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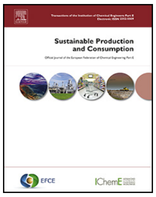

Research article

# The life cycle environmental impacts of negative emission technologies in North America

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ABSTRACT

Negative emission technologies (NETs) could play a key role in ensuring net-zero and longer-term net negative emission ambitions are met. However, greenhouse gas emissions (and other pollutants) will occur over the life cycle of a NET and will need to be taken into consideration when developing schemes to roll out their use. We compare five NETs: afforestation/reforestation (AR), enhanced weathering (EW), mangrove restoration (MR), bioenergy and direct air capture with carbon storage (BECCS and DAC), using life cycle assessment to determine their environmental impacts (global warming, freshwater, toxicity etc.). We find that there is a wide range in the environmental impacts estimated across the NETs and the context in which they are used will directly impact which NET has low or high environmental impacts. This is an important aspect to consider when deciding which NET to prioritise in strategies to roll out their use on large scales. If consistent removal of $CO_2$ from the atmosphere is the goal, then AR and MR have the lowest environmental impacts. However, if large and quick $CO_2$ removal is the goal then EW, DAC and BECCS have similar, if not lower, environmental impacts.

## 1. Introduction

The need to reduce greenhouse gas (GHG) emissions is well established if global temperature rises are to be held well below 2 °C by the end of the century. To achieve this, many countries have pledged to cut their GHG emissions to net-zero by 2045 to 2070 (Energy and Climate Intelligence Unit, 2021). In addition to decarbonising by phasing out fossil fuels and improving energy efficiency, negative emission technologies (NETs) are an important aspect of net-zero strategies (IEA, 2021). In the IPCCs Sixth Assessment Report they found that NETs will be crucial for meeting net-zero and net negative emission goals. This is because non-carbon dioxide ($CO_2$) GHGs (particularly methane) will likely not reach net-zero and to balance global emissions (regions/countries can emit GHGs if others are negative emissions) (IPCC, 2022). A NET is a technology which permanently removes $CO_2$. They do this by either physically removing it from the atmosphere by chemical processes or by increasing the size of natural reservoirs which absorb it (carbon sinks), such as vegetation (National Academies of Sciences, 2019). While many efforts have been made to reduce GHG emissions, decarbonisation is lagging behind the rate needed to keep global temperature rises to below 1.5 °C or 2 °C (IPCC, 2021). Therefore, NETs will likely have a crucial role in meeting emission targets, even if emissions reduction is pursued to the greatest degree possible (Grant et al., 2021).

However, emissions and other environmental impacts will occur over the life cycles of all NETs, from activities before, during and post operation. If, for example, the direct and imbodied GHG emissions from energy use and materials consumed by NETs are large, then they could significantly hinder or negate their emission reduction purpose. Also, additional unintentional burdens could be placed on the environment. For biomass-based NETs, monoculture would reduce adaptability and resilience to climate change, as well as worsen impacts to water availability and biodiversity (IPCC, 2022; Pörtner et al., 2022). At the time of writing, NETs are not widely deployed but many emission abatement and climate scenarios project a large uptake of them in the near to mid-term future (IEA, 2021, Rogelj et al., 2018, EASAC, 2018, CCC, 2019). As they are a somewhat new concept, there is not much known about their environmental impacts, beyond the amount of $CO_2$ they could reduce national and international levels by. There have been studies which have investigated the environmental impacts of NETs using life cycle assessment (LCA) and other tools (Bennett et al., 2019; Cambria and Pierangeli, 2012; Deutz and Bardow, 2021; Lefebvre et al., 2019; Lefebvre et al., 2021; Terlouw et al., 2021b; Toochi, 2018). However, there is not yet a study which compares various NETs on their environmental impacts to assess which have low/high impacts, an important gap in the literature as identified by Terlouw et al. (2021a).

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As NETs are likely to become increasingly important in meeting future emission targets, it is crucial their life cycle impacts be understood to ensure they are being used responsibly and sensibly, as well as to prevent any unintended harm to ecosystems e.g., a reduction in $CO_2$ but at the cost of increased strain to water or terrestrial resources.

In this paper we aim to estimate the life cycle environmental impacts of multiple NETs, deployed in North America (USA and Canada), and compare them on their environmental performance. The USA and Canada were selected because of their large GHG emissions ($CO_2$ and other GHGs) (IEA, 2022, The World Bank, 2022) and because a wide variety of NETs can be deployed there. This is the novelty of this paper, as we are the first to assess various NETs on their environmental impacts using LCA and compare them to one another so that it can be determined which have low/high environmental impacts. While there is already a considerable number of LCA studies which have assessed NETs, these only consider one NET and do not attempt to compare their results to other NET LCA studies and thus, at the time of writing it is uncertain which NETs have low environmental impacts and which could potentially cause negative impacts to the environment if deployed on a large scale. The work presented in this paper can be used in evaluating NETs chosen to be used in emission reduction strategies in the USA and Canada, so would be of interest to policy makers and NET developers in these two countries, as well as other academics who work in future energy systems or climate modelling. While specific to the USA and Canada, our results can also give an indication of the likely environmental impacts of the NETs modelled if deployed in other countries. The rest of the paper is structured as follows: a literature review of the NET literature, followed by the methods used in the LCA modelling and then the results and discussion. The paper will finish with areas for future work and the conclusions drawn.

## 2. Literature review

As we move further into the 21st century, the needs to reduce $CO_2$ emissions into the atmosphere will become increasingly important if 1.5 °C or 2 °C targets are to be met (IPCC, 2021). The primary mechanism used to reduce emissions is decarbonising the energy sector, with the focus of transitioning away from fossil fuels to renewable energy. However, when comparing decarbonisation plans submitted by countries who are parties to the Paris Agreement to the cuts in carbon emissions needed to meet Paris Agreement goals (estimated by the IPCC), there is a significant gap (IPCC, 2021). Therefore, there may be the need to physically remove $CO_2$ from the atmosphere using technology.

At the time of writing, it is clear NETs will play an import role in tackling climate change, but it is uncertain which NETs will be widely deployed. This is because, apart from afforestation/reforestation, they are not mature technologies and have not been deployed on large scales (National Academies of Sciences, 2019). Despite this, there is much interest in developing NETs, and governments around the world are looking into them as a way of achieving net zero targets (UK Parliament, 2021; World Resources Institute, 2020). NETs encompass a range of technologies which can remove (permanently) atmospheric $CO_2$, ranging from those which make carbon sinks bigger to those which can chemically remove it (EASAC, 2018; National Academies of Sciences, 2019). Within NETs, there is an overlap in their categorisation. NETs which increase the size of carbon sinks can be referred to as natural climate solutions, soil carbon sequestration and terrestrial carbon removal and sequestration. This can make categorising NETs difficult but broadly speaking there are seven types (EASAC, 2018, National Academies of Sciences, 2019): afforestation/reforestation (AR), bioenergy with carbon capture and storage (BECCS), blue carbon restoration (BC), direct air capture (DAC), carbon mineralisation (CM), land management (LM) and ocean fertilisation (OF).

AR removes $CO_2$ through plants absorbing $CO_2$ to grow. Typically, it involves planting trees and improving forestry management. LM removes $CO_2$ by increasing the amount of carbon in soils. Many technologies fall under LM, including agricultural practices and applying biochar to soil to improve soil conditions. BC is the restoration of aquatic ecosystems, such as mangroves, salt marshes and seagrass beds and removes $CO_2$ through the same mechanism as AR. BECCS is when the $CO_2$ produced in bioenergy production (e.g., electricity, biofuels) is captured and sequestered underground. CM is when minerals react with $CO_2$ to remove it from the atmosphere, including mineralisation of calcium/magnesium rich rock with a $CO_2$ rich fluid or gas, reaction with a solid or alkali solution rich in calcium/magnesium, or by injecting $CO_2$ into calcium/magnesium rich rock. DAC is when $CO_2$ in air is chemically removed by contacting it with chemicals which bind with $CO_2$. The $CO_2$ is then separated through further chemical processes and sequestered. OF is when chemicals such as iron, urea and phosphorus are added to the upper layer of the ocean to increase photosynthesis activity of phytoplankton.

At the time of writing, only AR is being utilised on large scales (EASAC, 2018, National Academies of Sciences, 2019). There are pilot DAC plants and small scale BECCS plants in operation with larger plants under construction (EASAC, 2018, National Academies of Sciences, 2019). LM is being utilised but is often used to improve soil conditions and agricultural yields rather than to solely sequester $CO_2$. There are no dedicated EW projects in operation, but projects have been proposed. BC is being carried out with both community and NGO led projects to replant mangrove forests and restore wetlands. OF is effectively banned until the effects of manipulating the oceans' food chains are better understood (Tollefson, 2008; Lukacs, 2012; Tollefson, 2017).

When assessing the environmental impacts of NETs (for $CO_2$ removal only), there are several studies in the life cycle assessment (LCA) literature, but these have focussed largely on BECCS (12), DAC (4) and biochar (36). There are numerous LM studies (35), but these assess agricultural systems and do not set $CO_2$ sequestration as the primary focus. There are four LCA studies which assess AR and one which assesses CM. Terlouw et al. (2021a) conducted an in-depth review of NET LCA papers and thus a detailed overview of the studies covered in their paper will not be presented in this paper. They found that in the NET LCA literature there were many shortcomings, specifically on how negative emissions are defined, the lack of multiple indicators used to assess NETs and transparency in the LCA data and LCA modelling. Readers may refer to Terlouw et al. (2021a) for an overview of NET LCA studies. For BC and OF, as far as the authors are aware, there have been no LCA studies conducted for OF, wetland and seagrass restoration but Moriizumi et al. (2010) assessed mangrove restoration and compared it to charcoal production.

Elsewhere in the NET environmental assessment literature, a series of papers written in 2018 (Minx et al., 2018; Fuss et al., 2018; Nemet et al., 2018) compared multiples NETs. These focussed on the cost and potential of each NET and only qualitatively touched on the potential side effects. Our paper aims to fill this gap with quantitative results on many environmental impacts. A separate paper by Smith et al. (2016) does quantitively compare multiple NETs on several environmental factors. However, this work utilises a full LCA to calculate the impacts of each NET. By utilising LCA modelling we can more transparently compare NETs in the same region. Moreover, this paper considers more environmental impact factors and different NETs, such as mangroves.

Based on the NET literature available, despite the shortcomings, they indicate that NETs are beneficial in $CO_2$ removal as they removed more $CO_2$ than GHGs emitted over their life cycle. However, there are numerous factors which impact their life cycle GHG emissions, such as energy use, energy source, chemicals consumed and transport needs. From the existing NET LCA literature it is unclear which NETs are more effective as the studies use different functional units (e.g., kWh electricity generated, t of biomass, t product, ha land). Also, none compare different NETs against one another. Harmonisation can be applied to allow multiple LCA studies to be compared to one another, but Terlouw et al. (2021a) found that because of a lack in transparency in the LCA data and modelling, harmonisation is not possible (at the time of writing).

In this work we consider five NETs: AR, enhanced weathering (EW), mangrove restoration (MR), BECCS and DAC. These were chosen as literature data were available which enabled LCA models to be built. Biochar was excluded as there are numerous routes of producing it (feedstocks, pyrolysis technologies/processes) and we were uncertain which route would be most favourable in $CO_2$ removal. Other LM methods were also excluded because data on agricultural management and practices were limited, and we were unable to find a specific project we could base a LCA model on. OF was excluded because it is effectively banned. Other BC methods were excluded as we were unable to find projects to base LCA models on.

## 3. Methodology

An LCA was conducted to estimate the life cycle environmental impacts of five NETs: AR, EW, MR, BECCS and DAC with carbon storage. These were selected as they are prominent in the NET literature, have high $CO_2$ removal potential (EASAC, 2018; IEA, 2021; IPCC, 2021; National Academies of Sciences, 2019) and have ample literature data available to build LCA models. Please note that our LCA modelling is based on literature data only and no primary data from operators were used. The LCA conducted followed the steps outlined in the ISO 14040/104044 (ISO, 2006a; ISO, 2006b). Several parameters were varied in the models, which are described in the following section and an

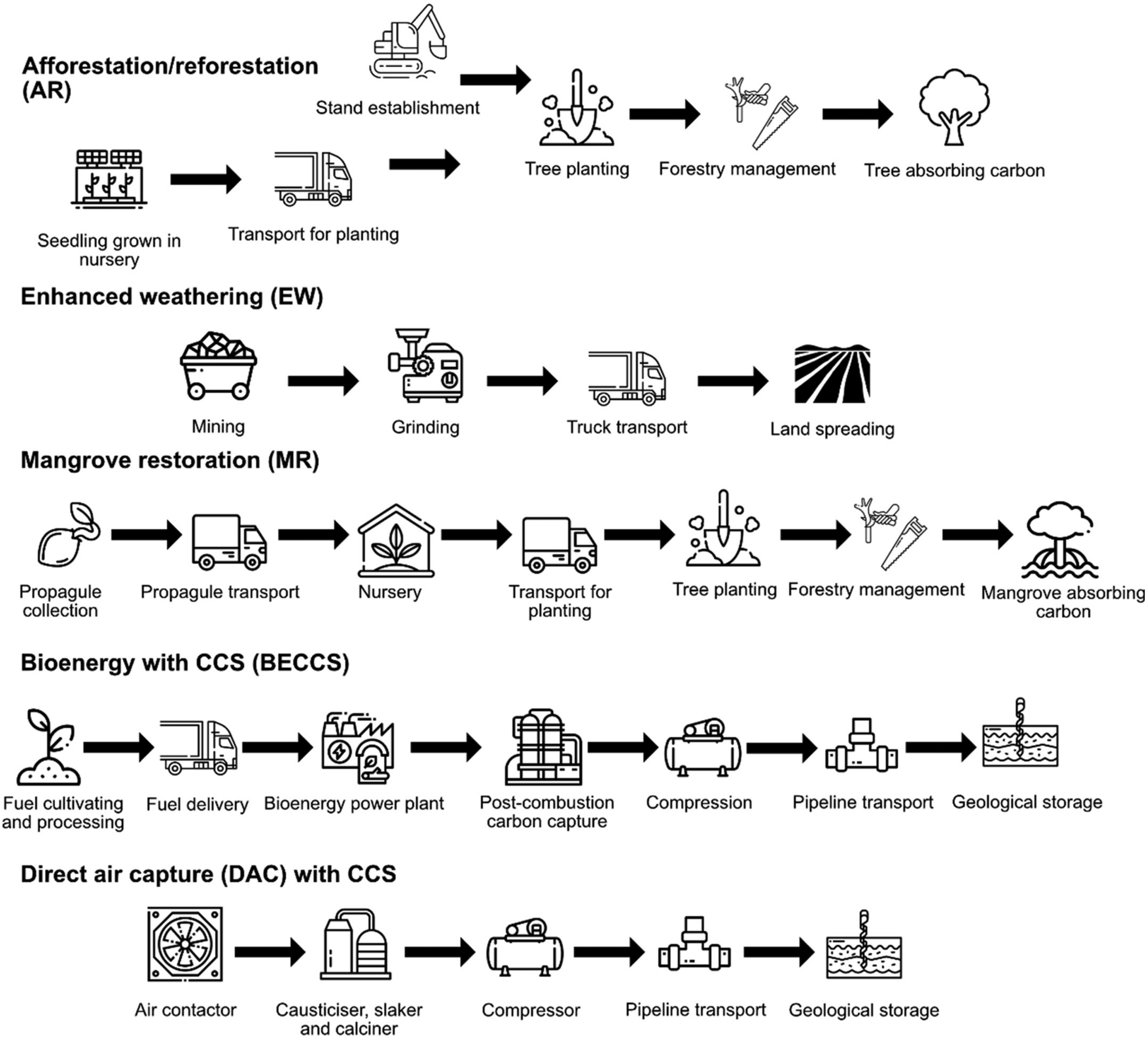


**Fig. 1.** Life cycle system boundaries of the negative emission technologies (NETs): afforestation/reforestation, enhanced weathering, mangrove restoration, bioenergy with carbon capture and storage (CCS) and direct air capture (with CCS), considering a cradle to grave system boundary. End of life activities are excluded. Please note that the $CO_2$ sequestered is included in our system boundary.

overview of the scenarios modelled can be found in Tables S4 and S5 in the Supporting Information (SI).

### 3.1. Life cycle assessment (LCA)

#### 3.1.1. Goal and scope

To determine the life cycle impacts of the five NETs, a LCA was carried out, considering a predevelopment to $CO_2$ sequestration (cradle to grave) system boundary, as shown in Fig. 1. We compare the NETs using two functional units (FU): one tonne of $CO_2$ (1000 kg $CO_2$; 1 t $CO_2$) sequestered and 1 t $CO_2$ sequestered in a year. Two FU were considered as the NETs have varying $CO_2$ removal rates.

All LCA models are based on being in North America (USA and Canada). DAC is Canadian specific as the project we base our model on is Canadian (Keith et al., 2018). BECCS is also Canadian specific (because of LCI data availability). EW is USA specific as the USA has the larger market for rocks such as olivine (Kremer et al., 2019). MR is USA specific as part of the USA (Florida) is tropical. AR is also USA specific because we consider hardwood trees which are more common in USA forests than in Canadian forests. We exclude end of life activities because of inconsistencies and uncertainties in what activities are likely to occur for each NET. This is because for the AR, EW and MR it is uncertain what end of life means. The wood produced in AR and MR can be used as fuel or used to make wood products while in EW land use changes or soil management will impact end of life. For DAC and BECCS end of life is more straightforward- the site and facilities are decommissioned, and equipment and materials recycled or disposed of. The life cycle inventory data are presented in Tables 1 and 2 and a detailed inventory of the data can be found in the SI.

*3.1.1.1. Afforestation/reforestation (AR).* In this NET, trees are planted in an area with or previously covered by trees. We consider all stages from growing the seedling and planting, through to the growth of the tree to maturity (Fig. 1). We estimate the $CO_2$ sequestering rate based on tree $CO_2$ absorption rate data (Forestry Research, 2018; Toochi, 2018; Urban Forestry Network, 2020) to determine the range in capacity. This was then used to determine the quantities of land, forestry and seedling management needed (Tables 1 and 2). We consider hardwood trees as they have a longer lifespan and higher absorption rate and only consider $CO_2$ absorbed by the tree; soil $CO_2$ and methane are not considered. It should be noted that we assume a constant rate of $CO_2$ removal; the rate of removal slows down when the tree reaches maturity. The application of fertiliser was excluded. We assume native species are planted and the areas planted are suitable for forests/woodlands. Fertiliser can be applied but the fertiliser application strategy will depend on various factors, such as soil type and condition and if boosting the size of the trees is an objective. These will impact how frequently the fertiliser will be applied, as well as the type of fertiliser applied. As we combined data from multiple literature sources to build our AR LCA models, we were unable to determine what the appropriate fertiliser application strategy would be and therefore it was excluded from our LCA models. We consider lifespans of 10 to 100 years. A range in the project lifespan, tree density and $CO_2$ absorption rate were considered in a sensitivity analysis.

*3.1.1.2. Enhanced weathering (EW).* This is a CM route where $CO_2$ is removed from the atmosphere by the decomposition of rock via weathering (mineralisation route). We consider olivine, a silicate rock which has good $CO_2$ absorption capabilities. It decomposes in the presence of $CO_2$ to form carbonate minerals. For our EW LCA models, we consider ex-situ CM where olivine is mined and ground into small pieces before being transported to an area where it is spread over land. Once spread, the olivine will react with $CO_2$ in the air to form carbonate minerals. Land spreading was chosen over coastal spreading as it has been found to have a faster dissolution rate (Hangx and Spiers, 2008). The amount of $CO_2$ absorbed is 1 t $CO_2$ per 1.6 t to 3.7 t (average 2.7 t) of olivine (Hangx and Spiers, 2008; Mazzotti et al., 2008). We consider all stages in the life cycle from mining through to land spreading (Fig. 1 and Table 2). The sensitivity analysis considered the impact of rock purity (60 % and 100 %), truck distance and electricity used for grinding.

*3.1.1.3. Mangrove restoration (MR).* Mangroves are shrubs and trees which grow in salt water, typically in tropical and subtropical coastal areas. While the mechanism of $CO_2$ removal is the same as AR, MR differs in that the area restored is wetland rather than forest. The $CO_2$ absorption rate of mangroves is high, but their life span is short in comparison to hardwood trees. In MR, mangrove seeds (propagules) are collected and then transported to a nursery. In the nursery, the propagule can be placed in salt water and roots develop, or planted and grown into a sapling. We consider both, with a small-scale nursery producing propagules with roots and a large-scale nursery producing mangrove saplings. For more information on how the mangrove nurseries were modelled, see Section 2 in the SI. Fertiliser was excluded from our LCA models, in both the nursery and planting. We were unable to find data on whether fertiliser is used in mangrove restoration, so it was excluded. From the nursery the propagule or sapling is transported to the site being restored where they are planted by hand. We consider lifespans of 10 to 50 years, as well as variations in tree density in a sensitivity analysis.

*3.1.1.4. Bioenergy with carbon capture and storage (BECCS).* In this NET $CO_2$ from a biomass power plant is captured. For this, we modelled a

**Table 1**
Life cycle inventory data: overview of negative emission technologies (NETs) modelled in this work.

| NET | Project lifespan (years) | $CO_2$ removal capacity[b] | $CO_2$ removal units | Based on | Geography |
|---|---|---|---|---|---|
| Afforestation/reforestation | 10 to 100 | 5 (1.4 to 22) (Toochi, 2018, Forestry Research, 2018, Urban Forestry Network, 2020) | kg per year per tree | Panama reforestation project and UK forestry projects (Carbonfund.org, 2020, CCC, 2019, Forest Carbon, 2020) | USA |
| Enhanced weathering | <1 to 10[a] | 2.7 (1.6 to 3.7) (Hangx and Spiers, 2008, Mazzotti et al., 2008) | t olivine per t $CO_2$ | Project Vesta (Project Vesta, 2020), Hangx and Spiers (2008) | USA |
| Mangrove restoration | 10 to 50 | 12.3 (Eden Reforestation Projects, 2019) | kg per year per tree | Moore (2009), Clarke and Johns (2002), SKinno News (2019) and Chimbi (2022) | USA |
| Bioenergy with CCS | 25 | 4 (Drax, 2018) | Mt per year | Drax BECCS plant (Drax, 2018) | Canada |
| Direct air capture (with CCS) | 25 | 1 (Keith et al., 2018) | Mt per year | Carbon Engineering (Keith et al., 2018) | Canada |

[a] Own calculation based on Hangx and Spiers (2008).

[b] Average rate of removal across project lifetime. Please note that the removal rate of trees and olivine saturates and will vary because of environmental and other factors e.g., temperature, climate.

**Table 2**
Life cycle inventory data: parameters and life cycle inventory (LCI) data modelled in GaBi-data and assumptions.

| NET | Data |
|---|---|
| Afforestation/reforestation | Nursery<br>ecoinvent data used (RER: tree seedling produced in heated greenhouse)<br>Transport from nursery<br>100 km between nursery and planting site (Nicese et al., 2021)<br>Transported in freight lorry (7.5–16 metric ton, EURO5)<br>Sapling weight assumed to be 45 kg (root ball plus container weight) (Johnsons of Whixley, 2021)<br>Stand establishment<br>Adapted from Nicese et al. (2021)<br>Activities: excavation, ploughing, tilling and harrowing<br>Energy requirements are from Nicese et al. (2021)<br>Planting<br>1400 (333 to 2500) trees per hectare, tree density (Carbonfund.org, 2020, CCC, 2019, Forest Carbon, 2020)<br>85 % survival rate of saplings to maturity (Cambria and Pierangeli, 2012)<br>Plastic tree protector use- 2 kg weight assumed for plastic tree protector<br>Fertiliser not applied and wood residue left onsite<br>Forestry management<br>Adapted from Nicese et al. (2021)<br>Energy requirements for forestry management and stand establishment taken from Nicese et al. (2021) |
| Enhanced weathering | Olivine mined in quarry and crushed and ground into sand onsite<br>Mining<br>ecoinvent basalt quarry data adapted for olivine<br>2 kWh/t material energy requirement (Hangx and Spiers, 2008)<br>100 % ore purity and 60 % ore purity were assessed in a sensitivity analysis<br>Grinding<br>Mined olivine transported to be crushed and then to be ground<br>2 kWh/t material (Hangx and Spiers, 2008) crushing energy<br>3.5 kWh/t material (Hangx and Spiers, 2008) energy to transport crushed olivine to the grinder<br>173 kWh/t material (Hangx and Spiers, 2008) to grind 10 μm diameter olivine grains (Hangx and Spiers, 2008)<br>Grid electricity (USA) and diesel is used to meet energy demands<br>Renewable electricity (hydroelectricity and wind) for grinding were assessed in a sensitivity analysis<br>Transport<br>20 t truck used to transport ground olivine to location for land spreading<br>43 km distance (one way) by truck was assumed<br>Doubling the transport distance was assessed in a sensitivity analysis<br>Land spreading<br>ecoinvent manure loading and spreading by hydraulic loader data assumed<br>Temperatures of 25 °C and 15 °C are assumed<br>Time to reach saturation calculated based on Hangx and Spiers (2008) |
| Mangrove restoration | Propagule collection and transport<br>Propagules are collected manually<br>Propagules are transported to the nursery in a light commercial vehicle (30 km (Moriizumi et al., 2010))<br>Nursery<br>Grid electricity (FRCC) is used to provide power in the nursery<br>In small scale nursery, propagules are placed in buckets filled with seawater for two weeks (Moore, 2009)<br>The water in the buckets is changed every three days and this is done using an electric pump (Moore, 2009)<br>The number of propagules per bucket is 50 (Moore, 2009)<br>Plastic bucket is 10-gal capacity, and a weight of 1 kg is assumed<br>In large scale nursery, the propagule is planted in ponds<br>Ponds are flooded and drained of sea water (to mimic the tide) twice per day (Clarke and Johns, 2002)<br>6 months assumed to grow propagule into sapling (Clarke and Johns, 2002)<br>10-year life span assumed for large scale nursery<br>Total of 90,000 propagules planted over nursery life span<br>Transport for planting<br>The propagule with fine roots transported out of the nursery is assumed to weigh 43 g (Rabinowitz, 1978; Lin and Leonel da, 1995)<br>Propagules and saplings transported from nursery to planting site in a light commercial vehicle (36 km (Moriizumi et al., 2010))<br>Sapling is assumed to weight 1.8 kg (Schultz, 1997)<br>Tree planting<br>Tree density: 930 to 4850 (2500 average) (Dey and Kar, 2012) (Moriizumi et al., 2010) (Zoological Society of London, 2019)<br>Survival rate: 30 % for propagule with fine roots planted and 90 % for saplings grown from propagules (Costa et al., 2016; Wodehouse and Rayment, 2019; Kodikara et al., 2017; Marchand, 2008)<br>Planting site preparation is manual and no heavy machinery or equipment is used<br>Metal fencing is not used at planting site<br>Forestry management<br>Forestry management is assumed to be the same as in AR |
| Bioenergy with CCS | Fuel production and processing<br>ecoinvent data for miscanthus pellets and woodchips<br>Grid electricity and renewable electricity in miscanthus pellet production is considered<br>ecoinvent data used for feedstock supply chain<br>Fuel delivery<br>ecoinvent data for miscanthus transport and woodchips transport<br>Bioenergy power plant<br>Based on Drax power plant for $CO_2$ capture potential (Drax, 2018)<br>Miscanthus pellets and woodchips are considered as fuel<br>ecoinvent data used for bioenergy plant<br>0.6 kg miscanthus pellets/woodchips per kg $CO_2$ based on ecoinvent data<br>CCS process<br>CCS process based on Cuéllar-Franca and Azapagic (2015) and Koornneef et al. (2008)<br>Scrubber: 24 kWh/t $CO_2$ (Koornneef et al., 2008)<br>Stripper: 1390 kWh/t $CO_2$ (Koornneef et al., 2008)<br>$CO_2$ compression: 111 kWh/t $CO_2$ (Koornneef et al., 2008)<br>2.34 kg MEA consumption per t $CO_2$ (Koornneef et al., 2008)<br>CCS pipeline distance: 80 km (Global CCS Institute, 2012)<br>CCS pipeline sensitivity distances: 1.9 and 500 km<br>7 kWh/t $CO_2$ for injecting $CO_2$ (Koornneef et al., 2008, Petrescu and Cormos, 2017) |
| Direct air capture (with CCS) | $CO_2$ from air removed via liquid absorbent (KOH) and based on Carbon engineering process (Keith et al., 2018)<br>DAC process<br>Energy inputs (Keith et al., 2018):<br>• 82 kWh/t $CO_2$ by the contactor<br>• 27 kWh/t $CO_2$ by the causticer<br>• 3584 kWh/t $CO_2$ by the slaker<br>• 1458 kWh/t $CO_2$ by the calciner<br>• 213 kWh/t $CO_2$ in auxillary needs<br>A variety of heat and power configurations were modelled in the sensitivity analysis:<br>• Grid electricity[a] (Canadian) with natural gas for heat<br>• Grid electricity[a] (Canadian) for heat and power<br>• Natural gas CHP for heat and power<br>• Biogas CHP for heat and power<br>• Woodchips CHP for heat and power<br>• Renewable (wind) electricity[a] for heat and power |

**Table 2** (*continued*)

| NET | Data |
|---|---|
| | • Renewable (wind) electricity[a] for power and biogas for heat<br>Material flows (Keith et al., 2018):<br>• Air: 251,000 t/h<br>• KOH: 286 t/h<br>• $K_2CO_3$: 352 t/h<br>• $Ca(OH)_2$: 189 t/h<br>• CaO: 143 t/h<br>• $CO_2$: 112 t/h<br>CCS<br>CCS pipeline distance: 80 km (Global CCS Institute, 2012)<br>CCS pipeline sensitivity distances: 1.9 and 500 km<br>7 kWh/t $CO_2$ for injecting $CO_2$ (Koornneef et al., 2008, Petrescu and Cormos, 2017) |

[a] Carbon intensity of grid electricity: 0.207 kg $CO_{2eq.}$/kWh; carbon intensity of renewable (wind) electricity is 0.017 kg $CO_{2eq.}$/kWh (ecoinvent, 2017).

bioenergy power plant which uses monoethanolamine (MEA), an amine solvent, to remove $CO_2$ from the flue gas post-combustion. Miscanthus and woodchips (from wood waste) are considered. The carbon capture and storage (CCS) process, including pipeline transport and injection into a underground storage facility, is based on Cuéllar-Franca and Azapagic (2015) and Koornneef et al. (2008) for the material and energy flows. We built our LCA model using the Drax plant (Drax, 2018) as our reference and assume a power plant lifespan of 25-years (Tables 1 and 2). All of the carbon capture energy needs are met using the energy produced by the power plant. As more heat and power is generated than is needed to sustain the carbon capture process, subdivision is applied in our modelling. The impact of the length of the pipeline used to transport $CO_2$ to the geological storage site was assessed in a sensitivity analysis. $CO_2$ leakage in the pipeline and from the geological storage site were not included due to a lack in measurement studies which assess the potential of $CO_2$ leakage in CCS pipelines and storage facilities.

*3.1.1.5. Direct air capture (DAC) with carbon storage.* In this NET, $CO_2$ in the air is removed using a Lewis acid. We consider the DAC process developed by Carbon Engineering (Keith et al., 2018), which uses liquid potassium hydroxide (KOH) to remove $CO_2$. In this process, $CO_2$ is adsorbed by KOH in an air contactor to make potassium carbonate ($K_2CO_3$). The $K_2CO_3$ is then fed into the causticer where it reacts with calcium hydroxide ($Ca(OH)_2$) to make calcium carbonate ($CaCO_3$), which then goes through the slaker and calciner to separate out the $CO_2$ (and calcium oxide (CaO)), which is then compressed. For further details on this process, see Keith et al. (2018). The heat and power needs of the $CO_2$ capture process is met through a variety of methods: grid and renewable electricity (wind) for power only or for power and heat, natural gas and biogas for heat only and natural gas, biogas and woodchips CHPs for heat and power. We assume the captured $CO_2$ is sent for storage in a deep geological storage facility. A 25-year lifespan is considered for the facility and $CO_2$ transport via pipeline to offshore geological storage is considered (Tables 1 and 2). The heat and power source, as well as the length of the CCS pipeline were assessed in a sensitivity analysis. As with BECCS, $CO_2$ leakage from the CCS pipeline and storage site were not considered.

### 3.1.2. Life cycle inventory (LCI): data and assumptions

We built our LCA models using GaBi 10 software (Sphera, 2021a) using literature data and data from the ecoinvent v3.4 database (ecoinvent, 2017). Literature data was used to model each NET, as well as model processes not available in ecoinvent (Table 2). We built our LCA models based on projects either in operation or proposed (Table 1) and consider activities over the entire operating/project lifetime. In all of the NETs, data from multiple sources were used to build the models. This is because we were unable to collect the data needed to model each NET from a single source. The data used in the LCA models can be found in Table 2 and the SI (citing data sources). The $CO_2$ sequestered is included in the system boundary. Therefore, the results for global warming and climate change can be negative (−1000 kg $CO_{2eq.}$/t $CO_2$ sequestered would be if no GHG are emitted across the life cycle). In places where North American LCI data were unavailable, GLO (Global) data were assumed.

### 3.1.3. Life cycle impact assessment (LCIA)- estimating life cycle environmental impacts

The IPCC AR and ReCiPe 2016 impact assessment methods are used to estimate the life cycle environmental impacts. We used the IPCC AR method to estimate the impacts to global warming and climate change as it uses more up to date $CO_2$ equivalences (Sphera, 2021b). For the other environmental impacts: air (photochemical ozone formation and stratospheric ozone depletion), freshwater (consumption, ecotoxicity and eutrophication), marine environments (marine ecotoxicity and eutrophication), human health (human toxicity, cancer and non-cancer), resources and land (terrestrial acidification and ecotoxicity, land use and metal depletion), ReCiPe was used. ReCiPe was chosen over other LCIA methods as it calculates numerous environmental impacts including those that could be important for NETs in decision making e.g., land use and water impacts. TRACI is a LCIA method which is more commonly used in North America but does not calculate impacts to land use or water so was not used.

## 4. Results

The results of the LCA will be presented in the order given in Section 3.1.3, followed by a comparison to the literature.

### 4.1. Impacts to global warming and climate change- global warming potential (GWP)

There is a net benefit in $GWP_{100}$ and $GWP_{20}$ across the NETs with AR and MR being the most effective (Fig. 2). In AR the project lifespan and tree density are the key parameters as under the ten-year and lowest tree density scenario, the $GWP_{100}$ reduces to −364 kg $CO_{2eq.}$/t $CO_2$ sequestered (compared to −992 kg $CO_{2eq.}$/t $CO_2$ sequestered in the 100-year and average tree density scenario). This is because under this scenario, the largest amount of land is needed which results in more material and energy inputs being needed during the tree planting (plastic tree protector and trucks to deliver saplings) and forestry management stages. For MR, the project timespan and the type of nursery are the key parameters, but there is little difference between the scenarios ($GWP_{100}$–988 to −996 kg $CO_{2eq.}$/t $CO_2$ sequestered). BECCS is the third most effective NET with the biomass/feedstock consumed in the power plant having an impact (Section 6 in the SI) because of materials and energy consumed when producing and processing the fuel. The least effective NETs are DAC and EW. DAC requires larger quantities of heat and power per t $CO_2$ removed in comparison to BECCS and it is this that accounts for the majority of GWP impacts. In $GWP_{100}$, there is no net carbon removal when all power needs are met through grid electricity (Canadian) and all heat through natural gas. When the $GWP_{20}$ is considered, all scenarios which use grid electricity and or natural gas have no net carbon removal (Fig. 2). For EW, the main GWP hotspot is grinding of the olivine to 10 μm diameter grains (Section 4 in the SI). When grinding is decarbonised (renewable electricity for grinding) the GWP greatly improves. It is not necessary to grind the olivine to grains of 10 μm diameter, but the rate of $CO_2$ removal is dependent on the grain size; larger sized pieces of olivine will remove $CO_2$ at a slower rate (see Section 1 in the SI).

When the functional unit of 1 t $CO_2$ in a year is considered, EW, DAC and BECCS are on par with AR and MR on a $GWP_{100}$ basis (Fig. 2). AR, while still effective, results in no net benefits when the tree density is

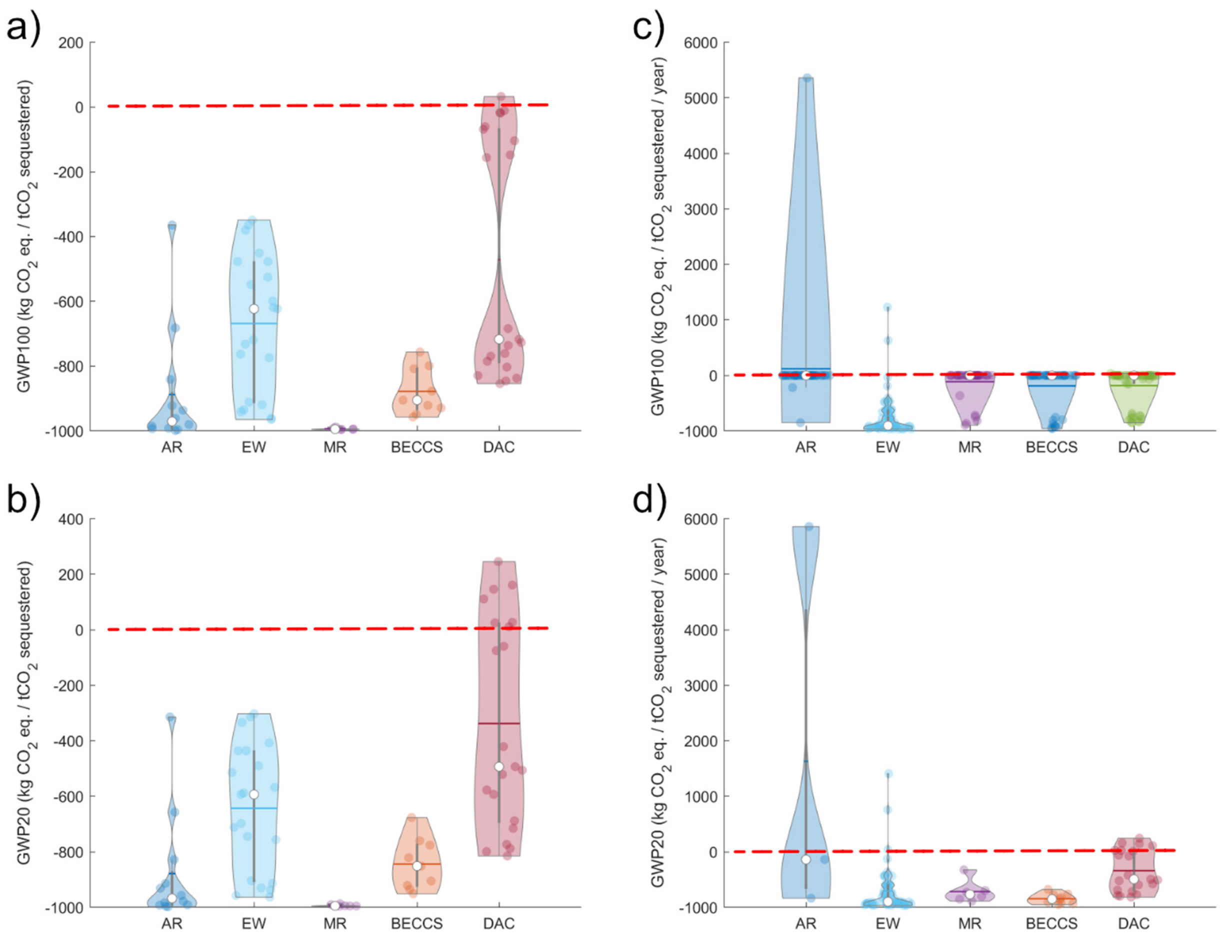


**Fig. 2.** Impacts to global warming and climate change of the NETs (global warming potential (GWP)): afforestation/reforestation (AR), enhanced weathering (EW), mangrove restoration (MR), bioenergy with carbon capture and storage (BECCS) and direct air capture (DAC) with carbon storage. a) GWP$_{20}$ per t CO$_2$ sequestered b) GWP$_{100}$ per t CO$_2$ sequestered. c) GWP$_{20}$ per 1 t CO$_2$ sequestered in a year. d) GWP$_{100}$ per 1 t CO$_2$ sequestered in a year. The red line indicates where there is no benefit in GWP- below the line there is a net benefit. The figures are violin plots, which are a hybrid of kernel density plots and box and whisker plots. Each scenario is shown by a coloured dot and the density of scenarios is represented by the width i.e. the thicker the more scenarios. The lines on the violin plot show the mean (horizontal) and interquartile range (vertical) and the median is indicated by the white dot.

low. This highlights the importance of CO$_2$ removal rate. While over its project lifetime, per t CO$_2$ sequesters, AR is much more effective than EW, DAC and BECCS. As its CO$_2$ removal rate is much lower, long-time spans and/or large numbers of trees need to be planted to match the CO$_2$ removal capacity of DAC and BECCS. When GWP$_{20}$ is considered, AR has higher impacts than the other NETs, with EW, MR and BECCS having much lower impacts.

### *4.2. Impacts to air: photochemical ozone formation and stratospheric ozone depletion*

In impacts to air, AR and MR have the lowest impacts (Fig. 3). In AR, the plastic tree protector causes most of the impacts towards photochemical ozone formation and forestry management (sawing) is responsible for most of the impacts towards stratospheric ozone depletion (Section 3 in the SI). For MR, the forestry management (sawing) is the main impact hotspot. EW also has low impacts, particularly in stratospheric ozone depletion with most impacts to photochemical ozone formation and stratospheric ozone depletion being caused by mining (waste produced and blasting) and electricity used for grinding. DAC and BECCS have the highest impacts, largely attributed to heat and power needs (Sections 6 and 7 in the SI). For AR, impacts to photochemical ozone formation are high when a shorter project lifespan is considered because, under the ten-year project lifespan more trees are needed, and consequentially more plastic tree protectors are needed.

When 1 t CO$_2$ in a year is considered, MR is still effective but EW, DAC and BECCS are now on par in photochemical ozone formation (Fig. 3). In stratospheric ozone depletion EW has the lowest impacts followed by MR, while AR, BECCS and DAC have similar impacts. AR has the highest impacts when a low tree density is considered. The shift in the impacts is caused by AR having low CO$_2$ removal rates, as larger quantities are needed to remove 1 t CO$_2$ in a year in comparison to EW, DAC and BECCS.

### *4.3. Impacts to freshwater: freshwater consumption, ecotoxicity and eutrophication*

When impacts to water are considered, AR and MR have the lowest impacts, but impacts are high in AR when shorter project lifespans are considered (Fig. 4); plastic tree protector, truck to deliver seedlings and forestry management are the main impact hotspots (Section 3 in the SI). Fertiliser can induce high impacts to eutrophication and ecotoxicity (Goglio et al., 2012; Tonini and Astrup, 2012). Had it been included in our LCA models, the impacts to freshwater eutrophication and ecotoxicity would be higher. However, as fertiliser application depends on various factors, including soil type and condition, we cannot speculate how much higher the impacts would be. BECCS has the second lowest impact, with impacts mostly from producing and processing the biomass/fuel. EW and DAC have higher impacts, particularly in freshwater consumption. In freshwater eutrophication, EW has higher impacts

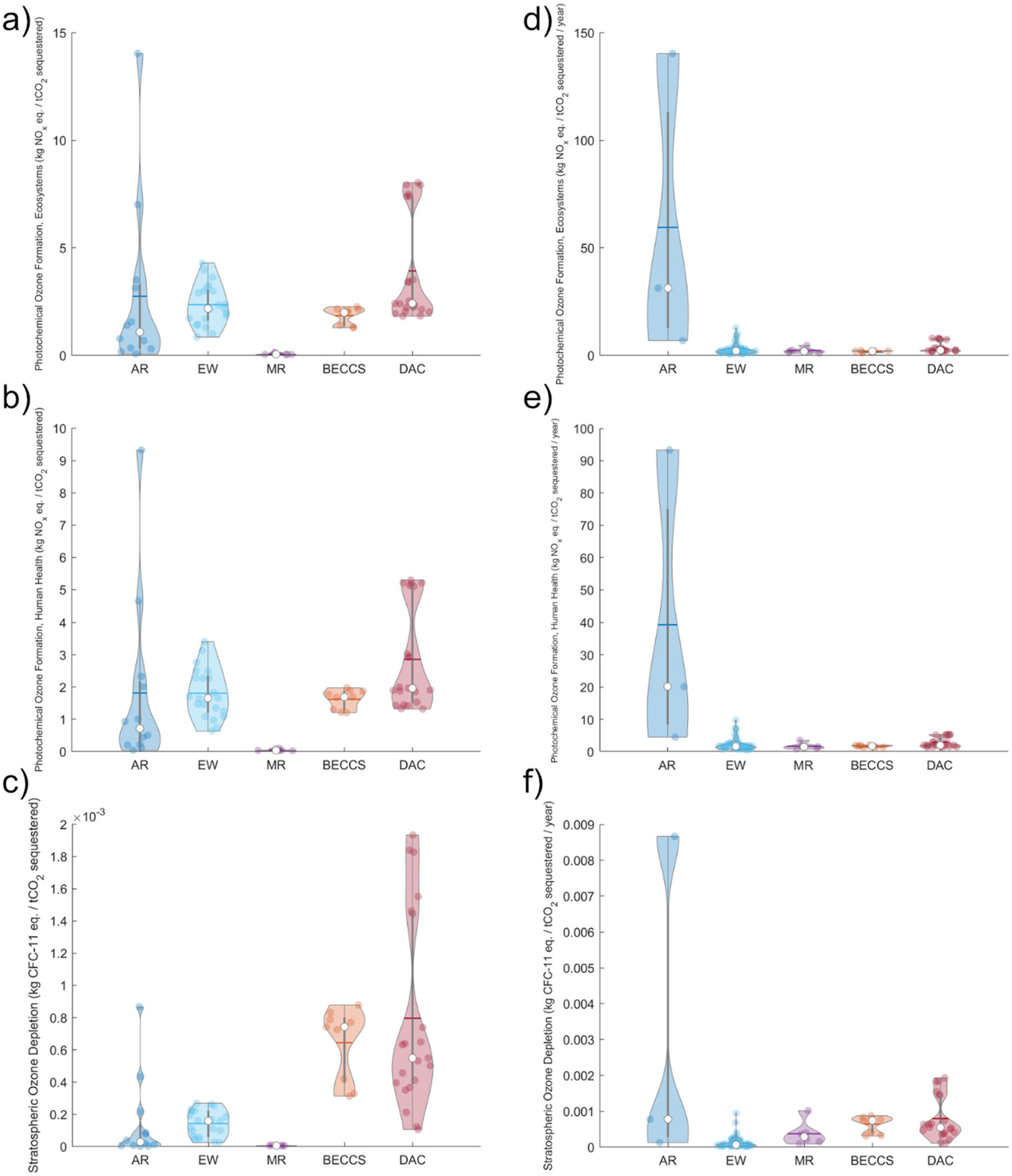


**Fig. 3.** Impacts to air of the NETs: afforestation/reforestation (AR), enhanced weathering (EW), mangrove restoration (MR), bioenergy with carbon capture and storage (BECCS) and direct air capture (DAC) with carbon storage. a) Photochemical ozone formation, ecosystems per t $CO_2$ sequestered. b) Photochemical ozone formation, human health per t $CO_2$ sequestered. c) Stratospheric ozone depletion per t $CO_2$ sequestered. d) Photochemical ozone formation, ecosystems per 1 t $CO_2$ sequestered in a year. e) Photochemical ozone formation, human health per 1 t $CO_2$ sequestered in a year. f) Stratospheric ozone depletion per 1 t $CO_2$ sequestered in a year. Refer to Fig. 2 for how to read the violin plots.

than the other NETs. The heat and power requirements of the DAC process account for most of the impacts to freshwater consumption and eutrophication while in EW it is the electricity needed to grind the olivine that is the main hot spot. An interesting result is that for EW, freshwater consumption increases when renewable energy is used for grinding, but this is only when hydroelectricity is used. When wind power is used, the impacts to freshwater consumption are greatly reduced.

When comparing on a 1 t $CO_2$ in a year basis, there is no shift in impacts to freshwater consumption but in freshwater eutrophication, EW, BECCS and DAC have similar impacts to MR and lower impacts than AR (Fig. 4). In freshwater ecotoxicity, BECCS and DAC have similar impacts to MR and lower impacts than AR. However, EW still has much higher impacts than the other NETs. AR and DAC can also have high impacts (to freshwater consumption, eutrophication and ecotoxicity), but these are much lower than EW.

### *4.4. Impacts to marine environments: marine ecotoxicity and eutrophication*

In impacts to marine environments, AR, MR and BECCS have the lowest impacts while EW and DAC have high impacts (Fig. 5). In AR,

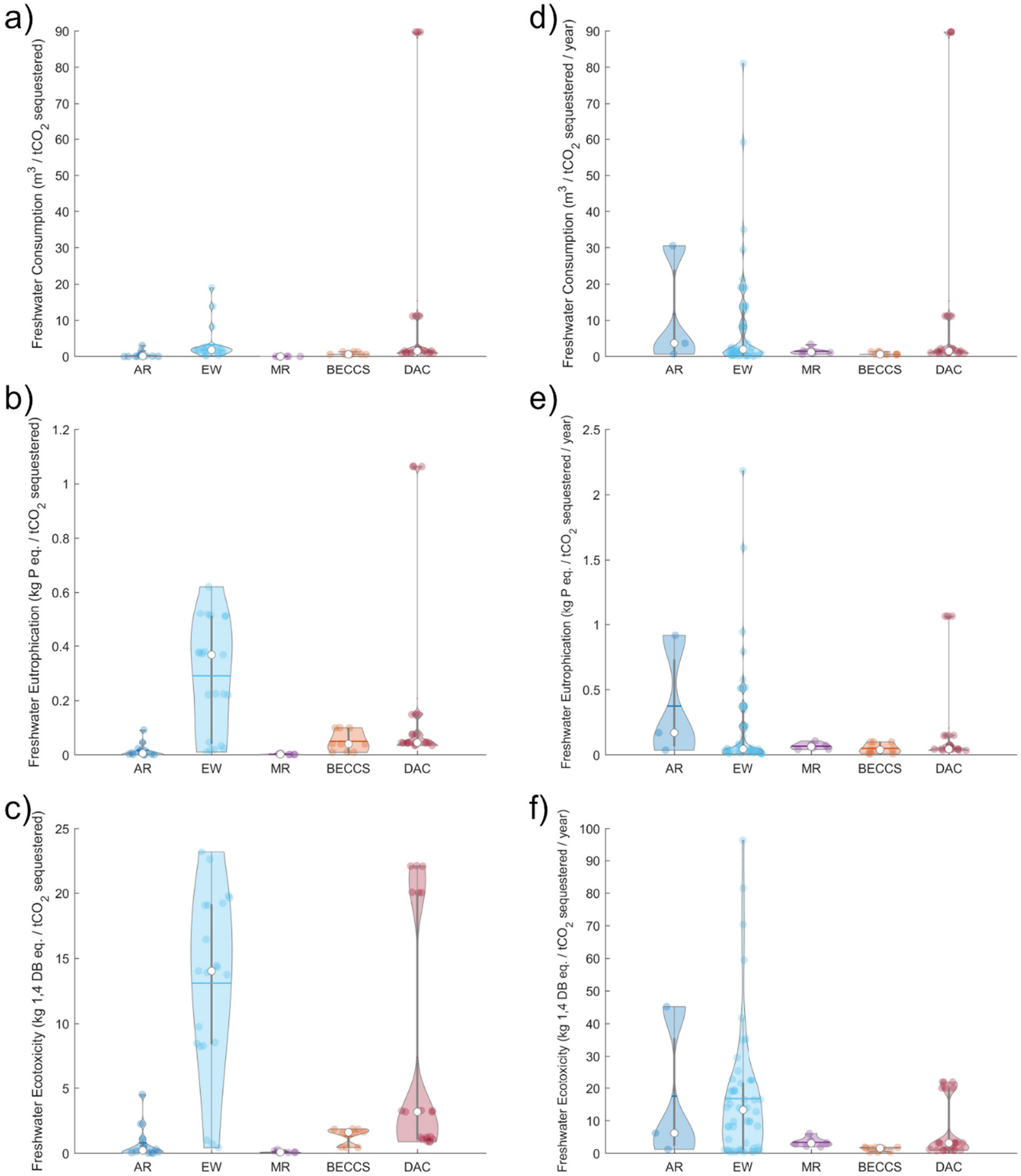


**Fig. 4.** Impacts to freshwater for the NETs: afforestation/reforestation (AR), enhanced weathering (EW), mangrove restoration (MR), bioenergy with carbon capture and storage (BECCS) and direct air capture (DAC) with carbon storage. a) Freshwater consumption per t $CO_2$ sequestered. b) Freshwater ecotoxicity per t $CO_2$ sequestered. c) Freshwater eutrophication per t $CO_2$ sequestered. d) Freshwater consumption per 1 t $CO_2$ sequestered in a year. e) Freshwater ecotoxicity per 1 t $CO_2$ sequestered in a year. f) Freshwater eutrophication per 1 t $CO_2$ sequestered in a year. Refer to Fig. 2 for how to read the violin plots.

the main impact hotspots are the plastic tree protector (ecotoxicity) and sawing in forestry management (eutrophication). In MR, the main hot spots are transporting the sapling and electricity used in the nursery (ecotoxicity) and sawing in forestry management (eutrophication) (Section 5 in the SI). As was mentioned previously, the impact of fertilisers is not included in our results. Had fertiliser been included, the impacts to marine ecotoxicity and eutrophication would be higher. In BECCS, the main emission hotspots are the feedstock and power plant (ecotoxicity) and feedstock and MEA (eutrophication). In marine ecotoxicity, EW has much higher impacts, mostly caused by mining and the electricity used for grinding. DAC has lower impacts, but impacts can be as high as EW when grid electricity and natural gas are used for heat and power. In marine eutrophication EW and BECCS have the highest impacts but AR can be much higher under a 10-year lifespan.

When 1 t $CO_2$ in a year is considered, DAC is as effective as BECCS and both have lower impacts than AR and MR (Fig. 5). EW has lower impacts in marine eutrophication but higher impacts in marine ecotoxicity. AR

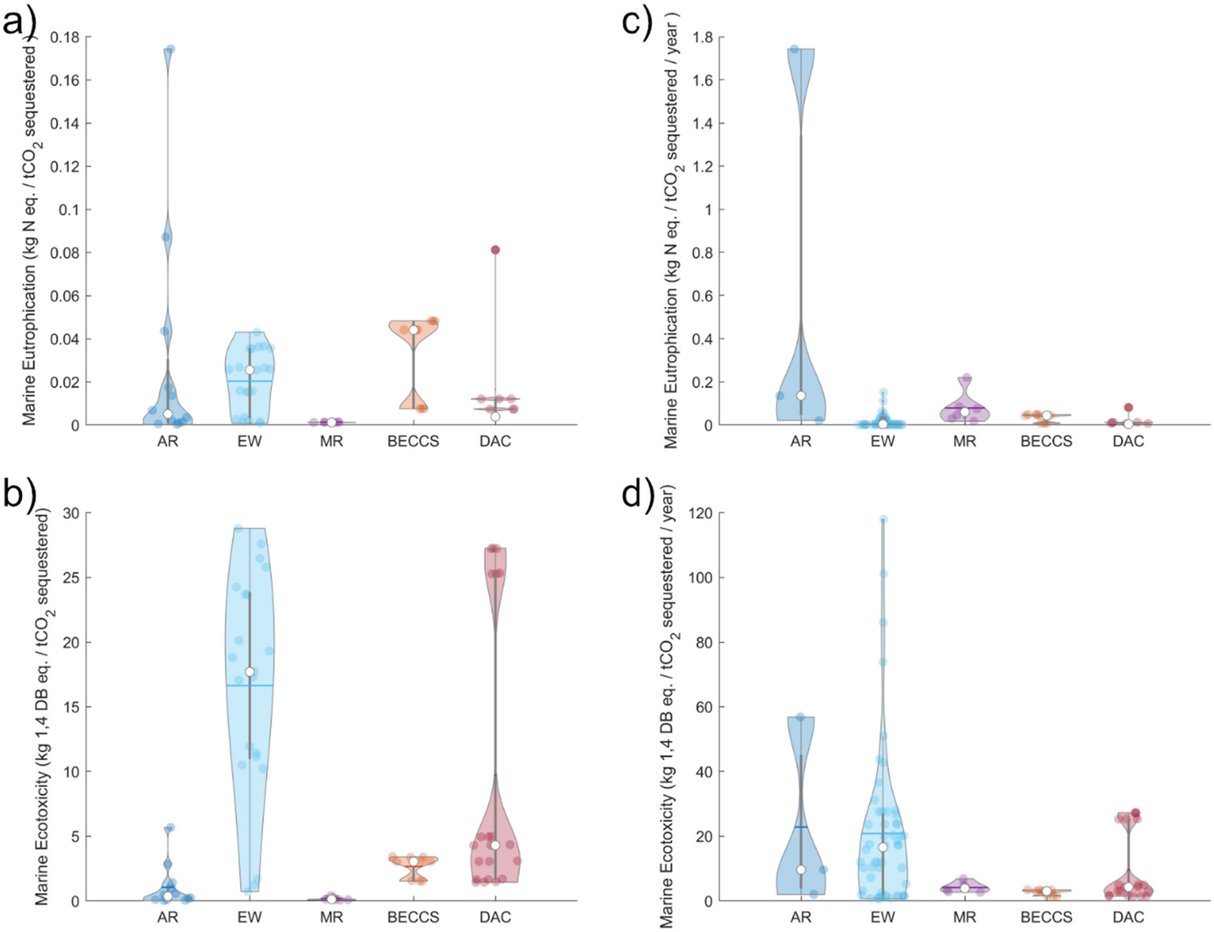


**Fig. 5.** Impacts to marine environments of the NETs: afforestation/reforestation (AR), enhanced weathering (EW), mangrove restoration (MR), bioenergy with carbon capture and storage (BECCS) and direct air capture (DAC) with carbon storage. a) Marine ecotoxicity per t $CO_2$ sequestered. b) Marine eutrophication per t $CO_2$ sequestered. c) Marine ecotoxicity per 1 t $CO_2$ sequestered in a year. d) Marine eutrophication per 1 t $CO_2$ sequestered in a year. Refer to Fig. 2 for how to read the violin plots.

can have high impacts when the tree density is low in both marine eutrophication and ecotoxicity but EW has the highest impact in marine ecotoxicity.

### *4.5. Impacts to human health: human toxicity, cancer and non-cancer*

When impacts to human health are compared, AR and MR have much lower impacts than the other NETs, except for human toxicity, non-cancer, where BECCS has lower impacts (Fig. 6) mostly due to benefits to human toxicity during miscanthus growing (removal of chemicals harmful to humans). In AR, the plastic tree protector is the main hotspot, while in MR it is transport (propagule and sapling). Across the toxicity indicators, EW is the NET with the highest impacts, mostly from the grinding, transporting and spreading the olivine. DAC has high impacts, mostly from heat and power needs and chemicals consumed ($CaCO_3$ in the pellet reactor).

On a 1 t $CO_2$ in a year basis, DAC has lower impacts than AR and MR, while BECCS has the lowest impacts across the NETs (Fig. 6). EW also has lower impacts than AR and MR and similar impacts to DAC. This is a significant shift in impacts and highlights how the context in which a NET is used can impact the LCA results.

### *4.6. Impacts to resources and land: terrestrial acidification and ecotoxicity, land use and metal depletion*

When impacts to resources and land are considered, MR has the lowest impact (Fig. 7). The forestry management and transport stages are the main emission hotspots (Section 5 in the SI). In comparison to BECCS, the other NETs have low impacts in land use; AR and EW have low impacts in metal depletion; AR and MR have low impacts in terrestrial acidification; MR has low impacts in terrestrial ecotoxicity. EW and DAC have high impacts in terrestrial acidification and ecotoxicity because of grinding and land spreading and grid electricity and natural gas for heat and power, respectively. In land use, BECCS has the highest impact because of agricultural needed- this indicator is a measure of the species lost (flora and fauna) incurred from land transformation, occupation and relaxation (Huijbregts et al., 2017). The impacts are calculated based on the species lost incurred relative to annual crop production (agriculture) (Huijbregts et al., 2017) and because of this, it is the biomass production stage which accounts for most of the impacts in this indicator. When woodchips are used the impact is reduced by 10 %, but in comparison to the other NETs, the impact is still high. In DACs the heat and power needs are the primary hot spot, while in EW it is mining (operating the quarry) and forestry management (sawing and soybean oil used during sawing) in AR. In metal depletion, the main emission hotspots are sawing, transport and the plastic tree protector in AR; grinding and land spreading in EW; heat and power in DACs; feedstock and power plant in BECCS.

When 1 t $CO_2$ in a year is considered, EW, BECCS and DAC see improvements (Fig. 7). In impacts to terrestrial acidification, DAC and AR have higher impact than the other NETs. In terrestrial ecotoxicity, AR has higher impacts than the other NETs while DAC has the lowest impact. In land use, BECCS is still the NET with the highest impacts, but AR and MR now have higher impacts than EW and DAC. In metal depletion, EW and DAC have the lowest impact, while AR, MR and BECCS have similar (higher) impacts. Across the impacts to metal depletion, land

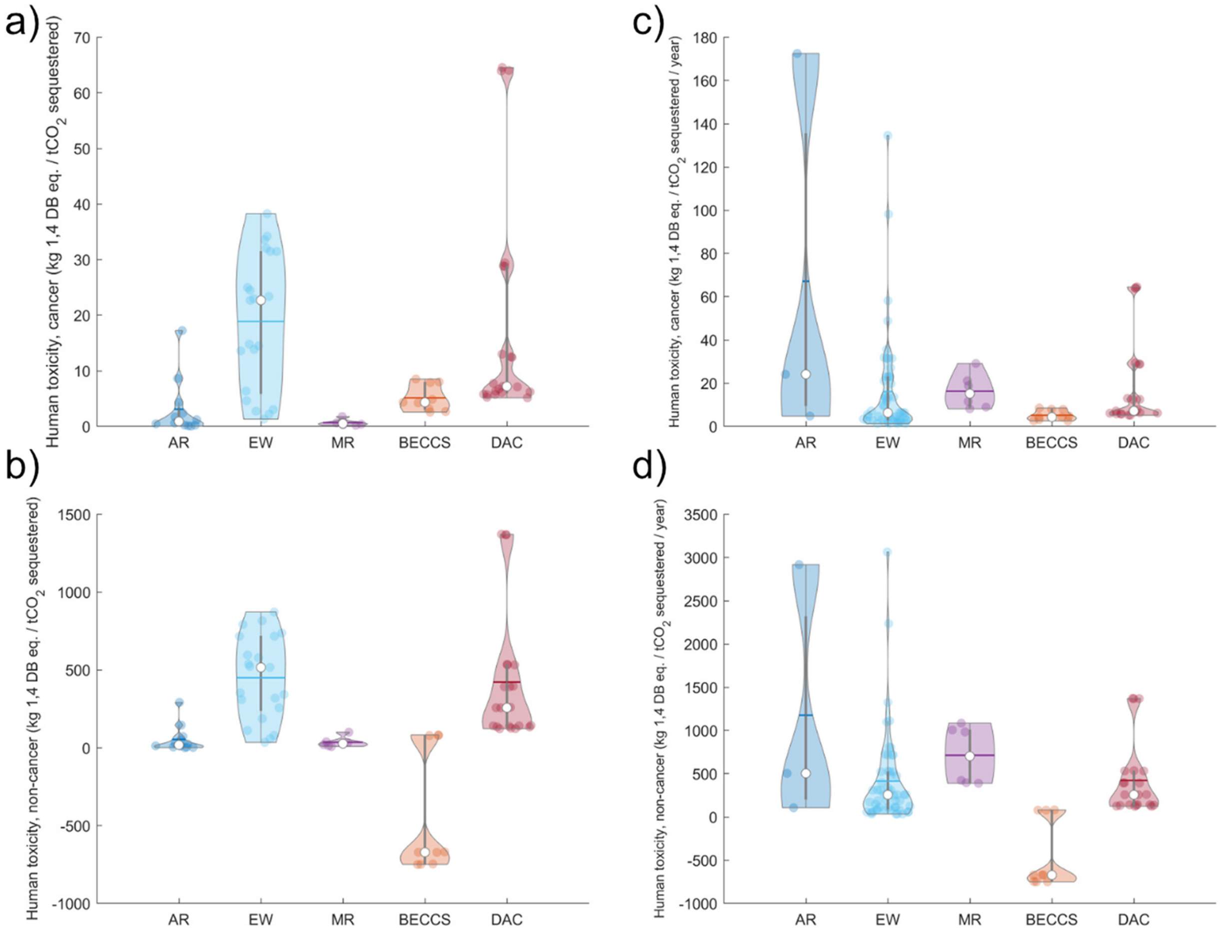


**Fig. 6.** Impacts to human health of the NETs: afforestation/reforestation (AR), enhanced weathering (EW), mangrove restoration (MR), bioenergy with carbon capture and storage (BECCS) and direct air capture (DAC) with carbon storage. A) Human ecotoxicity, cancer per t $CO_2$ sequestered. B) Human ecotoxicity, non-cancer, per t $CO_2$ sequestered. C) Human ecotoxicity, cancer per 1 t $CO_2$ sequestered in a year. d) Human ecotoxicity, non-cancer per 1 t $CO_2$ sequestered in a year. Refer to Fig. 2 for how to read the violin plots.

use and terrestrial acidification and ecotoxicity, AR has the highest impact when the tree density is low.

### 4.7. Comparison to the literature

Our results are in agreement with what other LCA studies have found. However, most studies only calculated impacts to climate change and global warming (GWP), so comparisons based on other environmental impacts are limited. For AR, our results are in agreement with the GWPs Gaboury et al. (2009) and Brunori et al. (2017) estimated (Fig. S81 in the SI). There are other AR LCA studies which have been conducted but these did not calculate the GWP. In MR, as far as the authors are aware, only Moriizumi et al. (2010) has conducted a LCA of mangrove plantations and found the GHGs emitted across the life cycle to be negligible (Table S7 in the SI). While we did estimate MR to be highly effective, we found GHG emissions to be small but not insignificant. The difference in the GWP estimates are the result of different activities included in the system boundary; Moriizumi et al. (2010) did not include forestry management and they consider mangroves planted in Thailand.

For EW, as far as the authors are aware, only Lefebvre et al. (2019) has conducted a EW LCA. They considered EW in Brazil and did not grind olivine to 10 μm grains, as well as other differences in system boundaries. When we omitted grinding and adjusted our GaBi model to match their parameters (e.g. Brazil electricity mix, 65 km truck distance etc.), our results (GWP and human toxicity) are comparable but higher (Table S6 in the SI). The difference can be attributed to how the land spreading is modelling. We modelled land spreading using ecoinvent data for the loader while Lefebvre et al. (2019) used USLCI data and consequentially diesel consumption is higher in our model.

Our results for DAC are in agreement with those of Terlouw et al. (2021b), Deutz and Bardow (2021), Madhu et al. (2021), de Jonge et al. (2019) and Liu et al. (2020) (Table S9 in the SI). We also find that the heat and power source is the key factor in the cradle to grave GWP. Of particular importance is that our results are in agreement with Madhu et al. (2021) and Liu et al. (2020), who also built their DAC LCA models based on the Carbon Engineering plant. Terlouw et al. (2021b) estimated impacts to freshwater consumption, eutrophication, photochemical ozone formation and human toxicity and our results are comparable (Table S10 in the SI). Deutz and Bardow (2021) estimated the impacts to freshwater eutrophication, photochemical ozone formation and toxicity but for the sorbents used in DAC rather than the DAC process.

In BECCS, our results are in agreement with the LCA studies by Bennett et al. (2019), Pour et al. (2018) and Cheng et al. (2020) (Table S8 in the SI) and found the feedstock/fuel to be the key factor which impacts the GWP. There are other BECCS LCA studies but these either modelled co-fired biomass or municipal solid waste incineration power plants. Bennett et al. (2019) estimated the water use but this is not comparable with our results as we estimate impacts to freshwater consumption (water that has evaporated, been incorporated into products, transported to other watersheds or disposed of to the sea i.e. water no longer available to the watershed (Huijbregts et al., 2017)) rather than use.

## 5. Discussion

As emission targets tighten, carbon offsetting via NETs will play an increasing important role in ensuring we do not overshoot Paris Agreement targets (IEA, 2021, CCC, 2020). However, as emission targets become increasingly tight, there is less contingency available in emissions accounting. This is important to NETs, as GHG emissions will occur throughout a NET's life cycle. If these are not accounted for in emission budgets, they could have dire consequences to meeting Paris Agreement goals and other environmental targets.

Our LCA found that out of AR, EW, MR, DAC and BECCS, AR and MR are the most effective, on a 1 t $CO_2$ basis, as they have the lowest impacts (MR has the lowest impact in 15/16 indicators and AR is second lowest in 10/16) across the indicators considered in this paper. Focussing on climate change and global warming, they are much more effective than the other NETs, but their rate of $CO_2$ removal is much slower and on a 1 t $CO_2$ in a year basis DAC and BECCS have similar impacts while EW has lower impacts. This is an important aspect to consider in emission reduction strategies and strategies to roll out NETs. Of the NETs, EW and DAC are the least effective on a 1 t $CO_2$ basis, as they have higher impacts across most of the indicators (EW has the highest impacts in 5/16 indicators, DAC the highest in 8/16). On a 1 t $CO_2$ in a year basis, the impacts shift in favour of EW, BECCS and DAC. This is because the rate of $CO_2$ removal is slow in AR and MR. Therefore, when devising strategies to roll out NETs, the context in which a NET is used is important i.e., are they to remove large quantities of $CO_2$ in short time periods or to remove $CO_2$ consistently over long timeframes. This is important as it directly impacts which NET is the most effective. If large $CO_2$ removal over short timeframes is the desired output, then EW, DAC and BECCS are the NETs which have the lowest environmental impacts, but if longer time horizons are considered i.e., over a project's lifespan, then AR and MR have the lowest impact.

Another consideration to NET rollout is land occupation and $CO_2$ sequestration costs. AR, MR, EW and BECCS will have a higher impact to land occupation than DAC due to the need to plant trees, produce feedstock and spread olivine. If cost is considered, DAC and BECCS are large industrial processes and will have higher capital, operating and maintenance costs than AR, MR and EW. Resource availability is another important factor to consider as olivine is not found in all countries and shipping it long distances would likely be expensive and significantly impede its $CO_2$ removal capacity.

A further consideration, applicable to DAC and BECCS only, is that despite DAC having higher impacts, its capability to remove $CO_2$ is (technically) unlimited as it removes $CO_2$ directly from the atmosphere, while BECCS removes atmospheric $CO_2$ via photosynthesis. This means that DAC is not limited in its $CO_2$ removal capacity as its $CO_2$ feedstock is not hindered by factors such as supply chain issues, price, or transport issues. However, it is more energy and materially (more chemicals consumed) intensive to sequester $CO_2$ via DAC than BECCS and thus it will likely have higher environmental impacts under many scenarios. Also, the effectiveness of DAC to remove $CO_2$ is dependent on the concentration in the air; in areas where the concentration of $CO_2$ is below average background levels, DAC may be less efficient. This would similarly apply to AR, MR and EW, as they also remove $CO_2$ directly from the atmosphere.

The results of the LCA show that there is a wide range in the environmental impacts of the NETs considered. AR and MR are better over longer time periods but if large and quick $CO_2$ removal is needed then EW, DAC and BECCS are the better options. However, if comparing on a one-to-one basis which NET to prioritise in negative emission and emission offsetting strategies, then our LCA results infer that MR is the most effective as it has low impacts across the indicators in both functional units. However, if impacts to climate change and global warming are prioritised, then EW, DAC and BECCS can be the NETs to prioritise if large and quick $CO_2$ removal is needed. However, our results should be interpreted with caution as we consider a limited number of NETs and base our models on literature data rather than primary data.

### 5.1. Limitations of the work

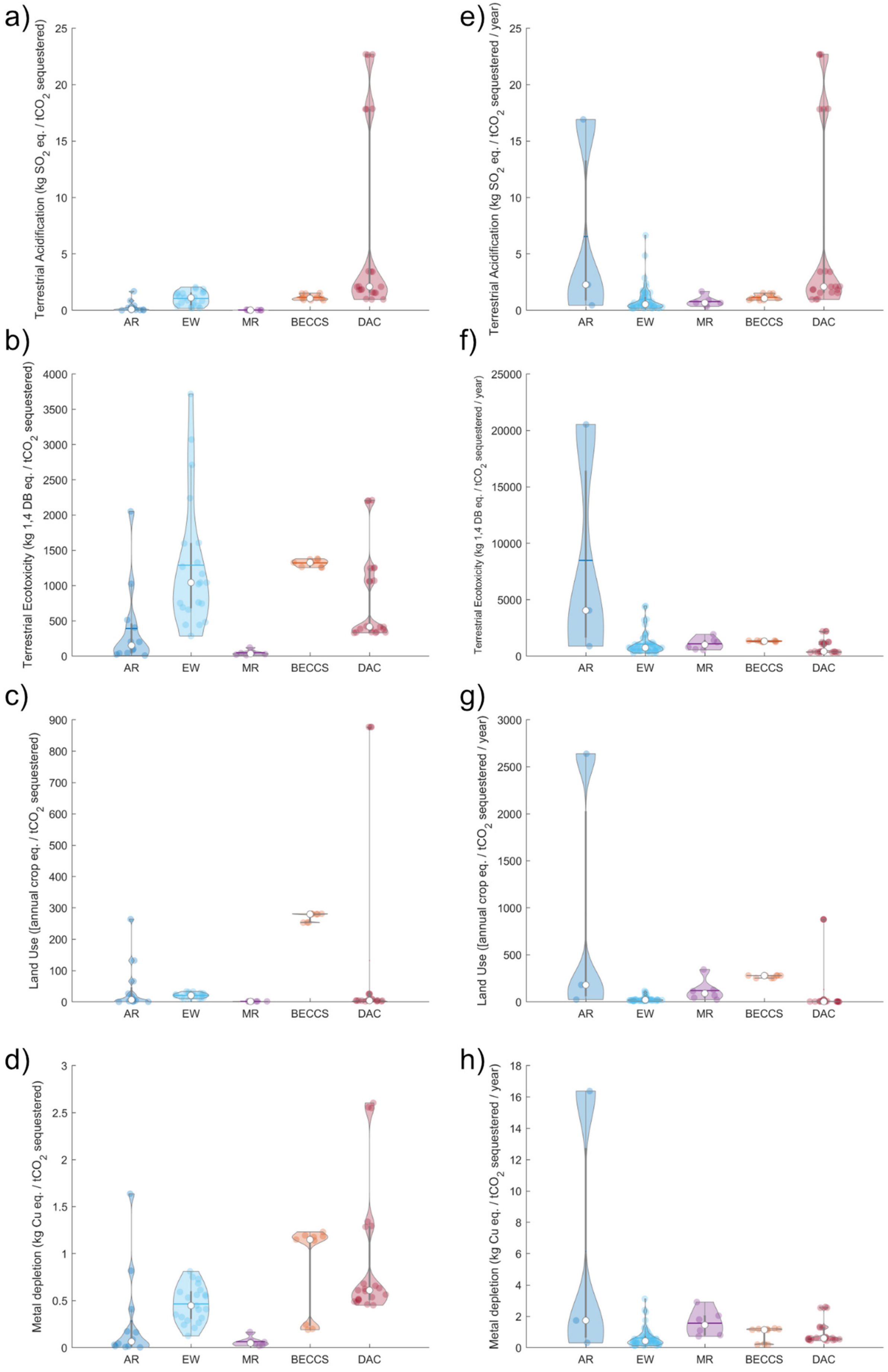
While we have assessed the life cycle environmental impacts of five NETs and measured the impacts through 17 indicators, there are several limitations to the work. The reliance on literature/secondary data is the main limitation. As we built our LCA models using only literature data, several assumptions were made, such as no fertiliser used in AR and MR and transport distances for vehicles and pipelines. A further limitation to using literature data is that the data was inconsistent in factors such as temporal scale and geography as we had to used multiple sources to collect enough data to build our LCA models. The reliance on ecoinvent data is also a limitation. Whilst this dataset is extensive and high quality, it does not provide data specific to the USA and Canada for all of the energy and materials used in the LCA models and consequentially, data for other geographies were used instead. As a result, our LCA models are not completely representative of NETs being/to be deployed in the USA and Canada. We do consider a wide variation in parameters in our sensitivity analyses and explore a range in potential NET configurations through different scenarios. Despite this, because of the limitations listed there will be uncertainties in our results that we were unable to capture/fully account for.

### 5.2. Future work

Our LCA models are based on literature data and as such, a number of assumptions were made in our modelling. All of the NET models were built using data from multiple sources and consequentially there is a degree of inconsistency in the data. Therefore, future studies should use data collected directly from NET projects if possible, or from fewer studies/sources to improve consistency in the data. We have attempted to take into account temporal impacts by estimating impacts per t $CO_2$ sequestered in a year in addition to per t $CO_2$ sequestered. To better account for temporal effects, future studies should include carbon discounting factors, as well as more detailed $CO_2$ removal rates (for AR and MR as we assume a constant rate).

Given that literature data was used to build our LCA models, and we consider deployment in the USA and Canada only and the small number of LCA studies conducted for some of the NETs modelled. More LCA studies should be conducted on the NETs considered in this work to better understand and characterise their environmental impacts. While we have estimated the life cycle environmental impacts of five NETs, there are other NETs and NET configurations not considered in this paper. There are alternative DAC processes available, such as solid amine instead of liquid and BECCS encompasses a variety of bioenergy processes such as biofuel production. There are also other means of increasing the size of the carbon sink besides AR and MR; soil carbon sequestration (such as grassland, improved soil management and farming practices) and blue carbon (preservation and restoration of other coastal/marine habitats) will also increase the size of the carbon sink. There are also other NETs not considered in this paper, such as biochar and land management, and while we have assessed the environmental impacts using 17 indicators, there are impacts which will be important that we have not considered, including water footprint and resource depletion. Therefore, future studies

**Fig. 7.** Impacts to land and metal depletion of the NETs: afforestation/reforestation (AR), enhanced weathering (EW), mangrove restoration (MR), bioenergy with carbon capture and storage (BECCS) and direct air capture (DAC) with carbon storage. a) Terrestrial acidification per t $CO_2$ sequestered. b) Terrestrial ecotoxicity per t $CO_2$ sequestered. c) Land use per t $CO_2$ sequestered. d) Metal depletion per t $CO_2$ sequestered. e) Terrestrial acidification per 1 t $CO_2$ sequestered in a year. f) Terrestrial ecotoxicity per 1 t $CO_2$ sequestered in a year. g) Land use per 1 t $CO_2$ sequestered in a year. h) Metal depletion per 1 t $CO_2$ sequestered in a year. Refer to Fig. 2 for how to read the violin plots.


a)
Terrestrial Acidification (kg SO2 eq. / tCO2 sequestered)
0
5
10
15
20
25
AR
EW
MR
BECCS
DAC
e)
Terrestrial Acidification (kg SO2 eq. / tCO2 sequestered / year)
0
5
10
15
20
25
AR
EW
MR
BECCS
DAC
b)
Terrestrial Ecotoxicity (kg 1,4 DB eq. / tCO2 sequestered)
0
500
1000
1500
2000
2500
3000
3500
4000
AR
EW
MR
BECCS
DAC
f)
Terrestrial Ecotoxicity (kg 1,4 DB eq. / tCO2 sequestered / year)
0
5000
10000
15000
20000
25000
AR
EW
MR
BECCS
DAC
c)
Land Use ([annual crop eq. / tCO2 sequestered)
0
100
200
300
400
500
600
700
800
900
AR
EW
MR
BECCS
DAC
g)
Land Use ([annual crop eq. / tCO2 sequestered / year)
0
500
1000
1500
2000
2500
3000
AR
EW
MR
BECCS
DAC
d)
Metal depletion (kg Cu eq. / tCO2 sequestered)
0
0.5
1
1.5
2
2.5
3
AR
EW
MR
BECCS
DAC
h)
Metal depletion (kg Cu eq. / tCO2 sequestered / year)
0
2
4
6
8
10
12
14
16
18
AR
EW
MR
BECCS
DAC

should also focus on the NETs and environmental impacts not covered in this paper. These are all needed to build a better picture of how environmentally sustainable NETs are.

Whilst understanding the environmental impacts of NETs is important, it is only one of the three spheres of sustainability. To understand how sustainable NETs are, economic and social life cycle studies are needed, in addition to studies which combine the three spheres of sustainability to calculate the overall life cycle sustainability. Therefore, future studies are needed to assess the economic, social and overall life cycle sustainability of NETs.

## 6. Conclusions

As we move further into the 21st century negative emission technologies (NETs) will become increasingly important in meeting emission targets. However, emissions of greenhouse gases (GHGs), and other chemicals that cause harm to the environment, will be emitted throughout the life cycle of a NET. Therefore, it is crucial the life cycle environmental impacts be understood, as otherwise NET rollouts could risk being less effective and incompatible with well below 2 °C targets. In this paper we have compared five NETs: afforestation/reforestation (AR), enhanced weathering (EW), mangrove restoration (MR), bioenergy with carbon capture and storage (BECCS) and direct air capture (DAC), using life cycle assessment (LCA) to determine which have lower/higher environmental impacts.

We find that AR and MR are the most effective when a functional unit of 1 t $CO_2$ sequestered is considered across most of the indicators considered. MR has the lowest impact in 15/16 indicators and AR has the second lowest impacts in 10/16 indicators. For this functional unit, BECCS has higher impacts than AR and MR in 13/16 indicators, but lower impacts than DAC and EW in 10/16 indicators. EW has the highest environmental impacts in $GWP_{20}$, freshwater and marine eutrophication and ecotoxicity and human health, mostly from the electricity needed to grind the olivine to 10 ɥm diameter grains. However, when a functional unit of 1 t $CO_2$ in a year is considered, the impacts shift in favour of EW, BECCS and DAC; EW has the lowest impacts in 8/16 indicators, DAC the lowest in 7/16 and BECCS the lowest in 7/16. Thus, the context to which NETs are used is important as it directly effects which NET is preferable.

Overall, we find that there are distinct differences in the life cycle environmental impacts of NETs which need to be taken into consideration in net-zero strategies. If these are not considered, and measures taken to reduce the impacts, countries risk not meeting their pledges made to meet Paris Agreement goals, as well as causing other damage to the environment. However, our results are limited to a small selection of NET configurations and are specific to the USA and Canada. Therefore, future work should focus on the NETs considered in this work (explore impacts when deployed in other countries), as well as the NETs not studied. These studies are needed to build a better understanding of the life cycle environmental impacts of NETs, to improve our understanding on how to use them responsibly and effectively.

## CRediT authorship contribution statement

**Jasmin Cooper:** Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Visualization, Project administration. **Luke Dubey:** Conceptualization, Investigation, Writing – original draft, Writing – review & editing, Visualization. **Adam Hawkes:** Conceptualization, Writing – review & editing.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

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