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Environmental sustainability of negative emissions technologies: A review

Harish Kumar Jeswani*, Djasmine Mastisya Saharudin and Adisa Azapagic*

Department of Chemical Engineering, The University of Manchester, Manchester M13 9PL, UK

Corresponding authors: harish.jeswani@manchester.ac.uk; adisa.azapagic@manchester.ac.uk

Abstract

Negative emissions technologies (NETs) are expected to play a significant role in mitigating climate change. However, there are also concerns that a large scale deployment of NETs may cause various environmental impacts due to the use of land, water and energy resources. A number of studies have assessed the environmental performance of various NETs; however, a comprehensive review comparing a range of different NETs is not available in the literature. To address this research gap, this paper compares life cycle assessment (LCA) studies of the following options for which the data were available in the literature: bioenergy with carbon capture and storage (BECCS), biochar incorporation into soil, afforestation and reforestation, soil carbon sequestration, building with biomass, direct air carbon capture and storage (DACCS), enhanced weathering and mineral carbonation. It is evident from this review that these technologies can have net negative life cycle GHG emissions, ranging from -603 kg CO_2 eq./t CO_2 removed for building with biomass to -1173 kg CO_2 eq./t CO_2 removed for biochar incorporation into soil. However, the estimates of GHG removal potentials vary widely among the studies for each technology as well as among the NETs owing to technological differences, methodological choices and differing assumptions. For example, the net global warming potential (GWP) of biochar varies among the reviewed studies between a net positive impact of 1710 to a net negative GWP of 3300 kg CO_2 eq./t CO_2 removed, depending upon the feedstock, pyrolysis technology and the assumptions for credits for co-products and co-benefits. Overall, biochar used as soil amendment has the lowest GWP per tonne of CO_2 removed, followed by soil carbon sequestration, while building with biomass ranks last. The review also reveals that the removal of CO_2 by these technologies could lead to a significant increase in other environmental impacts. Especially, the use of energy in non-bio NETs (DACCS, enhanced weathering and mineral carbonation) leads to relatively high fossil depletion, acidification and human toxicity. These impacts can be reduced if the energy demand of NETs is met by renewables instead of fossil fuels. The paper also identifies several methodological issues and challenges in conducting LCA of NETs and provides recommendations to address them.

Keywords: Climate change; Greenhouse gas removal technologies; Carbon dioxide removal; Net zero; Life cycle assessment; Carbon capture and storage

1 Introduction

Negative emissions technologies (NETs), also sometimes referred to as carbon dioxide removal (CDR) or greenhouse gas removal technologies (GGRTs), are expected to play an important role in efforts to mitigate climate change and achieve 'net zero' targets. It is predicted that the negative emissions (removal) of an order of 100-1000 Gt of CO_2 will be needed by the end of the century to limit the global temperature increase to 1.5 °C above that of the pre-industrial era (IPCC, 2018). The existing and potential NETs are bioenergy with carbon capture and storage (BECCS), biochar incorporation into soil, afforestation and reforestation (AR), soil carbon sequestration (SCS), building with biomass (BwB), direct air carbon capture and storage (DACCS), enhanced weathering (EW), mineral carbonation (MC), ocean fertilisation, ocean alkalinisation, and wetland, peatland and coastal habitat restoration. These technologies differ widely in terms of technological readiness level, greenhouse gas (GHG) removal potentials, costs, risks, co-benefits and trade-offs. In addition, deployment of NETs at such a large scale can pose serious challenges in terms of the use of land, water and energy resources and may be associated with other environmental implications (Fuss et al., 2018, Smith et al., 2019). Therefore, it is vital to assess the associated environmental risks of NETs prior to their large-scale deployment to ensure that the removal of GHG is not carried out at the expense of other environmental issues. Life cycle assessment (LCA) is commonly used for such evaluations and has a broad acceptance among academic and industrial practitioners as well as policy makers.

A number of LCA studies have considered the potential impacts of some NETs, particularly BECCS and biochar, while there is limited literature on others. Several review papers have also discussed LCA impacts of NETs, but these have largely focused on methodological aspects (Goglio et al., 2020, Matuštík et al., 2020, Terlouw et al., 2021). Specifically, Matuštík et al. (2020) reviewed the differences in functional units, system boundaries and impact assessment methods in LCA studies of biochar used as soil amendment. A short review by Goglio et al. (2020) identified that the key issues in LCA studies of NETs are in defining the functional unit and system boundaries, data availability and consideration of temporal aspects of GHG emissions and removals. The paper also proposed a methodological LCA framework to facilitate comparisons of diverse NETs. Finally, Terlouw et al. (2021) conducted a more detailed review of the methodological choices in LCA studies of several NETs. The authors identified several shortcomings in the studies, such as lack of transparency in reporting data and assumptions, lack of consideration of indirect effects (e.g. indirect land use change (iLUC), soil quality changes, effect on biodiversity and albedo changes) and incorrect consideration of avoided emissions through system credits. Even though the review by Terlouw et al. (2021) covered most NETs, it did not present a quantitative analysis and discussion of the life cycle impacts of NETs as reported in the published studies.

Therefore, to the best of the author’s knowledge, a comprehensive review comparing the environmental performance of a range of different NETs is not available in the literature. This review paper aims to close this gap by analysing and comparing the life cycle impacts of NETs based on the results of published LCA studies. Firstly, the environmental impacts of each technology are reviewed and compared for different feedstocks, process routes and other parameters, and then the environmental performance of different NETs is compared. Furthermore, two additional NETs (mineral carbonation and building with biomass), which were not included in other reviews, are also analysed here. Thus, the novelty of this review lies in a quantitative evaluation of life cycle environmental performance, providing to date the most comprehensive analysis of eight different NETs. The main objective is to provide a better understanding of the environmental impacts associated with NETs with the aim of informing future policy and technology developers. A further objective is to identify the key methodological challenges in conducting and comparing LCA studies of diverse NETs and provide recommendations to address those issues.

To set the context, the next section provides an overview of the reviewed NETs. The methodology used in conducting this review is described in Section 3. This is followed in Section 4 by a review of LCA studies, discussing the impact of each NET in turn and then comparing the impacts across the reviewed technologies. Section 5 focuses on key methodological challenges in assessing the environmental impacts of NETs and the ways to address them. The study conclusions can be found in Section 6.

2 Overview of negative emissions technologies

A number of technologies can remove CO_2 from the atmosphere and store it over long periods (The Royal Society and Royal Academy of Engineering, 2018). The removal of CO_2 is achieved through capture of CO_2 from air or flue gases, increasing biological uptake or enhancing organic reactions with rocks. The removed CO_2 can be stored in permanent forests, soils, deep oceans, geological reservoirs or built environment. These technologies are discussed below.

2.1 Bioenergy with carbon capture and storage (BECCS)

BECCS is a combination of two well-known technologies for climate change mitigation: bioenergy and carbon capture and storage (CCS). Broadly, BECCS comprises sequestration of CO_2 from the atmosphere during the growth of biomass, which is then used to provide energy via combustion or another process, with the resulting CO_2 emissions captured and stored in geological formations. Several routes have been proposed for integrating bioenergy with post- or pre- combustion CCS options, as shown in Figure 1. In post-combustion BECCS routes, CO_2 is captured from the flue gas of dedicated biomass-fired or co-fired heat and/or power plants. Pre-combustion BECCS systems capture CO_2 produced during the bioenergy transformation process, such as biomass gasification to produce syngas and fermentation for bioethanol production. The net emissions from BECCS can be negative if the amount of CO_2 stored is greater than that of GHG emitted during biomass production, transport, conversion and utilisation.

Various technology routes for bioenergy are already well established, while CCS is largely at the demonstration stage. Currently, there are five BECCS facilities in operation worldwide, which capture 1.5 Mt CO_2 per year from bioethanol plants (Global CCS institute, 2019). Since BECCS simultaneously provides energy and can reduce atmospheric CO_2 concentration, it is considered as one of the most promising NET and is now included in Integrated Assessment Models (IAMs) by many climate scientists in the modelling “pathways” to meet 1.5 or 2 ^{o}C emission trajectories (IPCC, 2018). The mitigation potential of BECCS is estimated to be between 0.4 and 11.3 bn t of CO_2 eq. per year (IPCC, 2020). However, there are a number of potential challenges associated with the widespread use of BECCS, primarily around scale and land availability (Gough et al., 2018). In particular, there are concerns that the large-scale bioenergy production can have adverse effects on food security, land degradation, biodiversity and water provisioning (IPCC, 2020).

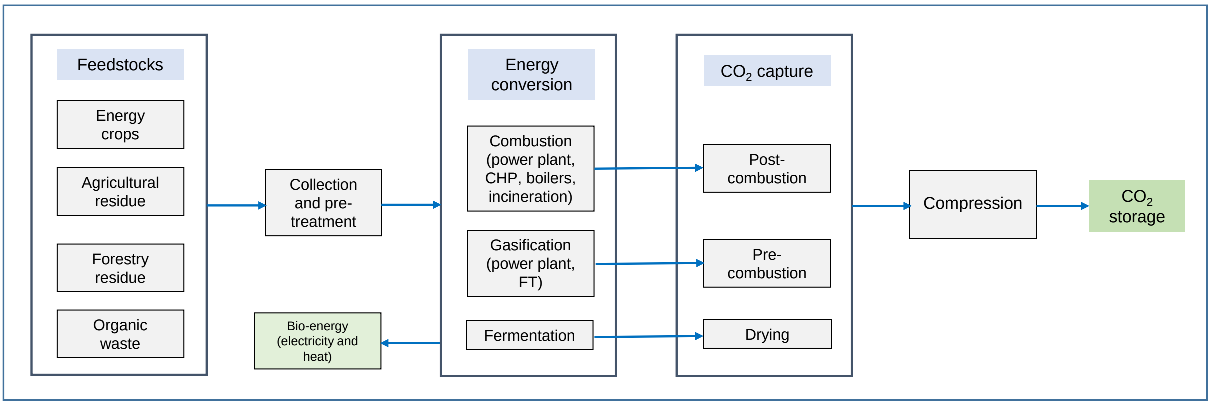


Figure 1 An overview bioenergy with carbon capture and storage for different feedstocks and technologies
(CHP: combined heat and power; FT: Fischer Trospsch)

2.2 Biochar incorporation into soil

Biochar is a carbon-rich material produced by thermochemical conversion of biomass in the absence of oxygen. As indicated in Figure 2, it can be produced from various biomass feedstocks, such as wood, agricultural residues, forestry residues and organic waste (Vijayaraghavan, 2019). Several techniques, including gasification, flash carbonisation, hydrothermal carbonisation, slow pyrolysis, fast pyrolysis and torrefaction, can be used, but the most common and widely employed method to produce biochar is pyrolysis (Wang and Wang, 2019). Pyrolysis converts biomass into solid (biochar), liquid (bio-oil) and gaseous (syngas) products (Figure 2). Numerous factors, such as operating parameters (heating rate, temperature, duration) and feedstocks, affect the yield and quality of biochar. In general, slow pyrolysis has higher (25-35%) biochar yield, while fast pyrolysis is more suited for energy recovery purposes as it favours production of bio-oil or syngas with lower (10-25%) yield of biochar (Ganesapillai et al., 2020).

Biochar technologies can contribute to climate change mitigation in two main ways: by supplying biochar for soil amendment, and bioenergy generation via biochar co-products (bio-oil and syngas). When applied to soil, biochar can act as a long-term carbon storage of a large fraction of carbon in soils in a recalcitrant form, while providing a range of other soil quality benefits, such as improved water and nutrient retention and higher crop yields (Kavitha et al., 2018). However, these effects depend on various factors, including biomass feedstock type, biochar production conditions and its application rates, soil properties and local climate conditions (Kavitha et al., 2018, Tisserant and Cherubini, 2019). It is estimated that biochar addition to soil can potentially mitigate 0.03 - 6.6 bn t of CO_2 eq. per year and could provide moderate benefits for food security by improving yields in the tropics or through improved water-holding capacity and nutrient-use efficiency (IPCC, 2020). Moreover, biochar is technologically mature, can be deployed at lower costs and has multiple co-benefits (see Figure 2). However, like BECCS and other bio-based technologies, the large-scale production of biomass required as feedstock for biochar can cause additional pressure on land, water and biodiversity (IPCC, 2020).

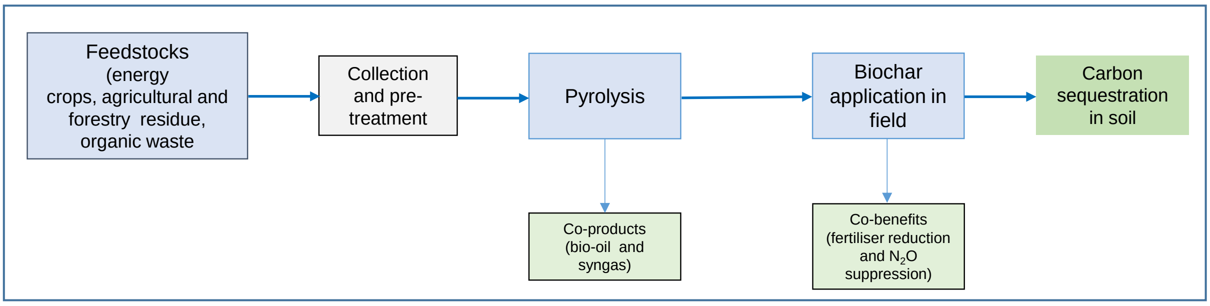


Figure 2 An overview of biochar production and application to soil

2.3 Afforestation and reforestation (AR)

Afforestation is defined as an activity leading to forest creation on a land that was not previously a forest, while reforestation refers to an activity leading to the re-establishment of lost forests (Watson et al., 2000). AR is one of the simplest, if not the simplest NET for removing CO_2 from the atmosphere (Leung et al., 2014). It does not require advanced infrastructure to implement and can be deployed on a large scale using existing technology and equipment (The Royal Society and Royal Academy of Engineering, 2018, Fuss et al., 2018). Forests can store CO_2 in above-ground and below-ground biomass (e.g., stems, branches, trunks, roots, leaves), as well as in soil for decades up to centuries. This is considered 'short-term' storage when compared with a more stable geological storage that can last over thousands of years. One of the issues with AR is that the stored carbon is prone to both human and natural disturbances, such as illegal deforestation, forest fire or drought. The other issues include land competition with the agricultural and energy sectors, land-use change (LUC) effects, natural biodiversity disruptions and decrease of albedo effects in high latitudes (Fuss et al., 2018). The estimated sequestration potential for AR varies among studies from 143 to 536 Gt CO_2 by 2100, depending on the assumptions on the availability and suitability of land for AR (FAO, 2016). It is estimated that by 2030, AR could contribute to reductions of 4.05 Gt CO_2 eq. per year at carbon prices up to US$100 per tonne CO_2 eq. (FAO, 2016).

2.4 Soil carbon sequestration (SCS)

SCS involves transfer of atmospheric CO_2 into stable soil organic carbon through improved land management practices (Lal, 2013). Several management practices can be effectively applied to ecosystems that have high carbon sequestration potentials, such as croplands, grazing lands and degraded soils (Sykes et al., 2020). In croplands, carbon can be sequestered in soil through improved farm management practices, such as the conversion of conventional tillage to reduced or no tillage, application of manure and cover crops, diversification of crop rotation and more efficient water management (Lal, 2013, Sykes et al., 2020). In grazing lands, controlled grazing, management of rangeland fire and improving forage species can promote SCS (Lal, 2013). Pathways for SCS in degraded soils include restoration through soil-erosion control and afforestation on marginal land.

Global estimates of the SCS potential vary considerably, but a recent review suggests the annual technical potential stands between 2.3 and 5.3 Gt CO_2/year (Fuss et al., 2018). The main advantage of SCS is that 20% of the global potential can be realised at net-negative cost (net saving), and the remainder at a low cost (US$ 0-100/t CO_2 removed) (Smith, 2016). Furthermore, the additional land requirement for SCS is almost negligible. However, similar to AR, a drawback of this method is the reversibility of stored carbon as it is prone to human and natural disturbances. Thus, protecting soil carbon storage is dependent on preventing future land-use changes (Petersen et al., 2013). Another downside of SCS is sink saturation, which can be reached as quickly as in ten years, depending on the soil type and climate region (The Royal Society and Royal Academy of Engineering, 2018). After that, the sequestration potential of soil could decrease to zero.

2.5 Building with biomass (BwB)

BwB as a NET relies on incorporation of biomass in various applications in construction to provide storage for CO_2 captured through forestry and agriculture. Biomass, such as wood and straw, has been used in construction since time immemorial. Nowadays, the most commonly used biomass is wood, which is typically made into building frames, claddings and other components. Other biomass, such as bamboo, straw and hemp, are also used for various purposes, including wall panels, flooring, scaffolding and insulation (Kuittinen et al., 2021). Furthermore, the plant fibres such as hemp, can be used in making concrete (Arrigoni et al., 2017, Ip and Miller, 2012). The storage of the sequestered carbon through BwB can be up to several decades, depending on the lifespan of the building component and its end-of-life treatment (Element Energy, 2021). Furthermore, the timber used in construction, if sourced from sustainably managed forests, can reduce the embodied GHG emissions of buildings by replacing GHG-intensive materials, such as cement or steel (Andersen et al., 2021). It is estimated that BwB is capable of removing 0.5-1 Gt CO_2 per year (McLaren, 2012). However, this would require active reforestation efforts to maintain carbon storage in forests (Churkina et al., 2020).

2.6 Direct air capture and carbon storage (DACCS)

Direct air capture (DAC) is an emerging technology, which can play an important role in climate change mitigation as it can capture directly CO_2 from the atmosphere (Breyer et al., 2019). The proposed DAC processes are mainly based on using either an aqueous solution, such as NaOH or KOH (Keith et al., 2018), or solid sorbents, including amine-based anion exchange resin (Lackner, 2009), as the capture media. Following CO_2 capture, the capturing medium is regenerated to produce a near-pure stream of CO_2, which can be stored into geological formations, e.g. depleted oil and gas reservoirs (Figure 3). Since the CO_2 concentration in air is 100 to 300 times lower than in point sources (e.g. flue gases from power or cement plants), the DACCS process is more energy intensive than conventional CCS. Furthermore, the regeneration of capture media requires additional energy, which depends on the type of DAC process. The aqueous process is more energy intensive as the regeneration of hydroxides is carried out at 800-900 ^{o}C (Daggash et al., 2020). On the other hand, solid-sorbent DAC has lower energy needs as relatively lower temperatures (80-100 ^{o}C) are required for the regeneration of solid sorbents (Fasihi et al., 2019). Furthermore, solid-sorbent DAC can be a net producer of water, while the water demand in aqueous DAC systems could be very high (Fasihi et al., 2019). An advantage of DAC plants, regardless of the medium used, is that they can be employed next to suitable geological storage sites, hence eliminating the need for long-distance transportation. To date, several privately and publicly funded projects have developed demonstration and small commercial DACCS plants (Fasihi et al., 2019).

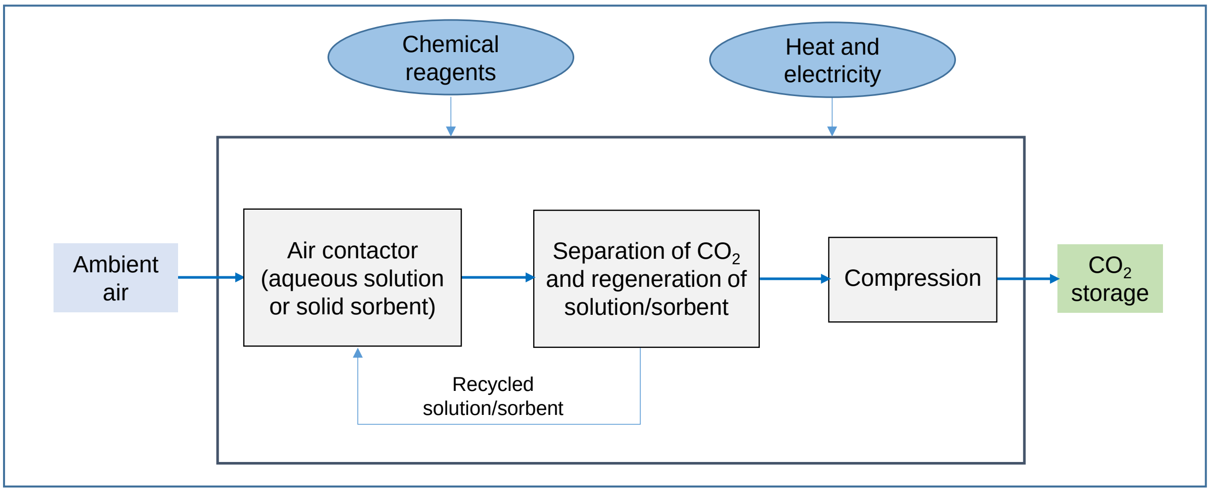


Figure 3 An overview of direct air carbon capture and storage

2.7 Enhanced weathering (EW)

EW is based on the acceleration of naturally-occurring weathering reactions between CO_2 and alkaline minerals to sequester CO_2 from the atmosphere on a much shorter timescale than in nature (Beerling et al., 2018, Fawzy et al., 2020). This is achieved by grinding silicate rocks to enhance their mineral dissolution rate and applying them on the land surface (Figure 4). These silicate minerals exist naturally in igneous rocks, such as dunite and basalt, or can be found in the by-products of certain industrial processes, for example, the manufacture of steel, iron, cement and lime (Renforth, 2012). Weathering of basalt rock on soils improves crop growth and soil fertility, hence can reduce the environmental impacts of agriculture (Beerling et al., 2018). Furthermore, co-deployment of EW with feedstock crops for BECCS and biochar could enhance the feasibility and carbon sequestration potential of these methods (Amann and Hartmann, 2019, Beerling et al., 2020). Unlike bio-based NETs, EW can sequester CO_2 without disrupting food production. However, the development of widespread mining, grinding and spreading operations would lead to negative environmental impacts through increased energy consumption and emissions of particulate matter (Taylor et al., 2016). These concerns may limit its social acceptability.

The carbon removal potential of EW will depend on various site-specific factors, including the type of rock used, its application rate and the geology of the application area considered (Beerling et al., 2018, Moosdorf et al., 2014). The best suited locations are warm and humid areas, such as those found in Brazil, China, India and the USA (Beerling et al., 2020, Strefler et al., 2018). Most Ca- and Mg-rich silicate rocks have the capacity to sequester >1 t CO_2/t rock, while the capture potential of basic rocks, including basalts, ranges from 0.2–0.8 t CO_2/t (Kantola et al., 2017). Theoretical estimates of CO_2 capture and sequestration schemes involving global croplands and silicate rocks are very uncertain. Based on a literature review, Fuss et al. (2018) estimated that EW is capable of sequestering 2-4 Gt CO_2 per annum by 2050. Like other large-scale NETs, EW is relatively immature and requires further research and demonstration across a range of crops, soil types, climates and regions (Beerling et al., 2018). Although alkaline and silicate minerals are available in abundance, the main barriers for the application of EW would be land constraints and costs (Strefler et al., 2018, Tan and Aviso, 2021).

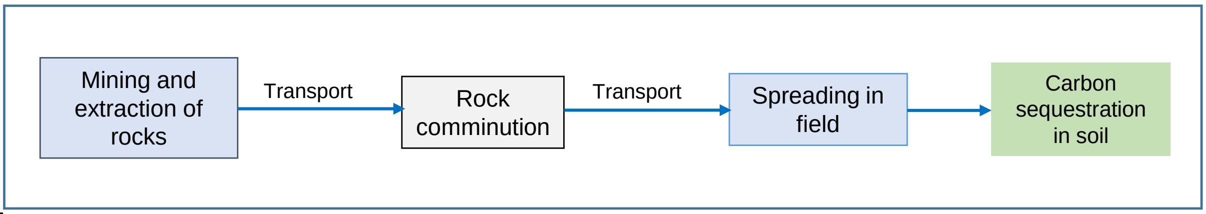


Figure 4 An overview of enhanced weathering

2.8 Mineral carbonation (MC)

Similar to enhanced weathering, MC mimics the natural weathering process of rocks to sequester CO_2 but at a faster rate than in nature. As indicated in Figure 5, unlike enhanced weathering, MC converts CO_2 chemically into stable carbonates by reacting it with minerals, such as calcium, magnesium and iron. These elements occur in high concentrations in silicate rocks (e.g. forsterite, serpentinite and wollastonite) as well as in some industrial wastes, such as steel slag, cement kiln dust and fly ash (Liu et al., 2021, Galina et al., 2019). This reaction can take place either *in situ* (below ground) or *ex situ* (above ground). The *in-situ* process involves the injection of CO_2 into permeable rock, while the *ex situ* approach requires rock mining (for natural minerals) and comminution before reacting with CO_2. Chemicals used for these purposes include sodium bicarbonate, ammonium sulphate, hydrochloric and sulphuric acids (Sanna et al., 2014). Since mining and grinding of natural minerals are energy-intensive processes, the use of pre-ground material, such as industrial waste or mine tailings, reduces the energy demand of this process. The resulting carbonate products can be potentially used as construction materials.

Mineral carbonation is largely considered as an alternative to conventional CCS since it uses a CO_2-rich gas from industrial processes. A number of companies are working actively in the UK, Europe and the USA to commercialise the MC process (Khoo et al., 2021). Although it has a huge sequestration potential (>36,000 Gt CO_2) owing to the abundance of silicates around the globe, its application is currently limited because of high costs associated with the energy use and material handling, which range from \$50 to \$300 per t CO_2 sequestered (Sanna et al., 2014).

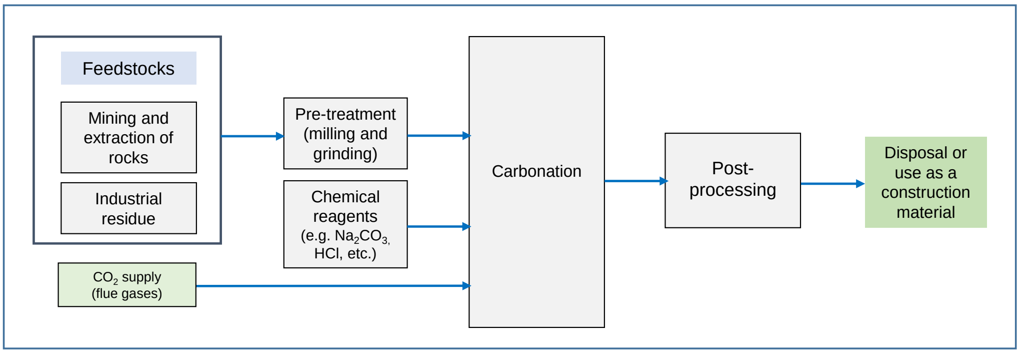


Figure 5 An overview of mineral carbonation (*ex situ*)

2.9 Other negative emissions technologies

There are several other possible NETs, such as ocean fertilisation, ocean alkalisation, and wetland, peatland and coastal habitat restoration, which can remove and permanently store CO_2 (The Royal Society and Royal Academy of Engineering, 2018). These are not considered in this review as there are no LCA studies for these techniques.

3 Methods

A systematic literature search has been performed in different databases (Web of Science, Science Direct, Scopus and relevant academic journals) to identify academic, peer-reviewed studies on LCA of NETs. The following keyword strings have been used: "('life cycle assessment' OR 'LCA' OR 'life cycle') AND ('greenhouse gas removal' OR 'GGRT' OR 'carbon dioxide removal' OR 'CDR' OR 'negative emission' OR NET)". This was then followed by including the keywords for the specific technology (e.g. 'bioenergy with carbon capture and storage' OR 'BECCS', biochar, etc.) in the search string. All search results were screened based on their title and abstract and excluded if irrelevant. In addition to the above, the additional relevant papers were identified via the 'snowball approach' (Wohlin, 2014) by checking the references in the previous reviews and other studies identified and shortlisted in the database search. In total 175 papers had been found initially, but after detailed screening on their relevance, 82 papers on LCA of different NETs have been selected for the review. To enable comparison of the LCA impacts of different NETs, the LCA results from the reviewed studies have been converted to the functional unit of 1 t of CO_2 removed as per the methods summarised in Table 1. The studies which have not provided the required information for such conversion are excluded from the comparative analysis. Furthermore, some of the reviewed studies have included additional processes or functions in the system boundaries as explained later in the relevant sections (such as co-firing in BECCS and additional agricultural processes after incorporation of biochar in soil in biochar studies); hence, these are also excluded from the comparisons.

The findings of the literature review of different NETs are presented in the next section. First, the environmental impacts of each technology are discussed in turn in Sections 4.1-4.8, followed by a comparison of their impacts in Section 4.9. For details on the LCA impacts of each technology, see Supplementary Information (SI).

4 Life cycle environmental impacts of negative emissions technologies

4.1 Environmental impacts of BECCS

Several LCA studies have assessed the environmental impacts of BECCS technologies. As shown in Table 2, these cover a wider range of regions, feedstocks, technologies, functional units and impact assessment methods. The regional coverage spans Europe, India, the USA, Brazil, China and Australia. Over ten feedstocks have been evaluated, such as agricultural and forestry residues, energy crops and municipal solid waste (MSW). The two most-studied BECCS technologies are biopower and biogasification power plants. The other options include bioethanol, biohydrogen and combined heat and power (CHP) plants, as well as electricity from landfill biogas. For carbon capture, most studies have considered post-combustion capture in monoethanolamine (MEA), but some evaluated other options, including Selexol, hot potassium carbonate and membrane separation. Studies on bioethanol and biogasification have analysed pre-combustion capture from fermentation or gasification processes.

Table 1 Methods used to convert LCA impacts reported in the literature to 1 t of sequestered, removed and/or stored CO_2

NETs	Actual functional unit in literature	Formulae used in the current study to convert impacts in LCA studies to the functional unit of 1 t of CO_2 (sequestered, removed and/or stored)
Bioenergy with carbon capture and storage (BECCS)	1 kWh (or MWh or MJ of energy)	$\frac{\text{LCA impacts of electricity with CCS per kWh} - \text{LCA impacts of electricity without CCS per kWh}}{\text{t } CO_2 \text{ stored per kWh}}$
Biochar	1 t of feedstock	$\frac{\text{LCA impacts per t of feedstock}}{\text{t } CO_2 \text{ sequestered per t of feedstock}}$
	1 t of biochar	$\frac{\text{LCA impacts per t of biochar}}{\text{t } CO_2 \text{ sequestered per t of biochar}}$
	1 ha of agricultural land use	$\frac{\text{LCA impacts per ha}}{\text{t } CO_2 \text{ sequestered per ha}}$
	1 year operation of the production plant	$\frac{\text{LCA impacts per year}}{\text{t } CO_2 \text{ sequestered per year}}$
Afforestation and reforestation (AR)	1 ha	$\frac{\text{Net LCA impacts of AR per ha}}{\text{t } CO_2 \text{ sequestered per ha}}$
Soil carbon sequestration (SCS)	1 t (or kg) of crops or animal products	$\frac{\text{LCA impacts of improved agricultural practices per t} - \text{LCA impacts of conventional agricultural practices per t}}{\text{t CO2 sequestered from improved agricultural practices per t} - \text{t CO2 sequestered from conventional agricultural practices per t}}$
Building with biomass (BwB)	1 m^2	$\frac{\text{LCA impacts per } m^2}{\text{t } CO_2 \text{ sequestered per } m^2}$
	1 m^3	$\frac{\text{LCA impacts per } m^3}{\text{t } CO_2 \text{ sequestered per } m^3}$
	1 kg	$\frac{\text{LCA impacts per kg}}{\text{t } CO_2 \text{ sequestered per kg}}$
Direct air carbon capture and storage (DACCS)	1 kWh of electricity (or MWh or MJ of energy)	$\frac{\text{LCA impacts of electricity with DACCS per kWh} - \text{LCA impacts of electricity without DACCS per kWh}}{\text{t } CO_2 \text{ stored per kWh}}$
Enhanced weathering	1 t of rock	$\frac{\text{LCA impacts per t of rock}}{\text{t } CO_2 \text{ sequestered per t of rock}}$
Mineral carbonation (MC)	1 MWh of electricity (if electricity impacts included)	$\frac{\text{LCA impacts of electricity with MC per MWh} - \text{LCA impacts of electricity without MC per MWh}}{\text{t } CO_2 \text{ sequestered per MWh}}$
	1 MWh of electricity (if electricity impacts excluded	$\frac{\text{LCA impacts of electricity per MWh}}{\text{t } CO_2 \text{ sequestered per MWh}}$
	1 t of feedstock	$\frac{\text{LCA impacts per t of feedstock}}{\text{t } CO_2 \text{ sequestered per t of feedstock}}$

Table 2 Summary of BECCS LCA studies

Study (by year)[a]	Region	Feedstocks	Bioenergy technology	Carbon capture and storage (CCS) option	Functional unit	System boundary	LCIA method and impacts[b] assessed
Spath and Mann (2004)	US	Energy crops	IBGCC[c]	Post-combustion absorption in MEA[d]	1 kWh	From biomass production to CO_2 storage	GWP only
Carpentieri et al. (2005)	Europe	Poplar	IBGCC[c]	Post-combustion absorption in MEA[d]	1 MJ	From biomass production to CO_2 compression	Eco-indicator 95 (GWP, ODP, AP, EP, summer smog, energy)
Laude et al. (2011)	France	Sugar beet	Fermentation	Pre-combustion (for fermentation); amine-based post-combustion (for co-generation)	1 hl of ethanol	From biomass production to CO_2 storage	Impact 2002+ (GWP and energy use)
Jana and De (2016)	India	Agricultural residue (rice straw)	Gasification	Pre-combustion	1 kg of ethanol and 1 MJ of electricity	From biomass collection to CO_2 storage	GWP only
Susmozas et al. (2016)	Germany	Poplar	Biomass gasification	Post-combustion membrane	1 kg of hydrogen	From biomass production to CO_2 compression	CML 2001 (GWP, CED, ODP, POFP, AP, EP and land competition)
Gibon et al. (2017)	Global	Forestry residue, SRC[e]	CHP[f]	Not mentioned	1 kWh (electricity)	From biomass production to CO_2 storage	ReCiPe 1.08 (GWP, AP, EP, POCP, HTP, FETP, MDP, PMFP, land occupation)
Oreggioni et al. (2017)	Norway	Spruce woodchips	Biomass gasification CHP[f] with CCS	Pre-combustion and post-combustion	kWh_e	From biomass production to CO_2 storage	ReCiPe 1.08 (GWP, FDP, HT, PMFP, TAP)
Pour et al. (2018)	Australia	Landfill gas, bagasse and forestry residue	Gas turbine (landfill gas); Others: circulating fluidised bed power plant	Post-combustion absorption in MEA[d]	1 kWh	Up to CO_2 storage	ALCAS (GWP, ADPf, ODP, AP, EP, POCP, HTP (cancer, non-cancer), FETP, PMFP)
Yang et al. (2019)	China	Mix of residues and switch grass	Co-firing power plant	Post-combustion absorption in MEA[d]	1 MWh	From biomass production to CO_2 storage	CML 2001 (GWP, ADPf, ODP, POCP, AP, EP, HTP, FAETP, MAETP, TETP)
Zang et al. (2020)	Europe	Pine wood	IBGCC[c]	Post-combustion absorption in MEA[d] and Selexaol	1 kWh	From biomass production to CO_2 storage	CML 2001 (GWP, AP, EP, HTP, ODP)
Lask et al. (2021)	Croatia	Miscanthus	Fermentation	Pre-combustion	1 vehicle-km (also 1 MJ)	From biomass production to CO_2 storage	GWP only

[a] The following studies did not provide the required information to convert the results to the functional unit of 1 t of CO_2, hence were excluded from the analysis: Fajardy and Mac Dowell (2017), Cumicheo et al. (2019), Bello et al. (2020) and Hammar and Levihn (2020).

[b] LCIA: Life cycle impact assessment; GWP: Global warming potential; ODP: Ozone depletion potential; AP: Acidification potential; EP: Eutrophication potential; CED: Cumulative energy demand; POFP/POCP: Photochemical oxidants formation/creation potential; HTP: Human toxicity potential; FETP/FAETP: Freshwater aquatic ecotoxicity potential; MDP: Metal depletion potential; PMFP: Particulate matter formation potential; TAP: Terrestrial acidification potential; FDP: Fossil depletion potential; ADPf: Abiotic depletion potential (fossil); MAETP: Marine aquatic ecotoxicity potential; TETP: Terrestrial ecotoxicity potential.

[c] IBGCC: Integrated biomass gasification combined cycle.

[d] MEA: Monoethanolamine.

[e] SRC: Short rotation coppice.

[f] CHP: Combined heat and power.

The LCA studies on BECCS also differ in terms of scope, system boundaries and functional unit. One aspect common to all studies that considered energy crops as feedstock is that they include agricultural processes in the system boundary. Similarly, for agricultural and forestry residues, the system boundary in all studies starts with the collection process. Also, most of the studies account for transport and geological storage of captured CO_2. However, some (Carpentieri et al., 2005, Susmozas et al., 2016) have excluded these two final steps stating that they have negligible effect on the overall results. Although most studies consider carbon neutrality of biogenic CO_2, Oreggioni et al. (2017) argue that due to the temporal gap between biogenic CO_2 emission and corresponding sequestration by biomass, the climate change impact related to biogenic carbon should be included. To account for this, they use a characterisation factor for biogenic CO_2 based on a study by Guest et al. (2013). Furthermore, Yang et al. (2019) have accounted for soil carbon sequestration (SCS) in their study, while Lask et al. (2021) have analysed the effect of SCS in a sensitivity analysis. The functional unit is expressed in energy units (kWh, MWh, GWh or MJ) in all except three studies, which instead consider vehicle-km (Lask et al., 2021), hl of bioethanol (Laude et al., 2011) and kg of biohydrogen (Susmozas et al., 2016). The last two studies exclude the use of bioenergy from the system boundary.

Figure 6 compares the global warming potential (GWP) of different BECCS options; for further details, see Table S1 in the SI. As can be seen from the figure, the average net GWP (per t CO_2 removed) for all BECCS technologies is net negative, with the values ranging from -12 kg CO_2 eq./t CO_2 removed for LFG to -1013 kg CO_2 eq./t CO_2 removed for bioethanol. The average net GWP of all BECCS technologies, except electricity from LFG, is below -830 kg CO_2 eq./t CO_2 removed. However, it is important to note that none of these studies have considered the effect of land-use changes (LUC). According to Fajardy and Mac Dowell (2017), BECCS carbon efficiency (ratio of geologically stored CO_2 by BECCS to the sequestered CO_2 by the biomass used in BECCS) is reduced from 62% (marginal land) to 46% (grassland) when adding direct LUC and indirect LUC, with the latter accounting for over 26% of the carbon leakage. Thus, if the production of feedstock is associated with LUC, BECCS may no longer be carbon negative. This emphasises the fact that, although efforts must be made throughout the BECCS supply chain to reduce carbon leakage (related to chemicals, transport and carbon capture), a better understanding and control of LUC will be necessary to maximise BECCS carbon efficiency and ensure that they are net-negative.

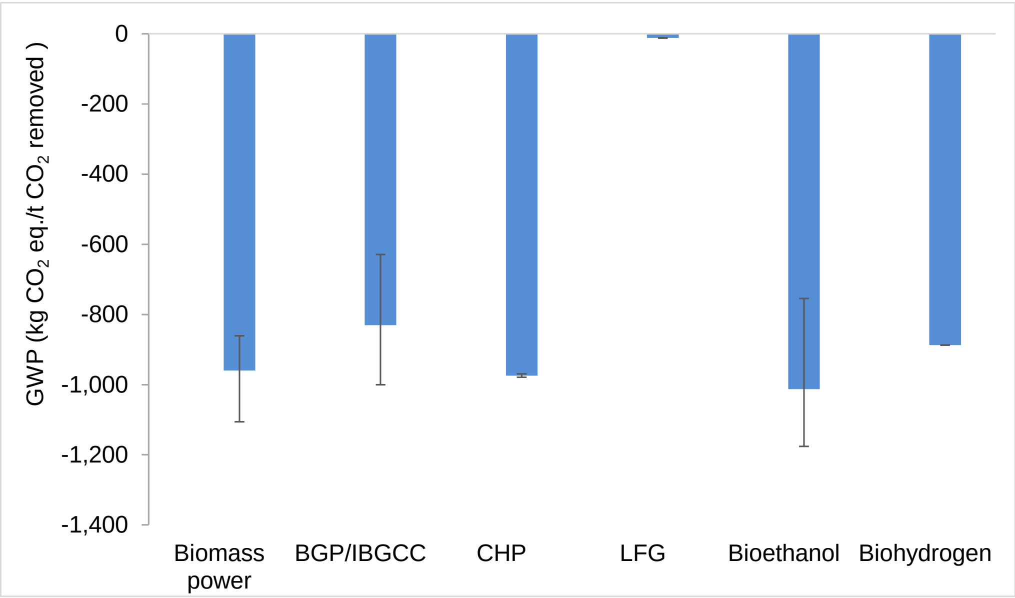
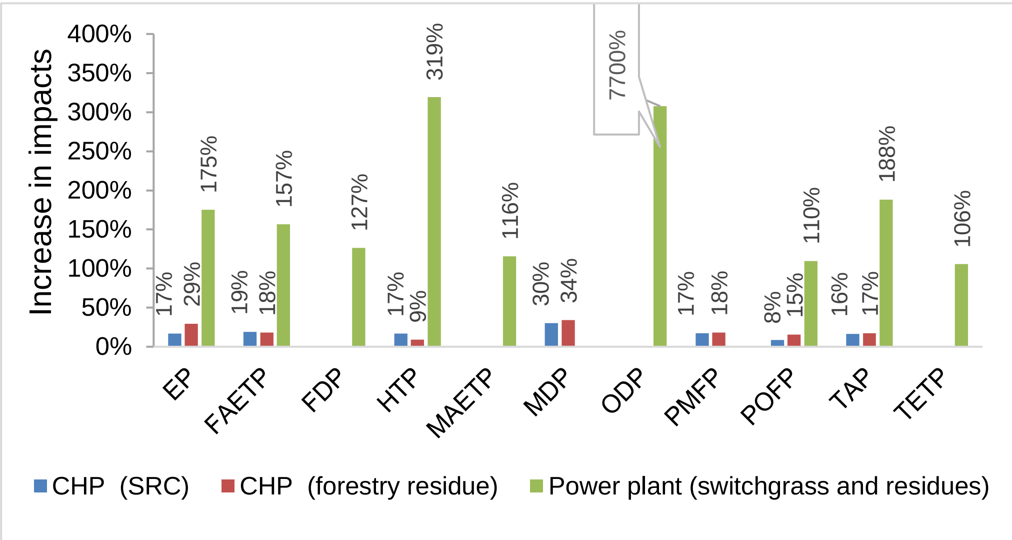
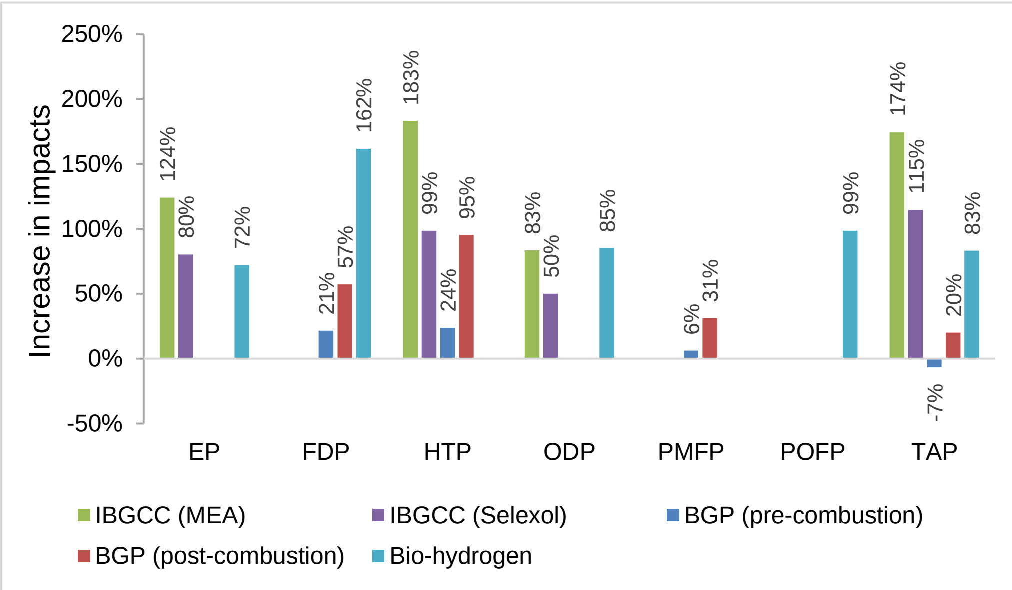


Figure 6 Global warming potential (GWP) of BECCS options

[For data and their sources, see Table S1 in the SI. BGP: Biomass gasification power; IBGCC: Integrated biomass gasification combined cycle; CHP: combined heat and power; LFG: landfill gas.]



(a) Power plant and CHP (Gibon et al., 2017, Yang et al., 2019)



(b) Gasification (Oreggioni et al., 2017, Susmozas et al., 2016, Zang et al., 2020)

Figure 7 Increase in LCA impacts of BECCS in comparison to bioenergy without CCS
[EP: Eutrophication potential; FAETP: Freshwater aquatic ecotoxicity potential; FDP: Fossil depletion potential; HTP: Human toxicity potential; MAETP: Marine aquatic ecotoxicity potential; MDP: Metal depletion potential; ODP: Ozone depletion potential; PMFP: Particulate matter formation potential; POFP: Photochemical oxidants formation potential; TAP: Terrestrial acidification potential; TETP: Terrestrial ecotoxicity potential; SRC: Short rotation coppice; CHP: Combined heat and power; BGP: Biomass gasification power; IBGCC: integrated biomass gasification combined cycle.]

As indicated in Table 2, eight out of 11 studies have assessed a range of other LCA impacts in addition to GWP; however, they have used different impact assessment methods. As a result, they cannot be compared directly. Instead, it is only possible to consider the relative increase in impacts due to CCS. As shown in Figure 7a, the LCA impacts of electricity from CHP increase by 8-34% with CCS (Gibon et al., 2017). On the other hand, the study by Yang et al. (2019) on power plants has found that the increase in impacts due to CCS is much higher, varying from 106-116% for marine aquatic ecotoxicity potential, photochemical oxidant formation potential (POFP) and terrestrial ecotoxicity potential, to 7700% for ozone depletion potential (ODP). This is mainly due to a 30% decrease in the net efficiency of the plant (due to the high power demand of the CO_2 capture unit) and the emissions from the supply chain of MEA. Furthermore, Oreggioni et al. (2017) report that the incorporation of the post-combustion CCS in biomass gasification plants increases fossil depletion potential (FDP) by 57%, human toxicity potential (HTP) by 95%, terrestrial acidification potential (TAP) by 20% and particulate matter formation by 31%. However, with the pre-combustion CCS, there is a small (7%) decrease in TAP while the other impacts increase by 6-24% (see Figure 7b). Moreover, Zang et al. (2020) find that the eutrophication potential (EP), HTP, ODP and TAP of electricity would be 83-183% higher for IBGCC (using MEA) with CCS than without it. Similarly, in the same study the authors state that if Selexol is used to capture CO_2, the increase in impacts due to CCS is in the range of 50-115%. Finally, the study on hydrogen production using biomass gasification (Susmozas et al., 2016) suggests that inclusion of CCS leads to 72-162% increase in EP, FDP, ODP, POFP and TAP of biohydrogen due to the increased electricity demand of the CCS system.

4.2 Environmental impacts of biochar

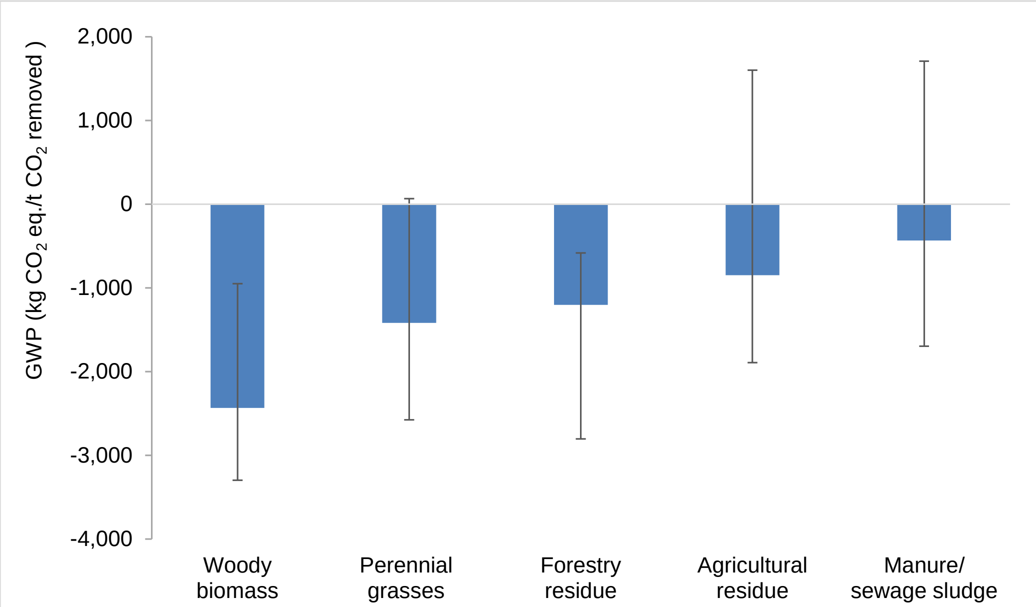
Studies assessing the life cycle environmental impacts of biochar as a NET have considered over 30 different feedstocks, such as energy crops, agricultural and forestry residues, sewage sludge and animal manure (Table 3). Besides feedstocks, various other parameters, including pyrolysis process conditions and effects of biochar on soil, can affect the impacts of biochar (Matuštík et al., 2020). The LCA studies vary in terms of the pyrolysis conditions and techniques, as indicated in Table 3. Moreover, the chemical properties and associated differences in recalcitrance and agronomic benefits (carbon sequestration, reduction in N_2O emissions, crop yield improvements) of biochar vary depending on the feedstock and the thermo-chemical conversion pathways (Field et al., 2013). The majority of the studies have considered pyrolysis temperatures between 450 and 600 °C, whilst some studies also included lower temperatures (200-400°C). The yield of biochar is lower at high temperatures, but the produced biochar contains a higher percentage of fixed carbon (Lu and El Hanandeh, 2019). Since the pyrolysis temperature has a significant impact on the characteristics and the quantity of produced biochar, as well as other co-products (bio-oil, syngas), some LCA studies (e.g. Lu and El Hanandeh (2019)) have compared the impacts for different temperature conditions as discussed further below.

Furthermore, published LCA studies on biochar also differ in terms of system boundaries, functional unit and the allocation methods used to apportion impacts between biochar and co-products. Studies focusing on the utilisation of waste or residual biomass have defined the functional unit based on the amount of feedstock, i.e. t of biomass (Bartocci et al., 2016, Ji et al., 2018), t of manure (Kreidenweis et al., 2021) or m^3 of sludge (Cao and Pawłowski, 2013). Some other studies, such as Brassard et al. (2018), Muñoz et al. (2017) and Rajabi Hamedani et al. (2019), have assessed the impacts using the amount of biochar produced as the functional unit. Moreover, Thers et al. (2019) have assumed the perspective of biochar as soil amendment and its effects on crops, defining the functional unit as the amount of crop produced on the treated field. In yet another study, the authors (Peters et al., 2015) have considered the use of agricultural land over a certain time period (ha/year) as the functional unit. Similar to the BECCS LCA studies, the agricultural processes are included in the system in biochar LCA studies when the feedstock is an energy crop, while for agricultural and forestry residues, manure and sludge, the system boundary starts with the collection process.

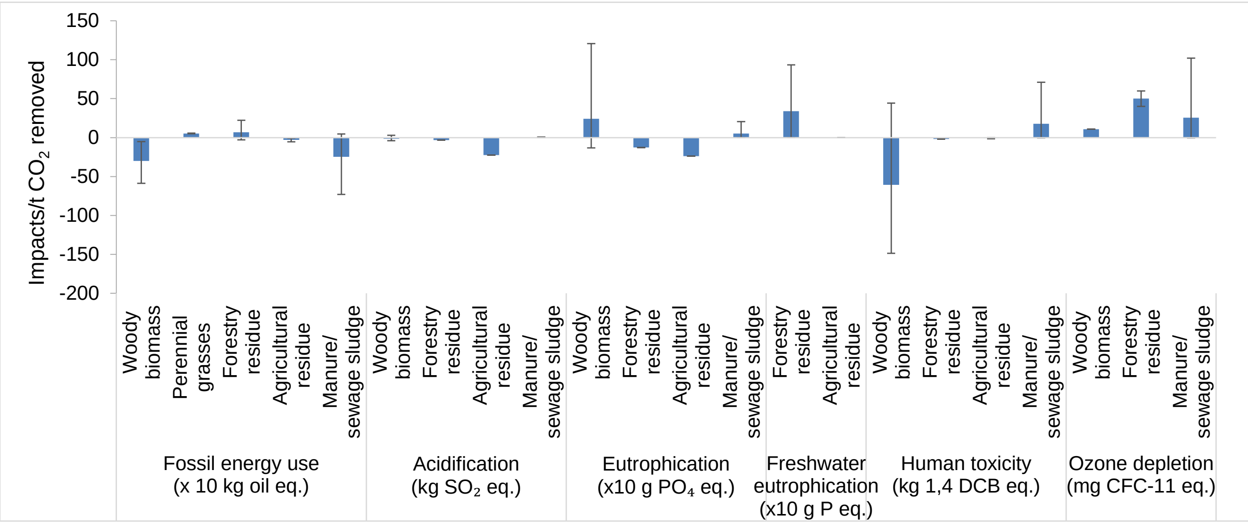
Figure 8a compares the GWP of biochar used as a soil amendment produced from different feedstocks as reported in LCA studies; see also Table S2 in the SI. As for BECCS, the original results have been recalculated for the functional unit of 1 t of CO_2 removed using the methodology detailed in Table 1. As the figure reveals, the GWP of biochar varies significantly between feedstocks as well as among the studies for the same feedstock. On average, biochar from woody biomass has the highest GHG removal benefits (-2433 kg CO_2 eq./t CO_2 removed), followed by the perennial grasses (Miscanthus and switchgrass; -1418 kg CO_2 eq./t CO_2 removed), while manure and sewage sludge have the lowest GHG removal capacity (-433 kg CO_2 eq./t CO_2 removed). These differences are due to differing assumptions in the studies, such as pyrolysis conditions affecting the yield of biochar and co-products (bio-oil and syngas), stable carbon content in biochar (affecting carbon sequestration in soil), consideration of co-benefits of biochar application to soil (reduction in fertiliser needs, reductions in soil N_2O emissions and increase in crop yields), substitution credits for the co-products and other methodological aspects. For example, the earlier mentioned study by Lu and El Hanandeh (2019), which considered a range of temperatures (300-600^oC) for pyrolysis of hardwood, found that the net benefit in GWP per t of feedstock is highest at 300 ^oC and lowest at 600 ^oC, due to three-times higher biochar yield at lower temperatures (51.2% vs 16.3%). However, when the results are converted to per t of CO_2 removed, the biochar produced at higher temperatures has a better GWP removal rate due to higher credits for the bio-oil and syngas.

Furthermore, the pyrolysis conditions also affect the carbon content in biochar and its stability in soil. In the reviewed studies, different assumptions have been made to estimate the carbon sequestration potential of biochar in the soil, which varies from 0.08 to 3.4 t CO_2 eq./t biochar (Table 3). In particular, some studies on biochar from agricultural residues (Llorach-Massana et al., 2017, Robb and Dargusch, 2018), manure and sewage sludge (Cao and Pawłowski, 2013, Kreidenweis et al., 2021, Rajabi Hamedani et al., 2019) have estimated a lower sequestration potential of biochar, resulting in lower net GHG benefits or even net-positive GHG emissions per t CO_2 removed (Figure 8a). The majority of studies have applied the system expansion approach to deal with the pyrolysis co-products, such as syngas and bio-oil, but in some, the co-products are either used within the pyrolysis plant to meet its energy needs or not recovered (Kreidenweis et al., 2021, Mohammadi et al., 2016b, Robb and Dargusch, 2018). Only one study (Llorach-Massana et al., 2017) has used the allocation based on the carbon content in products and co-products instead of substitution, hence estimating the lower net GHG benefits. Although the reduction in N_2O emissions from soils due to biochar application is one of the most uncertain aspects of biochar systems (Brassard et al., 2018), a number of studies have accounted for this benefit (see Table 3). Most of the studies have excluded LUC; however, the analysis by Roberts et al. (2010) suggested that, if the production of feedstock is associated with LUC, then the biochar could have net-positive rather than net-negative GHG emissions.

As indicated in Table 3, only a small number of studies have assessed other LCA impacts in addition to GWP, often using differing LCA methods. Figure 8b reveals that, on average, the use of biochar as NET results in the net savings/benefits for fossil energy use (-29 to -300 kg oil eq./t CO_2 removed), acidification (-1.6 to -22.4 kg SO_4 eq./t CO_2 removed) and human toxicity (-1.1 to -60.7 kg dichlorobenzene eq./t CO_2 removed), if it is produced from agricultural residues and woody biomass. Biochar from agricultural residues also leads to net savings for eutrophication, while biochar from forestry residue has net savings for acidification, eutrophication and human toxicity. However, as can be seen in Figure 8b (and Table S3 in the SI), there is a large variation between the feedstocks, as well as for the same feedstocks among the studies, where some have estimated a net impact and others a net benefit. There are several reasons for these variations. For example, Rajabi Hamedani et al. (2019) finds that producing biochar from pig manure is less beneficial for fossil depletion and ozone depletion than the biochar produced from willow due to the high amount of energy used in the pre-treatment step. On the other hand, the production of biochar from willow has significantly higher eutrophication, acidification and human toxicity due to the use of fertiliser and diesel in the cultivation stage. A study by Muñoz et al. (2017), comparing agricultural and forestry residues for biochar production, has reported that the syngas (co-product) produced from the latter has higher calorific value and hence receives higher credits for the avoided natural gas. As a result, biochar produced from forestry residues has lower impacts than biochar from agricultural residues. Furthermore, the study has also found that the net environmental benefits for both feedstocks increase with higher pyrolysis temperature due to the higher yield of co-products (syngas and bio-oil), which in turn bring higher system credits. A similar effect of temperature on the net environmental impacts has also been observed by Lu and El Hanandeh (2019), who have assessed biochar production from woodchips for a temperature range of 300-600 ºC. However, it should be noted that the higher yield of co-products happens at the expense of biochar yield, which in turn means that more feedstock is required to produce the biochar. This can also be a limiting factor for the biochar production if there are constraints on the availability of land.



(a) Global warming potential



(b) Other life cycle impacts

Figure 8 LCA impacts of biochar as soil amendment for different feedstocks
[For data and their sources, see Tables S2 and S3 in the SI.]

Table 3 Summary of biochar LCA studies

Study (by year)[a]	Region	Feedstocks	Pyrolysis conditions	System boundary	Functional unit	No. of case studies	Cseq. (t CO_2/t biochar)	Soil N_2O reduction	Energy credits	Credits for fertilisers	Other credits	LCIA method and impacts assessed[b]
Roberts et al. (2010)	USA	Crop residue swtichgrass, yard waste	Slow pyrolysis (450 ºC)	Up to the use stage	1 t of dry feedstock	5	1.9 -2	✓	✓	✓	-	GWP and energy
Hammond et al. (2011)	UK	Agro-forestry residues	Pyrolysis	Up to the use stage	1 t of feedstock	10	1.32	✓	✓	✓	For increase in soil organic carbon (SOC) levels	GWP only
Cao and Pawłowski (2013)	Poland	Sewage sludge	Fast pyrolysis (500 ºC)	Up to the use stage	500 m^3 of sewage sludge/day	1	0.8	✓	✓	✓	-	GWP and energy
Field et al. (2013)	USA	Pine wood residue and spent grains	Slow (500 ºC)	Up to soil application	1 t of dry biomass	1	2.6	✓	✓	✓	-	GWP only
Clare et al. (2015)	China	Straw	Slow pyrolysis	Up to the use stage	1 odt of straw	1	2.1	✓	✓	✓	-	GWP only
Peters et al. (2015)	Spain	Poplar	Slow pyrolysis (450 ºC)	Includes all agricultural production processes until the final product substitution	1 ha/yr	1	2.2	✓	✓	✓	Increase in yield, increase in SOC	CML 2001 (GWP, ADP, AP, EP, energy use)
Bartocci et al. (2016)	Italy	Miscanthus	Slow pyrolysis	Cultivation, transformation, packaging, distribution and use	1 t of feedstock	1	1.1	-	✓	-	-	GWP only
Mohammadi et al. (2016b)	Vietnam	Rice straw	Drum oven	Up to the production of rice	1 t of dry rice straw		1.3	✓	-	-	-	GWP only
Llorach-Massana et al. (2017)	Spain	Tomato plant residues	400 ºC	Up to the use stage	1 t of biochar	9	0.08 - 0.32	-	-	-	-	GWP only
Muñoz et al. (2017)	Chile	Oat hulls and pine bark	Pyrolysis (300, 400, and 500 ºC)	Up to soil application	1 t of biochar	6	2.3	✓	✓	✓	-	ReCiPe (GWP, FD, HTP, FEP)
Brassard et al. (2018)	Canada	Switchgrass	460 ºC and 590 ºC	Up to soil amendment	1 t of biochar	2	1.15 & 1.92	✓	✓	-	-	GWP & energy balance
Ji et al. (2018)	China	Straw	Slow pyrolysis	Up to the use stage	1 odt of straw	1	3.4	✓	✓	✓	-	GWP only
Oldfield et al. (2018)	Spain	Oak residue	Slow pyrolysis (650 ºC)	Up to the application of biochar to soil	1 kg product and 1 ha/yr	1	1.27	-	✓	✓	-	CML (GWP, AP, EP)

Study (by year)[a]	Region	Feedstocks	Pyrolysis conditions	System boundary	Functional unit	No. of case studies	Cseq. (t CO_2/t biochar)	Soil N_2O reduction	Energy credits	Credits for fertilisers	Other credits	LCIA method and impacts assessed[b]
Robb and Dargusch (2018)	Indonesia and Australia	Oil palm empty fruit bunch	280 °C	Up to the use stage	1 t of boichar	2	0.38	✓	-	✓	-	GWP only
Azzi et al. (2019)	Sweden	Forest residues	Slow pyrolysis (700 °C)	Up to the use stage	1 t (dry weight) of woodchips	3	2 – 2.6	✓	✓	✓	SOC changes; dairy farm (feed & manure management)	GWP only
Lu and El Hanandeh (2019)	Australia	Hard wood	300 – 600 °C	Up to the use stage	1 t of green logs	7	2.5 - 2.9	-	✓	✓	-	CML 2001 (GWP, AP, FDP, HTP, EP)
Mohammadi et al. (2019a)	Sweden	Sludge (paper mill)	Pyrolysis and hydrothermal carbonation	Up to the use stage	1 t of dry sludge	2	1.5 (pyro-char) & 0.3 (hydro-char)	✓	✓	✓	-	GWP, AP & EP
Mohammadi et al. (2019b)	Sweden	Sludge (paper mill)	Anaerobic digestion with pyrolysis	Up to the use stage	1 t of dry sludge	1	1.5	✓	✓	-	-	GWP, energy use
Mohammadi et al. (2019c)	Sweden	Sludge (paper mill)	Anaerobic digestion with pyrolysis	Up to the use stage	1 t of dry sludge	3	1.5	✓	✓	-	-	CML 2001 (GWP, ADP, ADPf, AP, EP, FAETP, HTP, MAETP, TETP, ODP, POCP)
Rajabi Hamedani et al. (2019)	Belgium (Europe)	Pig manure and willow woodchips	500 °C	From biomass production to the application of biochar to soil	1 t of biochar	2	2.2 (willow) & 0.98 (pig manure)	✓	✓	✓	-	CML & Impact 2002+ (GWP, AP, EP, FETP PCOP, HTP, IRP, MDP, PMFP, land occupation)
Thers et al. (2019)	Denmark	Rapeseed straw	400 and 800 °C	Up to the production of rapeseed	1 t of dry seed	2	1.5 and 1.6	✓	✓	-	-	GWP only
Cheng et al. (2020)	USA	Agricultural and forest residues, sludge	Slow pyrolysis (400 °C, 550 °C, and 700 °C)	Up to the use stage	1 t (dry weight) of feedstock	9	1- 2.7	-	✓	✓	-	GWP and energy

Study (by year)[a]	Region	Feedstocks	Pyrolysis conditions	System boundary	Functional unit	No. of case studies	Cseq. (t CO_2/t biochar)	Soil N_2O reduction	Energy credits	Credits for fertilisers	Other credits	LCIA method and impacts assessed[b]
Kreidenweis et al. (2021)	Germany (Europe)	Broiler manure	400 °C	Up to the application of biochar to soil	1 t of manure	1	0.85	-	-	-	-	GWP only
Papageorgiou et al. (2021)	Sweden	Wood waste	Slow pyrolysis (700 °C)	Up to the use stage (as soil remediation)	1 year of operation of the pyrolysis plant	2	2.4	-	-	-	Heat to compensate for lower heat generation	ILCD 2.0 (GWP, ADP, ADP_f, EP (freshwater), FAETP, ODP, POFP, TAP)
Sahoo et al. (2021)	USA	Forestry residue	Gasifier, kiln, air curtain burner	Up to the use stage	1 t of biochar	10	1.6-2.4	-	-	-	-	GWP only
Yang et al. (2021)	China	Agricultural residues	Slow pyrolysis (300-800 °C)	Up to the use stage	1 t of feedstock	1	1.8	✓	✓	✓	Crop yield increase, SOC	CML (GWP, ADP, ADP_f AP, FAETP, HTP, MAETP, TETP, POCP)

[a] The following studies did not provide the required information to convert the results to the functional unit of 1 t of CO_2 removed and hence were excluded from the analysis: Ibarrola et al. (2012), Sparrevik et al. (2013), Dutta and Raghavan (2014), Galgani et al. (2014), Sparrevik et al. (2014), Mohammadi et al. (2016a), Righi et al. (2016), Smebye et al. (2017), Owsianiak et al. (2018), Moghaddam et al. (2019), Barry et al. (2019), Uusitalo and Leino (2019) and Azzi et al. (2021b).

[b] ADP: Abiotic depletion potential; FEP: Freshwater eutrophication potential; IRP: Ionising radiation potential; For the nomenclature for the others impacts, see Table 2.

4.3 Environmental impacts of afforestation and reforestation

LCA studies related to forests have been published extensively over the years; however, most have focussed on the environmental impacts of forest products (Klein et al., 2015, Schweier et al., 2019). There are only a few publications that have investigated the AR projects from the GHG-removal perspective (Table 4). Although the functional unit in all the studies is the same (1 ha), they differ in terms of the age of trees/forest, plant species, tree density per ha, forest management activities (e.g. fertilisation, thinning and harvesting) and previous land use. As a result, they all report different net CO_2 removal. For example, Proietti et al. (2016) estimate the carbon sequestration during 14 years by the English oak planted on a previous mining site in Italy at 34.96 t CO_2 eq./ha. Another study (Brunori et al., 2017) of the same site but over 34 years, reports around ten times higher carbon sequestration (339.9 t CO_2 eq./ha). Proietti et al. (2016) have also compared the above-mentioned oak plantation with an olive grove and a poplar-walnut plantation finding that, if managed semi-intensively, the last has higher carbon sequestration (252.1 t CO_2 eq./ha) than the intensively managed olive grove (39.6 t CO_2 eq./ha) and the extensively managed oak plantation (34.96 t CO_2 eq./ha).

The biomass growth rate can also affect the carbon removal potential as found by Gaboury et al. (2009) who have reported low sequestration by black spruce trees (282 t CO_2 eq./ha in 70 years) in Canada due to their slow growth. Another factor that influences carbon sequestration is the rotation period. For instance, studying the larch plantation for a rotation forestry project in China, Lun et al. (2018) have estimated the net GHG removal by forest litter and soil at 137.6 t CO_2 eq./ha over the first 41-year rotation period. However, in the subsequent rotation periods, the net sequestration rate showed a declining trend, decreasing from 3.36 t CO_2 eq./ha·year in the first rotation period to 0.97 t CO_2 eq./ha·year after the five rotation periods (at the end of 205 years).

As shown in Table 4, the LCA studies of AR considered only GWP. Their findings are presented in Figure 9 per 1 t CO_2 removed, with the original results recalculated using the methodology in Table 1. The GWP of AR varies from -351 kg CO_2 eq./t CO_2 removed (for olive plantation) to -995 kg CO_2 eq./t CO_2 removed (for black spruce). Although the latter appears to have the greatest removal potential, it should be borne in mind that this is over a much longer period (71 years) than for the other tree species considered (see Table 4). The influence of the plantation age on the net GWP can also be inferred from Figure 9, with a greater sequestration potential achieved over the longer period, although the relationship does not appear to be linear. For instance, the net carbon removal by the oak plantation over 34 years is only marginally lower than for the same plantation over 14 years (-983 vs -912 kg CO_2 eq./t CO_2 removed) (Brunori et al., 2017, Proietti et al., 2016).

Table 4 Summary of LCA studies on afforestation and reforestation

Study (by year)	Region	Type of plantation	Tree species	Years after plantation	Functional unit	Impacts assessed
Gaboury et al. (2009)	Canada	Afforestation of open woodland	Black spruce	70	1 ha	GWP only
Proietti et al. (2014)	Italy	Afforestation (agricultural)	Olive grove	11	1 ha	GWP[a] only
Proietti et al. (2016)	Italy	Afforestation (of ex-mining site for oak)	Oak, olive grove, walnut and poplar	14	1 ha	GWP only
Brunori et al. (2017)	Italy	Afforestation of ex-mining site	Oak	34	1 ha	GWP only
Lun et al. (2018)	China	Managed plantation forest	Larch	41	1 ha	GWP only

[a] The study also discusses the contribution of different life cycle stages to some other LCA impacts but does not provide the values for those impacts.

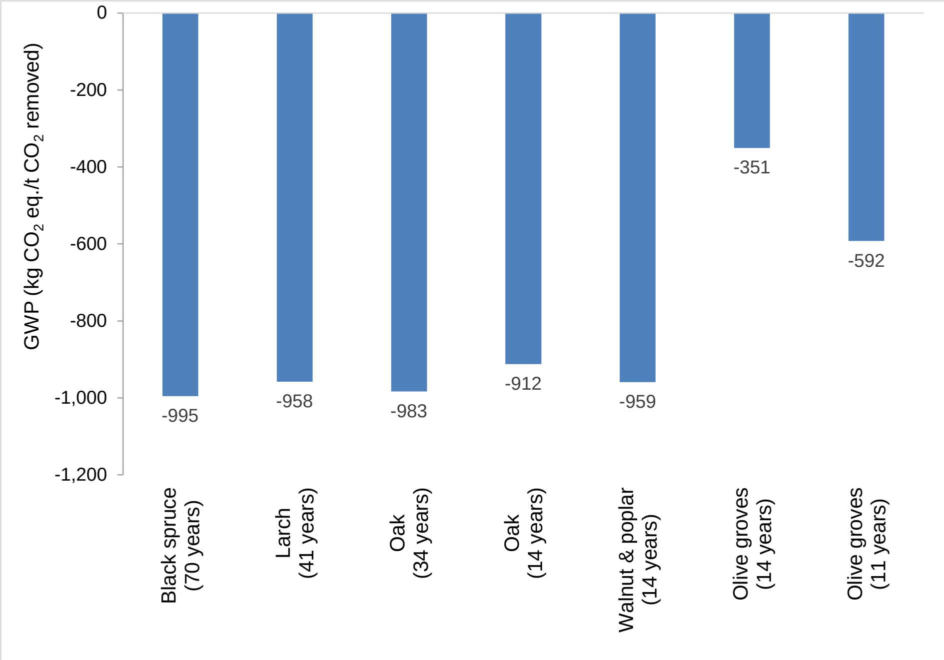


Figure 9 Global warming potential (GWP) of afforestation and reforestation for different tree species
[For data sources, see Table 4.]

The reviewed studies also report that the GWP of AR is directly related to the inputs required for the plantation growth and management. In the case of the olive plantation, the GHG emissions from the plantation represent more than 40% of the net carbon accumulation by the trees as they are intensively managed and require annual fertilisation. On the other hand, the GHG emissions from the activities related to the afforestation account for only 0.5% of the net carbon sequestration by the trees (Gaboury et al., 2009).

4.4 Environmental impacts of soil carbon sequestration

There are many LCA studies on agricultural products and practices but most of them do not include changes in the soil carbon due to the lack of well-defined methods (Goglio et al., 2015). Since the focus of this paper is on quantifying the impacts per unit of CO_2 removed, only studies which provided the required information for such calculations (i.e. GHG emissions of conventional and improved agricultural practices and net increase in soil carbon sequestration due to the improved agricultural practices), are included. As summarised in Table 5, these studies have assessed cradle-to-gate impacts of different crops and animal products (milk and meat). For soil carbon sequestration, they have investigated various management practices, including conservation (reduced or no) tillage, crop rotation, organic farming and changes in grazing. In addition to these differences, they have also used different approaches for estimating soil carbon sequestration. For instance, studies using the IPCC (2006) tier 1 method have considered a 20-years horizon for calculating changes in SCS, while some have used a 100-year timeline, based on Petersen et al. (2013). Comparing both time periods, Joensuu et al. (2021) have found that the annual SCS values for the 100-year horizon are around 60% lower than for 20 years.

Figure 10 compares the GWP reported in the LCA studies on SCS, recalculated here from the original values for the functional unit of 1 t CO_2 removed according to Table 1; for further details, see Table S4 in the SI. The average GWP of all improved practices is net negative, ranging from -453 kg CO_2 eq./t CO_2 removed (for grazing/feed changes) to -2174 kg CO_2 eq./t CO_2 removed (treatment using microbial phosphate inoculant). In the case of grazing changes, there is a large variation in GWP values, from a net-positive impact of 2111 kg CO_2 eq./t CO_2 removed to a net removal of -2600 kg CO_2 eq./t CO_2 removed. This is partly because these studies have considered very different systems with different carbon sequestration rates and partly due to the effect of changes in grazing on the meat or milk yield. For example, Stanley et al. (2018) have compared beef production for adaptive multi-paddock (AMP) grazing with the feedlot in the US and estimated a four-year carbon sequestration rate of 13.2 t CO_2/ha·year in the AMP grazed pastures, while in the feedlot scenario the soil erosion contributed to a small loss of 22.8 kg CO_2/ha·year. According to Arca et al. (2021), the transition from semi-intensive to semi-extensive dairy sheep farming in the Mediterranean would result in an increase in carbon removal by 0.32 t CO_2/ha·year. Both of these studies have also found that these changes in farming would reduce the carbon footprint of beef and milk. On the other hand, Sabia et al. (2020) have found that by changing the cattle feed in Alpine regions in Italy from high to low concentrate would results in a 20% increase in the carbon footprint of milk because of 40% reduction in the milk yield. The reduction in the yield due to improved agricultural practices is also observed in all except four (Kløverpris et al., 2020, Alam et al., 2019, Rahman et al., 2021, Fiore et al., 2018) studies. Improved agricultural practices, such as using microbial phosphate inoculant (Kløverpris et al., 2020) and organic farming (Aguilera et al., 2015), also reduce N_2O emissions from cultivation, hence have the lowest GWP per t CO_2 removed.

As indicated in Table 5, only two studies (Knudsen et al., 2019, Kløverpris et al., 2020) have also considered other LCA impacts of improved agricultural practices, but using two different impact assessment methods. The results in both studies (Figure 11) show that these practices would lead to net savings in all the impacts, except land use. According to Knudsen et al. (2019), the latter increases due to a reduction in the milk yield in organic cattle farming (Figure 11a). By contrast, Kløverpris et al. (2020) have found that using microbial phosphate inoculant would increase the yield and, hence, reduce land requirements (Figure 11b).

Table 5 Summary of LCA studies on soil carbon sequestration

Study (by year)	Region	Improved practices	Time horizon for SCS	Crop/ Product	Functional unit	System boundary	LCIA method and impacts assessed[a]
Archer and Halvorson (2010)	USA	No tillage and rotation	Not mentioned	Maize and beans	1 t of maize; 1 ha	Cradle to farm gate	GWP only
Aguilera et al. (2015)	Spain	Organic farming	100 years	Six crops (citrus, fruits, subtropical fruits, tree-nuts, grapes and olives)	1 kg of each crop	Cradle to farm gate	GWP only
Fiore et al. (2018)	Italy	Sustainable management (mulching of residues, no tillage, cover crops, application of compost)	20 years	Apricot, peach	1 t of fresh fruits; 1 ha	Cradle to farm gate	GWP only
Matsuura et al. (2018)	Japan	No tillage and organic farming	20 years	Eggplant	1 kg of eggplant	Cradle to farm gate	GWP only
Stanley et al. (2018)	US	Adaptive multi-paddock grazing	4 years	Beef	1 kg (carcass weight)	Cradle to farm gate	GWP only
Alam et al. (2019)	Bangladesh	Alternative cropping with higher residue retention	Not mentioned	Rice	1 t of rice	Cradle to farm gate	GWP only
Knudsen et al. (2019)	Western Europe	Organic farming	100 years	Milk	1 kg of FPCM[b]	Cradle to farm gate	PEF (GWP, AP, EP, FETP, ADP, land use and biodiversity)
Holka (2020)	Poland	Reduced and no tillage	100 years	Wheat	1 t of wheat	Cradle to the farm gate	GWP only
Holka and Bieńkowski (2020)	Poland	Reduced and no tillage	100 years	Maize	1 t of maize	Cradle to the farm gate	GWP only

Study (by year)	Region	Improved practices	Time horizon for SCS	Crop/ Product	Functional unit	System boundary	LCIA method and impacts assessed[a]
Kløverpris et al. (2020)	US	Adding of microbial phosphate inoculant	20 years	Maize	1 t of maize	Cradle to the farm gate	CML (GWP, AP, EP, POCP, land use, fossil energy)
Sabia et al. (2020)	Italy	Lowering of concentrated feed intake	100 years	Milk	1 kg of FPCM[b]	Cradle to the farm gate	GWP only
Arca et al. (2021)	Italy	Transition from semi-intensive to semi-extensive dairy sheep farming	Not mentioned	Milk	1 kg of FPCM[b]	Cradle to the farm gate	GWP only
Joensuu et al. (2021)	Finland	Cover crop	20 and 100 years	Wheat	1 kg of wheat	Cradle to farm gate	GWP only
Rahman et al. (2021)	Bangladesh	No tillage and minimum tillage	Not mentioned	Wheat	1 t of wheat	Cradle to farm gate	GWP only

[a] For the impacts nomenclature, see Table 2.
[b] FPCM: Fat and protein corrected milk.

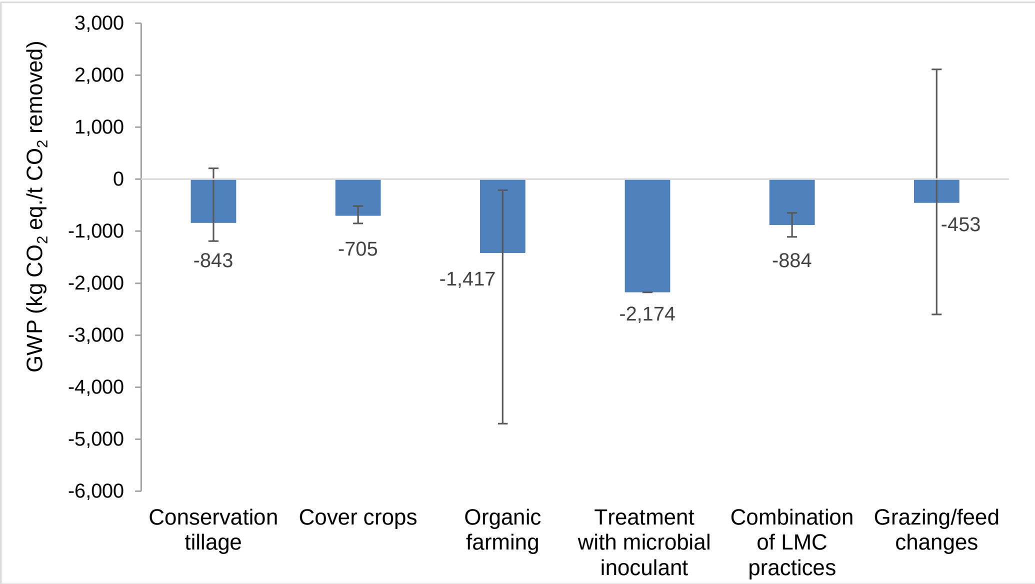
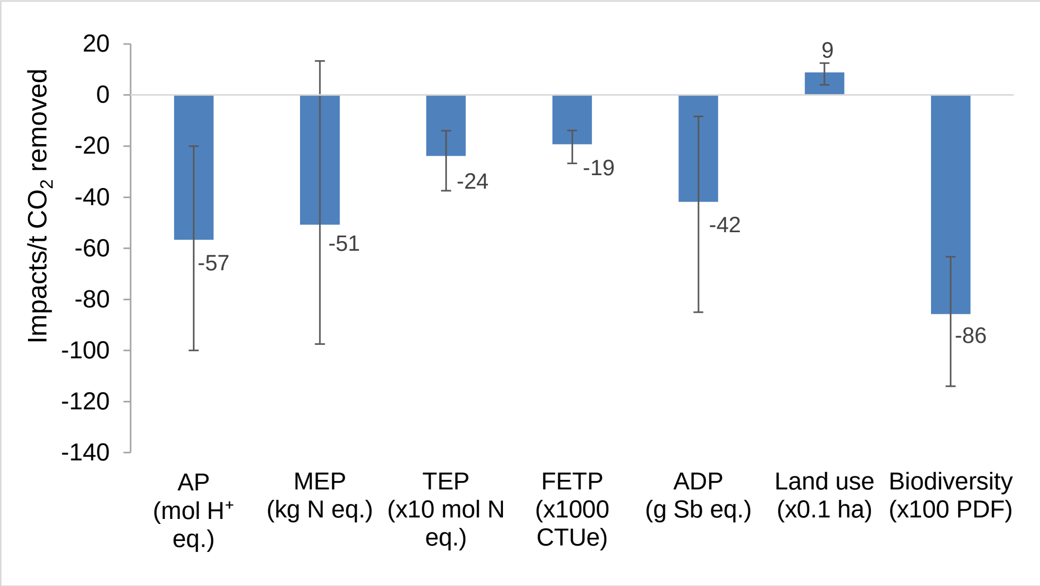
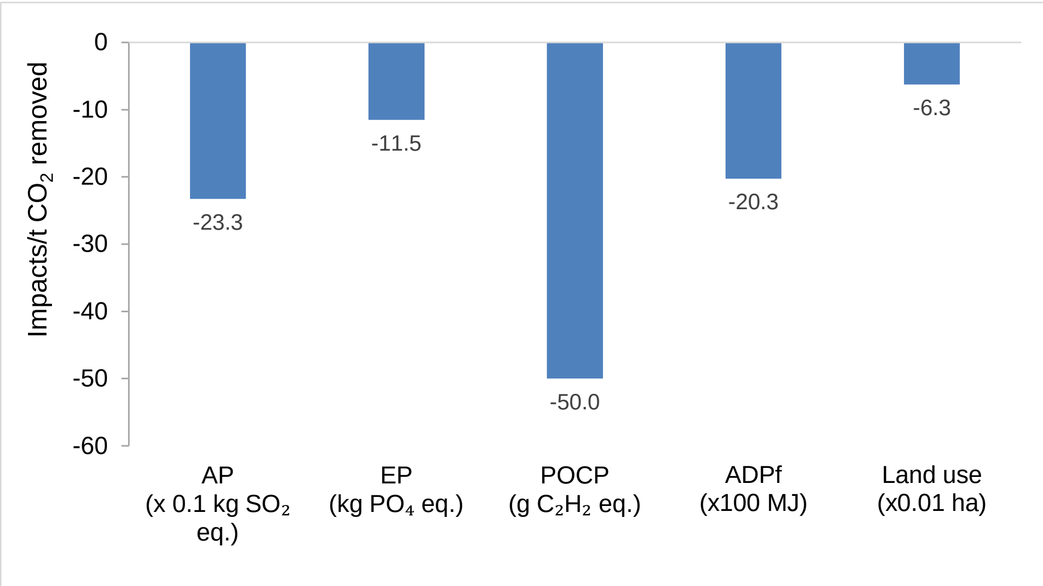


Figure 10 Global warming potential (GWP) of soil carbon sequestration for different improved agricultural practices
[For data and their sources, see Table S4 in the SI. LMC: Land management change]



(a) Soil carbon sequestration due to organic cattle farming (Knudsen et al., 2019)



(b) Soil carbon sequestration due to treatment with microbial phosphate inoculant (Kløverpris et al., 2020).

Figure 11 LCA impacts of soil carbon sequestration
[For the impacts nomenclature, see Figure 7.]

4.5 Environmental impacts of building with biomass

Many LCA studies are available on the built environment which have also considered bio-based materials, but most of these focus on whole buildings, often including within the system boundary the energy use in the building and its end of life (Arehart et al., 2021). Furthermore, most of these studies do not provide the required information on GHG removal and hence cannot be included in this review. As a result, only eight LCA studies on BwB are considered here as summarised in Table 6. Half of these are related to the production of bioconcrete using hemp and *Miscanthus* along with lime and other binder materials. The remaining half have assessed the use of biomass for building frames, flooring and insulation. Focusing on the frames, Malone et al. (2014) have found that traditional structures have lower cradle-to gate GWP in comparison to other frames, such as oriented structural boards and ply wood. Processing has been found a major contributor to the GWP of bamboo flooring (Gu et al., 2019). Unlike bamboo and timber, cultivation is the main GWP hotspot for hemp insulation (Scrucca et al., 2020, Zampori et al., 2013). As indicated in Table 6, only three studies, all on bioconcrete, have assessed other LCA impacts in addition to GWP.

The net GWP for different biomass-based building components, reported in the studies listed in Table 6 and recalculated here to the functional unit of 1 t CO_2 removed, is presented in Figure 12. As can be seen, in all cases considered, the GWP is net-negative but it varies significantly among the building components, depending on additional material inputs and required processing. For example, the GHG emissions from energy and other inputs (such as glue, polish, etc.) used for the production of bamboo flooring are much higher than for the production of timber frame. As a result, the former has the highest GWP (-56 kg CO_2 eq./t CO_2 removed) and the timber frame the lowest (-891 kg CO_2 eq./t CO_2 removed). Similarly, the production of hemp insulation has a significantly higher net GWP (-365 kg CO_2 eq./t CO_2 removed) than the timber frame because of the relatively higher GHG emissions from hemp cultivation and the production of insulation panels.

In the case of bioconcrete, besides the cultivation of hemp and *Miscanthus*, the choice of binder materials (lime, cement, dolomite, blast furnace slag, etc.) and the composition of concrete have significant influence on environmental impacts. For these reasons, the net GWP of bioconcrete varies widely, from -31 to -436 kg CO_2 eq./t CO_2 removed. Similar variation can also be seen in ADP, AP and EP (Figure 13 and Table S6 in the SI) due to the same reasons. Furthermore, unlike other bio-based NET, such as BECCS, biochar and SCS, the use of bioconcrete leads to net-positive impacts in all the assessed categories.

Table 6 Summary of LCA studies on building with biomass

Study (by year)	**Region**	**Biomass**	**Building component**	**Functional unit**	**System boundary**	**LCIA method and impacts assessed[a]**
Ip and Miller (2012)	UK	Hemp	Hemp lime wall	1 m^2 wall	Cradle to gate	GWP
Zampori et al. (2013)	Italy	Hemp	Insulation panel	1 m^2	Cradle to gate	GWP
Malone et al. (2014)	Canada	Timber and	Building frame	Floor area of 193 m^2	Cradle to gate	GWP
Pretot et al. (2014)	France	Hemp	Hemp concrete wall	1 m^2	Cradle to grave	French standard NF P 01-010 (PED, AP, POCP, EP, ADPf)
Arrigoni et al. (2017)	Italy	Hemp	Hemp concrete wall	1 m^2	Cradle to grave	CML (AP, ADPm, ADPf, EP, ODP, POCP, CED)
Gu et al. (2019)	China	Bamboo	Flooring	1 m^3	Cradle to gate	GWP
Scrucca et al. (2020)	France	Hemp	Hurd for insulation and other application	1 kg	Cradle to gate	GWP
Ntimugura et al. (2021)	UK	*Miscanthus*	*Miscanthus* concrete block	1 m^3	Cradle to gate	CML (AP, ADPm, ADPf, EP, FAETP, HTP, MAETP, ODP, POCP, TETP)

[b] For the impacts nomenclature, see Table 2.

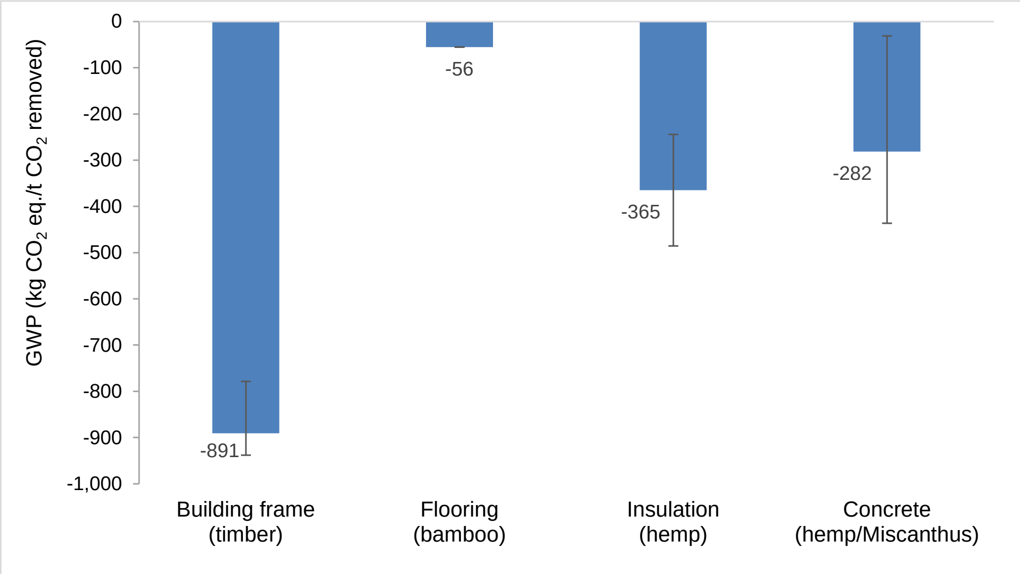


Figure 12 Global warming potential (GWP) of building with biomass for different materials and components

[For data and their sources, see Table S5 in the SI.]

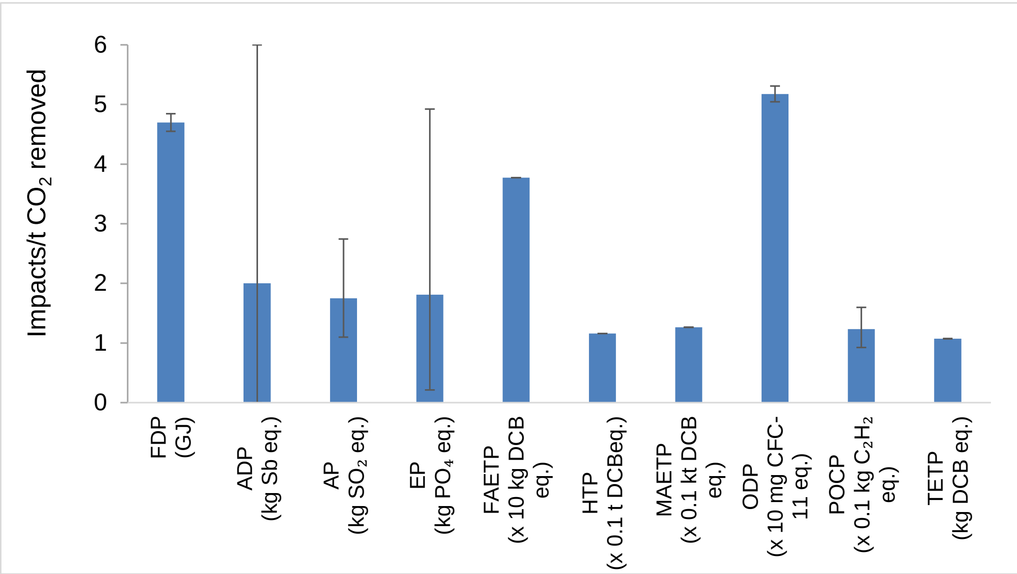


Figure 13 LCA impacts of building with biomass

[For the data and their sources, see Figure S6 in the SI. For the impacts nomenclature see Figure 7.]

4.6 Environmental impacts of DACCS

As can be seen from Table 7, only three LCA studies are available for DACCS, each considering a different carbon capture option: the anionic exchange resin humidity swing (HS) process (van der Giesen et al., 2017), absorption in NaOH (de Jonge et al., 2019) and adsorption on an amine-based sorbent (Deutz and Bardow, 2021). In addition to the different processes, the system boundaries and functional units also differ. For example, van der Giesen et al. (2017) have assessed the LCA impacts of DACCS for electricity from coal and hence used the functional unit of 1 kWh of electricity. Two electricity sources to power the DAC system have been considered in this study: coal and solar PV. The two other studies (de Jonge et al., 2019, Deutz and Bardow, 2021) have defined the functional unit as the mass (kg and t) of carbon captured and stored geologically. In terms of inventory data for the DACCS process, both van der Giesen et al. (2017) and de Jonge et al. (2019) have extrapolated data from laboratory prototypes, while Deutz and Bardow (2021) obtained data from two currently operating industrial plants in Iceland and Switzerland. In addition, Deutz and Bardow (2021) have also considered various scenarios for different sources of electricity and grid mix. For the LCA impacts, de Jonge et al. (2019) have focussed on GWP only, while the remaining two studies assessed a number of LCA impacts (see Table 7).

Table 7 Summary of LCA studies on direct air carbon capture and storage

Study	Country/ Region	DACCS process	Functional unit	System boundary	LCIA method and impacts assessed[a]
van der Giesen et al. (2017)	Not mentioned	Anion exchange resin; humidity-swing (HS-DAC)	1 kWh of coal electricity	Electricity generation (coal-fired and solar PV) combined with HS-DAC (to produce CO_2 ready for storage)	CML 2001 (AP, EP, FAETP, GWP, HTP, MAETP, ODP, POCP, TETP and water depletion)
de Jonge et al. (2019)	The Netherlands	NaOH sorbents	1 t of CO_2 stored	From capture to geological storage	GWP only
Deutz and Bardow (2021)	Iceland and Switzerland	Temperature–vacuum-swing adsorption	1 kg of CO_2 captured	From capture to geological storage	EF2.0 (GWP, EP (freshwater, marine, terrestrial), FAETP, HTP (cancer and non-cancer), IRP, MDP, ODP, POFP, PM, TAP, land use, energy use and water scarcity)

[a] IRP: Ionising radiation potential; PM: Particulate matter; For the nomenclature for the others acronyms, see Table 2.

Figure 14 (and Table S7) shows that all DACCS systems are net removers of CO_2 with the average GWP ranging from -220 to -977 kg CO_2/t CO_2 removed. The largest potential for GHG removal (-800 to -977 kg CO_2/t CO_2 removed) is when renewable energy sources are used to meet the energy demand of DAC operations. The emissions from DACCS plants depend on the GHG intensity of electricity used to operate the system, which in the reviewed studies varies from 23-200 kg CO_2 eq./t CO_2 removed for renewable energy to 80-1000 kg CO_2 eq./t CO_2 removed for electricity from fossil-fuel power plants or grid. The variation in GWP among different processes is also due to the significant variation in the energy demand for these processes, estimated at 378 kWh/t for HS (van der Giesen et al., 2017), 440-550 kwh/t for NaOH (de Jonge et al., 2019) and 700 kWh/t for the amine-based sorbent (Deutz and Bardow, 2021). Both NaOH and amine-based systems also require significant heat for sorbent regeneration, varying from 6282 MJ (de Jonge et al., 2019) to 16,600 MJ (Deutz and Bardow, 2021) per t of CO_2 captured, while HS-DAC utilises ambient-temperature heat for sorbent regeneration.

Figure 15 compares the LCA impacts (other than GWP) of the humidity-swing and the amine-based processes, using different electricity sources. As can be seen in Figure 15a, using coal electricity instead of solar PV in the humidity-swing process results in an increase in all the impacts except ODP, which decreases by 30%. The highest increase (an order of magnitude) is for abiotic resource depletion, eutrophication and marine aquatic eco-toxicity, while the other impacts are also significantly (2-8 times) higher. The LCA results for the amine-based system (Deutz and Bardow, 2021) also show that most environmental impacts are affected by the choice of electricity, with the use of renewable electricity having lower impacts than using fossil energy sources (Figure 15b). Thus, it is clear from these studies that the net removal of CO_2 occurs at the expense of other environmental impacts; however, their increase is highly dependent on the electricity source. The analysis by Deutz and Bardow (2021) suggests that to capture and store 1% of global annual CO_2 emissions using DAC plants would result in

<0.057% increase in other impacts if wind power is used and 0.5% if global grid mix is assumed.

As shown in Table 7, van der Giesen et al. (2017) and Deutz and Bardow (2021) have also assessed other LCA impacts but using different impacts assessment methods, preventing comparison of these two DAC processes (humidity and vacuum swing).

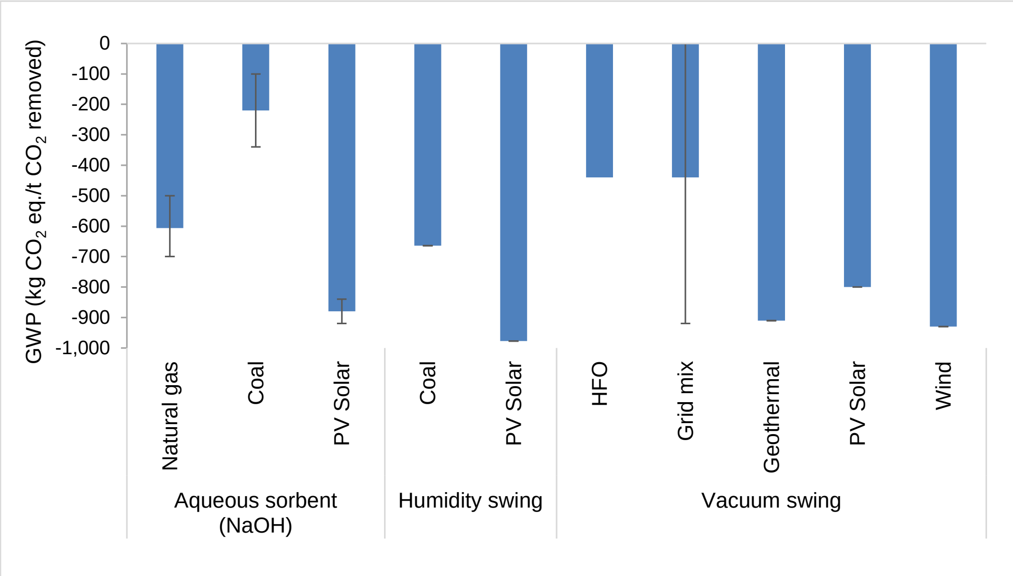


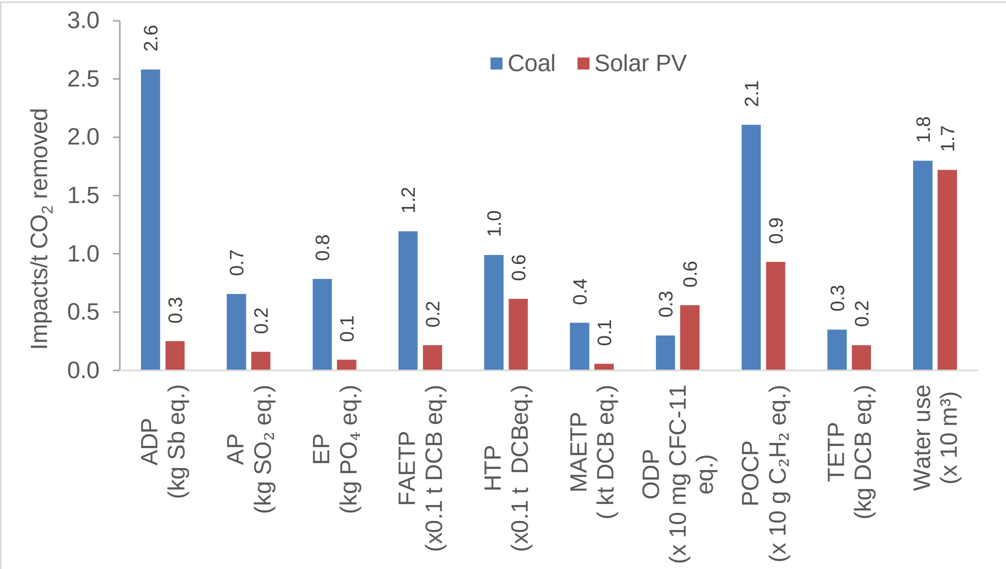
Figure 14 Net GWP of DACCS processes using electricity from different sources
[For data and their sources, see Table S7 in the SI. HFO: heavy fuel oil.]

4.7 Environmental impacts of enhanced weathering

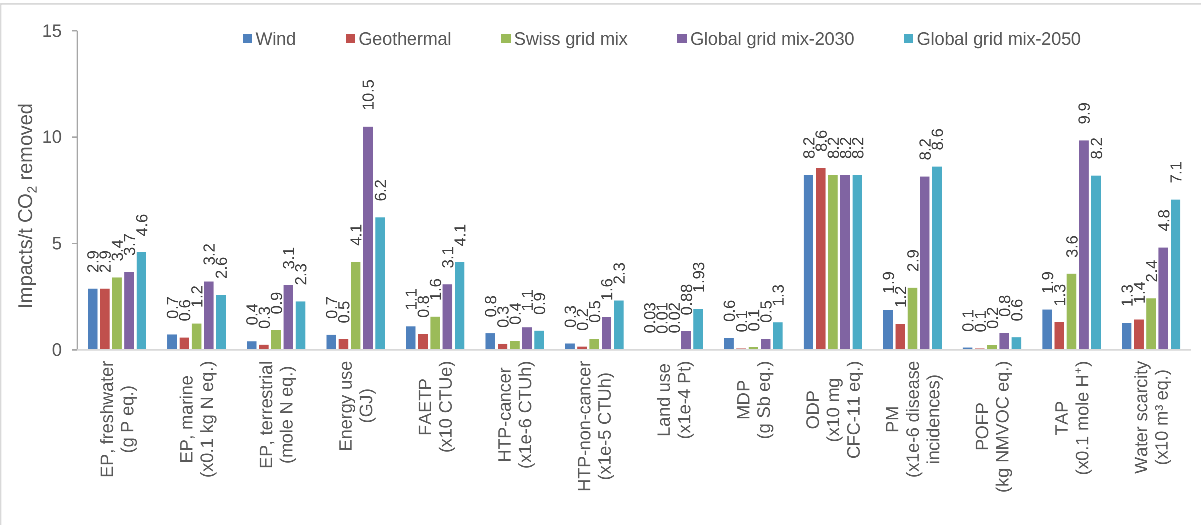
Three LCA studies are available on EW (Table 8). A UK-based study (Renforth, 2012) has reported the energy use and associated CO_2 emissions by EW activities, comprising extraction, comminution, transport and application of rocks. The comminution and material transport are found to be the most energy-intensive processes, accounting for 77–94% of the energy use, which ranges between 224 and 3501 kWh t/CO_2 removed. The study has also found that the energy requirements depend on the rock type, with ultrabasic rocks having much lower energy consumption than basic rocks. Moreover, ultrabasic rocks have higher sequestration potential than the basic (0.3 vs. 0.8 per t of rock) (Renforth, 2012). In a later study, Moosdorf et al. (2014) have conducted a similar assessment at a global level for ultramafic rocks by combining global spatial data sets of potential sources of rocks, transport networks and application areas. The third study by Lefebvre et al. (2019) have assessed a number of life cycle environmental impacts of CO_2 removal on agricultural land of Sao Paulo through EW of basalt rock.

The GWP and energy requirements of EW reported in these studies are compiled in Figure 16 (see also Table S8 in the SI). The net GWP savings vary between 453 and 948 kg CO_2 eq./t CO_2 removed, while the energy requirements are in the range of 0.8-12.6 GJ/t CO_2 removed. There are two main reasons for such large variations in the results: the energy used for rock grinding and the type of rock. The size reduction of rock by pulverising, thereby largely increasing their reactive surface, is the most energy consuming operation in EW. However, the energy requirements for grinding, which are influenced by the mineralogy and crushing practices, are variable. Both Renforth (2012) and Moosdorf et al. (2014) studies consider a broad range of energy requirements for grinding: 10-316 and 167-556 kWh/t rock, respectively. On the other hand, Lefebvre et al. (2019) have estimated the energy requirements for grinding at 5.5 kWh/t rock. In terms of the rocks, two types have been studied for EW: ultramafic, such as dunite and olivine, and mafic, such as basalt. Dunite and olivine have three times higher weathering efficiency than basalt (0.8-1.2 vs. 0.3-0.4 t CO_2/t rock). As a result, EW with ultramafic rocks has lower GWP and energy use per t CO_2 sequestered than mafic rocks. However, the former contain more harmful trace elements (specifically Ni and Cr) than basalt, which can potentially be released into the environment during dissolution. Moreover, basalts are rich in nutrients, such as phosphorus, magnesium and calcium, so their application on croplands could provide considerable additional benefit (Strefler et al., 2018).

In addition to the impacts per t of rock, Lefebvre et al. (2019) have also assessed the impacts per t CO_2 removed, which are summarised in Table 9. The study has found transportation to be the main contributor to the impacts, ranging from 35% for acidification to 88% for abiotic resource depletion; other operations, including grinding, have significantly lower contribution to these impacts. As mentioned earlier, this is largely due to the very low energy consumption assumed for grinding (5.5 kWh/t rock). Therefore, these impacts would significantly increase if a higher energy usage is considered for grinding.



(a) Humidity swing process (van der Giesen et al., 2017)



(b) Vacuum swing process (Deutz and Bardow, 2021)

Figure 15 LCA impacts of DACCS processes using electricity from different sources
[The impacts in (a) estimated using the CML 2001 method and in (b) by the Environmental Footprint method. For the impacts nomenclature, see Figure 7.]

Table 8 Summary of LCA studies on enhanced weathering

Study	Functional unit	Rock	Region	System boundary	LCA method and impacts assessed[a]
Renforth (2012)[b]	1 t of CO_2 removed	Ultramafic rock and basalt	UK	Extraction to field application	GWP and energy use
Moosdorf et al. (2014)[b]	1 t of rock	Ultramafic rock	Global	Extraction to field application	GWP and energy use
Lefebvre et al. (2019)	1 ha of agricultural land; 1 t of CO_2 removed	Basalt	Brazil	Extraction to field application	CML 2001 and USEtox (GWP, ADP, AP, CED, FET, HTP (cancer and non-cancer)

[a] For the impacts nomenclature, see Table 2.
[b] Screening study which provides estimates of GHG emissions and energy use.

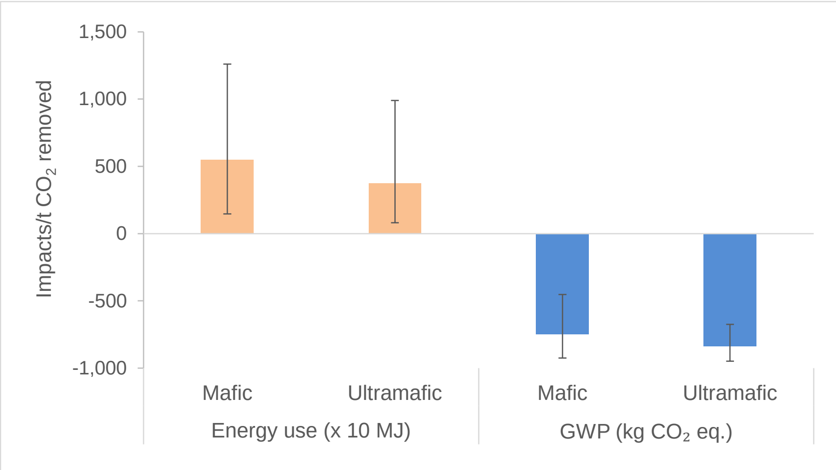


Figure 16 Net energy use and global warming potential of enhanced weathering
[For data and their sources, see Table S8 in the SI.]

Table 9 LCA impacts of enhanced weathering (Lefebvre et al., 2019)

Impact category	Impact per t CO_2 removed	Unit
Abiotic depletion	156	mg Sb eq.
Acidification	552	g SO_2 eq.
Freshwater eco-toxicity	289	CTUe[a]
Human toxicity, cancer	1.99×10^{-6}	CTUh[b]
Human toxicity, non-cancer	1.61×10^{-5}	CTUh[b]

[a] CTUe : comparative toxic unit for aquatic ecotoxicity;
[b] CTUh: Comparative toxic unit for human toxicity

4.8 Environmental impacts of mineral carbonation

As indicated in Table 10, the LCA studies of MC vary in terms of the source of CO_2, the material used for carbonation, the mineralisation process, the functional unit and the system boundaries. The CO_2 sources considered in the available studies include flue gases from power plants, steel mills and waste incinerators. All studies, except for Kirchofer et al. (2012), have accounted for environmental burdens of CO_2 separation from flue gases and its compression. The studies have considered various materials for carbonation, such as serpentinite, wollastonite and olivine rocks, pyroxene-rich tailings, steel slag and other industrial residues. These materials differ in terms of their CO_2 storage potential and energy requirements for pre-treating and activation. For example, Kirchofer et al. (2012) found that cement kiln dust has the highest net sequestration potential, followed by olivine, while serpentinite has the lowest CO_2 storage capacity. Focusing on energy for carbonation, another study (Giannoulakis et al., 2014) has determined that serpentinite has the highest and wollastonite the lowest energy consumption.

Some studies have also compared different carbonation processes. For instance, the study by Nduagu et al. (2012) have found that the Åbo Akademi University (ÅAU) process has almost the same energy intensity but a lower GWP than the National Energy Technology Laboratory (NETL) process. On the other hand, Giannoulakis et al. (2014) report that the current ÅAU process has much higher environmental impacts than the NETL process, but the optimised ÅAU process (with optimised heat exchangers and low solvent losses) would perform better than the NETL system on all the impacts. Ncongwane et al. (2018) have also assessed the ÅAU process alongside four others (the ammonium salts, direct aqueous, pH swing and Lackner's processes), reporting that all have higher CO_2 emissions than the amount sequestered. Among the options, Lackner's process has the highest GWP (due to high heat requirement and the make-up of chemical reagents) and the ÅAU process the lowest. Although in most studies the functional unit is related to the amount of CO_2 mineralised, in three studies (Khoo et al., 2011, Giannoulakis et al., 2014, Ghasemi et al., 2017), it is expressed per MWh of electricity generated. Some studies have applied credits for the use of carbonated material to substitute different construction materials (sand, aggregate or cement), or feed in steel production, as indicated in Table 10 and discussed further below.

Figure 17, which compares the GWP of different rocks and residues used in MC, shows that except pyroxene tailings, all other rocks and residues lead to a net removal of CO_2 (see also Table S9 in the SI). The average net sequestration potential of rocks varies between -445 kg CO_2 eq./t CO_2 removed for serpentinite rocks and -728 kg CO_2 eq./t CO_2 removed for wollastonite rocks. In the case of industrial residues, the highest average removal is for steel slag (-3711 kg CO_2 eq./t CO_2 removed), followed by cement kiln dust (-830 kg CO_2 eq./t CO_2 removed) and fly ash (-500 kg CO_2 eq./t CO_2 removed). However, the net GWP of CO_2 sequestration by steel slag varies from -50 to -6800 kg CO_2 eq./t CO_2 removed. The study by Pan et al. (2016), which assumes that the mineralised slag is used as cement and includes credits for the avoided burdens from cement production, estimates higher savings from MC (-4000 to -6800 kg CO_2 eq./t CO_2 removed), while other studies report significantly lower net removal (-50 to -1070 kg CO_2 eq./t CO_2 removed). The only MC method found to emit more CO_2 that it removes is that with pyroxene minerals (platinum-group metal tailings), with the GWP estimated at 354-17,295 kg CO_2 eq./t of CO_2 removed due to high heat requirements and chemical reagents (Ncongwane et al., 2018).

Out of ten LCA studies on MC, only three (Ghasemi et al., 2017, Giannoulakis et al., 2014, Julcour et al., 2015) have assessed other environmental impacts in addition to GWP. However, the first two have used different impact assessment methods (ReCiPe and CML, respectively), while Julcour et al. (2015) have included only three impacts (ADP, AP and POCP), making comparison of their results difficult.

The LCA impacts reported in Giannoulakis et al. (2014) and Julcour et al. (2015) are summarised in Figure 18a (see also Table S10 in the SI). Giannoulakis et al. (2014) have compared MC by the NETL processes involving wollastonite, olivine and serpentinite rocks with the ÅAU process utilising serpentinite. The findings suggest that the wollastonite-based process has the lowest environmental impacts and the one using serpentinite the highest. The main reason for wollastonite having a clear advantage over serpentinite and olivine is its high reactivity, which avoids the need for chemical additives. However, wollastonite is a relatively scarce mineral, whereas serpentinite is abundant and olivine is also widely available. Figure 18a also shows that for serpentinite, the NETL process has lower impacts than the current ÅAU process. However, as mentioned earlier, if the ÅAU (future) process is optimised to minimise the heat requirements and solvent losses, it would perform better than the NETL process on all environmental impacts. Figure 18a also shows that for olivine, the attrition leaching process assessed in Julcour et al. (2015) has a 30% higher acidification and 120% higher POFP than the NETL process in (Giannoulakis et al., 2014).

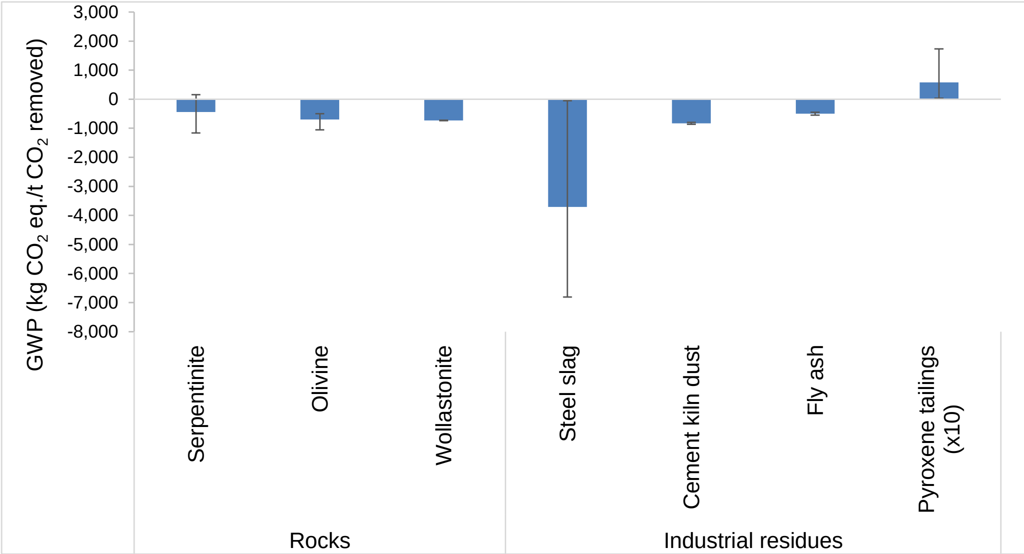


Figure 17 Global warming potential (GWP) of mineral carbonation
[For data and their sources, see Table S9 in the SI.]

The findings in the Ghasemi et al. (2017) study, presented in Figure 18b, suggest that CO_2 removal using the wet-slag route has higher impacts than the slurry route for all impact categories, except for ADP, for which the slurry option has a 33% higher impact. This is due to the wet route having lower carbonate conversion than the slurry one, hence more slag and energy for heating of the slag are required (Ghasemi et al., 2017).

4.9 Comparison of environmental impacts of NETs

This section compares the environmental impacts of different NETs discussed in the preceding sections for the functional unit of 1 t of CO_2 removed. The results for GWP and other impacts are shown in Figure 19 and Figure 20, respectively. As indicated in Figure 19, the net average GWP of all of the eight technologies is negative, ranging from -603 to -1173 kg CO_2 eq./t CO_2 removed. Biochar with an average GWP of -1173 kg CO_2 eq./t CO_2 removed ranks first, followed by SCS (-895 kg CO2 eq./t CO_2), while BwB (-603 kg CO_2 eq./t CO_2) ranks last. The main reason for biochar having the higher GHG avoidance is because, besides sequestering carbon in soil, it also receives the credits for the avoidance of fertilisers, reduction in N_2O emissions, as well as credits for the co-products (syngas and bio-oil). However, as the error bars in Figure 19 shows, the net GWP of biochar varies among different studies between a net-positive impact of 1710 to a net-negative GWP of 3300 kg CO_2 eq./t CO_2 removed, depending upon the feedstock, pyrolysis technology and the assumptions for credits for co-products and co-benefits, as discussed in Section 4.2.

As also indicated by the error bars in Figure 19, the net GWP of MC varies widely, ranging from a net-positive impact of 17,300 to the net removal of 6810 kg CO_2 eq./t CO_2 removed, depending on the material used and the assumptions for crediting the carbonated product, as discussed in Section 4.8. In the case of BwB, the net GWP depends on the additional processes (e.g. hemp cultivation) and other material inputs (such as lime in the case of bioconcrete) required in converting biomass into building materials. Furthermore, unlike bio-based NETs (BECCS, biochar, AR and SCS), MC, DACCS and EW are net consumers of energy; hence, the net GWP of these technologies depends largely on the source of energy used in these systems.

Table 10 Summary of LCA studies on mineral carbonation

Study[a]	Region	Rock	Process	CO_2 source	Functional unit	System boundary	Credits	No. of case studies	LCA method and impacts assessed[b]
Khoo et al. (2011)	Singapore	Ultramafic (serpentinite) rocks	Åbo Akademi University (ÅAU)	Natural gas power plant	1 MWh of electricity	Natural gas power plant, capture of CO_2 from flue gases to the mineralisation	None	4	GWP and energy use
Kirchofer et al. (2012)	USA	Olivine, serpentinite and industrial residues (cement kiln dust, fly ash and steel slag)	-	Outside of the system boundary (assumes input of pure compressed CO_2)	1000 t of CO_2 mineralised	Extraction of minerals to the mineralisation	Aggregate	10	GWP only
Nduagu et al. (2012)	Canada	Serpentinite rock	ÅAU and National Energy Technology Laboratory (NETL)	Coal power plant	1 t of CO_2 mineralised	Capture of CO_2 from flue gases to the mineralisation	Feed (FeOH and $Ca(OH)_2$) for sintering plant	2	GWP only
Giannoulakis et al. (2014)	Europe	Wollastonite, olivine and serpentinite rocks	ÅAU and NETL	Coal and natural gas power plant	1 MWh of electricity	Pulverised coal and natural gas power plant, capture of CO_2 from flue gases to the mineralisation	None	10	ReCiPe (GWP, EP (freshwater, marine, terrestrial), FAETP, FDP, HTP, IRP, MAETP, MDP, POFP, ODP, PMFP, TAP, TETP)
Julcour et al. (2015)	France	Olivine	Attrition leaching	Coal and natural gas power plant	1 MWh of electricity	Coal power plant, capture of CO_2 from flue gases to the mineralisation	None	1	Not mentioned (GWP, AP, POFP, energy use)
Pan et al. (2016)	China	Steel slag	High-gravity carbonation	Steel mill	1 t of slag input	Capture of CO_2 from flue gases to the use of carbonation product	Cement	9	GWP only and ReCiPe end-points
Ghasemi et al. (2017)	Europe	Steel slag	Slurry route and wet-slag route	Natural gas power plant	1 MWh of electricity	Capture of CO_2 from flue gases to the landfilling of carbonation product	None	2	CML (all impacts)
Ncongwane et al. (2018)	South Africa	Pyroxene minerals (platinum group metal tailings)	Ammonium salts, the direct aqueous, ÅAU, pH swing and Lackner	SASOL synfuels	1 t of CO_2 mineralised	From CO_2 capture to mineralisation	None	5	GWP only

Study[a]	Region	Rock	Process	CO_2 source	Functional unit	System boundary	Credits	No. of case studies	LCA method and impacts assessed[b]
Ostovari et al. (2020)	Europe	Olivine, serpentinite and industrial residues (steel slag)	Continuous stirred tank reactor (CSTR), rotary packed bed (RPB), ÅAU and Nottingham pathway	Steel mill	1 t of stored CO_2	From CO_2 capture to the use of carbonation product	Partial substitution of cement in blended cement	7	GWP only
Khoo et al. (2021)	Singapore	Ultramafic (serpentinite) mine tailings	-	Incineration plant	1 t of CO_2 mineralised	Capture of CO_2 from flue gases to the mineralisation	Sand	2	GWP only

[a] The study by Di Maria et al. (2020) does not provide the required information to convert the results to the functional unit of 1 t of CO_2; hence, it is excluded from the analysis.
[b] For the impacts nomenclature, see Table 2 and Table 3.

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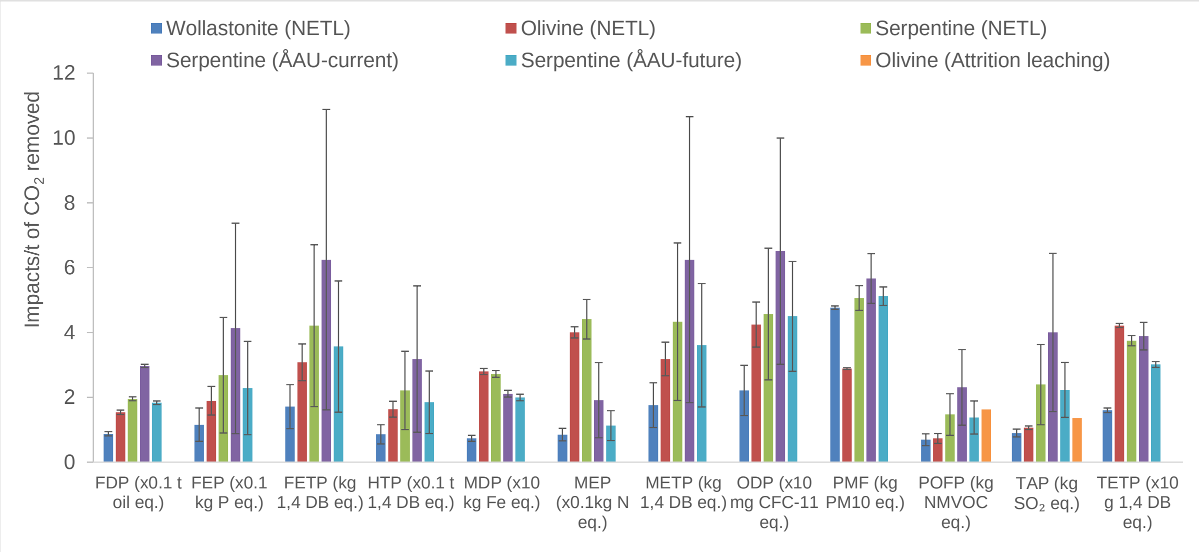
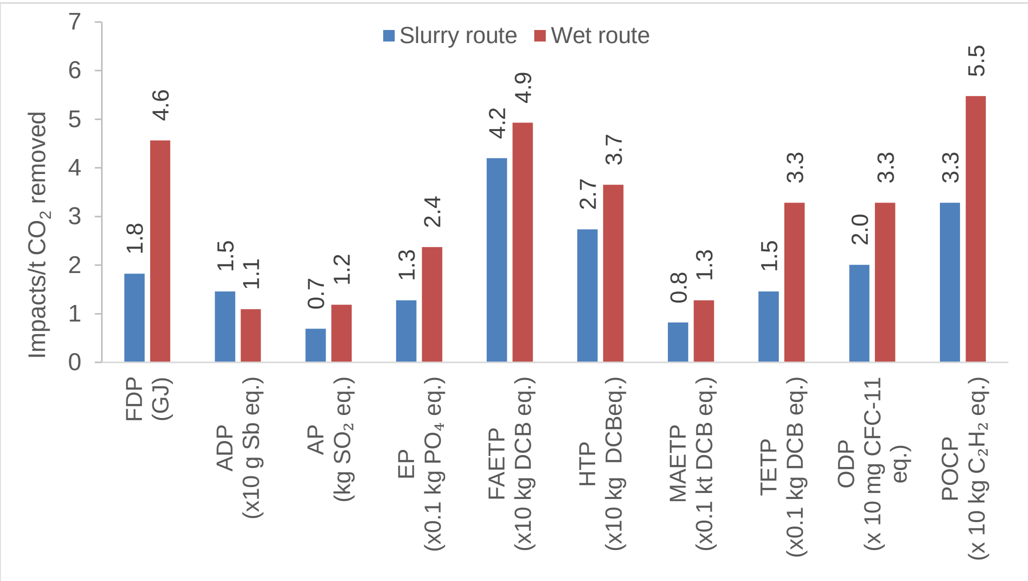


a) Mineral rocks (Giannoulakis et al., 2014, Julcour et al., 2015)
[For data, see Table S10 in the SI. Comparison of ADP is not possible as they are reported in different units.]



b) Steel slag (Ghasemi et al., 2017)

Figure 18 LCA impacts of mineral carbonation
[For impacts nomenclature see Figure 7.]

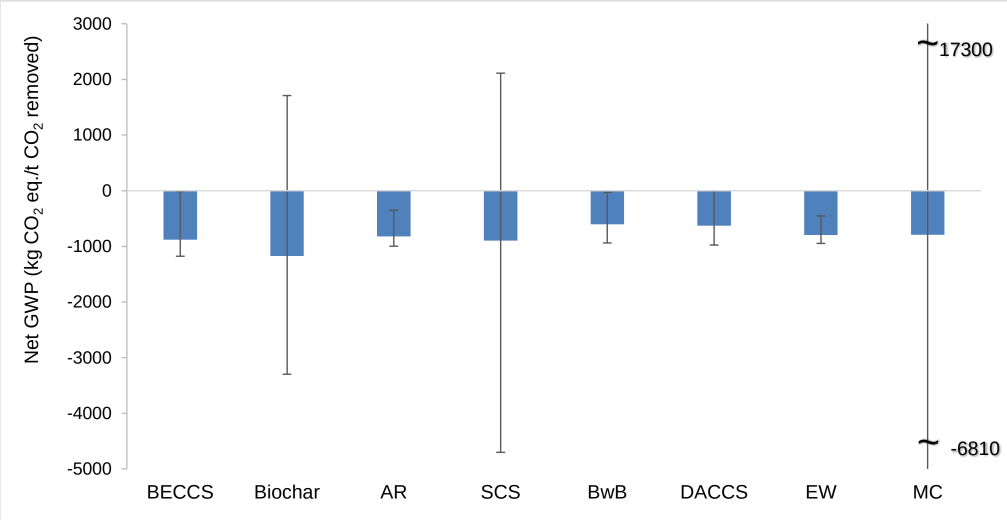


Figure 19 Comparison of global warming potential (GWP) of different NETs

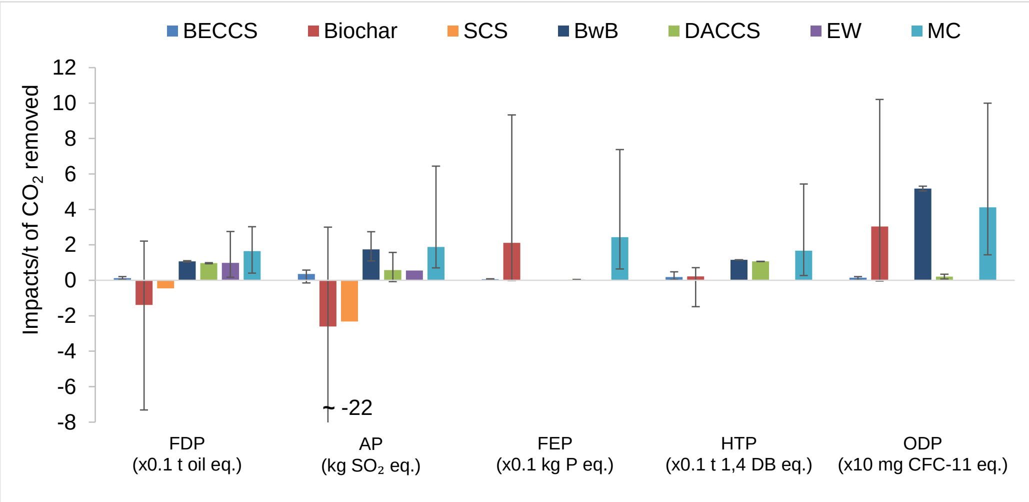


Figure 20 Comparison of selective LCA impacts of different NETs
[BECCS: Bioenergy with carbon capture and storage; SCS: Soil carbon sequestration; BwB: Building with biomass; DACCS: Direct air carbon capture and storage; EW: Enhanced weathering; MC: Mineral carbonation. For the impacts nomenclature see Figure 7.]

Since the published LCA studies have used different life cycle impact assessment (LCIA) methods (CML, ReCiPe, EF, ILCD, Impact 2002+, etc.), the results are not directly comparable for all the impacts. Therefore, the focus here is on the impacts calculated in most LCIA methods using the same methodology. These are: fossil depletion/energy use, acidification, freshwater eutrophication, human toxicity and ozone depletion.

As can be seen in Figure 20, these impacts for bio-based NETs (BECCS, biochar and SCS) are on average much lower than for the others (DACCS, EW and MC). Biochar, which receives the credits for the co-products and co-benefits, has net savings in fossil depletion, acidification and human toxicity. SCS also has net savings for fossil depletion and acidification. For non-bio NETs, as well as BwB, the net impacts are positive, with MC having the highest values. On the other hand, biochar has higher freshwater eutrophication and ozone depletion than BECCS, DACCS and EW. Although all the three non-bio based NETs are net consumers of energy, the different studies have considered different sources of energy, which has the significant influence on LCA results, as can be seen from the large ranges of values shown in the error bars. Overall, the review of LCA studies shows that the impacts of non-bio based

NET can be reduced if the energy demands are met through the use of renewable energy sources instead of fossil energy.

While biochar has the lowest environmental impacts, it also has higher energy penalty (Azzi et al., 2019) as these systems produce less heat and power in comparison to biomass combustion as part of the biomass energy remains in biochar. That loss of energy would then need to be produced using other sources. This is one of the key trade-offs between bioenergy and carbon sequestration via biochar which is often not considered in LCA studies.

5 Methodological differences, challenges and recommendations

This review has highlighted several methodological and other related issues in LCA studies, which make it difficult to compare the environmental performance of different NETs. These issues are discussed below, along with recommendations on how to address them.

5.1 Goal and scope

As discussed above, the LCA studies of NETs vary in terms of their scope, system boundaries and functional units. This is partly due to some inherent differences among the NETs in terms of their purpose, which affect their goal and scope. For example, in the case of BECCS, its main purpose is to produce energy; hence, in most studies the functional unit is defined in terms of energy (MWh, kWh, MJ or GJ). On the other hand, the LCA studies on multi-functional systems, such as biochar, have considered different functional units (t of biochar produced, t of feedstock used, t of crops produced, or ha/y of land treated). However, as the main function of NETs is to remove GHG from the atmosphere, it is important that all LCA studies on NETs also consider the unit mass of CO_2 eq. removed as a functional unit to enable their comparison.

Furthermore, there are inconsistences among studies in terms of system boundaries. For example, some (Carpentieri et al., 2005, van der Giesen et al., 2017) have excluded certain steps, such as transport of compressed CO_2 and its geological storage. Furthermore, several studies on biochar, assessing its influence on agricultural products, have included the production of crops utilising biochar as a fertiliser within the system boundaries (Mohammadi et al., 2016a, 2016b, Sparrevik et al., 2014, Uusitalo and Leino, 2019). On the other hand, most LCA studies of biochar and other bio-based NETs exclude LUC. In addition, the consideration of co-products and co-benefits (as discussed in the next section) is not consistent among the studies. All of these issues could significantly affect the overall LCA impacts of NETs.

5.2 Consideration for co-products and co-benefits

Some NETs, such as biochar and MC, can also produce co-products and/or have additional benefits. Therefore, for these technologies, the main issues which could affect their environmental performance, are the allocation of burdens between different functions of the systems (co-products) and the method for the consideration of co-benefits. In the case of biochar, most of the reviewed studies have used the avoided burden approach, in which the credits have been applied for the equivalent amount of the co-products (syngas and bio-oil) that would be replaced (natural gas, crude oil, electricity, etc.). Although the avoided burdens approach is commonly used in LCA, its application in biochar systems can be problematic. This is particularly the case when the pyrolysis process is designed (by setting temperature and other operating conditions) to produce more syngas and bio-oil and less biochar, whereby these systems would receive very high credits for the avoided fossil-based products. Since in this case biochar is not the main product and is not driving the existence of the process, the avoided burdens approach is inappropriate as the net impacts would be negative, suggesting the system is acting as a sink, which could be misleading (Tanzer and Ramírez, 2019, Müller et al., 2020). In such cases, the use of an allocation approach based on underlying physical or other non-physical relationships (ISO, 2006) would be more appropriate. Although allocation is theoretically capable of a fair assignment of the benefit to all functions, the distribution would depend on the allocation basis (carbon-content, mass, energy or the economic value). Mass allocation does not seem to be applicable in all cases; for example, if the co-products are heat and electricity, mass allocation cannot be used. Carbon content, energy or the economic value can also be applied for allocation. However, further case studies are needed to assess the suitability of these allocation criteria for biochar systems.

Some of the NETs have additional benefits which can reduce GHG emissions indirectly, reduce other environmental burdens or improve ecosystem services (Azzi et al., 2021a, Blanco-Canqui, 2021). For instance, as mentioned earlier, the application of biochar to agricultural fields can provide key nutrients and hence reduce the demand for fertilisers; it can also reduce N_2O emissions. One third of the reviewed studies on biochar have not considered the co-product credits and 20% have excluded the benefits of soil N_2O reductions. The remaining studies have used different assumptions and approaches for estimating the avoided fertiliser use and N_2O reduction. For example, Hammond et al. (2011) and Rajabi Hamedani et al. (2019) assume that biochar application of 30 t/ha for winter wheat crops can decrease N, P and K, fertilisers by 10%, 5%, 5%, respectively, and N_2O emissions by 25%. On the other hand, Oldfield et al. (2018), citing the results from field trials in Europe (which found that biochar application does not increase the availability of N and K), only credit the biochar system for the P fertiliser. Furthermore, the application of biochar to agricultural fields can improve ecosystem services, such as increase in water and nutrient retention and reduction in soil erosion through improved soil structure (Azzi et al., 2021a, Blanco-Canqui, 2021). Since these additional benefits depend on the type of biochar, as well as the conditions of the applied soil, crop type and climate, these benefits are highly uncertain. Therefore, it is important that future studies consider these factors while estimating those benefits.

Similarly, in the case of MC, the assumptions on the use of the carbonated product as a construction material, as well as system credits, vary across studies. Half of the reviewed LCA studies have not considered any use of the carbonated product, while the other half assume it as a replacement for different materials, such as cement (Ostovari et al., 2020, Pan et al., 2016), feed for sinter plant (Nduagu et al., 2012), sand (Khoo et al., 2011) and aggregates (Kirchofer et al., 2012) and hence credit the system accordingly. Since the production of some of the materials (e.g. cement) has much higher impacts than others (e.g. sand and aggregates), such choices – and the related credits – would have a very significant influence on the overall impacts of the system.

5.3 CO_2 storage, reversibility and the effect of delayed emissions

The permanency of the stored/sequestered carbon depends on the method of storage/sequestration. For example, the CO_2 stored via MC is irreversible. Similarly, the CO_2 stored in properly designed and managed geological storage sites is largely immobilised by various trapping mechanisms and could be retained for thousands of years (IPCC, 2005). However, in the case of ocean storage, the retention of CO_2 depends on the depth of the injection. It is estimated that up to 35% of captured CO_2 could be released after 100 years and up to 70% in 500 years for injection at a depth of 1000 m (IPCC, 2005). Similarly, most of the CO_2 sequestered or stored in the terrestrial biosphere through biochar incorporation into soil or soil carbon sequestration is non-permanent. Furthermore, other bio-based NETs, such as AR and BwB, could only potentially delay CO_2 emissions stored over a certain period of time. Similarly, CCU in chemical products offers only temporary carbon storage. The effect of non-permanent storage and delayed emissions are not often considered in LCA studies. Although various approaches have been suggested to deal with temporary storage and delayed emissions (Brandão et al., 2013, Jørgensen et al., 2015, Beloin-Saint-Pierre et al., 2020), there is no scientific consensus on how to account for these issues in LCA. Some methods, such as PAS 2050 (BSI, 2011) and the International Reference Life Cycle Data System (ILCD) Handbook (EC-JRC, 2010), allow accounting of the benefits of temporary carbon storage and delayed emission, while other carbon footprinting standards, such as ISO 14067 (ISO, 2013), GHG protocol (WRI and WBCSD, 2011) do not allow the inclusion of such benefits.

Furthermore, carbon neutrality is assumed for biogenic emissions in all of the reviewed LCA studies on bio-based NETs, except in Oreggioni et al. (2017), who have considered a dynamic approach and used time-dependent characterisation factors for biogenic carbon. Since the traditional LCA approach is based on static modelling, biogenic CO_2 emissions are considered as neutral, based on the assumption that the same amount of CO_2 that is released from biogenic sources is absorbed during the regrowth of biomass. However, some feedstocks, such as lignocellulosis from forestry, can take several years to recapture the emitted CO_2; hence, it is important to account for the time delay factor through dynamic modelling. Similar to temporary carbon storage and delayed emissions, there is also a need to develop scientific consensus on its accounting method.

5.4 Data availability and quality

Since most of the NETs are still at the development stage, the actual process-specific data are not available. Often, the inventory inputs and outputs are estimated based on laboratory experiments, stoichiometric reactions, mass and energy balances and process modelling. In addition, various assumptions are made while estimating the data through these methods. As a result, the uncertainty in data could be high, but most studies do not report data quality transparently. As transparency is one of the most important aspects in LCA, it is important that data estimation methods and assumptions are detailed in studies. There are several data quality assessment methods and frameworks available (e.g. Edelen and Ingwersen (2016); European Commission (2018)), which can be used for assessing and communicating the quality of life cycle inventory data. Furthermore, the influence of data variability and assumptions should be evaluated through sensitivity and uncertainty analyses to identify key variables affecting the results. The uncertainty analysis is particularly important for comparative studies to determine if the differences between the alternative options, processes or technologies are significant.

5.5 Impact assessment methods

Since the main driver for NETs is the removal of CO_2, the LCA studies usually focus only on GWP. However, like all others, these technologies will inevitably cause other impacts, such as depletion of resources, acidification, eutrophication, human and eco-toxicity, associated with the use of energy, materials, land and water. These impacts must also be evaluated to avoid shifting the problem from climate change to other environmental issues. Moreover, those LCA studies which do consider other environmental impacts, tend to use different LCIA methods, making comparisons between studies impossible. This is not helped by the fact that there is no consensus in the LCA community and relevant organisations on which LCIA method should be used. For example, the published studies on NETs have used ReCiPe, CML and Impact 2000+, while the Joint Research Centre of European Commission and the US EPA recommend the Environmental Footprint (EF) 3.0 (Zampori and Pant, 2019) and TRACI 2.1 methods (Bare, 2011), respectively. In recent years, significant efforts have been made on standardising the product category rules and environmental product declarations which recommend using the EF method (Zampori and Pant, 2019). As the latter method is gaining traction in Europe, it is therefore suggested that practitioners report the impacts using the EF method, at least in Europe, in addition to the method of their choice, to facilitate comparisons between different NETs and help decision makers make informed choices. Furthermore, studies on bio-based NETs should also include other relevant impacts which are typically not assessed in LCA, such as water footprint, biodiversity and land-use change (LUC). Direct LUC, which results from the direct transformation of previously uncultivated areas (such as grasslands and forests) into croplands for the biomass production, is relatively easy to calculate if the information on carbon stocks in soils and above-ground biomass is available for both before and after change (Ben Aoun and Gabrielle, 2017). However, estimating indirect LUC, which occurs when additional demand for biomass triggers displacement of food or feed crop production to new land areas previously not used for cultivation, is complex and highly uncertain and thus requires further research on developing credible models (De Rosa et al., 2016, Ben Aoun and Gabrielle, 2017).

6 Conclusions

It is predicted that the large scale deployment of NETs would be required to achieve net zero targets and to limit the increase in the average global temperature below 1.5°C. However, as most of these technologies are at an early stage of development, it is important to understand their potential environmental benefits along with any negative consequences to facilitate development of robust future policies and safe deployment of NETs.

It is evident from this review that the estimates of GHG removal potentials of NETs vary widely among the studies. These differences are partly due to technological differences and partly due to various assumptions and methodological choices. Despite this, it is apparent that all NETs reviewed in this study can have – on average – net-negative GHG emissions. In terms of GHG emissions per tonne of CO_2 removed, biochar as soil amendment performs best, followed by soil carbon sequestration, while building with biomass ranks last. This is due to biochar being a net producer of energy, while the production of biobased building materials requires energy and other inputs. DACCS, enhanced weathering and mineral carbonation also fall in the latter category, resulting in higher impacts compared to biochar (and BECCS, which is also a net energy producer). Furthermore, the biochar system benefits from the avoided use of fertilisers, reduction of N_2O emissions and co-production of syngas and bio oil. Soil carbon sequestration trails closely behind biochar due to additional benefits of improved agricultural practices, such as lower fertiliser needs and, in some cases, higher yield.

The LCA studies also show that the use of energy in DACCS, EW and MC leads to higher fossil depletion, acidification and human toxicity. These impacts can be reduced if the energy demands for these NETs are met through the use of renewables instead of fossil-fuel energy. Biochar is the only NET reviewed here that has net savings for these impacts, again due to the credits for the above-mentioned co-products and co-benefits.

Although some studies have considered impacts other than just GWP, the comparison of different NETs on these impacts is not possible due to different impact assessment methods used. In the absence of consensus on a particular method, it is recommended that studies use multiple methods to facilitate comparisons of NETs.

Furthermore, the review has highlighted several methodological and other related issues in LCA studies on NETs, which make it difficult to compare their environmental performance. Since the main function of NETs is remove GHG from the atmosphere, it is recommended that all LCA studies on NETs should also consider the unit mass (kg or t) of CO_2 eq. removed as the functional unit. In addition, there is a need to develop a consistent methodology for consideration of co-benefits and co-products, especially for biochar-based systems. Furthermore, there is a need to develop scientific consensus on consideration of non-permanent storage and delayed CO_2 emissions, as well as carbon neutrality for biogenic emissions from forestry feedstocks, to account for the time delay in assessing the performance of NETs.

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Environmental sustainability of greenhouse gas removal technologies: A review

Harish Kumar Jeswani, Djasmine Mastisya Saharudin and Adisa Azapagic

Department of Chemical Engineering, The University of Manchester, Manchester M13 9PL, UK

Supplementary Information

Table S1 Global warming potential (GWP) of different bioenergy with carbon capture and storage (BECCS) options

Technology	Study	Net GWP (kg CO_2 eq./t CO_2 removed)
Biomass power	Pour et al. (2018)	-1106
		-860
	Yang et al. (2019)	-912
	Average	*-959*
	Minimum	*-1106*
	Maximum	*-860*
Biomass gasification power (BGP) /Integrated biomass gasification combined cycle (IBGCC)	Spath and Mann (2004)	-894
	Carpentieri et al. (2005)	-1000
	Jana and De (2016)	-994
	Oreggioni et al. (2017)	-651
		-629
	Zang et al. (2020)	-812
		-883
		-796
		-864
	Average	*-830*
	Minimum	*-1000*
	Maximum	*-629*
Combined heat and power (CHP)	Gibon et al. (2017)	-979
		-969
	Average	*-974*
	Minimum	*-979*
	Maximum	*-969*
Landfill gas (LFG) electricity	Pour et al. (2018)	-12
Bioethanol	Laude et al. (2011)	-878
		-754
	Jana and De (2016)	-942
	Lask et al. (2021)	-1176
		-1170
		-1158
	Average	*-1013*
	Minimum	*-1176*
	Maximum	*-754*
Bio-hydrogen	Susmozas et al. (2016)	-887

Table S2 Global warming potential (GWP) of biochar as soil amendment for different feedstocks

Feedstock	Study	Net GWP (kg CO_2 eq./t CO_2 removed)
Woody biomass	Rajabi Hamedani et al. (2019)	-950
	Rajabi Hamedani et al. (2019)	-1818
	Hammond et al. (2011)	-2576
		-2576
		-2576
		-2424
	Lu and El Hanandeh (2019)	-1859
		-2191
		-2508
		-2726
		-2958
		-3167
		-3298
	Average	*-2433*
	Minimum	*-950*
	Maximum	*-3298*
Miscanthus/ Switchgrass	Bartocci et al. (2016)	-2003
	Hammond et al. (2011)	-2576
	Brassard et al. (2018)	-1835
		-1334
	Roberts et al. (2010)	-829
		68
	Average	*-1418*
	Minimum	*68*
	Maximum	*-2576*
Forestry residue	Oldfield et al. (2018)	-1522
	Hammond et al. (2011)	-2652
		-2803
	Field et al. (2013)	-1855
	Muñoz et al. (2017)	-1159
		-1169
		-1190
	Azzi et al. (2019)	-850
		-1500
		-1885
	Papageorgiou et al. (2021)	-673
	Papageorgiou et al. (2021)	-770
	Cheng et al. (2020)	-1430
		-1435
		-1377
	Sahoo et al. (2021)	-773
		-932
		-594
		-906
		-722
		-944
		-588
		-882
		-583
		-854
	Average	*-1202*
	Minimum	*-583*
	Maximum	*-2803*

Agricultural residue	Thers et al. (2019)	-1191
	Thers et al. (2019)	-1228
	Llorach-Massana et al. (2017)	1600
		1280
		1013
		16
		-111
		-211
		-368
		-445
		-503
	Hammond et al. (2011)	-1894
		-1894
		-1894
	Muñoz et al. (2017)	-1126
		-1155
		-1173
	Clare et al. (2015)	-1781
	Ji et al. (2018)	-681
	Robb and Dargusch (2018)	-1818
	Robb and Dargusch (2018)	-753
	Mohammadi et al. (2016)	-815
		-905
	Roberts et al. (2010)	-1470
		-1349
		-1545
	Cheng et al. (2020)	-1186
		-1109
		-1243
	Yang et al. (2021)	-1537
	Average	*-849*
	Minimum	*1600*
	Maximum	*-1894*
Manure/ Sewage sludge	Rajabi Hamedani et al. (2019)	-476
	Kreidenweis et al. (2021)	-734
	Cao and Pawłowski (2013)	-1462
	Cheng et al. (2020)	699
		524
		481
	Mohammadi et al. (2019a)	-998
		-838
	Mohammadi et al. (2019b)	-1336
	Mohammadi et al. (2019c)	-1697
		-1070
		1709
	Average	*-433*
	Minimum	*1709*
	Maximum	*-1697*

Table S3 Other LCA impacts of biochar as soil amendment for different feedstocks[a]

Feedstock	FDP[b] (kg oil eq.)	AP[b] (kg SO_2 eq.)	EP[b] (g PO_4 eq.)	FEP[b] (g P eq.)	HTP[b] (kg 1,4 DCB eq.)	ODP[b] (mg CFC-11 eq.)
Woody biomass	-107.85	3.00	1,056.25		32.50	10.77
	-149.71	-0.29	166.93		-3.70	-
	-52.70	-1.03	102.00		-46.61	-
	-147.59	-1.88	8.47		-90.19	-
	-256.42	-2.73	-44.09		-127.19	-
	-367.80	-3.49	-93.18		-148.78	-
	-473.37	-4.02	-133.08		-145.70	-
	-550.39	-4.17	-89.94		44.28	-
	-587.72	0.40	1,209.09			-
Average	*-299.29*	*-1.58*	*242.49*		*-60.67*	*10.77*
Minimum	*-587.72*	*-4.17*	*-133.08*		*-148.78*	*10.77*
Maximum	*-52.70*	*3.00*	*1,209.09*		*44.28*	*10.77*
Perennial grasses	57.80	-	-	-	-	-
	47.66	-	-	-	-	-
Average	*52.73*	-	-	-	-	-
Minimum	*47.66*	-	-	-	-	-
Maximum	*57.80*	-	-	-	-	-
Forestry residue	-18.57	-3.30	-126.87	-1.26	-1.61	60.00
	-22.30	-	-	-1.43	-1.74	40.00
	-30.09	-	-	-1.74	-2.04	-
	221.25	-	-	933.33	-	-
	189.96	-	-	766.67	-	-
Average	*68.05*	*-3.30*	*-126.87*	*339.11*	*-1.80*	*50.00*
Minimum	*-30.09*	*-3.30*	*-126.87*	*-1.74*	*-2.04*	*40.00*
Maximum	*221.25*	*-3.30*	*-126.87*	*933.33*	*-1.61*	*60.00*
Agricultural residue	-45.30	-22.36	-238.37	-0.78	-1.17	-
	-33.42	-	-	-1.22	-1.57	-
	-55.35	-	-	-1.30	-1.61	-
	-6.42	-	-	-	-0.02	-
	-17.20	-	-	-	-	-
	-18.64	-	-	-	-	-
Average	*-29.39*	*-22.36*	*-238.37*	*-1.10*	*-1.09*	-
Minimum	*-55.35*	*-22.36*	*-238.37*	*-1.30*	*-1.61*	-
Maximum	*-6.42*	*-22.36*	*-238.37*	*-0.78*	*-0.02*	-
Manure/ sewage sludge	-25.13	0.67	204.08	-	71.05	102.04
	46.92	0.00	-0.85	-	-0.44	-0.35
	-257.06	0.00	0.94	-	0.07	0.11
	-731.36	0.02	3.19	-	0.16	0.10
	-498.54	-	-	-		-
	-16.88	-	-	-		-
Average	*-247.01*	*0.18*	*51.84*	-	*17.71*	*25.48*
Minimum	*-731.36*	*0.00*	*-0.85*	-	*-0.44*	*-0.35*
Maximum	*46.92*	*0.67*	*204.08*	-	*71.05*	*102.04*

[a] Source: Roberts et al. (2010), Cao and Pawłowski (2013), Peters et al. (2015), Muñoz et al. (2017), Brassard et al. (2018), Oldfield et al. (2018), Lu and El Hanandeh (2019), Mohammadi et al. (2019a), (2019b, 2019c), Rajabi Hamedani et al. (2019), Cheng et al. (2020), Papageorgiou et al. (2021) and Yang et al. (2021).

[b] FDP: Fossil depletion potential; AP: Acidification potential; EP: Eutrophication potential; FEP: Freshwater eutrophication potential; HTP: Human toxicity potential; ODP: Ozone depletion potential.

Table S4 Global warming potential (GWP) of soil carbon sequestration

Improved agricultural practices	Study	Net GWP (kg CO_2 eq./t CO_2 removed)
Conservation tillage	Archer and Halvorson (2010)	-1191
		-1038
		-1054
	Holka (2020)	-315
		212
	Holka and Bieńkowski (2020)	-950
		-1152
	Rahman et al. (2021)	-1043
		-1056
	Average	*-843*
	Minimum	*212*
	Maximum	*-1191*
Cover crops	Joensuu et al. (2021)	-807
		-515
		-851
		-646
	Average	*-705*
	Minimum	*-515*
	Maximum	*--851*
Organic farming	Aguilera et al. (2015)	-1085
		-212
		-4700
		- 309
		-825
		-1369
	Average	*-1417*
	Minimum	*-212*
	Maximum	*-4700*
Treatment with microbial inoculant	Kløverpris et al. (2020)	-2174
Combination of land management change practices	Fiore et al. (2018)	-836
		-713
		-849
	Alam et al. (2019)	-979
	Archer and Halvorson (2010)	-1045
		-1112
	Matsuura et al. (2018)	- 889
		-647
	Average	*-884*
	Minimum	*-647*
	Maximum	*-1112*
Grazing/feed changes	Stanley et al. (2018)	-783
	Knudsen et al. (2019)	-1500
		-2600
		-1833
	Sabia et al. (2020)	2000
		2111
	Arca et al. (2021)	-564
	Average	*-453*
	Minimum	*2111*
	Maximum	*-2600*

Table S5 Global warming potential (GWP) of building with biomass

Building material	Study	Net GWP (kg CO_2 eq./t CO_2 removed)
Building frame (timber)	Malone et al. (2014)	-890
		-899
		-890
		-779
		-938
		-929
		-918
		-886
	Average	*-891*
	Minimum	*-779*
	Maximum	*-938*
Flooring (bamboo)	Gu et al. (2019)	-56
Insulation (hemp)	Zampori et al. (2013)	-485
	Scrucca et al. (2020)	-244
	Average	-365
	Minimum	-244
	Maximum	-485
Concrete (hemp/Miscanthus)	Ip and Miller (2012)	-436
	Pretot et al. (2014)	-31
	Arrigoni et al. (2017)	-351
	Ntimugura et al. (2021)	-308
	Average	*-282*
	Minimum	*-31*
	Maximum	*-436*

Table S6 LCA impacts of bioconcrete

Impacts[a]	Pretot et al. (2014)	Arrigoni et al. (2017)	Ntimugura et al. (2021)
FDP (GJ)	-	4.85	4.55
ADP (kg Sb eq.)	6.00	2.36E-05	3.42E-04
AP (kg SO_2 eq.)	2.75	1.10	1.41
EP (kg PO_4 eq.)	4.92	0.21	0.29
FAETP (x 10 kg DCB eq.)	-	-	3.77
HTP (x 0.1 t DCBeq.)	-	-	1.16
MAETP (x 0.1 kt DCB eq.)	-	-	1.26
ODP (x 10 mg CFC-11 eq.)	-	5.31	5.04
POCP (x 0.1 kg C_2H_2 eq.)	1.60	0.92	1.18
TETP (kg DCB eq.)	-	-	1.07

[a] FDP: Fossil depletion potential; ADP: Abiotic depletion potential; AP: Acidification potential; EP: Eutrophication potential; FETP: Freshwater aquatic ecotoxicity potential; HTP: Human toxicity potential; MAETP: Marine aquatic ecotoxicity potential; ODP: Ozone depletion potential; POFP: Photochemical oxidant formation potential; TETP: Terrestrial ecotoxicity potential.

Table S7 Global warming potential (GWP) of direct air carbon capture and storage

Technology	Study	Source of electricity	Net GWP (kg CO_2 eq./t CO_2 removed)
Aqueous sorbent (NaOH)	van der Giesen et al. (2017)	Natural gas	-620
			-700
			-500
		Average	*-607*
		Minimum	*-500*
		Maximum	*-700*
		Coal	-340
			-100
		Average	*-220*
		Minimum	*-100*
		Maximum	*-340*
		PV solar	-840
			-920
		Average	*-880*
		Minimum	*-840*
		Maximum	*-920*
Humidity swing	de Jonge et al. (2019)	Coal	-664
		PV solar	-977
Vacuum swing	Deutz and Bardow (2021)	Grid mix (Germany)	0
		Grid mix (Iceland)	-920
		Grid mix (Global 2030)	-400
		Average (grid mix)	*-440*
		Minimum (grid mix)	*0*
		Maximum (grid mix)	*-920*
		Geothermal (Iceland)	-910
		Heavy fuel oil	-440
		PV Solar	-800
		Wind	-930

Table S8 Global warming potential (GWP) and energy use of enhanced weathering

Rocks	Study	Net GWP (kg CO_2 eq./t CO_2 removed)	Energy use (GJ/t CO_2 removed)
Mafic	Lefebvre et al. (2019)	-925	1.47
	Renforth (2012)	-861	2.36
		-453	12.6
	Average	*-746*	*5.48*
	Minimum	*-453*	*1.47*
	Maximum	*-925*	*12.6*
Ultramafic	Renforth (2012)	-948	0.8
		-795	2.7
	Moosdorf et al. (2014)	-927	9.9
		-675	1.6
	Average	*-836*	*3.75*
	Minimum	*-675*	*0.8*
	Maximum	*-948*	*1.9*

Table S9 Global warming potential (GWP) of mineral carbonation

Rocks	Study	Net GWP (kg CO_2 eq./t CO_2 removed)
Serpentinite	Khoo et al. (2011)	-288
		-123
		-576
		-420
	Kirchofer et al. (2012)	-350
	Nduagu et al. (2012)	-483
		-317
	Giannoulakis et al. (2014)	-287
		153
		-355
		-453
		-186
		-478
	Ostovari et al. (2020)	-1150
		-1050
		-1166
		-445
	Khoo et al. (2021)	*-116*
		-366
	Average	*-445*
	Minimum	*153*
	Maximum	*-1166*
Olivine	Kirchofer et al. (2012)	-580
	Kirchofer et al. (2012)	-720
		-750
	Giannoulakis et al. (2014)	-541
		-550
	Julcour et al. (2015)	-502
	Ostovari et al. (2020)	-881
		-1052
	Average	*-697*
	Minimum	*-502*
	Maximum	*-1052*
Wollastonite	Giannoulakis et al. (2014)	-720
		-736
	Average	*-728*
	Minimum	*-720*
	Maximum	*-736*
Steel slag	Kirchofer et al. (2012)	-670
		-50
	Pan et al. (2016)	-6811
		-6808
		-6186
		-4592
		-4614
		-4272
		-4804
		-4041
		-6475
	Ghasemi et al. (2017)	-863
		-700
	Ostovari et al. (2020)	-1070
	Average	*-3711*
	Minimum	*-50*
	Maximum	*-6811*
Cement kiln dust	Kirchofer et al. (2012)	*-860*

		-800
	Average	*-830*
	Minimum	*-800*
	Maximum	*-860*
Fly ash	Kirchofer et al. (2012)	*-550*
		-450
	Average	*-500*
	Minimum	*-450*
	Maximum	*-550*
Pyroxene minerals	Ncongwane et al. (2018)	7798
		17,295
		354
		2126
		1364
	Average	*5787*
	Minimum	*354*
	Maximum	17,295

Table S10 Other LCA impact of mineral carbonation (Giannoulakis et al., 2014, Julcour et al., 2015)

Impacts[a]	NETL[b] (Coal)	NETL[b] (Natural gas)	NETL[b] (Coal)	NETL[b] (Natural gas)	NETL[b] (coal)	NETL[b] (Natural gas)	ÅAU1[c] (Coal)	ÅAU1[c] (Natural gas)	ÅAU2[c] (Coal)	ÅAU2[c] (Natural gas)	Attrition leaching
Rock	**Wollastonite**	**Wollastonite**	**Olivine**	**Olivine**	**Serpentine**	**Serpentine**	**Serpentine**	**Serpentine**	**Serpentine**	**Serpentine**	**Olivine**
FDP (x0.1 t oil eq.)	0.81	0.94	1.47	1.60	1.89	2.01	2.92	3.02	1.78	1.89	-
FEP (x0.1 kg P eq.)	1.67	0.64	2.34	1.46	4.46	0.90	7.37	0.88	3.73	0.85	-
FETP (kg 1,4 DB eq.)	2.39	1.03	3.64	2.51	6.70	1.72	10.88	1.61	5.59	1.54	-
HTP (x0.1 t 1,4 DB eq.)	1.15	0.56	1.88	1.39	3.42	1.00	5.44	0.92	2.81	0.88	-
MDP (x10 kg Fe eq.)	0.64	0.83	2.70	2.89	2.62	2.83	2.01	2.22	1.89	2.09	-
MEP (x0.1kg N eq.)	1.04	0.66	4.17	3.83	5.02	3.80	3.07	0.75	1.59	0.67	-
METP (kg 1,4 DB eq.)	2.45	1.07	3.70	2.67	6.76	1.91	10.65	1.83	5.51	1.70	-
ODP (x10 mg CFC-11 eq.)	1.44	2.99	3.55	4.94	2.54	6.60	3.02	10.00	2.80	6.19	-
PMFP (kg PM10 eq.)	4.82	4.71	2.91	2.86	5.44	4.68	6.43	4.90	5.40	4.84	-
POFP (kg NMVOC eq.)	0.87	0.51	0.89	0.58	2.11	0.83	3.47	1.14	1.89	0.86	1.62
TAP (kg SO_2 eq.)	1.02	0.78	1.12	1.01	3.63	1.15	6.44	1.56	3.08	1.38	1.36
TETP (x10 g 1,4 DB eq.)	1.53	1.67	4.15	4.28	3.91	3.59	4.31	3.46	3.10	2.93	-

[a] FDP: Fossil depletion potential; FEP: Freshwater eutrophication potential; FETP: Freshwater ecotoxicity potential; HTP: Human toxicity potential; MDP: Metal depletion potential; MEP: Mariner eutrophication potential; ODP: Ozone depletion potential; PMFP: Particulate matter formation potential; POFP: Photochemical oxidant formation potential; TAP: Terrestrial acidification potential; TETP: Terrestrial ecotoxicity potential.

[b] NETL: National Energy Technology Laboratory.

[c] ÅAU: Åbo Akademi University.

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