that macro-algae based BECCS could be a better long-term option than woody biomass energy crops, because achievable productivity is higher, freshwater demands are eliminated, and nutrient demands may be lower thanks to more efficient recycling systems. Macro-algal-based CDR thus warrants further research.

Direct Air Capture (DAC) escapes from most resource constraints except for the need for energy. Hence it has great physical potential but is estimated to be very costly. This means it is not expected to be deployed at scale until CCS is implemented on all concentrated point sources of CO_2 and probably not until BECCS of some form has been deployed at scale for decades.

All of the CDR technologies discussed (with the exception of afforestation and inadvertent nutrient fertilisation) are to varying degrees unproven, especially in terms of the engineering and infrastructure required to scale them up. Such scaling up, if feasible, is bound to produce a social reaction that could readily prevent it happening. Thus a more comprehensive assessment should include the full range of social, economic and engineering constraints on different CDR options, alongside the scientific constraints considered here.

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