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Sustainable scale-up of negative emissions technologies and practices: where to focus

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E-mail: gonzalo.guillen.gosalbez@chem.ethz.ch**Keywords:** negative emissions technologies, carbon dioxide removal, greenhouse gas removal, sustainability, scale-upSupplementary material for this article is available [online](#)

Abstract

Most climate change mitigation scenarios restricting global warming to 1.5 °C rely heavily on negative emissions technologies and practices (NETPs). Here we updated previous literature reviews and conducted an analysis to identify the most appealing NETPs. We evaluated 36 NETPs configurations considering their technical maturity, economic feasibility, greenhouse gas removal potential, resource use, and environmental impacts. We found multiple trade-offs among these indicators, which suggests that a regionalised portfolio of NETPs exploiting their complementary strengths is the way forward. Although no single NETP is superior to the others in terms of all the indicators simultaneously, we identified 16 Pareto-efficient NETPs. Among them, six are deemed particularly promising: forestation, soil carbon sequestration (SCS), enhanced weathering with olivine and three modalities of direct air carbon capture and storage (DACCS). While the co-benefits, lower costs and higher maturity levels of forestation and SCS can propel their rapid deployment, these NETPs require continuous monitoring to reduce unintended side-effects—most notably the release of the stored carbon. Enhanced weathering also shows an overall good performance and substantial co-benefits, but its risks—especially those concerning human health—should be further investigated prior to deployment. DACCS presents significantly fewer side-effects, mainly its substantial energy demand; early investments in this NETP could reduce costs and accelerate its scale-up. Our insights can help guide future research and plan for the sustainable scale-up of NETPs, which we must set into motion within this decade.

1. Introduction

The current global levels of anthropogenic CO₂ emissions—approximately 40 Gt a⁻¹ (UNEP 2021)—must urgently decline to net zero to avoid transgressing the carbon budget that would limit global warming to 1.5 °C above pre-industrial levels, i.e. 500 Gt CO₂ at the beginning of 2020 (50% likelihood, IPCC 2021). In addition to implementing stringent emissions reductions measures, most climate change mitigation pathways restricting the temperature increase to 1.5 °C rely on carbon dioxide removal (CDR) to either neutralise residual emissions that are hard

to prevent or offset a temporary temperature overshoot. The magnitude of the cumulative CDR needed throughout the 21st century is vast (median estimate in scenarios with no or limited overshoot: 584 Gt CO₂, IPCC 2022) but it is still unclear whether we will be able to amass the technical expertise or mobilise the resources needed to tackle this challenge in a timely manner (Lawrence *et al* 2018, Nemet *et al* 2018).

Multiple CDR companies have recently emerged (CarbonPlan 2022), and initiatives like the XPRIZE Carbon Removal competition (XPRIZE Foundation 2022) or the ClimAccelerator program (Carbon

Removal ClimAccelerator 2022) seek to promote the creation of novel start-ups and diversify the portfolio of available negative emissions technologies and practices (NETPs). Nonetheless, the effectiveness and sustainability implications of NETPs remain uncertain. Several studies point out that global net negative CO₂ emissions might be less effective at cooling than positive emissions are at warming (Vichi *et al* 2013, Jones *et al* 2016, Zickfeld *et al* 2016, Keller *et al* 2018, 2021), implying that the required amount of negative emissions might be higher than previously estimated. Moreover, the deployment of NETPs could raise other environmental concerns and lead to side-effects (Heck *et al* 2016, 2018 Fuss *et al* 2018, Cobo *et al* 2022a, Smith *et al* 2019b, Qiu *et al* 2022).

Here we build on earlier works (Fuss *et al* 2018, Minx *et al* 2018, Nemet *et al* 2018) to synthesise and critically discuss the growing body of knowledge on NETPs, including emerging NETPs that have previously been overlooked, e.g. marine NETPs. Based on five key performance indicators (KPIs), we assess the NETPs' performance level and conduct a Pareto analysis to identify those that could play a more important role in the future, and the areas where research and investments should focus. We screened the relevant academic papers in the Scopus database and used online resources—the CarbonPlan database (CarbonPlan 2022) and events organised by the OpenAir (OpenAir 2022a), AirMiners (Air Miners 2022) and Ocean visions (Ocean visions 2021) communities—to track the most recent developments in the greenhouse gas removal (GGR) space.

We found that none of the assessed NETPs simultaneously outperformed all the others in terms of the five KPIs, supporting the thesis that a portfolio of NETPs will likely be needed. Our analysis indicates that terrestrial and chemical NETPs are the most promising. Forestation and soil carbon sequestration (SCS) practices show an overall good performance level and are currently ready for deployment, offering potential co-benefits that could incentivise their implementation. However, they also pose risks, chiefly the possible release of the stored carbon. By contrast, most NETPs based on chemical processes can reduce the unintended impacts of negative emissions. Among them, direct air carbon capture and storage (DACCS) presents particularly appealing KPI values; thus, investing in DACCS appears to be a good strategy to accelerate the sustainable scale-up of CDR, if developed in parallel to the renewable energy system. Terrestrial enhanced weathering deploying olivine also exhibits a promising performance, but still needs further research to guarantee that the associated impacts are tolerable. Concurrently advancing the development of NETPs involving the degradation of greenhouse gases (GHGs), still quite immature, could also contribute to sustainably meeting the demand for negative emissions.

Our results suggest that bioenergy with carbon capture and storage (BECCS) and marine NETPs—the latter showing incipient development levels—should not be prioritised over more promising GGR strategies. Nevertheless, most climate change mitigation scenarios rely heavily on BECCS (IPCC 2022), which denotes the need to upgrade integrated assessment models to incorporate additional NETPs and better characterise their performance.

The paper is structured as follows: first, we revise the definition of NETPs and the state of the art. Then we assess their performance level and identify the best—i.e. Pareto-efficient—NETPs, to conclude with recommendations for their sustainable deployment.

2. NETPs definition

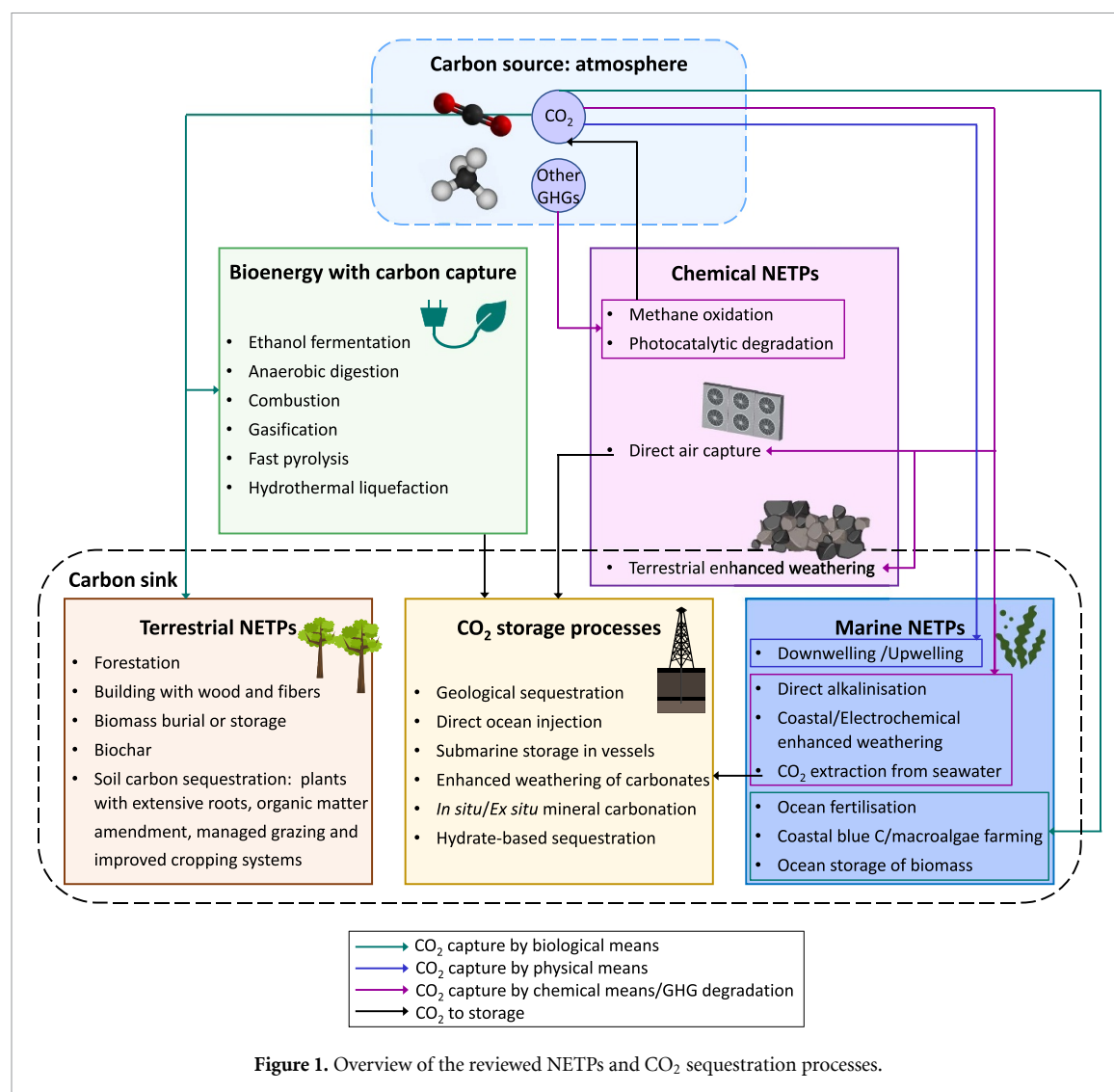
Although the terms *CDR* and *negative emissions* are often used interchangeably, the latter can refer to the removal of any GHG from the atmosphere. We define NETPs as the technologies and practices that attain negative emissions according to these three criteria:

- (a) The global warming impacts associated with the NETPs' life-cycle GHG emissions do not exceed the impacts prevented by withdrawing GHGs from the atmosphere.
- (b) The GHGs are either sequestered 'in a manner intended to be permanent' (Tanzer and Ramírez 2019)—i.e. in a sink that is not subject to foreseeable perturbations—, or permanently transformed into other compounds with lower global warming potentials.
- (c) NETPs achieve negative emissions relative to a baseline where they are not deployed, i.e. they must be additional to ongoing GGR. Hence, actions to preserve existing carbon sinks (e.g. forest conservation) are not classified as NETPs.

We can only verify that net negative emissions are achieved after quantifying the impacts of the GHGs emitted throughout the NETPs' entire life cycle (Terlouw *et al* 2021), a detailed analysis that is outside the scope of this study. Therefore, hereon we use the term NETPs more loosely, referring to those technologies and practices that could potentially generate negative emissions. On the other hand, some NETPs may have additional effects on the climate system, which could lead to a net warming effect despite achieving net negative emissions—e.g. forestation practices in high latitudes (Bala *et al* 2007, Bonan 2008).

3. State-of-the-art NETPs

Figure 1 provides an overview of the reviewed NETPs. We differentiate between terrestrial, marine, BECCS



and chemical NETPs. Terrestrial and marine NETPs enhance the CO₂ sequestration capacity of natural sinks, whereas BECCS NETPs capture and store the CO₂ generated in bioenergy production processes, which was previously taken up by biomass. NETPs relying on chemical processes either degrade GHGs into substances with lower global warming potentials or exploit the ability of CO₂ to react with specific compounds to separate it from the other air components. The CO₂ streams produced by BECCS and some chemical NETPs must be subsequently stored in the ocean or lithosphere, or transformed into carbonate products.

We do not consider the utilisation of atmospheric CO₂ for the production of chemicals as a NETP, given the uncertainties surrounding the carbon fate during the end-of-life treatment and its modest CDR potential (Hepburn *et al* 2019).

In this section, we review the NETPs within each of these four major categories. The CO₂ sequestration methods, integral to certain NETPs, are described in appendix A of the supplementary information.

3.1. Terrestrial NETPs

Terrestrial NETPs are the technologies and practices that increase organic carbon in the soil and land-based biological stocks. We distinguish between the terrestrial NETPs that predominantly sequester carbon in plants and forest products, soil organic matter and biochar.

3.1.1. Plants and forest products

The net carbon sequestered by plants is determined by the balance between the atmospheric carbon assimilated in the photosynthesis process, and the carbon losses related to plant and microbial respiration, organic matter decomposition, leaching, erosion, controlled burn or wildfire (Lal 2004, National Academies of Sciences Engineering and Medicine 2019). Although most of the captured carbon is stored in the plants and harvested plant products, the fraction of dead-root biomass and above-ground plant residues that is not subject to mineralisation or erosion losses becomes a source of carbon to the soil (Aalde *et al* 2006a, 2006b). The net

change in local soil organic carbon (SOC) depends on the environmental conditions (e.g. temperature and precipitation), edaphic factors (e.g. soil structure and C:N ratio), species characteristics and type of land-use change (LUC)—e.g. the conversion of grassland to short-rotation coppice such as willow or poplar plantations can result in net carbon emissions, i.e. the loss of SOC (Qin *et al* 2016).

Moreover, trees can produce trace GHGs, namely methane and nitrous oxide (Welch *et al* 2019), and forest ecosystems emit volatile organic compounds (VOCs) that control the formation of climate forcers responsible for warming (methane and tropospheric ozone) and cooling (aerosols). However, there is no consensus over the net climate impact of these VOCs (Unger 2014, Scott *et al* 2018).

The effect of these NETPs on the climate is also governed by the surface energy fluxes and the hydrologic cycle (Bonan 2008). Increasing the forest cover reduces the share of the incident solar radiation that is reflected. The change in the surface albedo varies greatly across regions, but it is particularly relevant in the boreal ecological zones frequently covered by snow, where it can counteract the benefits of CO₂ sequestration, leading to further warming. On the contrary, tropical forests show a net cooling effect because the increased warming associated with the reduced albedo is offset by the high evapotranspiration rates. The net climate forcing of temperate forests is extremely uncertain and location-dependent, but in general their climate benefits are considered marginal (Bala *et al* 2007, Bonan 2008).

Despite the enhanced precipitation that occurs in forested areas, their water demand can be substantial (Nolan *et al* 2021). Large-scale biomass plantations might also compete with the food system for land and nutrients, and they are chiefly constrained by land availability (Heck *et al* 2016, Boysen *et al* 2017, Ledo *et al* 2019). Moreover, the cultivation of non-native single species could have detrimental consequences for the local biodiversity if other ecosystems prevail in the area (Hulvey *et al* 2013, Liang *et al* 2016). Theoretically, trees could be genetically engineered to increase the photosynthesis rates (Jansson *et al* 2010) and improve nutrient and water use efficiencies (Jez *et al* 2016). It has also been suggested that the cultivation of plant varieties with light pigmentation—by selective breeding or genetic modification—could maximise the surface albedo (Ridgwell *et al* 2009). However, the potential risks of introducing genetically modified species into natural ecosystems are still largely unexplored.

Forest conservation is a crucial strategy to simultaneously mitigate climate change and preserve biodiversity (Popp *et al* 2012). However, we do not classify the activities preventing forest degradation and deforestation, such as those supported by the REDD+ program, (FAO 2022) as NETPs because they do not

involve the creation of new carbon sinks, as opposed to planting trees.

The global tree cover can be increased by integrating trees into agricultural systems and land with grazing livestock—i.e. through agroforestry and silvopasture practices—or, or establishing new forests. Planted forests currently constitute 7% of global forests (FAO and UNEP 2020). Some planted forests comprise native species and their structure is not defined by the silvicultural practices; they are usually planted to restore and protect the ecosystems. By contrast, plantation forests are a type of planted forests typically aiming at timber production and composed of intensively managed indigenous or introduced tree species, established by planting or seeding one or two tree species of even age with equal spacing (FAO 2018).

3.1.1.1. Forestation

The large-scale implementation of afforestation and reforestation practices—currently spurred by global initiatives like the trillion tree campaign (Trillion tree campaign 2021)—could reverse the global trend of forest loss, with average deforestation rates of 10 Mha a⁻¹ between 2015 and 2020 (FAO and UNEP 2020). Afforestation is the conversion of land that was previously not covered by forest to forestland, whereas reforestation takes place on land that has been recently deforested; i.e. afforestation implies a LUC, whereas reforestation occurs in an area classified as forest (FAO 2018).

Forests can maintain CDR for decades before it declines as the trees mature (Houghton *et al* 2015). Recent analyses cap the global CDR potential of forestation at 2.6 Gt a⁻¹ over 2025–2055 (Austin *et al* 2020) and 5.6 Gt a⁻¹ (average over 21st century, Favero *et al* 2020), with the largest potential being realised in the tropics (Doelman *et al* 2020). On the other hand, improved management practices in existing forests (e.g. reduced logging or extended rotations) could lead to the additional removal of up to 2 Gt a⁻¹ CO₂-eq (Smith *et al* 2020).

The advantage of forestation over other NETPs is that it is easy to deploy and cost-competitive (The Royal society 2009), with costs ranging between 5 and 53 US\$₂₀₂₀ t⁻¹ CO₂ removed (Fuss *et al* 2018). Nevertheless, the carbon sequestered in forests could return to the atmosphere due to human-induced LUC or unexpected perturbations such as pests, droughts or fires (Fuss *et al* 2018).

3.1.1.2. Building with wood and fibres

Agricultural residues and natural fibres like cork or hemp can be used as insulation materials, thereby sequestering the carbon contained in them (Kriegh *et al* 2021, Shen *et al* 2022a). Furthermore, the mechanical properties of certain engineered wood products—such as glued laminated timber (glulam) and cross-laminated timber—allow them to replace

steel and concrete as structural materials (D'Amico *et al* 2021). Bamboo—a fast growing grass with properties comparable to those of steel and concrete—is also an appealing material because of its ability to sequester carbon rapidly and thrive in degraded lands (Project Drawdown 2021a). The main climate benefits of these products stem from the substitution of construction materials (National Academies of Sciences Engineering and Medicine 2019, Shen *et al* 2022a); the GHGs emitted throughout the life cycle of wood and bamboo products are lower than those associated with the functionally equivalent amounts of steel and concrete (Upton *et al* 2008, Sathre and Gustavsson 2009, Sathre and O'Connor 2010, Zea Escamilla *et al* 2016). Nevertheless, the duration of the carbon sequestration in these materials is usually temporal, dependent on their lifetime and end-of-life treatment.

It has been estimated that deploying timber as a construction material could sequester up to 0.04–2.5 Gt a⁻¹ CO₂ by 2050 (Churkina *et al* 2020) at a low cost (McLaren 2012). Updating building codes to guarantee higher energy efficiencies in buildings—which would favour wood over other materials with worse insulating properties—could help to reach the full potential of wood as a construction material (Wimmers 2017).

3.1.1.3. Wood burial or storage

Wood can be harvested and buried under anaerobic conditions or stored above-ground (Zeng 2008, Zeng *et al* 2013). It has been estimated that this CDR strategy could sequester 3.7–11 Gt a⁻¹ CO₂ (Zeng *et al* 2013). The results of the field tests conducted to date (Carbon lockdown 2022) show that this NETP can significantly slow biomass decomposition, although it does not completely halt it (Adair *et al* 2010). Despite the low cost of this NETP (9–33 \$ t⁻¹ CO₂) and its easy implementation (Zeng 2008), some of its potential adverse side-effects—including nutrient lockup in the stored biomass, and disturbance of the local biodiversity—could hinder its large-scale deployment (Zeng 2008).

3.1.2. SCS

The NETPs in this category aim at increasing the amount of carbon sequestered in soil organic matter. The organic carbon content in soils is the difference between the added carbon—i.e. the carbon in roots, plant residues, and soil amendments such as compost or manure—and the carbon lost due to organic matter decomposition, microbial respiration, leaching and erosion (Paustian *et al* 2019).

Agricultural lands—which have lost 50–70% of their original SOC stocks (Zomer *et al* 2017)—show the greatest potential for SCS (Minasny *et al* 2017). Ideally, SCS does not compete with the food system for land (Nolan *et al* 2021); indeed, enhancing SCS in

vulnerable soils with low carbon stocks can improve crop yields and the soil's water-holding capacity, which may reduce irrigation and fertiliser requirements (Lal *et al* 2021). Additional co-benefits include improved habitats for the soil biota and reduced erosion (Paustian *et al* 2019, Bossio *et al* 2020). If implemented in bare soils, SCS could reduce the risk of nutrient leaching (INRAE 2021), although mismanagement could also lead to the loss of nitrogen and phosphorus—whose content in soil increases as the SOC pool is enhanced (van Groenigen *et al* 2017)—in the water runoff (Fuss *et al* 2018), causing eutrophication problems.

The SCS costs are highly variable and NETP-specific, typically ranging between 0 and 105 \$ t⁻¹ CO₂ removed (Fuss *et al* 2018). Currently, there are very few projects engaged in SCS practices, notably those operating under the Australian Carbon Farming Initiative, a voluntary carbon offsets scheme (von Unger and Emmer 2018). Nevertheless, recently launched programs such as the 4 per 1000 initiative (2021), which seeks to increase the SOC stocks in the first 30–40 cm of soil by 0.4% per year, or the carbon farming plan within the European Green Deal (European Commission 2022) could incentivise the adoption of SCS.

The technical CDR potential of SCS practices could reach 2–5 Gt a⁻¹ CO₂ by 2050 (Fuss *et al* 2018). However, these CDR rates can only be sustained for 2–3 decades in mineral soils, until SOC levels reach a new equilibrium (West and Six 2007, Paustian *et al* 2019). The main risk associated with this carbon sink is its reversibility; the SOC management practices must be indefinitely maintained to avoid losing the sequestered carbon (Bossio *et al* 2020). Nonetheless, even if these practices are permanently implemented, the rise in temperatures due to global warming is likely to drive the net loss of SOC to the atmosphere (Crowther *et al* 2016, Melillo *et al* 2017).

Restoring the hydrology of organic soils and transitioning from conventional tillage to no-till agriculture may increase SOC relative to a scenario with no change in land management—although the efficiency of the latter is heavily debated (Ogle *et al* 2019). However, we do not classify them as NETPs because they do not involve the sequestration of atmospheric carbon, but rather the prevention of SOC losses (Paustian *et al* 2019). We analyse the main SCS methods next.

3.1.2.1. Plants with extensive roots

Roots are usually the main contributor to plant SCS; they are five times more likely than above-ground litter to be stabilised as soil organic matter (Jackson *et al* 2017). Plants with deeper and larger roots—e.g. perennial vegetation such as trees and grasses—could remove on the order of 1 Gt a⁻¹ CO₂-eq (Paustian *et al* 2016). However, these crops should be planted

on low-carbon soils, otherwise the carbon loss due to the cultivation and land conversion processes could offset the carbon gains associated with the root system and the above-ground plant residues (Whitaker *et al* 2018).

While the roots and above-ground plant residues are left in the field to increase the SOC pool, biomass can be harvested and used for bioenergy generation or as a construction material (Shen *et al* 2022b), which provides the cultivation of these crops with an economic edge over other NETPs. Another advantage of extensive root systems is that they improve the plants' ability to retain nutrients and water, providing resistance to droughts and fertiliser runoff (Kell 2011).

Current research efforts focus on developing crops with larger and deeper roots through selective breeding or genetic engineering techniques (Kell 2011, Paustian *et al* 2019). Most notably, the Land Institute is working on the perennialisation of cereal grains and other annual crops, some of which are already commercialised (The Land Institute 2021).

3.1.2.2. Organic matter amendment

The carbon present in soil amendments (such as compost produced from municipal organic waste) contributes to increasing the SOC levels. Furthermore, organic amendments can improve the soil properties and nutrient availability, which stimulates plant productivity and additional carbon uptake (Paustian *et al* 2019). The CDR capacity of this NETP depends on the original fate of the materials—e.g. manure application does not sequester additional carbon compared to a baseline scenario where it is left in the field (Powlson *et al* 2011, Leifeld *et al* 2013)—, and on the decomposition rate of the added carbon. The latter is typically slower than that of fresh plant residues and dependent on the soil properties and amendment characteristics (Diacono and Montemurro 2011, Paustian *et al* 2016).

Spreading light-coloured residues such as cereal straw on the soil could provide cooling benefits by increasing the surface albedo (Smith 2016). By contrast, dark soil amendments like compost could have the opposite effect (Meyer *et al* 2012). Furthermore, the application of organic matter to the soil may increase nitrous oxide emissions (Smith *et al* 2001), which could offset the climate benefits of carbon sequestration. Moreover, the leaching of the nutrients contained in the organic amendments could pollute water bodies, whereas the heavy metals and organic pollutants in the composted organic wastes could pose a risk for human health (Cobo *et al* 2018).

3.1.2.3. Managed grazing

The practices under this category seek to maximise the SOC inputs from plant roots and residues in grazing lands by maximising forage production or controlling grazing intensity (Paustian *et al* 2019).

Introducing more productive species with deeper roots, such as legumes, and adjusting the water and fertiliser inputs to the plants' demand can increase forage production (Smith *et al* 2008). On the other hand, techniques to manipulate the grazing intensity include decreasing the number of animals in a given area (to prevent overgrazing), and implementing rotational or multi-paddock grazing systems, which allow the land that has already been grazed to recover (Project Drawdown 2021b).

The main co-benefits of these practices are the improvements in biological diversity and soil properties (Bossio *et al* 2020). However, the carbon sequestration rates are highly location-dependent, i.e. subject to the climate, soil and vegetation characteristics (Conant *et al* 2017). It has been estimated that the global sequestration potential of grazing lands ranges between 0.3 and 1.4 Gt a⁻¹ CO₂-eq (Henderson *et al* 2015).

3.1.2.4. Improved cropping systems

Maximising the time during which soil is covered by vegetation can increase the SOC stocks and simultaneously enhance soil quality and fertility. Fallow frequency can be reduced by planting seasonal cover crops—whose global CDR potential is estimated to be 0.4 Gt a⁻¹ CO₂-eq (Bossio *et al* 2020)—and diversifying crop rotations, specifically by introducing perennials, legumes and species that produce large amounts of residues (National Academies of Sciences Engineering and Medicine 2019, Paustian *et al* 2019). The costs of these practices vary greatly across locations and methods; the available estimates in the literature range from 23 to 147 \$ t⁻¹ CO₂-eq (Tang *et al* 2016).

3.1.3. Biochar

Biogenic carbon can be sequestered in the biochar produced as biomass is subjected to certain thermochemical processes, namely pyrolysis, gasification, hydrothermal carbonisation and flash carbonisation (Fawzy *et al* 2021). Slow pyrolysis is the main process used for biochar production, given the good product properties and high yields that it can achieve. In this process, biomass is heated in the absence of oxygen or in a low-oxygen environment, which allows around 50% of the carbon content in biomass to be transferred to the biochar (Schmidt *et al* 2019). The energy released in the combustion of the other pyrolysis products (bio-oil and pyrogas) is enough to satisfy the heat demand of the pyrolysis process and provide heat for other applications (Peters *et al* 2015a). The CO₂ generated in the combustion process could be separated from the flue gases and subsequently stored at the expense of substantially raising the energy penalty.

Biochar can be used as an aggregate in construction materials (Gupta and Kua 2017), but its main application is as a soil amendment. Although the

biochar applied to soils may release the nutrients originally contained in the biomass feedstock, these are not sufficient to replace conventional fertilisers. This limitation could be overcome by co-composting the biochar—i.e. using it as a compost additive—which has been demonstrated to improve the compost performance (Schmidt *et al* 2021).

The fertility and productivity of acidic soils typically improve after the application of biochar, whereas the crop yields of alkaline soils are more likely to decrease (Tisserant and Cherubini 2019). Given its high reactivity and specific surface area, biochar can adsorb nutrients and pollutants, contributing to soil remediation and water purification. Furthermore, it can increase the soil's water-holding capacity (Smith *et al* 2019a). However, its ability to immobilise chemicals may have detrimental consequences, such as a reduction in the efficiency of herbicides and pesticides. Biochar can also be a source of contaminants, such as heavy metals, organic pollutants, particulate matter, carbon black, etc (Tisserant and Cherubini 2019). Conversely, it reduces nitrous oxide and nitrogen oxide emissions from fertilised soils (Tisserant and Cherubini 2019). Ammonia and methane emissions rates are also affected by the soil application of biochar, but these can either decrease or increase, depending on the soil and biochar properties (Tisserant and Cherubini 2019, Schmidt *et al* 2021).

Biochar application to soils is likely to reduce the surface albedo, which decreases the climate mitigation benefits of this NETP (Genesio *et al* 2012, Meyer *et al* 2012). Moreover, the biochar carbon content can degrade to CO₂ in a timeframe ranging from a few years to millennia, contingent on the soil and climate conditions, and the characteristics of the feedstock and pyrolysis process (Gurwick *et al* 2013); e.g. at a soil temperature of 15 °C, between 18 and 37% of the carbon will be mineralised after 100 years (Woolf *et al* 2021). Hence the importance of meeting biochar quality standards, such as those set by the European biochar certificate (European Biochar Certificate 2022).

It has been estimated that 0.44–2.62 Gt a⁻¹ CO₂ could be sequestered as solid carbon in biochar without occupying additional land; the area needed to grow the biomass would equal the land reduction associated with the increase in crop yields due to the biochar application (Werner *et al* 2022). The costs of this NETP range between 32 and 127 \$ t⁻¹ CO₂ (Fuss *et al* 2018), and one of its main advantages is that it can be implemented at both small and large scales (Werner *et al* 2018). However, despite the high maturity level of this technology, pyrolysis plants at industrial scale are currently scarce, (Aines *et al* 2021) e.g. the one in Stockholm, which produces district heating and biochar from biomass residues (Bloomberg Philanthropies 2021).

3.2. Marine NETPs

We define marine NETPs as the set of technologies and practices that seek to maximise the long-term storage of carbon in the ocean. We classify marine NETPs as physical, chemical or biological, based on their underlying mechanism. Figure 2 illustrates the CDR mechanisms of marine NETPs.

Most marine NETPs rely on the manipulation of ocean processes. The main drivers of the oceanic carbon cycle are the *solubility pump*—the transport of dissolved carbon from the surface to the deep ocean by downwelling currents—and the *biological pump*, which is based on the fixation of the carbon dissolved in the surface waters by photosynthetic organisms (Scott-Buechler and Greene 2019).

Chemical weathering also contributes to CO₂ sequestration in the ocean. The chemical weathering processes that occur over geological timescales break down rocks, releasing cations that are transported to the ocean and react with the dissolved CO₂, producing carbonate minerals that are stored on the ocean floor (Berner 2003).

Enhancing the natural rate of CO₂ sequestration in the deep ocean would draw a shift in the equilibrium between the CO₂ concentrations in the atmosphere and the ocean surface water, leading to the transfer of atmospheric CO₂ to the ocean. However, this process is not instantaneous; it takes about one year for the CO₂ concentrations in the atmosphere and the seawater to reach an equilibrium, depending on the regional oceanographic properties (National Academies of Sciences Engineering and Medicine 2021).

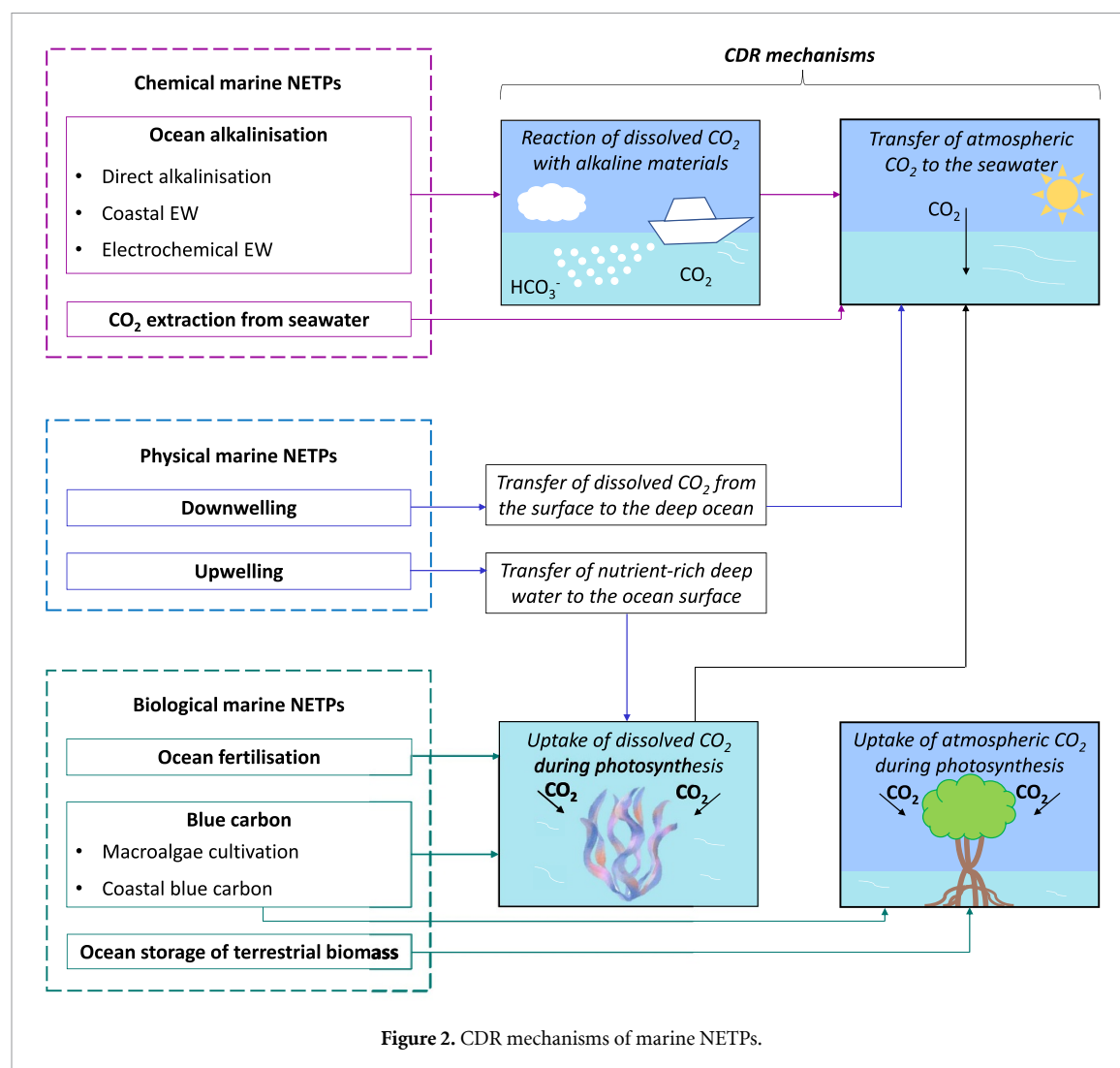
The NETPs involving the placement of materials in the ocean are subject to the London Protocol (IMO 2006a), which aims to prevent marine pollution. This protocol was amended to allow the storage of CO₂ in sub-seabed geological formations (IMO 2006b) and prohibit ocean fertilisation (IMO 2013), although research activities focusing on the latter may be considered for a permit (IMO 2013). The protocol also establishes a framework to regulate additional marine geoengineering activities in the future (IMO 2013).

3.2.1. Physical marine NETPs

These marine NETPs only require unit operations involving physical changes, like the movement of fluids and heat exchange.

3.2.1.1. Downwelling

Zhou and Flynn (2005) assessed the implications of enhancing the solubility pump by cooling the ocean surface water, which would increase downwelling currents. They concluded that it is unlikely that this technology will ever become a competitive carbon sequestration method; cooling 1 Mm³ s⁻¹ of seawater from 6 °C to 0 °C would require a heat flux



of 25 TW and capture only 35 Mt a⁻¹ CO₂, while the estimated costs could range between 258 and 5826 \$ t⁻¹ CO₂. Moreover, they emphasised the need for modelling research, suggesting that upwelling currents releasing more CO₂ than the amount captured could offset downwelling currents. To the best of the authors' knowledge, experimental downwelling has never been conducted before.

3.2.1.2. Upwelling

Lovelock and Rapley (2007) proposed to use vertical pipes to pump up nutrient-rich deep waters to fertilise algae in the ocean surface and increase the transfer rate of organic carbon to the deep ocean via the biological pump. A 300 m long pipe was deployed by Maruyama *et al* (2011) to investigate the effects of artificial upwelling driven by the difference in salinity and temperature at both ends of the pipe. They found that the chlorophyll concentration at the pipe outlet was much higher than in the surrounding surface water, which suggests an increase in the CO₂ absorption rate. Nonetheless, a recent analysis shows

that approximately 70% of the carbon exported to the deep ocean after increasing ecosystem productivity is transported back to the surface ocean within 50 years (Siegel *et al* 2021).

Earth system simulations reveal that this NETP could contribute to cooling the Earth's surface—due to the lower temperature of the ocean's bottom waters—and enhancing terrestrial carbon storage. However, discontinuing the upwelling could lead to rapid warming (Oschlies *et al* 2010, Keller *et al* 2014). Furthermore, the decrease in evaporation and evapotranspiration rates would lead to a decline in precipitations. Other side-effects include increased acidification, and a reduction in sea-ice loss (Keller *et al* 2014).

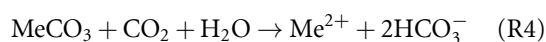
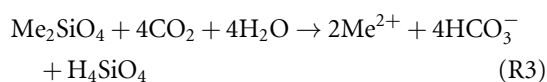
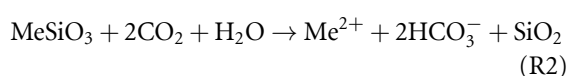
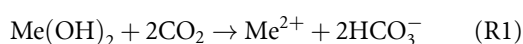
The global CDR potential of artificial upwelling is likely limited to 50–100 Mt a⁻¹ CO₂ (Kowek 2022); it has been estimated that upwelling 1 Mm³ s⁻¹ of seawater could only sequester 59 Mt a⁻¹ CO₂ (Lenton and Vaughan 2009). However, the results of the model developed by Yool *et al* (2009) showcased that pumping up water in some regions could lead to net CO₂ emissions to the atmosphere.

3.2.2. Chemical marine NETPs

These strategies aim at manipulating the pH of the seawater to either extract the dissolved carbon or transform it into other chemical species.

3.2.2.1. Ocean alkalinisation

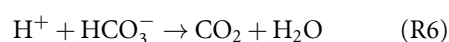
This marine NETP relies on the reaction of alkaline substances with the CO₂ dissolved in the seawater. The enhanced weathering (EW) reactions that occur as a result of adding synthetic chemicals or minerals to the ocean are shown below (Harvey 2008, Renforth 2019). Here, Me represents a divalent cation, typically calcium or magnesium.



While the alkaline materials react with the dissolved CO₂ quite rapidly, the subsequent transfer of atmospheric CO₂ to the surface ocean is slower (National Academies of Sciences Engineering and Medicine 2021). Moreover, although the residence time of bicarbonate ions in the ocean is virtually permanent, an increase in the alkalinity levels would lead to mineral carbonation reactions that produce solid carbonate minerals and release part of the previously captured CO₂ (Renforth 2019):



The reaction of acidic species (other than carbonic acid) with the produced bicarbonate ions could also lead to the emission of the captured CO₂, in accordance with reaction (R6) (Zhang *et al* 2022):



On the other hand, the nutrients released during the dissolution of the minerals could stimulate biological productivity, leading to additional carbon sequestration (Hartmann *et al* 2013, Hauck *et al* 2016). Nevertheless, the side-effects related to the dissolution of the trace metals present in the minerals on the marine biota are still largely unknown (Meysman and Montserrat 2017, Bach *et al* 2019, Gore *et al* 2019).

3.2.2.1.1. Direct alkalinisation

Lenton *et al*'s (2018) simulations indicate that adding olivine ((Mg,Fe)₂SiO₄) to the seawater could counteract ocean acidification and lead to the cumulative sequestration of 524–676 Gt CO₂ between 2020 and 2100, whereas Feng *et al* (2017) provided a conservative sequestration potential of 971 Gt CO₂ over that period.

The direct addition of synthetic chemicals to the ocean could also speed up the carbon sequestration process (The Royal society 2009, Renforth *et al* 2013). Renforth *et al* (2013) performed a techno-economic analysis of an ocean liming system based on the production of calcium and magnesium oxides via the calcination of limestone and dolomite. To ensure a net negative carbon balance, the study considered that the CO₂ generated in the calcination process was captured and stored in a geological reservoir. They concluded that an energy input of 0.7–6.8 GJ would be needed to achieve the net removal of 1 t CO₂ from the atmosphere, and the costs could range between 82 and 181 \$ t⁻¹ of atmospheric CO₂ removed.

Alternatively, alkaline wastes could be used (Renforth 2019). Davies (2015) proposed to decompose desalination reject brine using solar thermal energy to produce magnesium oxide, which can be subsequently added to the seawater. This process would require 13.8 GJ t⁻¹ CO₂ removed from the atmosphere. By contrast, treating the desalination reject brine with electrolysis to produce magnesium hydroxide could reduce the energy consumption to 1.8 GJ t⁻¹ CO₂. However, 13.7 m³ of water would be consumed per t CO₂ removed with this method (Davies *et al* 2018).

3.2.2.1.2. Coastal enhanced weathering

Coastal EW constitutes an alternative CDR strategy to directly releasing alkaline minerals in the seawater, whereby the minerals are spread on beach environments, and further comminuted and transferred to the ocean due to the action of the waves on the beach (Hangx and Spiers 2009, Montserrat *et al* 2017). Project Vesta (2022), a non-profit organisation aiming to advance the deployment of coastal EW, estimates that the cost of this NETP could range between 34 and 50 \$ t⁻¹ net CO₂ removed if implemented at scales above 100 Mt, with costs falling below 100 \$ t⁻¹ at the 1–10 Mt scale (Green 2022). Nonetheless, the CDR potential of coastal EW deploying olivine grains is limited by the toxicological effects associated with the nickel and chromium contained in the olivine; 0.51–37 Gt CO₂ could be sequestered until 2100 without putting benthic organisms at risk (Flipkens *et al* 2021).

3.2.2.1.3. Electrochemical enhanced weathering

Other authors have suggested the deployment of electrochemically mediated EW processes.

House *et al* (2007) estimated that, under an optimistic scenario, 3 GJ of renewable electricity would be required to sequester 1 net t CO₂ in the ocean by adding the NaOH produced via the electrolysis of an artificial brine. To manage the generated chlorine, they suggested producing hydrochloric acid and neutralising it with silicate rocks. The current CDR costs of Heimdal's pilot plant, based on a similar configuration (Heimdal 2022b), are below 500 \$ t⁻¹ (Heimdal 2022a).

Another approach that reduces chlorine generation as a byproduct is based on the integration of water electrolysis and the mineral weathering of carbonate (Rau 2008) and silicate minerals (Rau *et al* 2013). The protons generated at the anode dissolve the minerals. The metal cations move towards the cathode to form metal hydroxides, and the carbonate and silicate anions migrate to the anode to form carbonic acid, silicic acid, or silica. The hydroxides react with the dissolved CO₂, enabling its capture as metal bicarbonate or carbonate. The energy consumption of this process (approximately 7 GJ t⁻¹ CO₂ captured) could be reduced by oxidising the hydrogen generated in the cathode. The total costs of this technology, highly dependent on the energy source, could reach 614 \$ t⁻¹ CO₂ (Rau *et al* 2018). The company Planetary Technologies (2022) is currently working on the scale-up of an electrochemical EW process using mine tailings as a source of alkalinity (OpenAir 2022c).

A novel CDR strategy based on the electrochemical splitting of water into proton and hydroxide solutions has recently been proposed (Tyka *et al* 2022). Pumping the acidic water to the deep ocean would induce the dissolution of calcite deposits—accelerating reaction (R4)—, whereas the basic solution would be released at the surface water, speeding up the ocean's CO₂ uptake. The authors estimate that 3.7–11 t a⁻¹ CO₂ could be sequestered with this method while limiting the pH decrease in the deep water by 0.2, at a cost of 89–285 \$ t⁻¹ CO₂ (Tyka *et al* 2022).

3.2.2.2. CO₂ extraction from seawater

Eisaman *et al* (2012) described an experimental set up to extract up to 60% of the carbon dissolved in seawater with bipolar electrodialysis membranes. This process is based on the premise that returning the CO₂-depleted water to the ocean would draw the absorption of more atmospheric CO₂ into the seawater. The dilute acid and base products derived from the electrodialysis process are used to alter the pH equilibrium in the processed seawater and extract the dissolved carbon. Two process configurations were proposed: in the *acid* process, the produced acid is used to lower the pH of the treated water and convert the dissolved inorganic carbon to CO₂ gas, which should be sequestered or mineralised to attain carbon negative emissions. In the *base* configuration, the pH of the processed water is increased with the

produced base, which causes the precipitation of the dissolved inorganic carbon as calcium carbonate. Before returning the water to the ocean, the pH is restored with the produced chemicals (Eisaman *et al* 2018). The associated energy consumption of the acid and base configurations is 11.3 and 15.8 GJ t⁻¹ CO₂ extracted, respectively (Eisaman *et al* 2018). Capture costs between 393 and 637 \$ t⁻¹ of extracted CO₂ were estimated (Eisaman *et al* 2018).

This is an incipient research area with potential for novel technological developments; an experimental prototype of an electrolytic cation exchange process capable of extracting 92% of the CO₂ present in the seawater and simultaneously produce hydrogen has also been presented (Willauer *et al* 2014).

3.2.3. Biological marine NETPs

These NETPs sequester carbon in the ocean by biological means, either by boosting the CO₂ uptake rate of marine photosynthetic organisms—in the case of ocean fertilisation and blue carbon—or, through the accumulation of terrestrial biomass in the deep ocean.

3.2.3.1. Ocean fertilisation

Iron availability limits the photosynthesis rate in around a third of the open ocean (Emerson 2019), whereas nitrogen and phosphorus are the limiting nutrients in the rest of the ocean (Williamson *et al* 2012). Hence, fertilising the ocean with site-specific deficient nutrients could lift the constraint on the biological pump.

Several small-scale iron fertilisation experiments have been carried out, confirming that iron promotes biomass productivity and CO₂ drawdown from the atmosphere in many regions (Williamson *et al* 2012, Emerson 2019). Nonetheless, field experiments on phosphorus fertilisation registered a slight decrease in phytoplankton and biomass chlorophyll, suggesting additional limitations to nutrient availability (Williamson *et al* 2012).

The costs of ocean fertilisation are dependent on the regional conditions; it has been estimated that the CDR costs via macronutrient (nitrogen and phosphorus) and iron fertilisation are 24 and 519 \$ t⁻¹ CO₂, respectively (Harrison 2013, Jones 2014). The cost difference between the two strategies can be mainly attributed to the low sequestration efficiency of the latter.

Zahariev *et al* (2008) calculated that the maximal CDR rate that could be achieved by means of iron fertilisation would be below 3.6 Gt a⁻¹. In contrast, the maximum CO₂ sequestration potential of combined nitrogen and phosphorus fertilisation has been estimated to be 5.5 Gt a⁻¹ (Harrison 2017). However, the permanence of the carbon sequestered in the deep ocean by enhancing the upper ocean biological productivity is very low: on average, only 32% of the carbon would remain sequestered after 50 years, and it would drop to 25% after 100 years (Siegel *et al* 2021).

According to Williamson *et al* (2012), the potential unintended impacts of large-scale ocean fertilisation may include:

- The production of gases that may affect the climate, such as nitrous oxide, methane and dimethyl sulphide (DMS). The latter can increase cloud condensation and reflectivity (albedo), leading to a cooling effect (Wingenter *et al* 2007).
- Far-field effects on primary productivity due to the depletion of non-limiting nutrients.
- Decrease of oxygen levels.
- Biodiversity impacts. Large-scale ocean fertilisation will likely change the relative abundance of different species.
- Increased acidification of deep waters.

3.2.3.2. Blue carbon

Blue carbon refers to the carbon sequestered in coastal and marine ecosystems.

3.2.3.2.1. Coastal blue carbon

The area occupied by vegetated coastal ecosystems (mangrove forests, seagrass beds, and salt marshes) is much smaller than that of terrestrial forests, but their current total contribution to long-term carbon sequestration (307–856 Mt a⁻¹ CO₂) is comparable (McLeod *et al* 2011). It has been estimated that restoring coastal vegetation would lead to a cumulative CDR of 95.3 Gt by 2100 (Gattuso *et al* 2021). Nonetheless, these ecosystems can act as net methane sources, offsetting the global warming impacts prevented by CDR (Al-Haj and Fulweiler 2020). The costs of this NETP exceed 200 \$ t⁻¹ CO₂ and are highly dependent on the type of ecosystem (Gattuso *et al* 2021).

3.2.3.2.2. Macroalgae cultivation

Macroalgae are the most productive marine macrophytes; they currently promote the storage of about 634 Mt a⁻¹ CO₂ in the deep ocean (Krause-Jensen and Duarte 2016). However, their net carbon sequestration capacity is uncertain; a recent study suggests that seaweed ecosystems may be a net carbon source (Gallagher *et al* 2022).

The cultivation of macroalgae forests (ocean afforestation or sea forestation) has been proposed as a CDR strategy where the natural drifting and sinking of a fraction of the macroalgal material allows carbon sequestration (Ocean visions 2022), with the costs of optimised farming systems at around 75–100 \$ t⁻¹ CO₂ (National Academies of Sciences Engineering and Medicine 2021).

Alternatively, macroalgae could be farmed and purposely sunk to the deep ocean. This CDR strategy could sequester up to 12.4 Gt a⁻¹ CO₂, and more than half of it could remain in the deep ocean for centuries (Wu *et al* 2022). Although recent economic evaluations indicate that the costs of this NETP could be

above 460 \$ t⁻¹ CO₂ (Coleman *et al* 2022, DeAngelo *et al* 2022), the company Running Tide (2021) is selling CDR through macroalgae farming and sinking at a price of 250 \$ t⁻¹ (CarbonPlan 2022), with estimated costs of 150–200 \$ t⁻¹ CO₂ (Calacanis 2020).

The CDR potential and costs of NETPs based on macroalgae cultivation, as well as their potential (positive and negative) effects on the local biodiversity are still highly uncertain (Campbell *et al* 2019, Ricart *et al* 2022). The latter may include a reduction in phytoplankton primary productivity and oxygen concentrations, and increased subsurface CO₂ and nutrient levels, which can lead to acidification and eutrophication impacts (National Academies of Sciences Engineering and Medicine 2021). Moreover, while GHG emissions—linked to net primary productivity in the open ocean (Weber *et al* 2019)—could counter the climate benefits, macroalgae could also increase the ocean albedo, reinforcing the cooling effect of this NETP (Bach *et al* 2021).

3.2.3.3. Ocean storage of terrestrial biomass

Metzger and Benford (2001) proposed collecting crop residues and depositing them (ballasted with stone) on the ocean floor as a carbon sequestration method. In a later work, the carbon sequestration efficiency of this practice was estimated to be 92.5% (Strand and Benford 2009). Thus, assuming a global generation of crop residues of 4.98 Gt a⁻¹ with a 40% carbon content, 6.75 Gt CO₂ could be sequestered annually, at the cost of 117 \$ t⁻¹ CO₂ (Strand and Benford 2009). The authors suggest that lignocellulosic materials are highly stable in the marine environment, yet it is hard to foresee the associated environmental impacts due to the lack of experimental studies. Biochar may be more suitable for this application than raw biomass, given that it is more resistant to microbial degradation and it would not require ballasting because of its higher density (Miller and Orton 2021).

3.3. BECCS

BECCS encompasses a wide set of technologies that derive various energy vectors—electricity, heat, methane, hydrogen, ethanol, oil and other biofuels—from biomass. The carbon released as CO₂, which was removed from the atmosphere during photosynthesis, is subsequently captured and sequestered.

Biomass can be sourced from forest and agricultural residues, although their overall CDR potential is limited because of their important role in maintaining SOC stocks and soil fertility (Smith *et al* 2016). Likewise, the energy valorisation of organic wastes coupled with CCS can provide additional opportunities for CDR (Pour *et al* 2018). Perennial energy crops including grasses such as switchgrass and *Miscanthus*, and short-rotation coppice like poplar are particularly promising feedstocks because of their high yields, ability to grown on marginal land and potential contribution to SCS (Canadell and Schulze 2014).

The cultivation of dedicated bioenergy crops in agricultural and forested lands could interfere with the global food supply and biodiversity conservation targets (Popp *et al* 2012, Smith *et al* 2016). Thus, BECCS is primarily constrained by land availability and crop productivity (Boysen *et al* 2017, Jones and Albanito 2020). Starting the deployment of BECCS early would allow exploiting the biomass resources to a greater extent compared to a scenario where BECCS is implemented later, thereby releasing the pressure on land (Galán-Martín *et al* 2021, Xu *et al* 2022).

Currently, the largest BECCS facility worldwide is an ethanol fermentation plant located in Illinois, which captures $1 \text{ Mt a}^{-1} \text{ CO}_2$. Only four other BECCS plants—also producing ethanol—were operating in 2019; the total capacity of these five facilities is $1.5 \text{ Mt a}^{-1} \text{ CO}_2$ (Global CCS Institute 2019). However, the global technical CDR potential of sustainable BECCS could reach $10.4 \text{ net Gt a}^{-1} \text{ CO}_2\text{-eq}$ by 2050 (Koornneef *et al* 2012). The CDR potential of BECCS strongly depends on the technology and the biomass cultivation site (Hanssen *et al* 2020); greater CDR rates are achieved in subtropical and warm temperate areas, where biomass yields are usually high and initial LUC emissions, low (Hanssen *et al* 2020). Nevertheless, the soil carbon loss induced by LUC could even offset the sequestered CO_2 under sub-optimal conditions (Fajardy and Mac Dowell 2017, Harper *et al* 2018). Furthermore, tree plantations may emit other GHGs, such as methane, nitrous oxide and VOCs (Unger 2014, Scott *et al* 2018, Welch *et al* 2019). Regarding the other impacts of BECCS on the climate system, it is commonly assumed that the decrease in the surface albedo linked to biomass plantations may further reduce the climate benefits, although a recent study indicates that bioenergy crops could induce a global cooling effect (Wang *et al* 2021).

By accounting for the LUC emissions of lignocellulosic crops over a 30 year period (typical plantation lifetime), a global spatially explicit analysis found that BECCS processes with a 90% carbon sequestration efficiency could remove $2.5 \text{ Gt a}^{-1} \text{ net CO}_2\text{-eq}$. By contrast, amortising the LUC emissions over 80 years (consistent with mitigation pathways towards 2100) could increase the CDR potential of BECCS to $40 \text{ Gt CO}_2\text{-eq/a}$ —at the expense of cultivating a considerable share of the crops on natural forests and grasslands (Hanssen *et al* 2020). If the substantial nutrient, freshwater and land demands of BECCS, and its impacts on the terrestrial biosphere integrity are factored in, the maximum net removal that would allow BECCS to remain within the Earth's safe operating space delimited by the planetary boundaries would be $0.2 \text{ net Gt a}^{-1} \text{ CO}_2$. CDR rates above $23 \text{ net Gt a}^{-1} \text{ CO}_2$ would imply a high risk for the Earth's stability (Heck *et al* 2018). Indeed, sequestering 1 Gt CO_2 through BECCS could lead to the extinction of over ten vertebrate species if crops were grown on extremely biodiverse areas (Hanssen *et al* 2021).

Replacing terrestrial biomass with algae could minimise some of the side-effects of land-based BECCS at the expense of increasing costs (Beal *et al* 2018). Nonetheless, algal BECCS is still an immature technology (Global CCS Institute 2019), and the net impacts of the large-scale cultivation of algae on the climate and marine ecosystems are not well understood yet (Campbell *et al* 2019, Bach *et al* 2021). An optimistic gross estimate suggested that $19\text{--}53 \text{ Gt a}^{-1} \text{ CO}_2$ could be sequestered by subjecting the macroalgae grown on 9% of the ocean's surface to anaerobic digestion (N'Yeurt *et al* 2012). However, transporting wet algae over long distances from the off-shore cultivation site to the BECCS plant could account for a great share of the energy penalty of algal BECCS, and even lead to net positive CO_2 emissions (Melara *et al* 2020).

On the other hand, the energy penalty linked to retrofitting existing bioenergy plants with CCS units would increase the energy production costs (IEA GHG 2009). The costs of BECCS are highly contingent on the biomass supply chain configuration, and region- and technology-specific (Fajardy and Mac Dowell 2020). Here we differentiate between two types of BECCS systems, namely those relying on biological and thermochemical processes. Figure 3 depicts the main mass and energy flows involved in different BECCS configurations.

3.3.1. Biological processes

BECCS systems based on biological processes couple fermentation or anaerobic digestion technologies with CCS to produce biofuels.

3.3.1.1. Ethanol fermentation

Fermentation is the process whereby microorganisms transform sugars into ethanol and CO_2 . As reaction (R7) shows, one third of the carbon in the glucose molecules subjected to fermentation is transformed into CO_2 that can be subsequently sequestered; the remaining carbon composes the ethanol and will be released as CO_2 when the ethanol is used as fuel. Certain substances with a high sugar content, such as the juice extracted from raw sugarcane or sugar beets, can be directly fermented (Laude *et al* 2011, Milão *et al* 2019). By contrast, lignocellulosic materials must previously undergo an enzymatic hydrolysis process, where polysaccharides are broken down to glucose and other fermentable sugars (Xiros *et al* 2013).



A purification unit is required to meet the commercial specifications of ethanol. Although several technologies such as membranes or molecular sieves could help reduce the energy consumption of the ethanol separation process, this step is typically carried out in a distillation unit, which accounts for 50–80% of the total energy input (Logsdon 2004). Therefore,

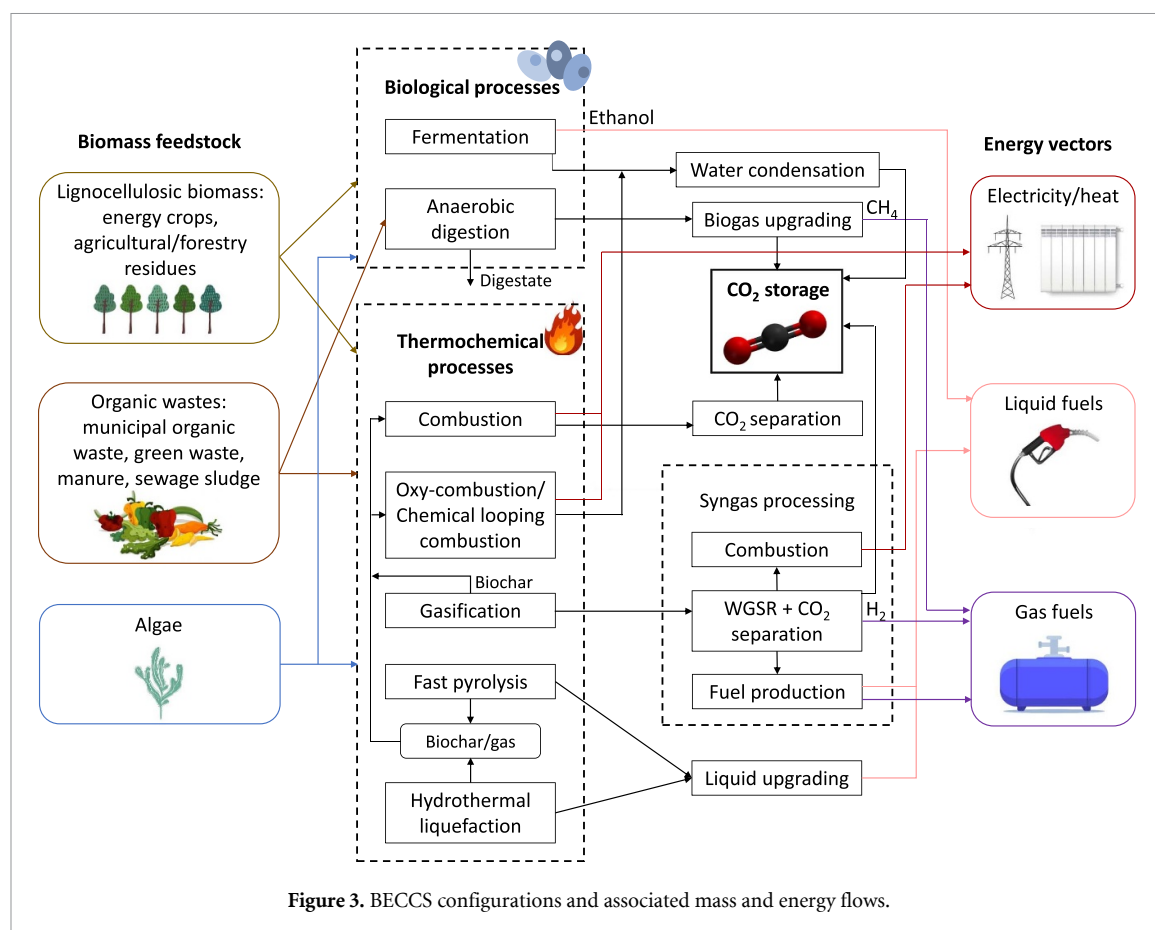


Figure 3. BECCS configurations and associated mass and energy flows.

the main CO₂ source in a conventional ethanol plant is the cogeneration unit, designed to supply heat and power to the system (Carminati *et al* 2019).

The CO₂ produced in the fermentation unit accounts for 11%–13% of the carbon emissions of an ethanol plant (Koornneef *et al* 2012); hence, sequestering this CO₂ stream alone does not make the overall process carbon negative (Laude *et al* 2011). However, most fermentation-BECCS plants—including the largest one to date, with a capacity of 1 Mt a⁻¹ CO₂—only capture the CO₂ released in the fermentation process (Global CCS Institute 2020). This CO₂ stream just needs to be dehydrated and compressed (Moreira *et al* 2016), whereas capturing the CO₂ produced in the cogeneration unit—which is diluted in the flue gas stream—is more energy-intensive and costly (Laude *et al* 2011, Bello *et al* 2020). The overall costs are likely to range between 21 and 185 \$ t⁻¹ CO₂ captured, with the upper bound corresponding to the capture of CO₂ from both the fermentation and cogeneration units (Fuss *et al* 2018). The CDR potential of fermentation-BECCS could reach 1 Gt a⁻¹ by 2050 (Koornneef *et al* 2012).

3.3.1.2. Anaerobic digestion

Under anaerobic conditions, bacteria can degrade certain types of biomass to biogas, which is mainly composed of methane (50%–70%) and CO₂ (Thrän *et al* 2014). The latter must be captured—via

CO₂ absorption in a liquid phase, pressure swing adsorption or membranes—prior to injecting the methane into the natural gas grid or using it as fuel in vehicles (Thrän *et al* 2014). Anaerobic digestion and biogas upgrading are well-established processes; in 2012 there were 277 biogas upgrading plants linked to anaerobic digesters, but none of them sequestered the separated CO₂ (Thrän *et al* 2014).

By 2050, 2.7 Gt a⁻¹ CO₂-eq could be removed from the atmosphere through the anaerobic digestion of energy crops, agricultural residues, municipal solid waste, sewage and manure (Koornneef *et al* 2013), although sequestering the CO₂ produced by burning the methane could provide additional negative emissions. A fraction of the biomass carbon content—contingent on the feedstock—is lost in the digestate, i.e. the remaining material that cannot be further degraded. This material is rich in nutrients and can be used for soil amendment, but it may require further treatment because of its high water content (WRAP 2012).

The CDR potential of this NETP is modest relative to other BECCS processes because feedstocks with high lignin contents cannot be subjected to anaerobic digestion; this technology is particularly well-suited for biogenic wastes such as the organic fraction of municipal solid waste, sewage sludge or manure (Koornneef *et al* 2013). Since these biomass resources are decentralised, the size of anaerobic

digesters is typically small, which can increase the costs of connecting these facilities to the natural gas and CO₂ pipelines (Koornneef *et al* 2013). The total costs of anaerobic digestion BECCS, which are highly dependent on the biomass source, could range between 235 and 557 \$ t⁻¹ CO₂ (IEA GHG 2013).

3.3.2. Thermochemical processes

In this type of BECCS, biomass is transformed into energy vectors through chemical reactions that occur at high temperatures, releasing CO₂ that is subsequently captured and sequestered.

3.3.2.1. Combustion

Theoretically, a global CDR potential of 10.4 Gt a⁻¹ could be sustainably achieved by 2050 with combustion-BECCS (Koornneef *et al* 2012). As biomass is burnt to generate heat and power, all the carbon in the feedstock is transformed into CO₂, whereas the non-combustible components of the biomass are transferred to the ashes. Several post-combustion capture processes (membrane separation, adsorption, etc) can separate the diluted CO₂ from the flue gas generated in the boiler. The chemical absorption of CO₂ into a monoethanolamine (MEA) solution followed by a desorption step is the most widespread of these processes (Bui *et al* 2018), and it can achieve CO₂ capture rates above 90% (Brandl *et al* 2021). Nevertheless, the associated energy penalty is substantial; e.g. the net energy efficiency of a 75 MW biomass power plant could drop from 36% to 23% when a CO₂ capture unit is added (IEA GHG 2009). The use of advanced commercial solvents based on amines blends could help reduce the energy penalty (Bui *et al* 2018).

Post-combustion capture BECCS is still in the pilot and demonstration phase, although several projects are expected to start sequestering on the order of Mt a⁻¹ CO₂ from biomass combustion plants within this decade (Global CCS Institute 2019, 2020). Capture costs between 224 and 266 \$ t⁻¹ CO₂ have been estimated for BECCS based on post-combustion capture (IEA GHG 2009). However, retrofitting existing combustion and waste-to-energy plants with CCS could lower the CDR costs; sequestering the biogenic CO₂ generated in the energy recovery processes of kraft pulp mills could lead to 135 Mt a⁻¹ of negative CO₂ emissions (Kuparinen *et al* 2019) at a cost of 62–79 \$ t⁻¹ (Onarheim *et al* 2017). Similarly, the incineration of the organic fraction of municipal solid waste produced worldwide coupled with CCS could produce negative emissions at the Gt scale (Pour *et al* 2018).

Oxy-combustion is an alternative technology to post-combustion capture where the feedstock comes into contact with pure oxygen instead of air, thereby releasing a stream of highly concentrated CO₂, which only has to be cooled in order to remove the water vapour prior to compression and storage (Cabral

et al 2019). Although the electricity consumed in the air separation unit is considerable, this technology can improve the combustion efficiency (Cabral *et al* 2019), generating more energy per unit of biomass than BECCS based on post-combustion capture (Mac Dowell and Fajardy 2016) at a lower CDR cost (Bhave *et al* 2017), estimated to be 155 \$ t⁻¹ CO₂ (Cabral *et al* 2019). Notwithstanding this, the feasibility of oxy-combustion BECCS has not been demonstrated at the pilot scale yet (Cabral *et al* 2019).

In BECCS processes based on chemical looping combustion, the biomass feedstock reacts with the oxygen in a metal oxide, generating a gas stream consisting mostly of CO₂ and water with a low energy penalty. The reduced metal oxide is then transported to another reactor where it is regenerated with air (Adánez *et al* 2018). This technology has been tested at semi-industrial scale, but the complete conversion of the fuel is hard to achieve due to the formation and loss of char (Adánez *et al* 2018). Furthermore, a third reactor is needed to fully oxidise the gas, increasing the CDR costs, which could range between 171 and 200 \$ t⁻¹ CO₂ (Keller *et al* 2019). A novel variant of this technology, chemical looping with oxygen uncoupling—in which the fuel reacts with the oxygen released by the metal oxide—, could achieve carbon capture rates over 99%, but it is still in early development stages (Cormos 2017).

3.3.2.2. Gasification

Biomass can be partially oxidised with a gasifying agent (air, oxygen or steam) to generate high yields of synthesis gas (syngas), mainly composed of carbon monoxide and hydrogen. Part of the biomass feedstock is transformed into biochar, which is usually burnt to provide the energy required to sustain the endothermic gasification reactions. The gasification process also produces a liquid fraction of heavy hydrocarbons (tars), which must be removed to prevent equipment fouling and plugging (McKendry 2002).

Steam is used to increase the CO₂ and hydrogen concentration in the gas phase through the water-gas-shift reaction (WGSR). The produced CO₂ is subsequently captured via pressure swing adsorption, absorption or membrane separation processes. Given the high concentration of CO₂ in the gas, this pre-combustion capture method consumes less energy than the alternative post-combustion capture (Shahbaz *et al* 2021).

After the CO₂ separation, hydrogen can be compressed and used as fuel off-site, or directly oxidised to generate heat and electricity. BECCS based on integrated gasification combined cycles (IGCCs)—which deploy both gas and steam turbines—can achieve higher energy efficiencies than combustion-BECCS at the expense of higher capture costs, between 175 and 364 \$ t⁻¹ CO₂ (Bhave *et al* 2017). Approximately half of the biomass carbon content can be recovered from

the syngas, (Rhodes and Keith 2005) but higher CDR efficiencies could be achieved by capturing the CO₂ generated in the biochar combustion and gas turbine. Hence, the technical CDR potential of IGCC-BECCS could amount to 10.4 Gt a⁻¹ by 2050 (Koornneef *et al* 2012). On the other hand, coupling biomass gasification with fuel cells could potentially double the power produced by IGCCs (Sadhukhan *et al* 2010), but this technology is still in its infancy (Bhave *et al* 2017).

A range of biofuels—including methane, methanol and dimethyl ether—can be synthesised after adjusting the carbon monoxide to hydrogen ratio in the syngas via the WGS and removing the CO₂ (Gassner and Maréchal 2009, Sikarwar *et al* 2017). Notably, syngas can be converted into syn-crude via the Fischer–Tropsch (FT) process, and further refined to produce synthetic gasoline, diesel or jet fuel, which would allow sequestering 57–74% of the biogenic carbon in the feedstock (Kreutz *et al* 2020), with costs ranging between 368 and 524 \$ t⁻¹ CO₂ captured (Larson *et al* 2020). The CDR potential of BECCS based on gasification and subsequent methanation could amount to 3.5 Gt a⁻¹ CO₂-eq (Koornneef *et al* 2013), whereas the FT-BECCS pathway could sequester up to 5.8 Gt a⁻¹ CO₂-eq by 2050 (Koornneef *et al* 2012). However, these figures may substantially drop (below 0.1 Gt a⁻¹ CO₂-eq) if LUC emissions are factored in (Hanssen *et al* 2020).

Although the feasibility of biomass gasification has already been tested at large scale (e.g. *GoBiGas* biomass-to-methane demonstration plant, Larsson *et al* 2018) and even reached commercialisation (*Enerkem* waste-to-methanol plant, Enerkem 2022) these facilities do not include CCS.

3.3.2.3. Fast pyrolysis

Heating biomass in oxygen-free or oxygen-restricted environments triggers the pyrolytic reactions that yield bio-oil, biochar and pyrogas. Fast pyrolysis systems, which require high temperatures (around 500 °C) and short residence times (below 2 s), can maximise the production of bio-oil (Perkins *et al* 2018). Although bio-oil is composed of highly oxygenated hydrocarbons, it can be directly combusted to generate heat and power for stationary applications (Czernik and Bridgwater 2004). However, using it as a drop-in transportation fuel requires upgrading via hydrotreatment and hydrocracking processes, which can be partially carried out with the hydrogen derived from the pyrogas. These processes produce a mixture of lighter hydrocarbons with a low oxygen content (Peters *et al* 2015b).

Under fast pyrolysis conditions, approximately 48% of the biomass carbon content is distributed between the biochar and pyrogas (Schmidt *et al* 2019). These products can be burnt to provide the heat required by the system, releasing the carbon as CO₂ that can be subsequently captured and stored (Meerman and Larson 2017). Hence, we estimate the

maximum technical CDR potential of fast pyrolysis as 48% of the CDR potential of combustion-BECCS, i.e. 5 Gt a⁻¹ (Koornneef *et al* 2012). It has been estimated that the costs of this BECCS configuration (including bio-oil upgrading) equipped with MEA post-combustion capture could amount to 404 \$ t⁻¹ CO₂ (Meerman and Larson 2017).

Several companies (*ENSYN*, *btg bioliquids*) have successfully commercialised the fast pyrolysis technology (Perkins *et al* 2018), but we are not aware of any bio-oil projects integrating the combustion of biochar and pyrogas with CCS. The company *Charm Industrial* has developed an alternative CDR pathway whereby bio-oil is not used to deliver energy, but injected into geological reservoirs. They offer this CDR service at a price of 600 \$ t⁻¹ CO₂-eq (Charm Industrial 2021).

3.3.2.4. Hydrothermal liquefaction

Processing biomass in an aqueous medium at high temperatures and pressures (250 °C–374 °C and 40–220 bar) leads to the disintegration of the polymers in the biomass and the production of bio-crude (Elliott *et al* 2015). This bio-crude is more viscous, less dense and less oxygenated than pyrolysis bio-oil. It can directly substitute heavy fuel oil, but using the bio-crude as a transportation fuel requires a previous hydro-treatment process (Elliott *et al* 2015).

The hydrothermal liquefaction of biomass also generates biochar and gas—mainly composed of CO₂ and hydrogen (Lozano *et al* 2020). Part of the hydrogen required for the hydrotreatment can be sourced from this gas stream, whereas the remaining combustible gases can be burnt along with the biochar to provide heat to the system. Although the process is almost self-sufficient, an external supply of hydrogen and energy is needed (Lozano *et al* 2020).

Altogether, the gas and solid products contain 26–32% of the carbon in the biomass (Lozano *et al* 2020, SundarRajan *et al* 2020). Thus, capturing and sequestering this carbon could contribute to the removal of up to 3.3 Gt a⁻¹ CO₂-eq, estimated as a fraction of the CDR potential of combustion-BECCS in 2050 (Koornneef *et al* 2012). This is a conservative assumption because hydrothermal liquefaction does not require a drying pretreatment and therefore can process a wide range of biomass feedstocks with high moisture contents (Tekin *et al* 2014).

Currently, hydrothermal liquefaction is at pilot and demonstration scale, with companies such as *Licella* and *Genifuel* at the forefront (Lozano *et al* 2020). However, none of these facilities incorporate carbon capture units. The estimated cost of this technology ranges between 583 and 1365 \$ t⁻¹ CO₂ captured (Lozano *et al* 2020).

3.4. Chemical NETPs

In this type of NETPs, chemical reactions between GHGs and other compounds enable the GGR. Ocean

alkalinisation and CO₂ extraction from seawater, classified as marine NETPs, also belong to this category.

3.4.1. Terrestrial enhanced weathering

Natural weathering processes can be accelerated by grinding silicate and carbonate rocks to small grain sizes and spreading them in warm and humid regions (Strefler *et al* 2018, Zhang *et al* 2022). The rock materials dissolve in the presence of water, reacting with CO₂ to produce bicarbonate anions and liberating base cations (e.g. Ca²⁺ and Mg²⁺), as shown in reactions (R2)–(R4). Runoff transports these ions to the oceans, where their residence time exceeds 100 000 years. Under certain soil conditions, some cations precipitate to form carbonate minerals or react with acidic species (reactions (R5) and (R6)), releasing part of the sequestered CO₂ (Beerling *et al* 2020, Zhang *et al* 2022). It has been estimated that between 7.1 and 21.3 Gt a^{−1} CO₂ could be sequestered globally through the enhanced weathering of basalt before reaction (R5) reduces the CDR efficiency of this NETP (Zhang *et al* 2022).

Although preliminary EW field experiments have been conducted, the long-term effects of this practice have not been quantitatively measured yet (Kantola *et al* 2017). The weathering rate is site-specific, but it can be accelerated by reducing the grain sizes. Based on theoretical dissolution kinetics, Stefler *et al* (2018) estimated that the global CDR potential of applying 20 µm grains of basalt and olivine to croplands is 4.9 and 95 Gt a^{−1}, respectively. Nonetheless, experiments simulating real soil environments (Amann *et al* 2020, Kelland *et al* 2020, Buckingham *et al* 2022, Cipolla *et al* 2022) tend to yield lower weathering rates than those assumed by Strefler *et al* (2018).

The CO₂ sequestration capacity of olivine-rich rocks is better than that of basalts (1.1 vs 0.3 t CO₂ per t rock, Strefler *et al* 2018), but olivine-rich rocks have higher concentrations of toxic heavy metals—chromium and nickel—that could leach into the soil (Beerling *et al* 2018). Moreover, finely crushed respirable grains pose health risks, which are particularly relevant if olivine contains asbestos (Taylor *et al* 2016). On the other hand, basalt may contain larger amounts of other metals such as lead and zinc, which could also lead to toxicity impacts for humans (Cobo *et al* 2022b, Wang *et al* 2020). Nonetheless, the soil application of basalt can help improve soil fertility—due to its greater phosphorus content—, hence boosting the storage of carbon in biomass (Goll *et al* 2021). Overall, this practice addresses the need to fertilise the soil with silica and other nutrients lost by harvesting (Beerling *et al* 2018), and to offset the effects of soil and ocean acidification (Beerling *et al* 2018, Vakilifard *et al* 2021). While enhanced weathering can partially abate the nitrous oxide emissions derived from the application of nitrogen fertilisers (Beerling *et al* 2018, Blanc-Betes *et al* 2021), applying inorganic nitrogen

fertilisers concurrently with the crushed minerals may be counterproductive, since in that case the weathering may be driven by nitric acid instead of the dissolved CO₂ (Hartmann *et al* 2013).

Costs between 81 and 211 \$ t^{−1} CO₂ captured with basalt, and 63 \$ t^{−1} CO₂ captured with dunite (mainly composed of olivine) have been reported (Strefler *et al* 2018, Beerling *et al* 2020). Deploying alkaline wastes as an alternative feedstock could help reduce the costs and improve the CDR potential of this NETP (Renforth 2019), (Bullock *et al* 2021, 2021).

3.4.2. Direct Air Capture

Direct air capture (DAC) enables the extraction of CO₂ from the atmosphere. It can only be considered a NETP (DACCS) if it is coupled with CCS. With the exception of DAC systems based on physical adsorption (Shekhah *et al* 2014), cryogenic (Agee *et al* 2013) or membrane separation processes (Castel *et al* 2021), DAC technologies rely on the chemical reaction of CO₂ with liquid or solid sorbents.

Multiple sorbents and system configurations have been proposed (Sanz-Pérez *et al* 2016), e.g. Metal Organic Frameworks (Sinha *et al* 2017) or electro-swing adsorption systems (Voskian and Hatton 2019)—the latter showing a low energy demand, i.e. 2.3–2.6 GJ t^{−1} CO₂ captured (Hemmatifar *et al* 2022, Seo *et al* 2022). Nevertheless, the majority of these systems remain either a theoretical proposition or a laboratory prototype (Erans *et al* 2022).

Climeworks operates the world's largest DACCS facility to date, with a CDR capacity of 4 kt a^{−1}, and they have announced the construction of a plant nine times larger (Climeworks 2021, 2022). On the other hand, Carbon Engineering is building a plant with a CDR capacity of 0.5–1 Mt a^{−1} (Carbon Engineering 2021), whereas CarbonCapture Inc. is planning for a 5 Mt a^{−1} project (CarbonCapture 2022). The technologies deployed by these commercial plants can be classified into two main groups (Fasihi *et al* 2019):

- High temperature liquid sorbent DAC (HTLS-DAC). CO₂ is absorbed into a basic solution, the regeneration of which requires high temperature heat. Carbon Engineering commercialises HTLS-DAC.
- Low temperature solid sorbent DAC (LTSS-DAC). CO₂ is adsorbed onto a solid sorbent that can be regenerated by temperature swing adsorption (TSA) with low temperature heat (80–100 °C), or by moisture swing adsorption (MSA). Climeworks, Global Thermostat and CarbonCapture Inc. are using this technology, and they rely on TSA.

Carbon Engineering's HTLS-DAC is based on two parallel cycles. The CO₂ entering the air contactor reacts with potassium hydroxide, forming potassium

carbonate. In the regeneration cycle, potassium carbonate reacts with a calcium hydroxide solution, producing calcium carbonate, which is calcined at 900 °C to release a pure stream of CO₂. The byproducts of these reactions are used as reactants in other stages of the process, thereby closing the loop. This process consumes 6.57–8.81 GJ t⁻¹ CO₂, a substantial energy demand compared to the thermodynamic minimum, approximately 0.5 GJ t⁻¹ (APS 2011). The current capture cost of this technology is roughly 300 \$ t⁻¹ (Direct Air Capture Summit 2021), but it has been estimated that the levelised CO₂ capture cost of scaled-up systems could range between 99 and 245 \$ t⁻¹ (Keith *et al* 2018). A variant of the *Carbon Engineering's* process where the solvent regeneration is carried out through bipolar membrane electrodialysis would allow reducing the energy consumed in the regeneration phase by 30% at the expense of substantially increasing the total costs (Sabatino *et al* 2020).

In LTSS-DAC systems, as air passes through a filter, CO₂ chemically binds to the sorbent. Once the solid is saturated, CO₂ is desorbed with low-temperature heat. *Climeworks* has patented sorbent materials based on amine-functionalised fibrillated cellulose (Maluszynska-Hoffman *et al* 2017) and potassium carbonate (Vargas *et al* 2019). Although the cost of LTSS-DACCS is roughly 600 \$ t⁻¹ (Fuss *et al* 2018), *Climeworks* expects this cost to drop to about 100 \$ t⁻¹ by 2030 (Beuttler *et al* 2019). The current total energy consumption—electricity and heat—of this DACCS configuration currently amounts to 14 GJ t⁻¹ CO₂, although scaled-up systems are expected to reduce the energy demand by half in the future (Deutz and Bardow 2021). Moreover, the low temperatures required to regenerate the sorbent allow the use of waste heat or heat derived from renewable geothermal energy. The carbon capture efficiencies of the two first commercial *Climeworks* plants are 85.4% and 93.1% (Deutz and Bardow 2021).

Another LTSS-DAC prototype deploys an anionic resin (amine ligands attached to polystyrene) that adsorbs CO₂ and releases it when it is exposed to moisture, following an MSA cycle (Lackner 2009). This device has been named ‘artificial tree’ because it does not require an external energy input to drive air through the filters or dry them in the regeneration stage. This feature results in a remarkably low energy consumption, between 1.36 and 2.25 GJ t⁻¹ CO₂ (Goldberg *et al* 2013, van der Giesen *et al* 2017). However, its reliance on wind conditions makes this technology's performance highly dependent on the local weather (van der Giesen *et al* 2017). A capture cost of 110 \$ t⁻¹ has been estimated for this NETP (McGlashan *et al* 2012).

These HTLS- and LTSS-DAC systems deploy synthetic sorbents that improve the CO₂ uptake

kinetics with respect to mineral sorbents. Nevertheless, slower DAC processes based on the successive calcination and carbonation cycles of mineral sorbents are gaining increasing attention. McQueen *et al* (2020) proposed to sequester the CO₂ generated in the calcination of mineral carbonates, and to subsequently spread the remaining metal oxides over land. In the presence of water, these are transformed into metal hydroxides, which subsequently react with atmospheric CO₂ to produce carbonates. After the carbonation process, the generated carbonates are collected and calcined again. Experimental results indicate that only 3–18% of the spread metal oxides would react in one year (Rausis *et al* 2022). The cost of this NETP could range between 47 and 161 \$ t⁻¹ CO₂ captured (McQueen *et al* 2020).

The company *Heirloom* (OpenAir 2022b) is developing a prototype passive air contactor (i.e. not reliant on forced air) that can carbonate 85% of the mineral sorbent—hydrated calcium oxide—in 2.5 days, with an energy demand (stemming mainly from the calcination process) of 5.4 GJ t⁻¹ CO₂ (McQueen *et al* 2022). *Calcite Carbon Removal* is also developing a technology based on calcination/ambient carbonation cycles (8 Rivers 2022). The CO₂ capture costs of mineral sorbent DAC using passive calcium hydroxide structures has been estimated to be 140–340 \$ t⁻¹ (Abanades *et al* 2020).

The general agreement in the literature is that DAC costs will drop as the installed capacity increases, following technology learning curves similar to those of photovoltaic panels and batteries (Breyer *et al* 2019, Fasihi *et al* 2019, Realmonte *et al* 2019, Baker *et al* 2020, 2020, Young *et al* 2022). Lackner and Azarabadi estimated that the capture of 1.5 Mt CO₂—which would require an investment of approximately 200 M\$—could bring down the cost of DAC below 100 \$ t⁻¹ (Lackner and Azarabadi 2021). They also suggest that small modular units could be more effective than high-output plants at accelerating the learning rate of this technology (Lackner and Azarabadi 2021).

Furthermore, because of the ability of DAC plants to be ramped up within minutes, they could benefit from the excess generation of cheap renewable electricity (Wohland *et al* 2018). Nonetheless, Breyer *et al* (2019) pointed out that a constant supply of electricity is needed to reduce costs; i.e. it is unlikely that future DAC plants will run solely on excess renewable power. In fact, the results of integrated assessment models suggest that the scale-up rate of DAC systems could be constrained by their energy consumption, which could reach up to 25% of global energy demand by the end of the century (Realmonte *et al* 2019). Other factors that could act as potential deployment barriers for DAC are the pollution associated with the large-scale production of the sorbents required for LTSS-DACCS (Realmonte *et al* 2019), and the high

water demand of HTLS-DACCS (Fuhrman *et al* 2020), LTSS-DACCS based on MSA (van der Giesen *et al* 2017) and DACCS relying on mineral sorbents (Mcqueen *et al* 2022). By contrast, some of the adsorbents used in LTSS-DACCS relying on TSA can co-adsorb water from the air; *Climeworks'* DAC configuration can produce up to 2 t water per t CO₂, depending on weather conditions and humidity (Fasihi *et al* 2019).

3.4.3. GHG degradation

The maturity level of technologies and practices aiming at the transformation of non-CO₂ GHGs into substances with lower global warming potentials is still low. Hence, further research is needed to establish the technical feasibility and effectiveness of these NETPs.

3.4.3.1. Methane oxidation

The atmospheric concentration of methane is low compared to CO₂ (1.9 versus 410 ppm in 2019, IPCC 2021), but its global warming potential is 34 and 86 times that of CO₂ for 100- and 20 year time horizons, respectively (Myhre *et al* 2013). Oxidising 1 Gt methane would prevent an increase in the global surface temperature of 0.21 ± 0.04 °C (Abernethy *et al* 2021). Furthermore, the averted formation of tropospheric ozone would entail benefits for human health and ecosystems.

The minimum thermodynamic energy required to separate methane from ambient air is almost five times that of CO₂ per unit mass (Boucher and Folberth 2010). However, expressed per t CO₂ equivalent, the minimum energy needed to separate atmospheric methane is 7 and 19 times lower than that of CO₂, considering 100- and 20 year time horizons (Jackson *et al* 2021). Moreover, the combustion of the recovered methane could provide 28% of the energy required for the separation (Boucher and Folberth 2010). Some zeolites and porous polymeric networks have been preliminarily identified as promising materials for methane capture because of their selectivity and sorption capacities (Kim *et al* 2013, Jackson *et al* 2019). Nonetheless, the feasibility of deploying copper-doped zeolites to oxidise methane at atmospheric concentrations has recently been demonstrated (Brenneis *et al* 2021), which could render the need to develop an intermediate separation step unnecessary.

Bioreactors exploiting the natural capability of methanotrophs to metabolise methane at atmospheric concentrations could be an alternative pathway to oxidise atmospheric methane. However, this strategy has only been proven for methane concentrations of about 300 ppm in the animal husbandry sector (Stolaroff *et al* 2012); it has been estimated that the average atmospheric methane concentration is too low to enable the survival of the methanotrophs within the bioreactor (Yoon *et al* 2009).

3.4.3.2. Photocatalytic degradation

The application of photocatalytic coats on the walls of buildings with enclosed animals has been demonstrated to be an effective measure to oxidise methane (de Richter *et al* 2017), but relatively modest reduction rates of nitrous oxide have been achieved under similar conditions (Ming *et al* 2016). The costs of applying photocatalysis on surfaces could be as low as 166 \$ t⁻¹ CO₂-eq by 2030 (Ming *et al* 2022). On the other hand, chlorofluorocarbons, and hydrochlorofluorocarbons have been successfully degraded to halogens, acid halides, and CO₂ in experimental photocatalytic reactors (de Richter *et al* 2016).

Another method proposed to photocatalytically degrade non-CO₂ GHGs is the injection of iron salt aerosols with a chloride content in the troposphere. Solar radiation would promote the formation of chlorine radicals, which could subsequently oxidise methane, VOCs, and carbon black (Dietrich Oestle *et al* 2017). Furthermore, the deposition of soluble iron over soils and oceans could lead to increased terrestrial and marine primary productivity (Ming *et al* 2021). Costs between 2 and 54 \$ t⁻¹ CO₂-eq have been estimated for this NETP (Ming *et al* 2022).

Relative to a baseline without GGR, this NETP can avert damage to human health and ecosystems through two pathways: the decrease in tropospheric ozone formation (due to the oxidation of methane and VOCs), and the prevention of stratospheric ozone depletion, linked to chlorofluorocarbons and hydrochlorofluorocarbons (Huijbregts *et al* 2017).

4. Critical assessment of NETPs and sequestration methods

Identifying the most appealing NETPs is critical to design optimal GGR pathways. To this end, we assess the performance level of the reviewed NETPs and sequestration processes based on five KPIs estimated with literature data:

- The technology readiness level (TRL)—a scale from 1 to 9 that rates the maturity of a given technology or practice.
- The maximum annual GGR potential.
- The cost of removing 1 t CO₂-eq.
- The number of negative side-effects. We identify the potential adverse consequences of NETPs—including unexpected impacts on the climate system, substantial resource use and pollutants emissions—in table 1.
- The number of co-benefits (i.e. positive side-effects) compiled in table 1, e.g. decrease in ocean acidification, the generation of co-products or improved crop yields.

We discuss the limitations of the selected KPIs in appendix B of the supplementary information. While more detailed analyses including a wider range

Table 1. Potential side-effects of the deployment of NETPs and CO₂ sequestration methods in accordance with the reviewed literature (positive effects: green cells, negative effects: pink cells, and either positive or negative effects, depending on location and system characteristics: blue cells).

	Delayed GGR	Release of stored C ^a	GHG emissions	Other climate effects ^b	Pollutant emissions	Ocean acidification	LUC	High fresh-water demand	High energy demand	Crop productivity	Biodiversity impacts ^c	Human health impacts ^d	GGR co-products ^e
TERRESTRIAL NETPs	Wood burial or storage.	Red	ΔSOC, VOCs.	Blue	Nutrient lock-up.	Red	Red	Red	Red	Blue	Red	Blue	Wood/fibers.
	Building with wood and fibers.		CH ₄ , N ₂ O.		Nutrients.								
	Biochar amendment.		ΔSOC, VOCs.		NO _x , NH ₃ , etc.								
	SCS.		N ₂ O, CH ₄		Nutrients, metals...								
	Forestation.		VOCs.		Nutrients.								
MARINE NETPs	Downwelling/Upwelling.	≈ 1 year.	CO ₂ .	Cooling.	Red	Red	Red	Red	Red	Red	Red	Air/soil emissions.	Biochar.
	Ocean fertilisation.	≈ 1 year.	CH ₄ , N ₂ O.	< Rain.									
	Blue carbon.	≈ 1 year.	CH ₄ , N ₂ O.	Green	DMS, nutrients.								
	Ocean alkalimisation.	s.t. grain size.	CH ₄ , N ₂ O.		DMS, nutrients.								
	CO ₂ extraction from seawater.	Reactions (R5)/(R6). ≈ 1 year.			Particles/metals.								
BECCS NETPs	Terrestrial BECCS.	Red	ΔSOC, VOCs.	Blue	Nutrients.	Red	Red	Red	Red	Blue	Red	Air/water emissions.	Energy.
	Algal BECCS.		CH ₄ , N ₂ O.		DMS, nutrients.								

(Continued.)

Table 1. (Continued.)

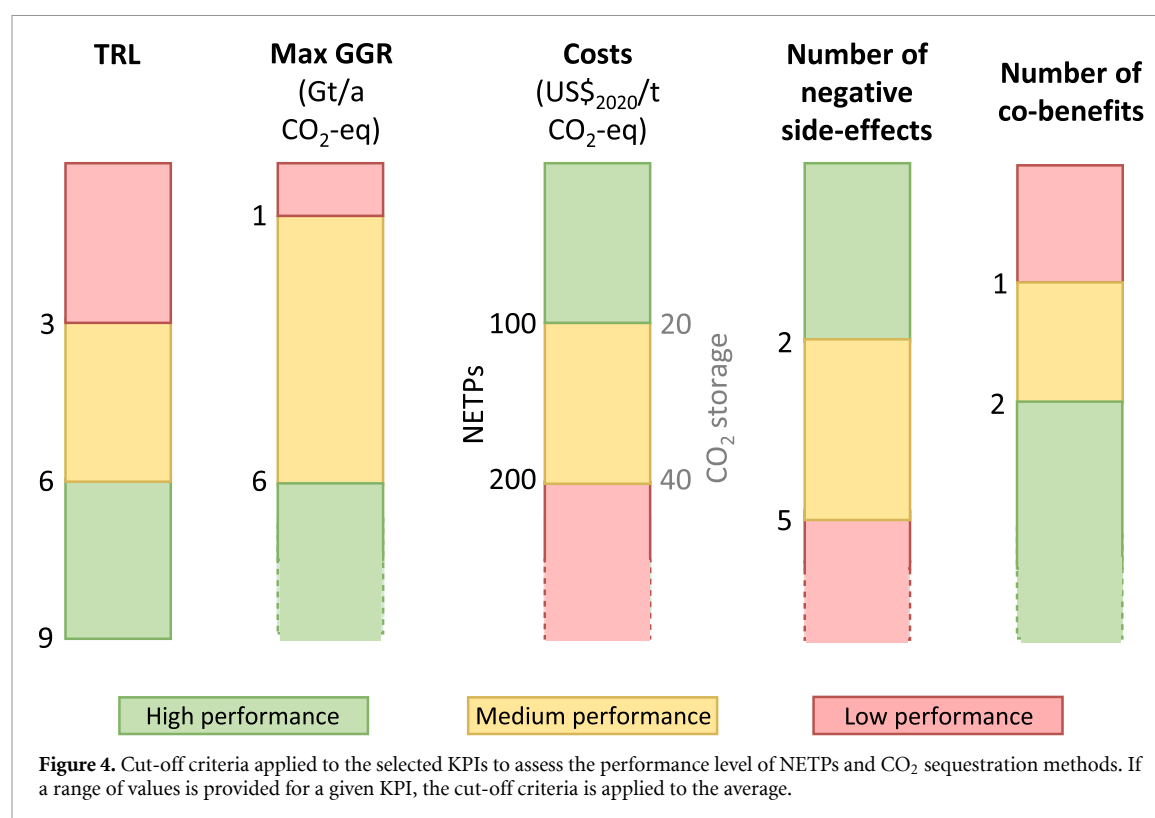
	Delayed GGR	Release of stored C ^a	GHG emissions	Other climate effects ^b	Pollutant emissions	Ocean acidification	LUC	High fresh-water demand	High energy demand	Crop productivity	Biodiversity impacts ^c	Human health impacts ^d	GGR co-products ^e
CHEMICAL NETPs													
Terrestrial EW.	s.t. grain size.	Reactions (R5)/(R6).	N ₂ O.		Particulates/metals.							Air/water emissions.	
Mineral sorbent DACCS (on land).	Years.												
Mineral sorbent DACCS (air contactor).	Days.												
GHG degradation.			Degraded products.	Fertilisation ^g .	Degraded products.							Avoided O ₃ formation/depletion.	
LTSS-DACCS (MSA).					Adsorbent.								
LTSS-DACCS (TSA).					Adsorbent.			Water extraction.					
HTLS-DACCS.													

(Continued.)

Table 1. (Continued.)

	Delayed GGR	Release of stored C ^a	GHG emissions	Other climate effects ^b	Pollutant emissions	Ocean acidification	LUC	High fresh-water demand	High energy demand	Crop productivity	Biodiversity impacts ^c	Human health impacts ^d	GGR co-products ^e
CO ₂ STORAGE													
Ex situ mineral carbonation.					Particles/metals.							Air/water emissions.	Metals.
Hydrate-based sequestration.					Additives.							Water emissions.	
Direct ocean injection.													
In situ mineral carbonation.												Seismicity.	
Geological sequestration.												Seismicity.	
EW of carbonates.													
Submarine storage in vessels.		Reactions (R5)/(R6).			Particles/metals.							Air/water emissions.	

^a Additionally, the amount of sequestered CO₂ is hard to meter.
^b Changes in the surface albedo, evaporation and evapotranspiration rates.
^c Excluding the biodiversity co-benefits of GGR (Hanssen et al 2021, Cobo et al 2022a).
^d Excluding the health co-benefits of GGR (Cobo et al 2022a).
^e Revenues from the sale of co-products/services, and avoided climate change impacts due to the substitution of other products/services.
^f Only downwelling.
^g Only iron salt aerosols; soil/ocean fertilisation.



of KPIs would be possible, they would require large amounts of data currently unavailable for many NETPs and sequestration processes. Hence, the KPIs above are intended to represent a good compromise between data availability and performance coverage. To facilitate the interpretation of the KPIs, we define three intervals for each of them—denoting low-, medium- and high-performance levels—according to the cut-off criteria shown in figure 4. NETPs with low potential in terms of their TRLs ($TRL \leq 3$) have not been validated in the laboratory yet. In contrast, high TRLs ($TRL \geq 7$) indicate that a system prototype has been at least demonstrated in an operational environment (European Commission 2014). The selection of the GGR cut-off criteria is based on the median annual CDR rates across 1.5 °C scenarios with no or limited overshoot for years 2030 and 2050; i.e. 1 and 6 Gt a⁻¹, respectively (IPCC 2022).

Concerning the economic performance, we consider 100 \$ t⁻¹ CO₂ as the threshold below which NETPs show a high economic potential, in line with other works (National Academies of Sciences Engineering and Medicine 2019, Lackner and Azarabadi 2021), and deem costs doubling that value uneconomical. We set 20 \$ t⁻¹—the highest cost reported for the most mature sequestration method (geological sequestration), according to the National Academies of Sciences Engineering and Medicine (2019)—as the lower bound below which the sequestration processes show a high economic potential, and consider that costs above twice that

value relegate a sequestration method to the low potential category in economic terms.

Regarding the number of negative side-effects—ranging from 0 to 7—, values lower than or equal to 2 denote low impact, whereas KPI values greater than or equal to 6 imply high impact. Finally, we define three bands for the co-benefits KPI: 0, 1 and 2 or more, indicating a low, medium and high number of positive side-effects, respectively. The number of negative and positive side-effects does not account for their magnitude or the probability of them occurring, and therefore, the implications of these KPIs are more uncertain. Following a more conservative approach, we applied stricter cut-off criteria to the side-effects KPIs in appendix C of the supplementary information.

Table 2 shows the performance level of NETPs and CO₂ sequestration methods according to the five studied KPIs. We include NETPs for which at least three KPIs are available, neglecting some configurations with very incipient development levels, e.g. certain DACCS modalities. Here we compare the overall performance of the main NETPs groups (terrestrial, marine, BECCS and chemical) and storage processes based on the KPIs compiled in table 2, with a special emphasis on the discussion of the side-effects listed in table 1.

We first focus on the NETPs reliant on terrestrial biomass. While the high TRLs and low costs of terrestrial NETPs could facilitate their quick implementation, the BECCS KPIs predominantly

Table 2. Performance level of NETPs and CO₂ sequestration methods according to the selected KPIs (high performance: green cells, medium performance: yellow cells, low performance: pink cells). Note that the KPIs of NETPs and sequestration methods with low TRLs may not be as robust as those of the more consolidated ones, given their incipient development levels.

NETPs/CO ₂ sequestration processes		TRL	Max. GGR (Gt a ⁻¹ CO ₂ -eq)	Cost (US\$ ₂₀₂₀ t ⁻¹ CO ₂ -eq)	No of Negative effects	No of positive effects
TERRESTRIAL	Wood burial or storage	4–6 ^a	3.7–11 ^{1,b}	9–33 ¹	6	0
	Building with wood	8–9 ²	0.040–2.5 ^{3,b,c}	Low ²	6	1
	Biochar amendment	7 ⁴	0.44–2.6 ^{5,b}	32–127 ⁶	5	1
	SCS	6–8 ^a	2.0–5.0 ^{6,b,c,d}	0–105 ⁶	5	2
	Forestation	8–9 ⁷	2.6–5.6 ^{8,9,b,e}	5–53 ⁶	5	0
MARINE	Downwelling	1–2 ^a	0.035 ^{10,b,f}	258–5,826 ¹⁰	7	1
	Upwelling	1–3 ^a	0.050–0.10 ^{11,b}	n/a	6	1
	Ocean fertilisation (Fe)	1–4 ²	3.6 ¹²	519 ¹³	6	1
	Ocean fertilisation (N and P)	2–3 ²	5.5 ¹⁴	24 ¹⁵	6	1
	CO ₂ extraction from seawater	2–3 ^a	g	393–637 ^{16,h}	3	0
	Coastal blue carbon	5–6 ²	1.2 ^{17,b,i,j}	>200 ¹⁷	5	1
	Ocean storage of terrestrial biomass	1–2 ^a	6.8 ^{18,k}	117 ¹⁸	n/a	n/a
	Coastal EW (olivine)	3–4 ^a	0.0064–0.46 ^{19,j,l}	34–100 ^m	5	1
	Electrochemical EW (acidity pumping)	1–2 ^a	3.7–11 ^{20,n}	89–285 ²⁰	5	1
	Electrochemical EW (alkalinity addition)	3–4 ^a	6.6–12 ^{21,22,j,o}	68–614 ^{26,p}	5	1
	Direct alkalinisation	3–4 ²	6.6–12 ^{21,22,j}	82–181 ^{23,q}	5	1
	Macroalgae farming and sinking	3–5 ^a	12 ^{24,b,j}	150–200 ²⁵	5	1
BECCS (terrestrial biomass)	Hydrothermal liquefaction	4–5 ^a	0.70 ^r –3.3 ^{b,c,s,t}	583–1,365 ^{27,h,u}	5	1
	Gasification-FT	4–5 ^a	0.10 ^r –5.8 ^{28,29,b,c,t}	368–524 ^{30,h,u}	5	1
	Fast pyrolysis	5 ^{a,v}	1.2 ^r –5.0 ^{b,c,t,w}	404 ^{31,u}	5	1
	Anaerobic digestion	8 ^{a,v}	2.7 ^{32,b,c}	235–557 ^{33,u}	5	1
	IGCC	5 ³⁴	2.5 ^{r,x} –10 ^{28,29,b,c,t}	175–364 ^{35,h,u}	5	1
	Chemical looping combustion	6 ³⁶	2.5 ^r –10 ^{28,29,b,c,t,x}	171–200 ^{37,u}	5	1
	Oxy-combustion	5 ³⁵	2.5 ^r –10 ^{28,29,b,c,t,x}	155 ^{38,u}	5	1
	Combustion	7 ^a	2.5 ^r –10 ^{28,29,b,c,t}	224–266 ^{39,h,u}	5	1
	Ethanol fermentation	8 ^a	0.0 ^r –1.0 ^{28,29,b,c}	21–185 ^{6,h,u}	5	1
CHEMICAL	Methane oxidation	1–2 ^a	g	n/a	2	2
	Photocatalytic coats	3–4 ^a	n/a	166 ⁴⁰	2	2
	Iron salt aerosols	1–2 ^a	n/a	2–54 ⁴⁰	2	2

(Continued.)

Table 2. (Continued.)

NETPs/CO ₂ sequestration processes	TRL	Max. GGR (Gt a ⁻¹ CO ₂ -eq)	Cost (US\$ ₂₀₂₀ t ⁻¹ CO ₂ -eq)	No of Negative effects	No of positive effects
Terrestrial EW (basalt)	3–5 ⁴¹	4.9 ^{42,y}	81–211 ^{42,43}	6	3
Terrestrial EW (olivine)	3–5 ⁴¹	95 ^{42,y}	63 ⁴²	6	3
Mineral sorbent DACCS (air contactor)	4 ^{44,45}	g	140–340 ^{46,h}	2	0
Mineral sorbent DACCS (on land)	1–2 ⁴⁷	g	47–161 ^{47,h}	2	0
LTSS-DACCS (MSA)	3–4 ^a	g	110 ^{48,h}	2	0
LTSS-DACCS (TSA)	7 ⁴⁹	g	≈600 ⁶	2	1
HTLS-DACCS	7 ⁴⁹	g	99–300 ^{50,51,h}	2	0
CO ₂ STORAGE					
<i>Ex situ</i> mineral carbonation	4 ⁵²	g,z	68 ⁵³	4	1
EW of carbonates	4–5 ⁵⁴	g,z	4–85 ⁵⁵	4	1
Hydrate-based sequestration	4–5 ⁵⁶	g,z	n/a	4	0
Direct injection into the ocean	1–2 ^a	13 ^{57,z,aa}	16–21 ⁵⁸	2	0
Submarine storage in vessels	1–2 ^a	g,z	18 ⁵⁹	0	0
<i>In situ</i> mineral carbonation	7 ⁴	g,z	19 ⁶⁰	2	0
Geological sequestration	7–9 ⁴⁹	g,z	1–20 ⁶¹	1	0

^a Authors' assessment, based on the reviewed literature. ^bMax CDR not indicative of climate effect, which is also correlated with the surface albedo change and/or evapotranspiration. ^cBy 2050. ^dThese CDR rates can only be sustained for 2–3 decades in mineral soils, until SOC levels reach a new equilibrium (West and Six 2007, Paustian *et al* 2019). ^eCDR rates decline to zero when forests reach maturity (Houghton *et al* 2015). ^fConsidering 1 Mm³ s⁻¹ of seawater. ^gLimited by resource use and/or scale-up rates. ^hExcluding sequestration costs. ⁱEcosystem restoration. ^jAssuming a constant CDR rate between 2020 and 2100. ^kConsidering crop residues. ^lTo limit risks for benthic biota. ^m34–50 \$ t⁻¹ of net CO₂ for cumulative CDR above 100 Mt, costs below 100 \$ t⁻¹ for 1–10 Mt (T. Green, personal communication, July 2022). ⁿTo limit the pH decrease in the deep water to 0.2. ^oCDR potential estimated for the direct addition of alkaline materials to the ocean. ^pEstimated with the data reported by Rau *et al* (2018), excluding revenues from the sale of hydrogen or the use of the produced hydrogen within the process. ^qVia calcination of limestone and dolomite. ^rConsidering only lignocellulosic crops and LUC emissions amortised over 30 years. ^sEstimated as 26–32% (Lozano *et al* 2020, SundarRajan *et al* 2020) of the CDR potential of combustion-BECCS. ^tMunicipal organic waste could increase the estimated CDR potential of this BECCS route. ^uCDR costs excluding energy revenues. ^vBioenergy pathway commercially available, but not integrated with CCS. ^wEstimated as 48% (Schmidt *et al* 2019) of the CDR potential of combustion-BECCS. ^xAssuming the carbon sequestration efficiency of post-combustion BECCS. ^yOn croplands. ^zFor the CO₂ storage processes, this KPI refers to the maximum sequestration potential. ^{aa}To limit the pH decrease to 0.1.

¹ Zeng *et al* (2013). ² McLaren (2012). ³ Churkina *et al* (2020). ⁴ NIRAS (2019). ⁵ Werner *et al* (2022). ⁶ Fuss *et al* (2018). ⁷ Lomax *et al* (2015). ⁸ Favero *et al* (2020). ⁹ Austin *et al* (2020). ¹⁰ Zhou and Flynn (2005). ¹¹ Koweek (2022). ¹² Zahariev *et al* (2008). ¹³ Harrison (2013). ¹⁴ Harrison (2017). ¹⁵ Jones (2014). ¹⁶ Eisaman *et al* (2018). ¹⁷ Gattuso *et al* (2021). ¹⁸ Strand and Benford (2009). ¹⁹ Flipkens *et al* (2021). ²⁰ Tyka *et al* (2022). ²¹ Lenton *et al* (2018). ²² Feng *et al* (2017). ²³ Renforth *et al* (2013). ²⁴ Wu *et al* (2022). ²⁵ Calacanis (2020). ²⁶ Rau *et al* (2018). ²⁷ Lozano *et al* (2020). ²⁸ Koornneef *et al* (2012). ²⁹ Hanssen *et al* (2020). ³⁰ Larson *et al* (2020). ³¹ Meerman and Larson (2017). ³² Koornneef *et al* (2013). ³³ IEA GHG (2013). ³⁴ Parkinson *et al* (2019). ³⁵ Bhave *et al* (2017). ³⁶ Adánez *et al* (2018). ³⁷ Keller *et al* (2019). ³⁸ Cabral *et al* (2019). ³⁹ IEA GHG (2009). ⁴⁰ Ming *et al* (2022). ⁴¹ Haszeldine *et al* (2019). ⁴² Streffler *et al* (2018). ⁴³ Beerling *et al* (2020). ⁴⁴ 8 Rivers (2022). ⁴⁵ Erans *et al* (2020). ⁴⁶ Abanades *et al* (2020). ⁴⁷ McQueen *et al* (2020). ⁴⁸ McGlashan *et al* (2012). ⁴⁹ Bui *et al* (2018). ⁵⁰ Keith *et al* (2018). ⁵¹ Direct Air Capture Summit (2021). ⁵² Slotte (2017). ⁵³ Gerdemann *et al* (2007). ⁵⁴ Kirchner *et al* (2020). ⁵⁵ Rau *et al* (2007). ⁵⁶ Rehman *et al* (2021). ⁵⁷ Adams and Caldeira (2009). ⁵⁸ Caldeira *et al* (2005). ⁵⁹ Caserini *et al* (2017). ⁶⁰ Gislason and Oelkers (2014). ⁶¹ National Academies of Sciences Engineering and Medicine (2019).

reflect intermediate performance levels. However, the BECCS cost estimates, which exclude revenues from the sale of energy, are quite conservative; allocating the costs among the CDR and energy supply services could make these NETPs more attractive from the economic viewpoint. Moreover, the produced bioenergy vectors can replace non-renewable energy, generating economic revenues and preventing the emission of more GHGs. Despite its higher costs, BECCS offers a greater annual sequestration potential than terrestrial NETPs. Furthermore, the risk of CO₂ leakages is reduced with BECCS, since the permanence of the carbon sequestered in the biosphere by terrestrial NETPs is subject to natural and human-made threats (pests, fires, etc).

Two terrestrial NETPs, namely SCS and biochar amendment in agricultural lands, may improve soil fertility. Conversely, BECCS and terrestrial NETPs based on forests and plantations are constrained by land availability, and therefore, they may compete between them and with the food system, posing a risk for food security. The use of water and fertilisers to increase crop yields further threatens the sustainability of the NETPs deploying terrestrial biomass. Additionally, the replacement of native species with productive monocultures will result in biodiversity losses, although proper forestation practices can bring biodiversity benefits.

These NETPs can also generate other impacts on the climate system. LUC can lead to either GHG emissions or SCS—the extent of which depends on the soil, environment, species and LUC characteristics—, whereas the albedo shifts and GHGs emitted by trees may offset the climatic benefits of the NETPs relying on terrestrial biomass. Algal BECCS could avoid some of these side-effects, but more research is needed to thoroughly analyse its deployment potential and sustainability implications.

Marine NETPs are still quite immature, with those involving physical operations (downwelling and upwelling) performing poorly in terms of most KPIs. The marine NETPs seeking to stimulate the biological productivity of marine organisms (blue carbon and ocean fertilisation) could contribute to climate change mitigation by increasing the ocean albedo, although the emissions of other GHGs could counteract the prevented global warming. Furthermore, other environmental issues may arise, namely increased ocean acidification and eutrophication linked to the release of nutrients, which may impact marine ecosystems. One of the main risks of enhancing the ocean's carbon sink capacity is the potential release of the stored carbon.

Although the environmental impacts of the ocean alkalisation practices are still quite uncertain, their ability to combat ocean acidification may pose an advantage over the other marine NETPs. Nevertheless, as with terrestrial EW, there is a lag between the time the alkaline materials are spread and the

carbon sequestration. Accelerating the CO₂ sequestration rates of ocean alkalisation and terrestrial EW requires small particle sizes, which entail a high energy consumption. However, the energy demand of coastal EW may be lower since the particle size can be reduced through natural processes. The rock extraction and grinding operations can be a source of particulate matter, which altogether with the potential leaching of heavy metals, can damage human health and biodiversity. On the other hand, the theoretical CDR capacity of terrestrial EW is substantial. Its potential co-benefits include improved crop productivity, the abatement of nitrous oxide emissions, and a reduction in soil and ocean acidification. The costs of this practice vary widely and depend on the type of rock used.

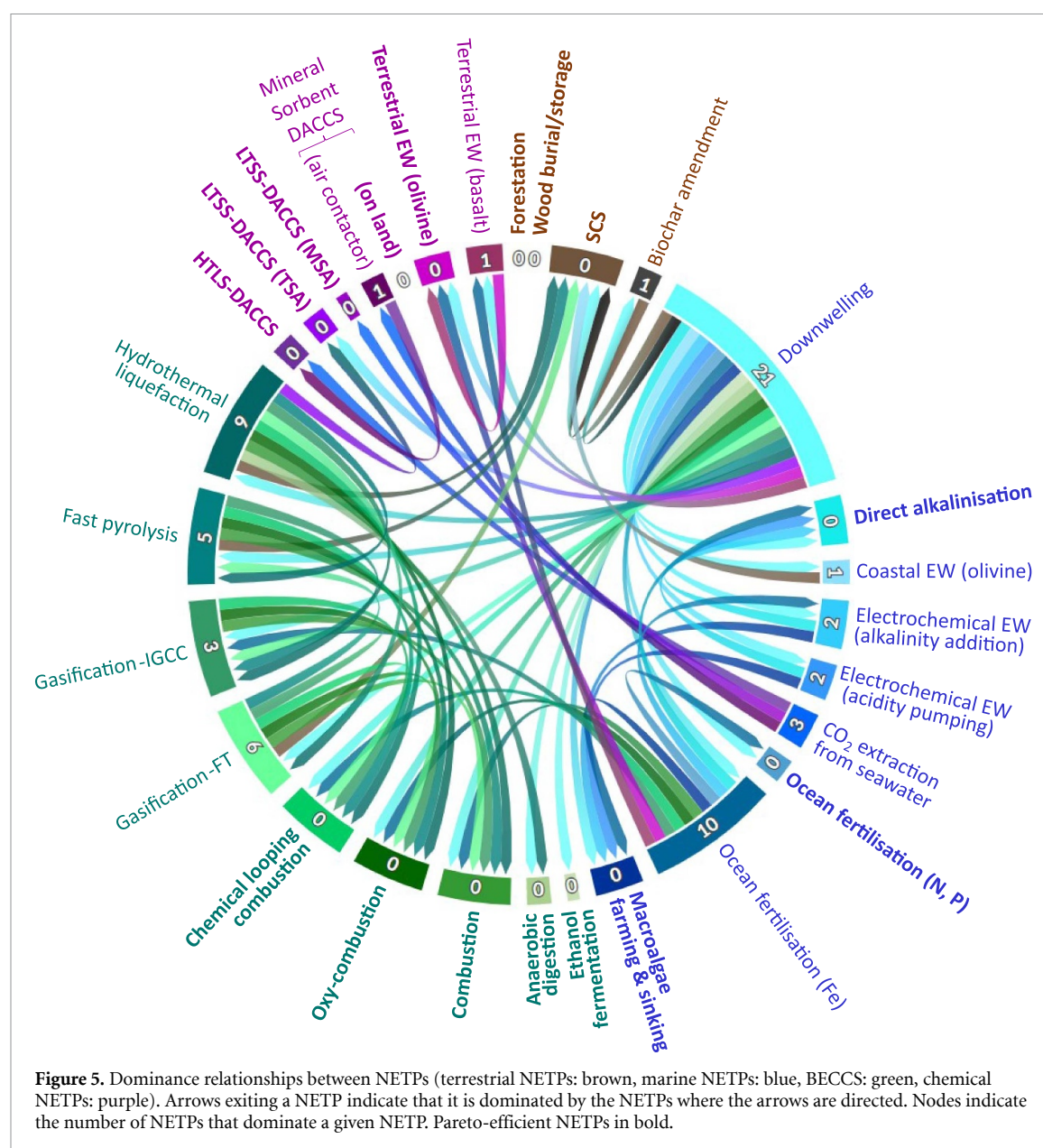
With the exception of terrestrial EW, the NETPs classified as chemical attain the lowest number of negative side-effects. Although DACCS cannot economically compete with other NETPs yet, it is much less constrained by biophysical limits; the main obstacle to its deployment is its high-energy demand—except for LTSS-DACCS based on MSA cycles, which uses water instead of energy for the desorption process. The water consumption could also hinder the scale-up of HTLS-DACCS and mineral sorbent DACCS deploying air contactors. Notably, LTSS-DACCS based on TSA can extract water from air—the only co-benefit across the DACCS configurations—, although its reported costs are the highest across the chemical NETPs group.

HTLS- and LTSS-DACCS (TSA) stand out as the most mature chemical NETPs. Mineral sorbent DACCS has recently appeared in the literature as a feasible CDR option, but it could avert the potential environmental problems associated with the production of synthetic sorbents at large scale. The degradation of GHGs is also an emergent area of research and therefore its impacts are not well characterised yet. However, it could simultaneously minimise the adverse side-effects of GGR and generate co-benefits for human health and biodiversity.

Finally, the CO₂ storage options show a wide range of KPI values. While none of them is limited by the storage capacity, *in situ* mineralisation and geological sequestration present high TRLs and low costs, indicating that they are ready for deployment.

5. Pareto-efficient NETPs and sequestration methods

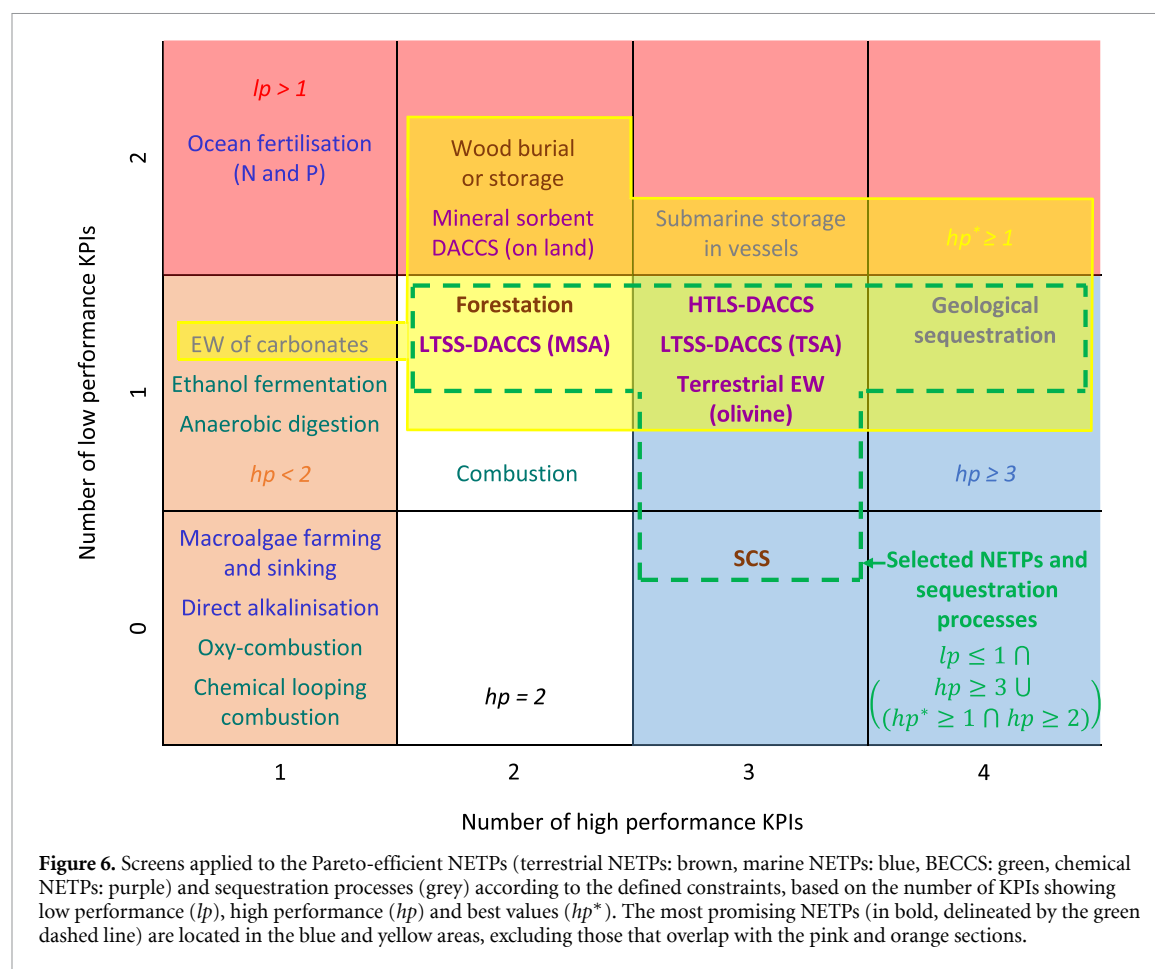
The analysis above highlights that multiple trade-offs arise between the considered NETPs and storage processes, making their relative assessment challenging. Ideally, a detailed optimisation model should be developed to identify the optimal region-specific portfolio of NETPs. This model should include a range of constraints limiting the NETPs' potential, which would require large amounts of data currently



unavailable or highly uncertain. This endeavour is out of the scope of this article; we instead apply the Pareto dominance concept to identify those NETPs that could most likely integrate the portfolio that concurrently optimises all the KPIs. The Pareto concept is widely used in multi-criteria decision-making to identify the best solutions within a set of alternatives. In essence, an alternative A is Pareto efficient if there is not any other alternative B improving A simultaneously in all the relevant criteria. Further analyses can then be performed based on a more detailed inspection of the Pareto solutions once the non-Pareto are dismissed, for example with Pareto filters (Antipova *et al* 2015), using weights to aggregate the criteria into one indicator (Cortés-Borda *et al* 2013), or applying the eco-efficiency concept (Schmidheiny 1992).

Figure 5 illustrates the dominance relationships between NETPs. Out of the 29 NETPs configurations

whose five KPIs are available, we found 16 Pareto-efficient NETPs that are not dominated by any others, i.e. their five KPIs cannot be concurrently improved by any other NETP (further details in appendix D of the supplementary information). These Pareto-efficient NETPs are (a) forestation, (b) wood burial or storage, (c) SCS, (d) direct ocean alkalisation, (e) ocean fertilisation with nitrogen and phosphorus, (f) macroalgae farming and sinking, BECCS deploying (g) ethanol fermentation, (h) anaerobic digestion, (i) combustion, (j) oxy-combustion and (k) chemical looping combustion, (l) HTLS-DACCS, LTSS-DACCS based on (m) TSA and (n) MSA, (o) mineral sorbent DACCS based on the land application of metal oxides, and (p) terrestrial EW deploying olivine. The Pareto-efficient sequestration processes are (a) geological sequestration, (b) submarine storage in vessels and (c) EW of carbonates (supplementary table 3).



In figure 5 we can see that SCS and combustion-BECCS (the modalities involving post-combustion capture, chemical looping combustion and oxy-combustion) are the most dominant (each of them dominating six other NETPs), which denotes an overall good performance relative to the other NETPs in terms of all the KPIs. By contrast, forestation, wood burial or storage and mineral sorbent DACCS (land application) are also Pareto-efficient NETPs, but they do not dominate any others. The reason is that these NETPs present the worst value for the co-benefits KPI (according to our KPI definition, which does not account for potential co-benefits if these can turn into negative side-effects when the NETP is not implemented properly) and, in the case of the DACCS configuration, the lowest TRL as well.

Regarding the inefficient NETPs, downwelling and ocean fertilisation with iron are dominated the most (by 21 and 10 NETPs, respectively), reflecting a poor performance. The intragroup dominance relationships are particularly relevant within the BECCS cluster, where fast pyrolysis, hydrothermal liquefaction and gasification coupled with either combined cycles (IGCC) or the Fischer-Tropsch process are heavily dominated—by five, nine, three and six NETPs, respectively, primarily other BECCS configurations.

We found that all the analysed terrestrial and chemical NETPs are either Pareto-efficient or

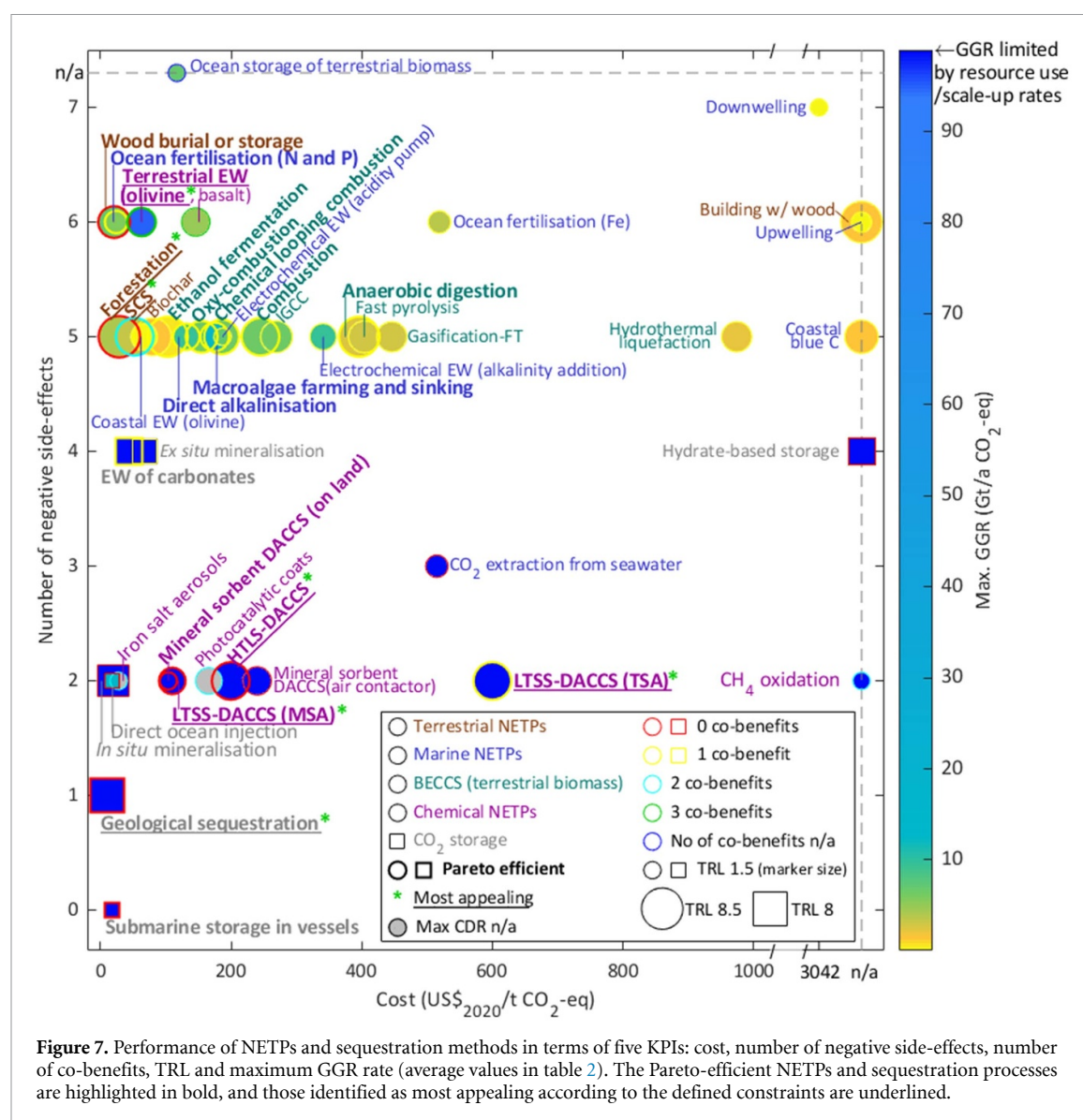
dominated by one Pareto-efficient NETP within their category. The latter include biochar amendment (dominated by SCS), mineral sorbent DACCS deploying an air contactor (dominated by HTLS-DACCS), and terrestrial EW with basalt (dominated by the same NETP using olivine). These results point towards the potential future role of terrestrial and chemical NETPs in optimal GGR portfolios.

The question remains as to which of the Pareto-efficient NETPs and sequestration processes are most promising. We further screened the Pareto-efficient NETPs by defining two constraints. First, the number of KPIs denoting a low performance (lp) must not exceed one. Second, one of these conditions must be met: either the number of KPIs showing a high performance (hp) is greater than or equal to three, or the number of KPIs with the best values across the KPI ranges (hp^*) is one or more, while hp is greater than or equal to two. The mathematical formulation of these constraints is as follows:

$$lp \leq 1 \quad \text{Constraint 1}$$

$$(hp \geq 3) \cup (hp^* \geq 1 \cap hp \geq 2) \quad \text{Constraint 2}$$

We graphically represent these constraints in figure 6, which illustrates how applying these filters reduces the pool of appealing Pareto-efficient NETPs



to these six: (a) SCS, (b) forestation, (c) HTLS-DACCS, LTSS-DACCS based on (d) TSA and (e) MSA, and (f) terrestrial EW using olivine. Moreover, these criteria render geological storage the most appealing sequestration process.

Out of the selected Pareto alternatives, SCS is particularly well balanced, as it does not attain the worst performance level in terms of any of the KPIs. Furthermore, its TRL, costs and co-benefits denote a high performance. Forestation, on the other hand, presents the best TRL and appealing costs, which will facilitate a quick deployment. The negative-side-effects and co-benefits of these two terrestrial NETPs are largely dependent on the NETPs' location and characteristics, and can therefore be optimised.

While the GGR potential of these terrestrial NETPs is medium, that of the DACCS configurations is theoretically much greater, mainly limited by the scale-up rates and, possibly, resource availability. The major appeal of DACCS is that it attains the lowest number of unintended side-effects. Nonetheless,

most DACCS modalities lack co-benefits to incentivise their deployment—except for LTSS-DACCS based on TSA, whose main bottleneck is its substantial cost. The preliminary cost estimates of LTSS-DACCS deploying MSA are much lower, despite its more incipient TRL. By contrast, the maturity level of HTLS-DACCS is within the upper end of the TRL scale (same as LTSS-DACCS using TSA), and shows intermediate costs.

Finally, terrestrial EW with olivine achieves the greatest number of co-benefits across the entire set of NETPs, in addition to appealing GGR potential and cost. Conversely, its potential number of adverse side-effects is considerable. Further research could help improve its TRL and ascertain the extent of the side-effects.

In general, research and progressive scale-up can improve poor TRLs and costs. Similarly, NETPs with low GGR potentials could still be integrated into a portfolio of NETPs. Nonetheless, the side-effects KPIs are more critical, since they could deter or

incentivise the adoption of a given NETP. Hence, we investigated the sensitivity of our results to the side-effects cut-off criteria. Figure 1 of the supplementary information reveals that only three chemical NETPs, namely HTLS-DACCS, LTSS-DACCS deploying TSA and terrestrial EW with olivine would be classified as most appealing if we considered stricter cut-off criteria for the side-effects KPIs. Furthermore, under a more conservative approach where the number of negative side-effects is limited, terrestrial EW—with the largest number of potential unintended consequences—would be excluded from this list. Thus, DACCS appears to be the most resilient NETP to successive screens.

It is worth noting that the KPIs of several NETPs have not been reported yet, and therefore these were not included in the Pareto analysis. However, as we can visualise in figure 7 (which summarises the performance of all the assessed NETPs and sequestration processes), the NETPs involving the degradation of GHGs preliminary show some promising KPIs despite their low TRLs and incomplete profile, especially in terms of their side-effects.

Figure 7 only provides a static picture of the status of NETPs and sequestration processes, since their performance will evolve as research and development intensify and scale-up continues. Hence, our results do not predict how the GGR space will progress; instead, they should be used as a guideline to help decision-makers prioritise investments and resources.

6. Conclusions and future work

Here we reviewed the most relevant NETPs, assessing their performance based on their technical maturity, economic feasibility, GGR potential and side-effects, including resource use and environmental impacts. Our results reflect the current state-of-the-art (or the best estimates based on current knowledge), but the costs and TRLs compiled here may improve with future research and development, while additional unexpected side-effects may also emerge. Despite these limitations, our Pareto analysis and subsequent screening of the optimal NETPs allow us to outline some of the strategies that could facilitate their sustainable scale-up.

Our findings point towards terrestrial and chemical NETPs as the most attractive GGR options. Forestation and SCS constitute easy-to-implement solutions that are ready for deployment. SCS practices are particularly appealing because of their potential co-benefits for crop productivity. However, these terrestrial NETPs could also generate adverse impacts; most notably, certain environmental conditions and human actions could induce the release of the stored carbon. Careful planning and continuous monitoring are needed to warrant that the undesired impacts of these NETPs are avoided or at least reduced.

The deployment of DACCS would minimise unintended side-effects, but removing CO₂ at the Gt scale with DACCS would require expanding the capacity of existing facilities by six orders of magnitude, a tremendous engineering challenge that would exert substantial pressure on the energy system. Early investments in DACCS could help drive costs down and enable its deployment at a pace consistent with 1.5 °C scenarios.

Our analysis also identifies terrestrial EW with olivine as one of the most promising NETPs. Despite its potential role in preventing additional environmental impacts and enhancing soil fertility, this NETP could also entail a considerable number of detrimental consequences, perhaps the most worrisome being the potential damage to human health. Further studies on EW are needed to better understand its risks and assess whether it can be safely deployed.

The NETPs relying on the degradation of GHGs are still quite immature and were not included in our Pareto analysis. Nevertheless, their preliminary KPIs are quite promising; investing in basic research on these NETPs could lead to negative emissions at a relevant scale with minimal trade-offs. By contrast, our results suggest that marine NETPs—also characterised by their overall low TRLs—should not be prioritised in the short term, although further research is still needed, especially to ascertain their environmental impacts.

Even though we identified several BECCS pathways as Pareto-optimal, they predominantly show intermediate performance levels, and none of them are included in the set of most attractive NETPs. Some of the impacts of the NETPs deploying terrestrial biomass are local- or region-specific and could therefore be minimised by carefully selecting the deployment locations. Nonetheless, because of the inability of large-scale biomass plantations to remain within biophysical limits, we do not foresee these NETPs representing the main share of sustainable GGR pathways.

Our findings suggest that climate change mitigation scenarios, which mostly rely on BECCS and forestation for CDR, should consider a wider range of NETPs. We recommend expanding the set of NETPs available within integrated assessment models to include those identified here as Pareto-efficient. This would improve our understanding of the interactions between NETPs and all economic sectors, and could help define detailed guidelines to prioritise future investments. However, drawing from the insights gained in this study, we can preliminarily propose two parallel investment plans focused on (a) companies and institutions commercialising the NETPs classified as most promising, and (b) basic research and development of the Pareto-efficient NETPs. This does not entail that investments in other NETPs should be discouraged, but rather that the Pareto-efficient NETPs should be prioritised.

This study represents a first step towards the design of optimal NETPs portfolios. Devising negative emissions pathways that capitalise on the synergies between NETPs and overcome their individual constraints while minimising overall collateral damage will require rigorous sustainability evaluations based on systematic life cycle assessment studies, multi-criteria decision-support tools and mathematical programming. Further research should build on spatially explicit data provided by Earth system models to develop regionalised NETPs portfolios adapted to location-dependent environmental and resource pressures.

The implications of this study are not only relevant for the climate modelling community investigating the environmental and socioeconomic consequences of GGR. Our results can also serve as a blueprint for scientists developing emergent NETPs or upgrading the more consolidated ones. Moreover, pinpointing the areas where further research and early investments would yield significant advances can help decision-makers draft climate policies and formulate sustainable business strategies to achieve carbon neutrality.

The decisions and investments made during this decade will determine the course of GGR throughout the 21st century; we should regard the 2020s as a training period to develop and upgrade NETPs, and ensure that they can be sustainably deployed at the needed scale. Thus, we echo other works calling for urgent action on negative emissions in addition to emissions reductions schemes.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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