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Assessment of technologies and economics for carbon dioxide removal from a portfolio perspective

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ABSTRACT

Carbon dioxide removal (CDR) is essential to achieve ambitious climate goals limiting global warming to less than 1.5°C, and likely for achieving the 1.5°C target. This study addresses the need for diverse CDR portfolios and introduces the LUT-CDR tool, which assesses CDR technology portfolios aligned with hypothetical societal preferences. Six scenarios are described, considering global deployment limitations, techno-economic factors, area requirements, technology readiness, and storage security for various CDR options. The results suggest the feasibility of large-scale CDR, potentially removing 500–1750 $GtCO_2$ by 2100 to meet the set climate targets. For a 1.0°C climate goal, CDR portfolios necessitate 12.0–37.5% more primary energy compared to a scenario without CDR. Remarkably, funding a 1.0°C target requires only 0.42–0.65% of the projected global gross domestic product. Bioenergy carbon capture and sequestration and rainfall-based afforestation play limited roles, while secure sequestration of captured CO_2 via direct air capture, electricity-based carbon sequestration, and desalination-based afforestation emerge as more promising options. The study offers crucial techno-economic parameters for implementing CDR options in future energy-industry-CDR system analyses and demonstrates the tool's flexibility through alternative assumptions. It also discusses limitations, sensitivities, potential trade-offs, and outlines options for future research in the area of large-scale CDR.

1. Introduction

Fossil fuel use accelerated during the industrial revolution and has continuously distorted the natural carbon cycle ([IPCC] 2013). This use was and still is, contributing to today's global warming (Steffen et al., 2018), causing us to enter the new geological epoch of the 'Anthropocene' (Lewis and Maslin, 2015). Notably, for the last three decades, global anthropogenic carbon dioxide (CO_2) emissions doubled (Friedlingstein et al., 2022), despite deep knowledge of key drivers and hazardous effects of global warming (Stoddard et al., 2021). The fact that global warming is human-caused is a scientific consensus (Cook et al., 2016, [IPCC] 2021). Vested interests of fossil fuels industry (Supran et al., 1979, Li et al., 2022, Kenner and Heede, 2021) hinder and urge the necessity of rapid defossilisation of all sectors in the energy system to curb the most severe impacts of climate change on humanity and the environment (Stoddard et al., 2021). There is growing understanding that net-zero CO_2 emissions by 2050 might not be sufficient to limit global warming to a sustainable level or even the 1.5°C aim stated in the Paris Agreement in 2015 ([IPCC] 2022, [IPCC] 2015, [UNFCCC] 2015), as lower atmospheric CO_2 levels are required for climate safety (Hansen et al., 2017, Azar and Rodhe, 1979). Hansen et al. (Hansen et al., 2017, Hansen et al., 2008) point out that climate safety requires an atmospheric CO_2 concentration of about 350 ppm corresponding to a climate target of about 1.0°C. Similarly, Rockström et al. (Rockström et al., 2023) conclude that a 1.0°C target is required for a safe climate respecting the Sustainable Development Goals. Although certainly ambitious, limiting global warming to 1.0°C by the end of the century is necessary to ensure a safe planet (Breyer et al., 2023). So far, no attempt was made to explore techno-economic system analyses to achieve such ambitious climate targets.

Delayed climate action might result in a significant temperature overshoot for several decades, possibly causing severe, irreversible consequences (Iyer et al., 2022) and disastrous climate tipping point (CTP) events (Lenton et al., 2008, Zhang et al., 1979), and cascades (Lenton et al., 2019, Liu et al., 2023, Wunderling et al., 2023). Therefore, immediate emissions reduction must be prioritised to maximise climate benefits and to avoid hazardous ramifications (Iyer et al., 2022,

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Nomenclature

AF	Afforestation
AF_{DB}	Desalination-based afforestation
AF_{RF}	Rainfall-based afforestation
AR	Reforestation
AR6	Sixth Assessment Report
BAWL	Buffered accelerated weathering of limestone
BC	Biochar production
BECCS	Bioenergy with carbon capture and sequestration
BECCU	Bioenergy with carbon capture and utilisation
BP	Balanced portfolio
Capex	Capital expenditures
CCS	Carbon capture and sequestration
CCU	Carbon capture and utilisation
CCUS	Carbon capture, utilisation, and sequestration
CDR	Carbon dioxide removal
CO_2	Carbon dioxide
COP	Coefficient of performance
CP	Conventional pyrolysis
CTP	Climate tipping point
DACCS	Direct air capture with carbon sequestration
DOG	CO_2 sequestration in depleted oil and gas fields
DSA	CO_2 sequestration in deep saline aquifers
EW	Enhanced weathering
e-CF	Electricity-based carbon fibre
e-SiC	Electricity-based silicon carbide
FE	Final energy
FED	Final energy demand
GDP	Gross domestic product
GHG	Greenhouse gas
GSAT	Global surface air temperature
HSP	High security portfolio
IAM	Integrated assessment model
IPCC	Intergovernmental Panel on Climate Change
IW	Industrial waste
LCOCDR	Levelised cost of carbon dioxide removal
LCA	Life cycle assessment
LCP	Low-cost portfolio
LEP	Low-energy portfolio
LUT-DEES	LUT delayed economic equality scenario
MEA	Monoethanolamine
MIN_{EX}	Ex-situ mineralisation
MIN_{IN}	In-situ Mineralisation
MR	Mined rock
NCS	Natural climate solutions
NET	Negative emission technology
OFF	Offshore
ON	Onshore
Opex	Operational expenditures
p	Process
PE	Primary energy
PED	Primary energy demand
PSC	CO_2 point source capture
PV	Photovoltaics
SCS	Soil carbon sequestration
TNE	Total negative CO_2 emissions
TFED	Total final energy demand
TPED	Total primary energy demand
TRL	Technology readiness level
WACC	Weighted average cost of capital
WGI	Working Group I
WGIII	Working Group III

[IPCC] 2022). Thereafter, ambitious CDR in the order of 11 - 39 $GtCO_2/a$ might be necessary to actively reduce the atmospheric CO_2 concentration to a sustainable level ([IPCC] 2022, Abbott et al., 2023, Breyer et al., 2021, Keiner et al., 2023, Smith et al., 2023). The definition of sustainability can often be ambiguous, and the dimensions focused on in this study are cost, energy demand, area demand, and permanence of CDR activities aligning in part with established sustainability definitions.

The current understanding is that negative CO_2 emission technologies (NET) (Fuss et al., 2014, Minx et al., 2018, Smith et al., 2016) will be required to enable carbon dioxide removal (CDR) from atmosphere to go net-negative, partially to offset unavoidable emissions from industry or agriculture and to actively decrease the atmospheric CO_2 concentration (Smith et al., 2023, Sovacool, 2021). To reach net-zero CO_2 emissions globally while satisfying the energy demand of humankind, the immediate scale-up of renewable energy and phase-out of fossil fuels must be prioritised. This transition ahead creates the necessity of investigating future energy-industry-CDR systems (Breyer et al., 2022). CDR is often associated with carbon capture and sequestration (CCS) operations and is criticised as a pretext to perpetuate the use of fossil fuels (Asayama, 2021). Therefore, it should be emphasised that CDR must not be a substitute for emission reductions but rather be used to complement the phase-out of fossil fuels and to account for unavoidable emissions (Asayama, 2021, Fankhauser et al., 2022).

Scenarios identified by Integrated Assessment Models (IAM) rely mostly on afforestation (AF) and bioenergy with carbon capture and sequestration (BECCS) deployment to reach climate mitigation goals ([IPCC] 2022). The projected scale might surpass the actual potential of these approaches (Köberle, 2019, Grant et al., 2022). While CDR and CO_2 sequestration options are discussed frequently in literature (Minx et al., 2018, Fuss et al., 2018, Nemet et al., 2018) (cf. Section 2), an assessment of comprehensive CDR portfolios based on a flexible CDR portfolio creation based on societal preferences is not yet available. The aim of this study is to provide a techno-economic assessment based on a flexible portfolio creation tool for an assessment of the techno-economics of various CDR portfolios in a system context. This study aims to tackle the following identified research gaps related to CDR portfolios by the following novelties:

- In-depth creation of CDR portfolios avoiding black box solutions such as direct air capture and sequestration (DACCS) or BECCS; instead of incorporating typical 5–7 NETs, the novel portfolio structure of this study includes more than 25 different CDR technology routes.
- Portfolio variation considering balanced, high-security, low-cost, low-energy, low-area, and high-TRL features, representing various societal preferences for possible future deployment of CDR with a newly introduced portfolio creation tool reflecting a climate target of 1.5°C as of the Paris Agreement but also 1.0°C for climate safety.
- Detailed techno-economic analysis of NETs and CDR portfolios within the context of global gross domestic product (GDP) and total primary energy demand (TPED) estimations.

The assessment is done by a harmonised identification of energy and mass balances, global potential, technology readiness levels (TRL), area demand, and costs of technologies that are vital for successful large-scale NET deployment in unprecedented detail and variety. This research provides a literature review on CDR portfolios in Section 2. Section 3 describes the methods applied in this study. The findings are presented in Section 4. Results are discussed in Section 5 and conclusions from this research are drawn in Section 6.

2. Literature review

Limiting global warming to 1.5°C requires about 420 - 1100 $GtCO_2$ of negative CO_2 emissions by 2100, according to calculations adapting findings of the Intergovernmental Panel on Climate Change (IPCC) ([IPCC] 2022, Smith et al., 2023) as presented by Keiner et al. (Keiner et al., 2023). Breyer et al. (Breyer et al., 2022) estimated that around 1483 $GtCO_2$, complying with the cumulative anthropogenic emissions until 2050, must be removed from the atmosphere to reach a level of atmospheric CO_2 concentration of about 350 ppm and limit average global warming to around 1.0°C as a required level of climate safety (Hansen et al., 2017, Azar and Rodhe, 1979, Rockström et al., 2023, Breyer et al., 2023, Clark et al., 2016), if fossil fuel use can be abated by mid-century.

The requirement for CDR portfolios and the benefits thereof, as well as the limitations of future BECCS deployment often presented as the sole or main NET, can be found in literature (Rickels et al., 2019, Rueda et al., 2021). Fuss et al. (Fuss et al., 2020) emphasise the benefit of promoting diverse CDR portfolios to reduce the risk related to the high dependence on large-scale deployment of few NETs and Fuhrman et al. (Fuhrman et al., 2023) point to benefits of diverse portfolios regarding cost, energy, and water demand as well as land system impacts. DACCS is discussed more in diverse CDR portfolios in the recent past (Realmonte et al., 2019, Breyer et al., 2019, Fuhrman et al., 2021), partially due to the limitations of land and water availability for BECCS and AF (Creutzig et al., 2019, Gambhir et al., 2019, Jones and Albanito, 2020). Rueda et al. (Rueda et al., 2021) create CDR technology portfolios using a framework that prioritises technologies based on weighted indicators for feasibility, effectiveness, and side impacts of NETs. Mainly ordinal data were used to determine the performance of technologies in the employed indices (Rueda et al., 2021), while only costs are based on continuous data taken from Fuss et al. (Fuss et al., 2018) and Minx et al. (Minx et al., 2018). Using the weighted sum method, a total score is calculated to prioritise the NETs thereafter before options are deployed to fulfil the CDR demand (Rueda et al., 2021). Rueda et al. (Rueda et al., 2021) consider up to six NETs for portfolio creation. Detailed specifications of NETs such as DACCS are missing. Fuhrman et al. (Fuhrman et al., 2023) incorporate DACCS, BECCS, AF, enhanced weathering (EW), biochar (BC) and direct ocean CO_2 capture and sequestration within the global change analysis model GCAM in a recent study. Abraham et al. (Abraham et al., 2022) investigate possible synergies of DACCS, BECCS, EW, BC, AF, and reforestation (AR) in terms of shared resources among the options. With this approach, the study claims the profit-optimal integrated NET system can create a profit of up to 315 USD/tCO_2 removed if carbon tax and subsidies of 544 USD/tCO_2 are applied (Abraham et al., 2022).

CDR is examined further in different ways: Babacan et al. (Babacan et al., 2020) examine DACCS and BECCS in terms of energy consumption, Kang et al. (Kang et al., 2021, Kang et al., 2022) review developments and innovations in CDR and CCS through patent analysis, Migo-Sumagang et al. (Migo-Sumagang et al., 2022) employ fuzzy mixed-integer programming to investigate CDR portfolios considering AR, BC, SCS, BECCS, EW, and DACCS, and Perdana et al. (Perdana et al., 2023) conduct an expert survey on perceptions on future mitigation potential and phase-in of DACCS and other technologies correlated with CDR. Further, Sovacool et al. (Sovacool et al., 2023) focus their survey on potential risks of NETs and McLaughlin et al. (McLaughlin et al., 2023) investigate the carbon capture, utilisation, and sequestration (CCUS) nexus, including CDR, from a sociotechnical perspective. However, Mertens et al. (Mertens et al., 2023) and Bruhn et al. (Bruhn et al., 2016) suggest separating the terms carbon capture and utilisation (CCU) and CCS due to different functions, business directions, role in the energy transition, and policy requirements. Nevertheless, some cases exist that justify CCUS as a correct term, since both CCU and CCS in the form of CDR can be fulfilled, as for electricity-based silicon carbide (e-SiC) (Mühlbauer et al., 2024) and electricity-based carbon fibre (e-CF) (Keiner et al., 2023).

The range and median of annual CO_2 sequestration, i.e., the capture with subsequent permanent sequestration or removal, potential, cost of CDR per tonne of CO_2 removed, and TRL as identified in the literature is visualised in Fig. 1. Consequently, Fig. 1 shows results of previous studies which are affected by scenario specifications and other assumptions not further assessed in this study rather than techno-economic assumptions which are derived from literature in Section 3.2. More detailed information on literature findings can be found in Tables S1-S5 in the supplementary material 1, note 1.

In current literature, there is a general uncertainty regarding the future potential of the different NETs. In addition, not all studies provide the year for which the authors present information. For Fig. 1, in such cases, the potential is assumed to be for 2100. Almost all NETs show a wide range of estimated potential. DACCS is estimated to be able to contribute 0.5 - 5.0 $GtCO_2$/a, with an average estimate of 2.8 $GtCO_2$/a, to the global CDR demand by 2050. Notably, the 2050 range for the global annual DACCS potential is consistent throughout the reviewed literature (Minx et al., 2018, Fuss et al., 2018, Brack and King, 2021). Minx et al. (Minx et al., 2018) suggest the high uncertainty in deployment results from different scale-up scenarios of relatively new technologies. In 2100, global DACCS potential estimates are 0.0 - 40.0 $GtCO_2$/a, with an average of 12.8 $GtCO_2$/a. Assuming no major sustainability issues arise, Fuss et al. (Fuss et al., 2018) anticipate that large-scale DACCS deployment will not be constrained and therefore assume that a DACCS deployment of around 40.0 $GtCO_2$/a, as also found by Chen and Tavoni (Chen and Tavoni, 2013), might be feasible. Sovacool (Sovacool, 2021), conversely, estimates minimal deployment of DACCS with 0.0 $GtCO_2$/a as a lower limit.

There seems to be a rough consensus on the potential of BECCS in 2050 with a range of 0.5 - 5.0 $GtCO_2$/a given in literature (Minx et al., 2018, Fuss et al., 2018, Brack and King, 2021, Hepburn et al., 2019). While Bui et al. (Bui et al., 2018) estimate a CDR potential of up to 20.0 $GtCO_2$/a, Jones and Albanito (Jones and Albanito, 2020) imply that there might not be any biomass available for BECCS applications, since sustainable biomass globally is limited to about 100 EJ (ca. 27.8 PWh). This sustainable potential includes energetic use for biofuels, etc. (Creutzig et al., 2015), and potentially further negative land-use implications (Harper et al., 2018) inducing significant land-use change emissions (Merfort et al., 2023). The average estimate identified for the BECCS potential is around 2.8 - 9.2 $GtCO_2$/a in 2050 and 2100, respectively.

The assumed maximum potential of EW ranges from 2.0 - 4.0 $GtCO_2$/a in 2050, again acknowledged throughout the literature (Minx et al., 2018, Fuss et al., 2018, Brack and King, 2021, Hepburn et al., 2019). The 2100 estimates for global EW are 0.5 - 27.0 $GtCO_2$/a with the lowest estimate given in Longman et al. (Longman et al., 2020) and the highest estimate provided by Brack and King (Brack and King, 2021) and with an average assumption of 6.0 $GtCO_2$/a. The AF 2050 potential ranges from 0.5 - 3.6 $GtCO_2$/a with an average assumption of the maximum AF deployment potential in 2050 of 2.1 $GtCO_2$/a. Fuss et al. (Fuss et al., 2018) note that by 2100 the AF carbon sink might be saturated; however, literature findings are 0.0 - 20.0 $GtCO_2$/a with an average of 4.5 $GtCO_2$/a. The global BC potential is assumed to be 0.3 – 2.0 $GtCO_2$/a with an average of 1.2 $GtCO_2$/a in 2050, and 0.0 - 35.0 $GtCO_2$/a with an average of 6.1 $GtCO_2$/a in 2100. Global 2050 SCS potential estimates range from 2.0 - 5.3 $GtCO_2$/a and the 2100 potential is assumed to be 0.5 - 11.0 $GtCO_2$/a. It should be particularly noted that NETs reliant on biomass compete for land-use resources, and, therefore, the maximum potentials identified in the literature cannot be additive.

Findings from literature lead to the conclusion that a portfolio approach is necessary to enable ambitious CDR deployment in the future. Only BECCS, ocean liming, and wetland restoration exceed the threshold of an average maximum potential in 2050 of more than 5.0

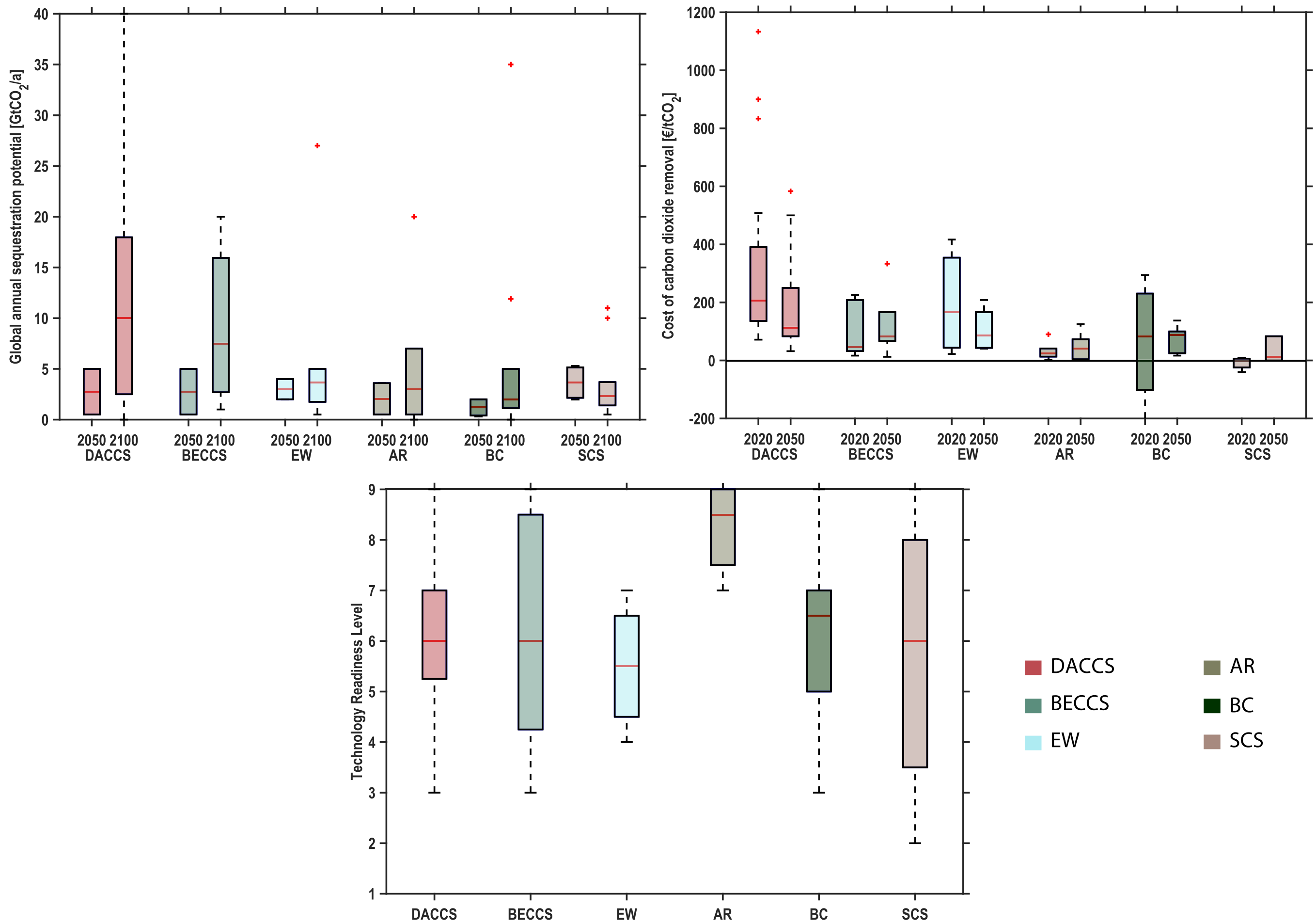

Fig. 1. Literature findings on NETs for global annual sequestration potential (top left), cost (top right) and technology readiness level (bottom). Red lines indicate the average between the first and third quartiles (black box). The black dashed lines show the normal range and red crosses are outliers.

$GtCO_2$/a as reported (cf. Fig. 2), and these options are partially evaluated to be uncertain in their potential (Smith et al., 2016). Consequently, to achieve ambitious climate change mitigation targets, a simultaneous deployment of NETs in a portfolio will be required to install the around 11 - 39 $GtCO_2$/a CDR estimated in Section 1 and further elaborated in Section 3.6.

The estimates of the current cost of CDR via DACCS range widely from 72 - 1133 €/tCO_2 with an average of 275 €/tCO_2. In 2050, the cost of DACCS is expected to be 32 - 583 €/tCO_2. Only Fasihi et al. (Fasihi et al., 2019) provide a specific cost development for DACCS for different cases. The findings suggest that there is high uncertainty when it comes to assessing the cost of DACCS (Madhu et al., 2021). One major reason is several fundamentally different approaches and process configurations are all summarised under the term DACCS (Erans et al., 2022).

Current BECCS cost are estimated significantly lower at 17 - 208 €/tCO_2 with an average cost assumption of 118 €/tCO_2 given in literature. In 2050 BECCS is estimated to cost 34 - 167 €/tCO_2. As the range is comparable to DACCS, the indication is that the latter could become cost competitive in the future (Madhu et al., 2021). EW shows a high range of cost with an upper limit of 1083 €/tCO_2 and a lower limit of 17 €/tCO_2 stated in literature for 2020, and the range narrows to 42 - 167 €/tCO_2 in 2050. The average estimation decreases from 422 €/tCO_2 in 2020 to 95 €/tCO_2 in 2050. AF cost assumptions in the literature range from 13 - 68 €/tCO_2 in 2020 and 2 - 125 €/tCO_2 in 2050. BC cost ranges from 17 - 138 €/tCO_2 in 2050. SCS estimations found in literature are between 0 €/tCO_2 and 83 €/tCO_2 for 2050 and even negative if valuable co-benefits are considered (Smith, 2016).

All CDR related approaches identified in the literature except AR are seen on an average TRL of about 6. This number is calculated as the average estimate and rounded to an integer value. AR is seen as more mature with an estimated average TRL of 8. The range in estimations can partially be explained by different publication years of the literature. Most approaches show a relatively narrow range of TRL and therefore can be assigned a low uncertainty.

This section discussed the research gap of lacking process specifications when NETs are presented in a portfolio context. In addition, there is a lack of techno-economic assessments providing energy and mass balances along the entire value chain making NETs suitable for energy-industry-CDR system transition modelling. Therefore, this study aims to give an overview on the most discussed NETs specifying each process chain with a subsequent techno-economic assessment to provide the basis for CDR portfolio creation considering societal preferences.

3. Methods and data

To create and evaluate CDR portfolios for societal preferences, several steps are taken, as visualised in Fig. 2. In the first step, all considerations are made for single technologies, before combining these technologies to NETs if applicable. For each technology considered, key assumptions, techno-economic parameters, future potential, current TRL, security of storage, and area demand were derived from literature. Key assumptions are made regarding the overall and annual CDR demand to limit global warming to 1.5°C and 1.0°C, the availability of technologies, global weighted average cost of capital (WACC), and the future development of electricity cost (Table S6 in supplementary material 1, note 2). Also, an initial deployment scenario must be assumed to

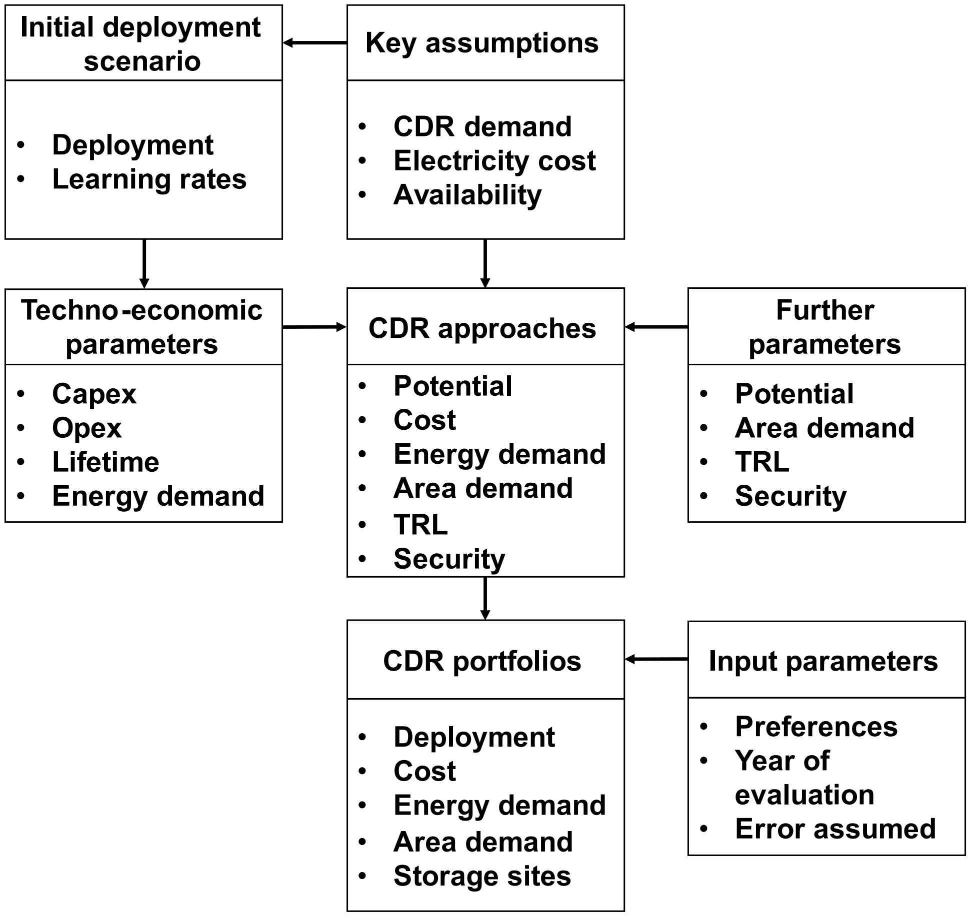

Fig. 2. Schematic overview of the methodology used to create CDR portfolios respecting societal preferences. Abbreviations: capex – capital expenditures, opex – operational expenditures, TRL – technology readiness level, CDR – carbon dioxide removal.

calculate exogenously the future cost development of technologies in case learning rates are considered. Details on the initial deployment portfolio can be found in Table S19 in the supplementary material 1, note 6.

Using the assumptions and considerations as mentioned above, the maximum potential, the levelised cost of CO_2 removal (LCOCDR), the final energy demand (FED) of CDR, the primary energy demand (PED) of CDR, the area demand, the TRL, and the security level of all technologies as well as for all NETs, representing a sequence of technologies, are determined. The results of these calculations are then used to create different CDR portfolio deployment scenarios. Furthermore, these results are used to calculate the total annualised cost, the total annual final energy demand (TFED), the TPED, and the gross and net land occupation of CDR for limiting global warming to 1.5°C by removing 500 $GtCO_2$ from the atmosphere until 2100. A second scenario resembling a total CDR of 1750 $GtCO_2$ until 2100, which may limit average global warming to about 1.0°C, is derived (cf. Section 3.6). In the following sections all steps for CDR portfolio creation and evaluation are examined in more detail.

3.1. Clustering of options for carbon dioxide removal

As shown in Fig. 3, there are three main ways to remove CO_2 from the atmosphere. The respective technologies considered in this work are listed in Table 1. Other natural climate solutions (NCS) methods such as soil carbon sequestration (SCS) are not addressed due to the lack of suitable techno-economic parameters.

It should be noted that IAMs are continuously updated and new NETs have been added recently (Fuhrman et al., 2023, Gidden et al., 2023). CDR pathways rely on different chemical reactions. One is the weathering process that occurs naturally when metal-bearing rocks are in contact with ambient air (Hampl et al., 2022). The principal overall reaction equation for weathering is given in Eq. (1) where *Me* represents metals like calcium (Ca) or magnesium (Mg).

$$CO_2 + MeO \rightarrow MeCO_3 + \Delta H \tag{1}$$

This reaction is exothermic but shows relatively slow kinetics (Kelemen et al., 2019). The EW approach employs this carbonation reaction and enhances the reaction kinetics by crushing and grinding suitable rocks to micro-scale to spread powdered rocks on open ground (Beerling et al., 2020, Strefler et al., 2018, Goll et al., 2021). Depending on the rock type, around 1.8 - 7.1 t of rock is required to sequester 1 tCO_2. Related literature findings can be found in Table S11 in the supplementary material 1, note 3. The bicarbonates and subsequently the carbonates produced are stable for geological timescales (Kelemen et al., 2019). Therefore, the CO_2 is stored safely for long periods. The weathering process reacting with atmospheric or ambient CO_2 is often referred to as EW. The same process used for storing concentrated CO_2 streams either underground in geologic rock formations or above ground in reactors is usually referred to as in-situ or ex-situ mineralisation, respectively (Kelemen et al., 2019, Snæbjörnsdóttir et al., 2020).

Another reaction utilised for CDR as portrayed in Fig. 3 is photosynthesis. Biomass takes up atmospheric CO_2 and produces hydrocarbons (Stirbet et al., 2020). Therefore, biomass acts as CO_2 storage. In contrast to the carbonates produced via EW or mineralisation mentioned above, the lifetime of the biogenic CO_2 storage is comparably low. The lifetime of terrestrial biomass often does not exceed hundreds of years (Chiquier et al., 2022). Also, there can be safety concerns raised

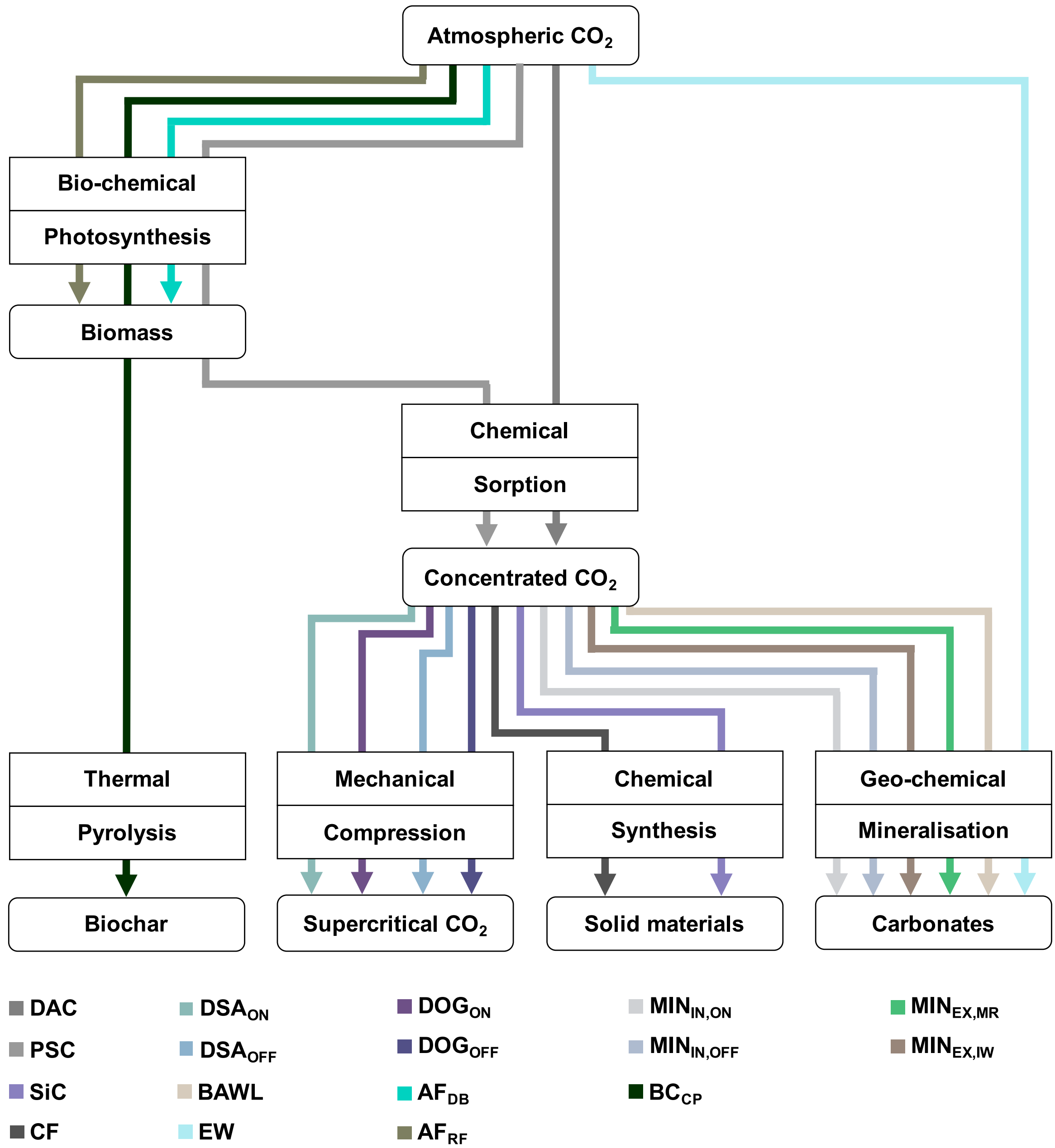

Fig. 3. Overview on CDR technologies examined in this study. Abbreviations: PSC – CO_2 point source capture, DSA – CO_2 sequestration in deep saline aquifers, DOG – CO_2 sequestration in depleted oil and gas fields, MIN_{IN} – in-situ mineralisation, ON – onshore, OFF – offshore, MIN_{EX} – ex-situ mineralisation, MR – mined rock, IW – industrial waste, SiC – electricity-based silicon carbide production, CF – electricity-based carbon fibre production, EW – enhanced weathering, AF_{DB} – desalination-based afforestation, AF_{RF} – rainfall-based afforestation, BC – biochar production, CP – conventional pyrolysis.

regarding the storage of CO_2 since forests or other accumulations of biomass can be exposed to hazardous wildfires (Chiquier et al., 2022). However, healthy rainforests, not impacted from deforestation or degradation, are more resilient to these major CO_2 discharge events (Caldera and Breyer, 2023, [WWF] 2020, Saatchi et al., 2021). In the context of CDR, biomass is often seen as intermediate CO_2 storage. One option is using biomass for energy supply or industrial applications and capturing the CO_2 stored in biomass from the exhaust gases as displayed in Fig. 3 (Fuss et al., 2018, Kemper, 2015). This approach is usually referred to as BECCS. BECCS can be used to simultaneously supply energy in the form of heat or electricity or to produce secondary energy carriers such as biogas (Tanzer et al., 2021) or biofuels (Burns and Nicholson, 2017). The concentrated CO_2 captured from the exhaust gas can subsequently be utilised for CCU (Mertens et al., 2023) or can be stored long term for CCS purposes (Vitillo et al., 2022). Therefore, forest cultivation can be seen as NET with immediate CO_2 storage for several decades or with subsequent long-term storage if biomass is processed further and the CO_2 is stored in geological underground formations or mineralised for millennia. Also, gaseous concentrated CO_2 can be used to produce solid, carbon-bearing materials such as e-CF or e-SiC (Mühlbauer et al., 2024, Keiner et al., 2023). Another approach is BC production to process biomass further enabling negative emissions. Feedstock biomass is usually pyrolysed to remove most of the constituents of the biomass besides carbon to produce biochar (Fawzy et al., 2021, Haeldermans et al., 2020). There are other procedures available to produce biochar such as torrefaction or hydrothermal carbonisation, among others (Sri Shalini et al., 2021). As a by-product of biochar production, gases or liquids are produced that can be either utilised or combusted for additional energy supply (Haeldermans et al., 2020). Combusting the by-product gas or liquid lowers the effective CDR

Table 1
Options for NETs clustered as investigated in this study. The IAM use is for the case of REMIND, IMAGE, MESSAGE, and GCAM. The numbers in parenthesis indicate how regularly NETs are included according to the IPCC's Sixth Assessment Report (AR6) WGIII contribution ([IPCC] 2022).

Capture	Storage	CO_2 storage technology	Physical condition	IAM implementation*
DAC/ PSC	Geological onshore	Deep saline aquifer onshore	gaseous, supercritical	DACCS, BECCS (3.5/4)
		Depleted oil and gas field onshore	gaseous, supercritical	
	Geological offshore	Deep saline aquifer offshore	gaseous, supercritical	DACCS, BECCS (3.5/4)
		Depleted oil and gas field offshore	gaseous, supercritical	
	in-situ onshore	in-situ mineralisation onshore	solid, minerals	not yet
	in-situ offshore	in-situ mineralisation offshore	solid, minerals	not yet
	ex-situ using mined rocks	ex-situ mineralisation mined rocks	solid, direct aqueous carbonation	not yet
	ex-situ using industrial solid waste	ex-situ mineralisation industrial waste	solid, direct aqueous carbonation	not yet
	BAWL	Buffered accelerated weathering of limestone	direct aqueous carbonation, only at coast	not yet
	SiC	e-SiC production	solid, useful material	not yet
	CF	e-CF production	solid, useful material	not yet
Bio-/geo-chemical	EW	Enhanced weathering of mined rocks	solid, minerals	EW (1/4)
	BC	Conventional biochar pyrolysis	biochar	not yet
	AF_{RF}	Rainfall-based afforestation	biomass	AF (4/4)
	AF_{DB}	Desalination-based afforestation	biomass, irrigated	not yet

* It should be noted that IAMs are being updated continuously as shown in recent publications (Fuhrman et al., 2023, Gidden et al., 2023).

potential per tonne of biomass, however, supplies energy.

Another way to capture atmospheric CO_2 is through sorption processes such as in DAC (Fasihi et al., 2019). There are several different methods of DAC such as low-temperature solid sorbent DAC or high-temperature liquid sorbent DAC (Fasihi et al., 2019). Solid sorbent DAC, e.g., uses a solid basic sorbent to capture atmospheric CO_2 with a subsequent temperature, pressure, or moisture swing to retrieve the reacted CO_2 from the air contactor (Fasihi et al., 2019, Madhu et al., 2021, Deutz and Bardow, 2021). There are also processes utilising the weathering reaction mentioned above in a circular way (Fasihi et al., 2019). The atmospheric CO_2 reacts with a metal-bearing rock slurry in the first process cycle (Fasihi et al., 2019). In the subsequent cycle, the slurry gets calcinated to release the bound CO_2 to produce a concentrated CO_2 stream (Fasihi et al., 2019). All DAC processes, similar to BECCS, produce concentrated CO_2 streams that can be either used for CCU (Mertens et al., 2023, Galimova et al., 2022) or CCS purposes (Vitillo et al., 2022). Therefore, effective sequestration options must be identified to enable an effective CDR potential.

3.2. Technology background and modelling

In this study, energy demand, as well as mass in- and output are normalised to the functional unit 1 tCO_2 removed. This normalisation implies an actual tonne of CO_2 removed from the atmosphere including carbon efficiency (losses or leakage of CO_2) along the entire process chain. The modelling done and data used in this study are based on an in-depth literature review of available CDR options. The full literature review can be found in the supplementary material, note 3.1. Dedicated techno-economic information derived from this review for all technologies assessed in this subsection can be found in Tables S7-S10 and Tables S12-S17 in the supplementary material 1, note 3.2. Both review and techno-economic information are available for CO_2 capture technologies (DAC, PSC), BECCS, transport of CO_2, geologic sequestration, in-situ and ex-situ mineralisation, production of carbon-bearing materials, enhanced weathering, rainfall-based and desalination-based afforestation, and biochar production. Future techno-economic estimates are uncertain and should be treated as such. The provided tool (supplementary material 2) enables readers to explore sensitivities of techno-economic input parameters on the results. In addition to the technologies presented in this section, other options would be available, but were excluded from this investigation due to low suitability for large-scale CDR or lack of data.

3.3. Sequestration potential

To estimate the upper boundaries of the NETs considered, the maximum technological deployment potential of all technologies within the process chains is derived from literature. The time it takes to reach the maximum potential level is either derived from literature or based on own assumptions. Logistic curves are used to project the maximum deployment potential of single technologies until 2100. For all technologies except DAC a logistic growth rate of 20% is applied while for DAC, due to its high scalability and to make portfolio creation more robust, a logistic growth rate of 30% is assumed. While the rapid exploration of suitable sequestration options for concentrated CO_2 could become a future bottleneck for DACCS or BECCS (Lane et al., 2021), this does not affect the assumption for DAC roll-out and its logistic growth rate. On the contrary, as DAC is mostly dependent on staple materials and an adequate sorbent industry to emerge (Deutz and Bardow, 2021), material demand for DAC manufacturing and electricity generation, dominated by solar PV (Breyer et al., 2022), is assumed to be no constraint for a rapid phase-in (Haegel et al., 1979, Goldschmidt et al., 2021, Zhang et al., 2021). All assumptions are summarised in Table S18 in the supplementary material 1, note 5. As described by Caldera and Breyer (Caldera and Breyer, 2023), the potential of AF_{DB} is linked to the average growth characteristic of a mixture of trees. In this study the potential estimations are based on initial plantation of trees in 2030 and the potential is not modelled as logistic curve as for all other technologies. Therefore, the final deployment is corrected with respect to the trees' actual growth characteristic. AF_{RF}'s potential is approximated through a sigmoid curve; however, the CDR deployment is corrected in alignment with the growth rate of trees proposed (Caldera and Breyer, 2023).

The calculation of the upper estimated potential for a NET as the lowest potential out of all maximum potentials of each process step in the NET process chain is shown in Eq. (2):

$$P_{CDR}^{max} = \min_{p}\left(P_{CDR,p}^{max}\right) \tag{2}$$

wherein P_{CDR} is the total CDR potential and p indicates each process step of a CDR process chain.

3.4. Techno-economic assessment

First, techno-economic parameters for each technology are derived from literature. Uncertainties in future estimates must be considered and are further discussed in Section 5.3. The provided tool can be used to explore the sensitivity of input parameters on the final results. Where applicable, the future cost development is projected using learning rates (Vartiainen et al., 2020). For DAC, a learning rate of 10% was derived from (Fasihi et al., 2019). Faber et al. (Faber et al., 2022) assign a learning rate of 10.6% to ex-situ mineralisation using mined rocks and for PSC and BC a learning rate of 10% and 5%, respectively, is assumed. Since PSC and conventional pyrolysis both are relatively well-established technologies, conservative learning rates of 10% and 5%, respectively, are assigned. To avoid an infeasibly steep cost decline during the initial scale-up of technologies, learning effects are considered only if the respective technologies surpass 1 $MtCO_2/a$ of total installed capacity. Fig. 4 shows the normalised cost reductions in capex for the technologies for that learning curves were assumed. Further details on the cost reduction through learning effects can be found in the supplementary material 1, note 6.

The final energy demand for technologies is derived from literature. Findings are also summarised in the supplementary material 1, note 3. In this study, it is assumed, that all final energy demand is covered by renewable electricity, by providing low-temperature heat through heat pumps and high-temperature heat by direct electric heating. For heat pumps an average coefficient of power (COP) of 2 (Eureka Luft 2022) is expected for a temperature difference of up to 80 K and it is assumed to grow linearly to 2.5 by 2070, then stabilise on that level. The NETs' specific and fully electrified final energy demand is calculated according to Eq. (3) as the sum of all sub-processes of the CDR process chain.

$$E_{FE,NET} = \frac{\sum_p E_{FE,p} \cdot m_{out,p}}{1 \text{ tCO}_2 \text{ removed}} \tag{3}$$

wherein $E_{FE,p}$ is the specific energy demand in MWh/tCO_2 of a process step and $m_{out,p}$ is the mass of material that must be reacted or treated to store 1 tCO_2, i.e. $m_{in,NET}$. The specific PED $E_{PE,CDR}$ is calculated by Eq. (4):

$$E_{PE,NET} = \frac{E_{FE,NET}}{\eta_{grid} \cdot \left[\frac{(1+\eta_{battery})}{2}\right]} \tag{4}$$

wherein η_{grid} is the efficiency of the transmission and distribution grid for electricity, and $\eta_{battery}$ is the average battery storage efficiency. Grid losses are adapted from Keiner et al. (Keiner et al., 2023) and are expected to decrease from 8.5% today to about 4.0% in 2100 leading to 91.5% and 96.0% transmission and distribution grid efficiency, respectively, while the battery storage efficiency is taken from Bogdanov et al. (Bogdanov et al., 2021) and increases from 91.0% in 2020 to 95.0% in 2040. Beyond 2040, it is assumed that the battery efficiency stays at 95.0%. It is assumed, that on average, 50% of generated electricity is stored in batteries. Further details are listed in the supplementary material 1, note 2.

The levelised cost of carbon dioxide removal $LCOCDR_{CDR}$ for a CDR route is calculated for all process technologies considered with available cost data, according to Eq. (5).

$$LCOCDR_{NET} = \sum_p \left[LCOP_{p,co} + H_{LT,p} \cdot LCOH_{LT} + \left(E_{FE,p} + H_{HT,p}\right) \cdot LCOE\right] \cdot \frac{m_{out,p}}{1 \text{ tCO}_2 \text{ removed}} \tag{5}$$

wherein the capex and opex, *co*, related levelised cost of process *LCOP* of each process *p* (excluding energy needs) is added to the cost for low-temperature heat and to the cost for electricity. The cost of low-temperature heat is the specific low-temperature heat demand $H_{LT,p}$ multiplied with the levelised cost of low-temperature heat $LCOH_{LT}$ which is calculated according to Eq. (8). The sum of specific electricity $E_{FE,p}$ and high-temperature heat $H_{HT,p}$ demand is multiplied by the levelised cost of electricity *LCOE* that already includes the cost for the grid and storage, and the efficiencies for the transmission and distribution grid η_{grid} and electricity storage $\eta_{battery}$. The cost of electricity is part of the key assumptions for this study presented in more detail in the supplementary material 1, note 2. The overall sum is then multiplied by the mass of output material $m_{out,p}$ of each process technology that is required to store 1 tCO_2. The products of each process steps are finally summarised to calculate the overall capex and opex related $LCOP_{p,co}$ for each technology. The $LCOP_{p,co}$ is calculated using the *WACC*, lifetime N_p, capacity cap_p and availability τ_p of each process *p*, according to Eq. (6a-c). For the e-materials, e-SiC and e-CF, the product value is considered with negative opex in units of €/tCO_2, while LCOCDR can be reduced to zero as an extreme. Some NETs have a positive impact on the local environment, such as soil quality or biodiversity conservation. As it is challenging to quantify, these possible impacts are not considered in this research.

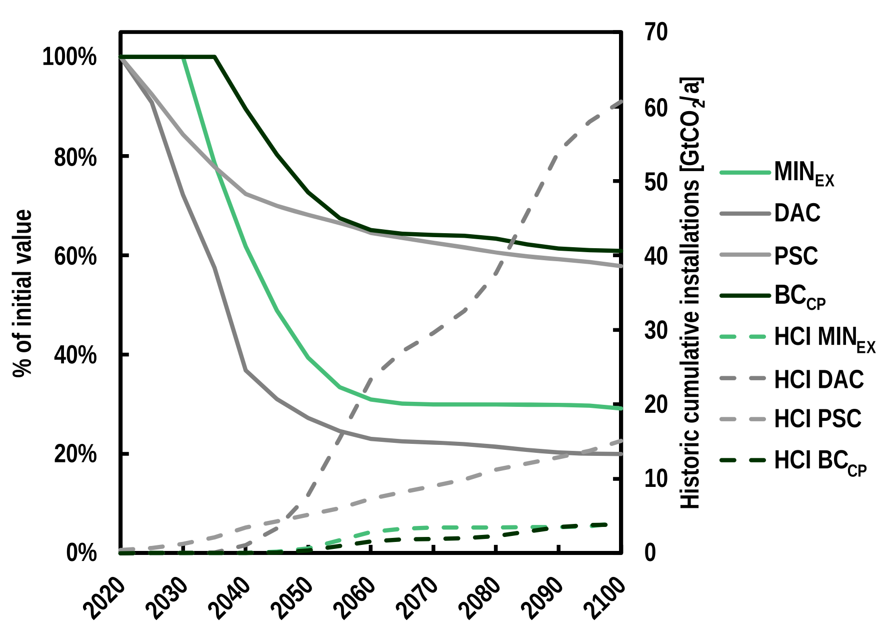

Fig. 4. Projected normalised capex development assumptions based on learning curves for DAC, PSC, MIN_{EX}, and BC_{CP} with the respective initial deployment assumption. Dashed lines indicate the historic cumulative installations (HCI) for the respective technologies.

$$LCOP_{p,co} = \frac{\left(CAPEX_p \cdot crf_p + OPEXfix_p\right) \cdot cap_p}{out_p} + OPEXvar_p \tag{6a}$$

$$crf_p = \frac{WACC \cdot (1 + WACC)^{N_p}}{(1 + WACC)^{N_p} - 1} \tag{6b}$$

$$out_p = cap_p \cdot \tau_p \tag{6c}$$

For calculating the $LCOP$ for process technologies with a capex based on power-based output Eq. (6) is used with capacity set to 8760 h/a.

The cost of low-temperature heat $LCOH_{LT}$ provided via heat pump HP is calculated using Eq. (7).

$$LCOH_{LT} = \frac{CAPEX_{HP} \cdot crf_{HP} + OPEXfix_{HP}}{8760 \cdot \tau} + OPEXvar_{HP} + \frac{LCOE}{COP} \tag{7}$$

wherein COP is the coefficient of performance of the heat pump.

Some of the techno-economic assumptions can be of decisive importance. For example, it has been shown that, for solar PV, the WACC can cause a major sensitivity for major technologies (Vartiainen et al., 2020). A sensitivity assessment for lower WACC assumptions is carried out and presented in Section 5.4.

3.5. Area demand, technology readiness, and security

As the global population increases, providing sufficient nutrition for all becomes increasingly challenging (Campbell et al., 2017, Steffen et al., 1979). With the effects of climate change endangering agricultural land further, efficient area utilisation will be crucial. Therefore, area occupation of NETs will be a limiting factor to large-scale deployment (Brack and King, 2021). The land occupation for all NETs considered is calculated according to Eq. (8):

$$a_{NET} = E_{PE,NET} \cdot a_{PE} + \sum_p a_p \tag{8}$$

wherein the specific area demand for CO_2 removal a_{CDR} with the unit $km^2/(GtCO_2 \cdot a)$ is the sum of the specific area demand for the process a_p and the specific area demand for primary energy a_{PE} multiplied by the PED of each process $E_{PE,p}$. The area occupation for all processes considered in this work and for primary energy production is listed in the supplementary material 2. PE supply is estimated with wind power and solar PV as the main energy resources of cost-optimised 100% renewable energy systems in the future. Exact shares are derived from Keiner et al. (Keiner et al., 2023). The power density of PV is assumed to increase linearly from 75 MW/km^2 in 2020 to 150 MW/km^2 in 2050 while the power density of wind power is assumed to be constant at 8.4 MW/km^2 (Keiner et al., 2023). In the case of wind power, it is furthermore distinguished between net area demand, comprising of the actual area covered by the turbines, maintenance area and access roads, and gross area demand, considering spacing between the turbines, which can still be used for farming or forestry, etc. The net area demand is set to 1% of the gross area demand for wind power (Denholm et al., 2009), for solar PV no difference between gross and net area demand is assumed.

The TRL of each NET is defined as the minimum TRL of the processes that constitute the respective CDR technology. Therefore, calculating the TRL of NETs is done according to Eq. (9),

$$TRL_{NET} = \min_p \left(TRL_p\right) \tag{9}$$

wherein TRL_{NET} is the TRL of the entire CDR technology and TRL_p is the TRL of each sub-process.

For considerations regarding the security of CDR, only the technical security of the storage is surveyed. While the potential for health issues and impacts on the environment by CDR technologies must be scrutinised, these are outside the scope of this work. The security score presented in this work describes the certainty of secure CO_2 storage and, therefore, the durability of the CDR option only. For single processes the risk of leakage in short term (<100 years) and in the long term (>100 years) is either derived from the literature or is set to 0% if CO_2 is subsequently stored in solid materials, i.e., carbonates, e-SiC, e-CF, or in forests that are actively irrigated (AF_{DB}). Since CDR for centuries is considerably more effective to reduce the atmospheric CO_2 concentration, more weight is given to short-term risk of technologies. Eq. (10a, b) shows the way the risk score for single technologies is calculated.

$$S_{NET} = \prod_p S_p \tag{10a}$$

$$S_p = 1 - (w_{ST} \cdot risk_{ST} + w_{LT} \cdot risk_{LT}) \tag{10b}$$

For NETs, the security scores of constituent technologies are multiplied as depicted in Eq. (10a) in order to obtain the security score for the whole CDR technology S_{NET}. In Eq. (10b), the security score of a single process S_p is calculated by subtracting the sum of the weighted short-term and long-term risk from unity, with w_{ST} the weight for short-term risk $risk_{ST}$ and w_{LT} the weight for long-term risk $risk_{LT}$. By default, the weight for short-term risk is set to 0.8 while the weight for long-term risk is set to 0.2 for this study. Only the risk for CO_2 leakage is examined using the risk factors listed in Table 2. No further risk regarding health or other environmental aspects are considered in this study. However, these crucial points should be addressed thoroughly in future studies.

For the security factor of CDR process chains, a weighted sum of short and long-term risk factors is calculated using the approach described above with the assumptions listed in Table 2.

3.6. Estimation of total negative emissions by 2100

A central element for the creation of CDR portfolios is the total amount of CO_2 that has to be removed from the atmosphere, or total negative CO_2 emissions (TNE), in order to ensure respective temperature targets by 2100. This study considers two central scenarios: Reaching the agreed 1.5°C target according to the Paris Agreement ([UNFCCC] 2015), and a more ambitious 1.0°C target as suggested by Hansen et al. (Hansen et al., 2017, Azar and Rodhe, 1979), limiting average global warming to these values by the end of the 21st century.

The basis for the TNE is the cumulative CO_2 emissions of the present legacy system by the point in time when global net zero CO_2 emissions are achieved in relation to the CO_2 balancing required for the respective scenario, as assumed to be in 2050 in this study. The remaining CO_2 emissions related to the energy-industry system are modelled with LUT-

Table 2
Short-term and long-term risk factors assumed for different technologies.

Technology	Short-term risk (<100 years)	Long-term risk (>100 years)	Reference
DAC	0%	0%	(Chiquier et al., 2022)
PSC	15.5%	15.5%	(Chiquier et al., 2022)
Geological sequestration	9.5%	9.5%	(Chiquier et al., 2022)
MIN_{IN}	0%	0%	own assumption
MIN_{EX}	0%	0%	own assumption
BAWL	0%	0%	own assumption
e-SiC	0%	0%	own assumption
e-CF	0%	0%	own assumption
EW	0%	0%	(Chiquier et al., 2022)
BC_{CP}	70%	95%	(Chiquier et al., 2022)
AF_{RF}	5%	50%	(Chiquier et al., 2022)
AF_{DB}	0%	0%	own assumption

DEMAND (Keiner et al., 2023) and can be estimated to be 680 $GtCO_2$ between 2020 and 2050. The basis for this assumption is a full defossilisation by 2050 as the main priority to be compliant with the 1.5°C target. Industrial emissions of the second half of the 21st century can be neglected, as it might be common practice to avoid these CO_2 emissions to reach the atmosphere with on-site point-source capture and subsequent safe sequestration (Leeson et al., 2017). For this, about 100 $GtCO_2$ can be accounted for (Keiner et al., 2023). To normalise the remaining CO_2 emissions to the time frame of 2015 - 2050, three years with each about 40 $GtCO_2$/a (Friedlingstein et al., 2022) are added to the remaining emissions, which leads to rounded rest emissions estimate of about 780 $GtCO_2$ from 2015 to 2050.

The remaining CO_2 emission budget for reaching the 1.5°C target according to the contribution of Working Group I (WGI) to the IPCC AR6 is 300 $GtCO_2$ between 2020 and 2100, given an 83% probability, i.e., the risk for not reaching this target shall be as low as possible ([IPCC] 2021). *Re*-normalised to the time frame 2015 - 2100 by adding five years with each 40 $GtCO_2$ (Friedlingstein et al., 2022) gives a remaining CO_2 emission budget of about 500 $GtCO_2$. For this remaining budget, insecurities of about 220 $GtCO_2$ can be considered ([IPCC] 2021), which leaves a remaining budget of about 280 $GtCO_2$ from 2015 - 2100 for reaching the 1.5°C target with high probability. This to be subtracted from the remaining emissions of 780 $GtCO_2$ of the legacy system from 2015 onwards, until reaching global net zero CO_2 emissions, thus leading to a CDR demand of about 500 $GtCO_2$ by 2100 that is required to ensure that global warming is limited to 1.5°C.

The TNE for a 1.0°C target is calculated based on a linear regression of C1 scenarios in the IPCC AR6 WGIII (cf. Figure 13, Section 5) ([IPCC] 2022, Byers et al., 2022), which indicate a slope of 250 $GtCO_2$/0.1 K of global warming (cf. subSection 5.5). Therefore, 0.5 K or 0.5°C less average global warming result in an additional CDR requirement of 1250 $GtCO_2$. In addition to the 500 $GtCO_2$ already calculated for the 1.5°C target, this estimation gives a CDR demand of 1750 $GtCO_2$ for limiting global warming to 1.0°C by end of the century. Basis is similar to the 1.5°C target calculation for CDR demand the phase-out of fossil fuel use by 2050 and, therefore, the 1.0°C is realised solely via CDR based on a 1.5°C energy transition.

3.7. Portfolio creation

After calculating the global CDR potential, energy demand, area demand, and LCOCDR of all NETs considered, these intermediate results are used to create CDR portfolios for different hypothetical societal preferences resembling potential dimensions of a stakeholder discourse. Further dimensions such as the environmental impact of CDR should be noted, though they are not quantified within this study. The methodology is explained in further detail in the supplementary material 1, note 7. It must be noted that while immediate security and long-term durability of CO_2 sequestration should be separated, both criteria are interwoven. A long-term durability assessment of CDR options beyond the time scale of this study in the year 2100 is considered out of scope at this point, and security considers mainly the immediate risk for leakage.

Preferences indicate the factors used to calculate a score for each NET with the weighted sum approach comparable to Rueda et al. (Rueda et al., 2021). In this work six portfolios are created:

- Balanced portfolio (BP): Equal distribution of weights over all societal factors without a preference.
- Low-cost portfolio (LCP): Preference of low-cost solutions and minor consideration of other factors.
- Low energy portfolio (LEP): Preference on solutions with low energy demand and minor consideration of other factors.
- High security portfolio (HSP): Preference on solutions with low risk of CO_2 leakage (high durability) and minor consideration of other factors.
- Low area portfolio (LAP): Preference on solutions with low area demand and minor considerations of other factors.
- High TRL portfolio (HTP): Preference on solutions with high TRL and minor considerations of other factors.

The weight distribution for the portfolios is shown in Fig. 5.

All NETs considered within this research are sorted according to their calculated score, and are used to calculate a deployment scenario. To resemble a realistic rollout, an error in the grading of the approaches can be chosen to group NETs with similar scores. These technologies are then deployed in parallel until their overall group potential is not anymore sufficient to cover the CDR demand. For this research an error in grading of 1.5% is assumed, which can be adjusted in the LUT-CDR spreadsheet tool. Technologies with similar score, i.e., within the error range are grouped and deployed in parallel as long as possible to resemble a realistic phase-in. The assumed error is chosen to group technologies to a reasonable extent; it can be adjusted in the spreadsheet LUT-CDR tool (supplementary material 2 and online https://doi.org/10.5281/zenodo.7657804). All technologies are evaluated for 2050 and results are put into greater context by comparing results with the delayed economic equality scenario (LUT-DEES, Figure S6-S8 in the supplementary material 1, note 9) as studied by Keiner et al. (Keiner et al., 2023). The results are furthermore corrected with respect to the growth rate of afforestation-based NETs. A detailed method description can be found in the supplementary material 1, note 7.

4. Results

Due to the broad variety of results obtained in this research, this section presents exemplary results only. In Section 4.1, the total global CDR potential of each NET is presented, followed by results for the energy demand in Section 4.2. Section 4.3 deals with the LCOCDR, Section 4.4 presents area demand and findings for the storage security. Finally, Section 4.5 presents findings for six exemplary CDR portfolios. Detailed results can be found and reproduced in the supplementary material 2. Compound NETs will be abbreviated in the following as capture technology with linked storage technology (CC•S), if applicable.

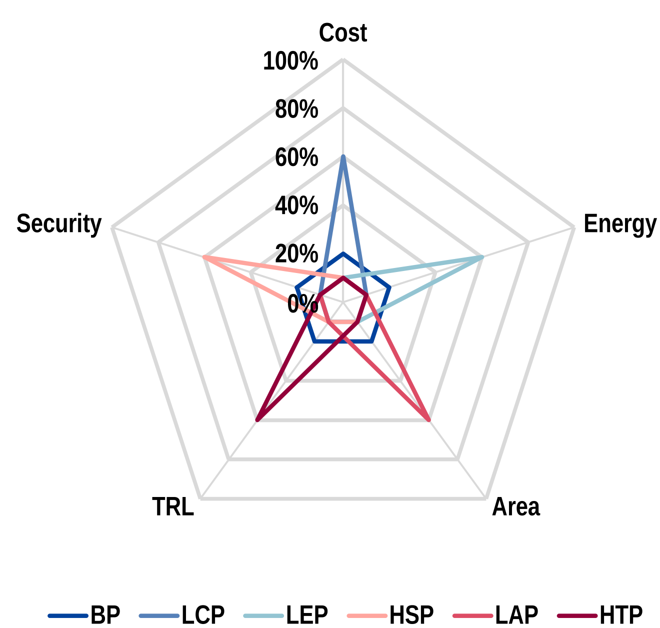

Fig. 5. Weight distribution on societal preferences for case scenarios considered in this study for portfolio creation.

4.1. Global sequestration potential

The global sequestration potential of different NETs considered in $GtCO_2$/a is compared in Fig. 6. It should be noted that all displayed maximum potential is for individual CDR technologies, i.e., actual potential in CDR portfolios might be decreased if a technology's potential of the CDR process chain is depleted already.

As can be seen, most NETs are assumed to have their respective maximum deployment potential in the range of 1 - 5 $GtCO_2$/a. Since DAC is expected to be highly scalable and the production of e-SiC does not seem to be constrained by any exogenous limit, the maximum potential of the CO_2 capture and permanent CO_2 sequestration process chain DAC with e-SiC production (DAC•SiC) is expected to be considerably high. For AF_{RF} and PSC the inflection point of the logistic curve is expected to be in 2030, for DAC it is assumed to be in 2045, and for EW the inflection point is reached in 2055. All other technologies considered reach their inflection point in 2035, and almost their maximum deployment potential by 2050.

4.2. Energy demand

The energy demand of DAC with CO_2 sequestration in onshore deep saline aquifers (DAC•DSA_{ON}), a representative of DACCS as discussed in literature (c.f. Section 2), DAC•SiC and AF_{DB} are shown as an example in Fig. 7. No further reduction of energy demand is assumed after 2050. As for DAC•DSA_{ON}, the energy demand of all options using low-temperature heat, is reduced when the whole process chain is electrified, and low-temperature heat is covered by heat pumps. A factor describing the discrepancy between FED and PED, reflecting grid and storage losses as derived from Keiner et al. (Keiner et al., 2023), is considered as described in Section 3.4.

The FED of DAC•DSA_{ON} decreases from currently 2.17 MWh/tCO_2 to 1.89 MWh/tCO_2, 1.65 MWh/tCO_2, and 1.44 MWh/tCO_2 in 2030, 2040, and from 2050 onwards, respectively. The energy demand reduction is due to efficiency improvements of DAC units. The share of electricity in the total FED slightly increases from 17.1% in 2020 to 21.0% in 2050 and beyond, due to the assumed heat pumps COP improvement and the decreasing heat demand of DAC units. The FED for a fully electrified process also decreases relative to the FED from current 1.27 MWh/tCO_2 to 1.08 MWh/tCO_2, 0.93 MWh/tCO_2, 0.80 MWh/tCO_2, and 0.76 MWh/tCO_2 in 2030, 2040, 2050 and from 2070 onwards, respectively, through the increasing COP that is assumed for heat pumps. The discrepancy between the electrified FED and PED decreases due to lower transmission grid and storage losses (Keiner et al., 2023) with the PED decreasing from current 1.46 MWh/tCO_2 to 1.20 MWh/tCO_2, 1.00 MWh/tCO_2, 0.85 MWh/tCO_2, and 0.76 MWh/tCO_2 in 2030, 2040, 2050 and 2070, respectively.

The FED of DAC•SiC is relatively high compared to DAC•DSA_{ON} due to the high energy demand for e-SiC production and decreases from 11.73 MWh/tCO_2 in 2020 to 10.99 MWh/tCO_2 in 2050. For similar reasons as for DAC•DSA_{ON} the electricity share in FED increases from 79.3% in 2020 to 84.0% in 2050. Due to the lower share in low-temperature heat in the FED, the relative reduction through electrification is lower than for DAC•DSA_{ON} with the FED for the fully electrified process decreasing from currently 10.83 MWh/tCO_2 to 10.31 MWh/tCO_2 from 2070 onwards. AF_{DB} is a fully electrified process chain requiring 2.17 MWh/tCO_2, 2.01 MWh/tCO_2, 3.15 MWh/tCO_2, and 2.07 MWh/tCO_2 in 2020, 2030, 2040, and 2050, respectively. Beyond 2050, the electricity demand, which follows the specific freshwater demand, decreases steadily to 1.0 MWh/tCO_2 in 2100. This is due to the increasing carbon sequestration rate of the trees and the stabilising water demand as the trees mature. No further efficiency improvements are expected. For none of the technologies, except heat pumps, efficiency improvements after 2050 are assumed.

4.3. Levelised cost of carbon dioxide removal

Fig. 8 shows the cost development of most NETs examined in this study in this century. AF_{DB} shows the highest initial cost of all bio-geo-chemical approaches displayed. However, cost decreases steeply when CO_2 uptake of the trees accelerates. PSC generally is lower in cost; however, DAC shows a major cost decrease due to learning effects towards 2050. AF_{RF} can be found to be the least cost solution a few decades after initial tree planting.

It should be noted that e-SiC and e-CF options are the most expensive with DAC•SiC having initial production cost of about 800 €/tCO_2 and DAC•CF having initial production cost of about 4965 €/tCO_2. However, for both options income through product sales is accounted for. For e-SiC, utilisation as partial construction sand substitute can decrease the total LCOCDR by 6.5 €/tCO_2 in 2020, which increases to about 27.6 €/tCO_2 from 2050 onwards (Mühlbauer et al., 2024). e-CF production can become profitable by 2050 if income of 11 €/kgCF through product sales is considered, as this is the expected amount to trigger high attractivity for industries such as automotive (Jones and Albanito, 2020). For mineralisation options, no revenue due to product sales is assumed. PSC, i.e., capturing CO_2 from biogenic combustion is lower in

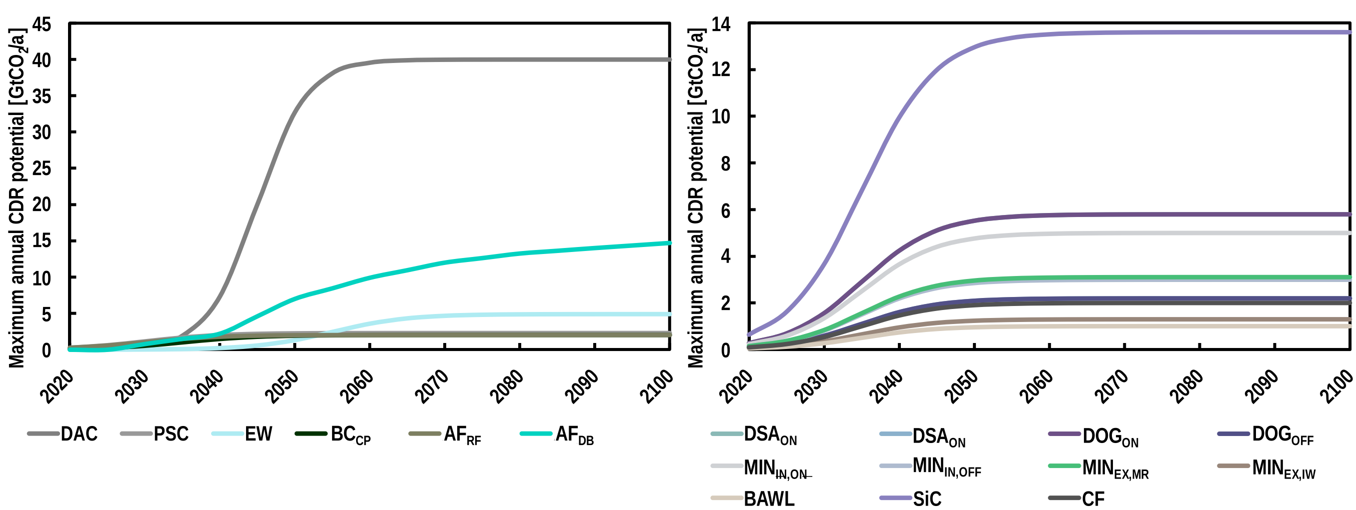

Fig. 6. Projection of the development of the maximum potential of capture technologies including bio-geo-chemical options (left) and storage technologies for concentrated CO_2 (right) for the different NETs. Please note, that DSA_{ON}, DOG_{ON} and $MIN_{IN,ON}$; as well as DSA_{OFF}, DOG_{OFF}, and $MIN_{IN,OFF}$ have equal potential development. Details are listed in the supplementary material 2.

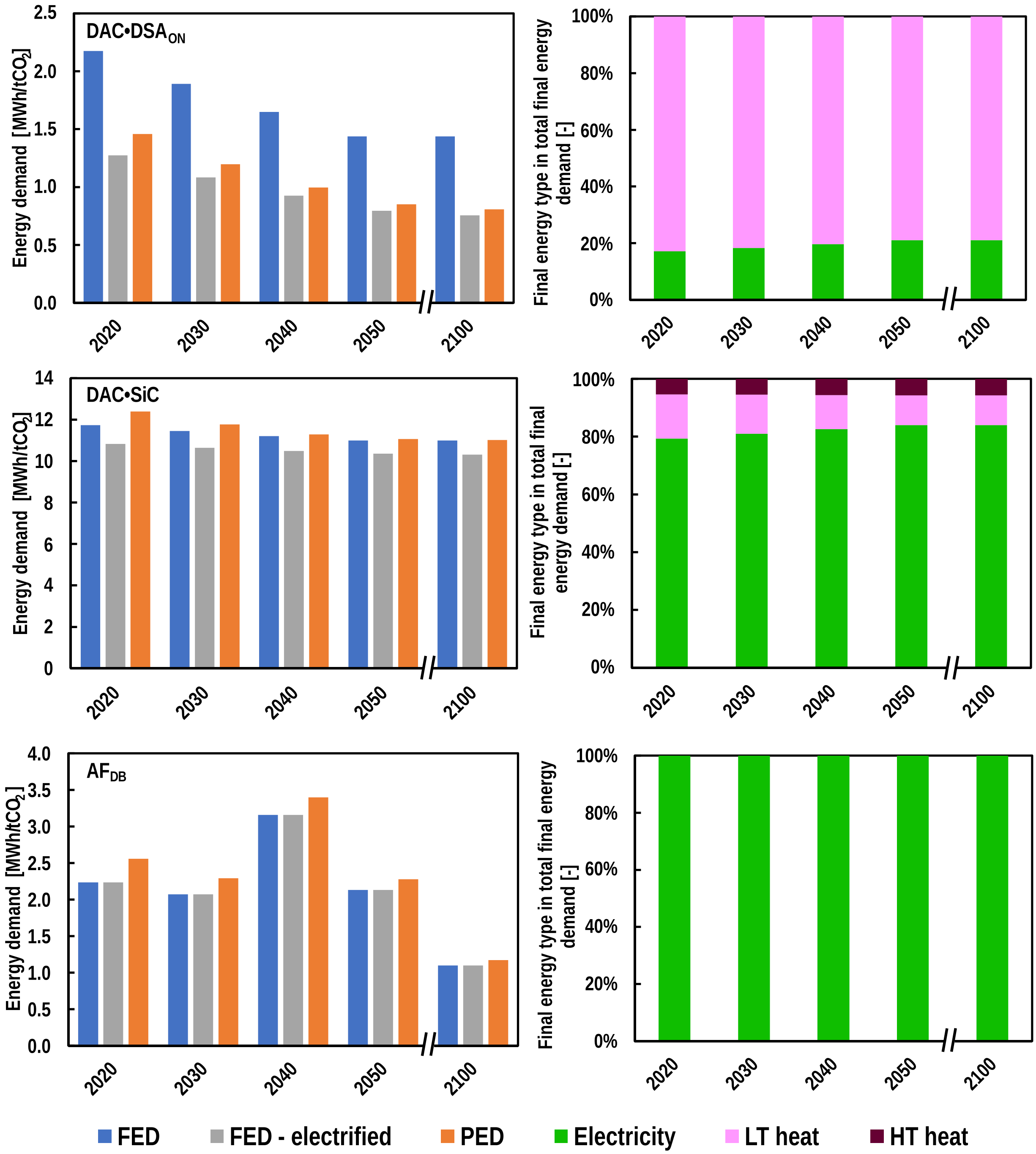

Fig. 7. Absolute final energy demand (left) and types of final energy in total demand (right) for DAC•DSA$_{ON}$ (top), DAC•SiC (centre), and AF$_{DB}$ (bottom). Ambient heat utilised by heat pumps is not considered as primary energy.

cost compared to DAC throughout the century, however the potential is limited to sustainable point sources. Most other NETs cost is around 200 €/tCO$_2$ from 2050 onwards. The initially calculated cost of AF$_{RF}$ is based on highly productive trees in best climatic locations (Caldera and Breyer, 2023). While AF$_{DB}$ can be applied there, AF$_{RF}$ is limited to humid areas and not able to promote optimal tree growth. Therefore, the initially calculated cost is normalised to literature findings to provide an average global cost that still bears a respective uncertainty. More details can be found in the supplementary material 2.

4.4. Land occupation

Results are shown in Table 3 for area demand of NETs excluding energy, the gross area demand for primary energy supply being included and the net area demand including land occupation for primary energy supply. Further details can be found in supplementary material 2 in the sheet '*Results_Area*'.

Biogenic NETs relying on photosynthesis, e.g., AF$_{RF}$, AF$_{DB}$ or BC$_{CP}$, and geo-chemical approaches, such as EW show significantly higher area demand compared to DAC- or PSC-based technologies. Even if area demand for primary energy is considered, energy-intensive options like PSC•SiC cover a fraction of the area that is required for biogenic technologies.

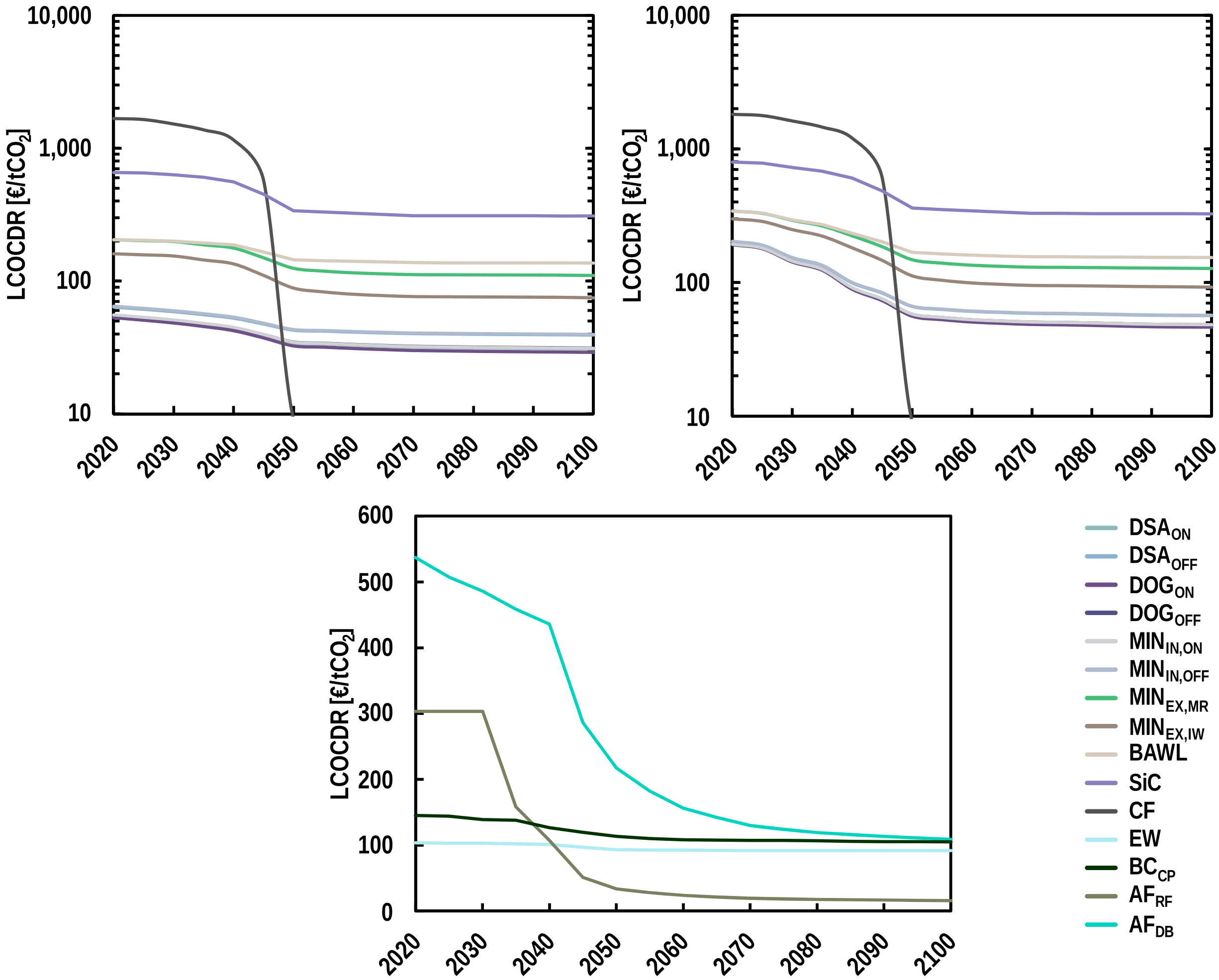

Fig. 8. LCOCDR of NETs considered within this research. Sequestration options for PSC (top left) and DAC (top right) as well as geo-chemical NETs (bottom) are displayed. The costs include the full CDR chain of capture, transportation and sequestration. Further information can be found in the supplementary material 2.

4.5. Carbon dioxide removal portfolios for societal preferences

In the following, results for the investigated CDR portfolios BP, LCP, LEP, HSP, LAP, and HTP, covering the ambitious CDR demand to limit global warming to 1.0°C (Figure S3 supplementary material 1, note 4), are examined in detail. Detailed results can furthermore be found in the supplementary material 2.

4.5.1. Portfolio technology composition

The portfolio composition for the deployment scenarios investigated in this study are presented in Fig. 9. Deployment results for the 1.5°C scenario can be found in Figure S9 in the supplementary material 1, note 10. The deployment scenarios are post-processed, i.e., corrected for the growth rate of afforestation-based NETs. Since the trees' growth cannot follow the assumed logistic curve of the CDR demand, afforestation initially exceeds the CDR demand and compromises the total installed CDR capacity later (c.f. negative and positive correction in Fig. 9). This characteristic is discussed further in supplementary 1, note 7–8.

For all portfolios created except HTP, both $MIN_{IN,ON}$ and $MIN_{IN,OFF}$ are the storage technologies deployed first. The in-situ mineralisation options have the highest overall score in the configurations used for portfolio creation, due to their relatively low cost and energy demand, i.e., comparable to geological underground sequestration, their low area demand, and high security score through mineralisation. However, due to the high annual CDR demand, DSA_{ON}, DSA_{OFF}, DOG_{ON}, and DOG_{OFF} using geological sequestration are deployed to full annual scale, which can be explained by the low cost, low area occupation, and low energy demand. Ex-situ mineralisation options are deployed in all portfolios except the LCP and the HTP to full scale while in the LCP $MIN_{EX,MR}$ is limited to about 1.1 $GtCO_2$/a and in HTP no ex-situ mineralisation is deployed. AF_{DB} is deployed in all portfolios except the LCP and the LAP, which can be explained by the relatively high cost in 2050, however never to its maximum potential.

In general, the LAP neglects all biomass-based NETs due to their high specific area demand. The LCP is the only portfolio where e-CF production is deployed to full scale with the zero CO_2 related cost resulting from product sales partially offsetting the high energy demand. EW is deployed in all portfolios except LAP and HTP indicating its attractiveness in many performance dimensions. It should be noted that with the configurations chosen for these case studies, bioenergy-based PSC is only deployed in the LCP and LAP. With the maximum annual sequestration potential of DAC surpassing all storage options for concentrated CO_2 and DAC being preferred in every portfolio in the year 2050, the deployment calculations do not prefer any bioenergy-based PSC to be introduced in other portfolios. This is due to the superior performance of DAC compared to PSC in security (carbon efficiency) and very similar cost and energy demand. However, once the portfolio is cost-optimised (LCP), PSC is preferred over DAC.

4.5.2. Energy demand for portfolios

Fig. 10 depicts the estimated annual PED for CDR and the specific average PED per tCO_2 for CDR required to realise a CDR sector to be added to the energy-industry system limiting global warming to 1.0°C and 1.5°C with different portfolios and respective implications.

Table 3
Area demand of NETs excluding and including area demand for primary energy supply via wind power and solar PV in 2050. PSC is actually bioenergy-related CO_2 captured at point sources. Assumptions for area demand for electricity generation are listed in the supplementary material 2 in the sheet '*Scenarios*'.

NET	Area excl. energy	Gross area incl. energy	Net area incl. energy
	$km^2/(MtCO_2 \cdot a)$	$km^2/(MtCO_2 \cdot a)$	$km^2/(MtCO_2 \cdot a)$
DAC•DSA_{ON}	1.4	9.2	4.4
DAC•DSA_{OFF}	1.4	9.2	4.4
DAC•DOG_{ON}	1.4	9.2	4.4
DAC•DOG_{OFF}	1.4	9.2	4.4
DAC•$MIN_{IN,ON}$	1.4	9.4	4.5
DAC•$MIN_{IN,OFF}$	1.4	9.4	4.5
DAC•$MIN_{EX,MR}$	1.4	22.2	9.5
DAC•$MIN_{EX,IW}$	1.4	19.7	8.5
DAC•BAWL	1.4	25.1	10.6
DAC•SiC	1.4	102.3	40.6
DAC•CF	1.4	544.7	212.7
PSC•DSA_{ON}	0.0	3.8	1.5
PSC•DSA_{OFF}	0.0	3.8	1.5
PSC•DOG_{ON}	0.0	3.8	1.5
PSC•DOG_{OFF}	0.0	3.8	1.5
PSC•$MIN_{IN,ON}$	0.0	4.0	1.6
PSC•$MIN_{IN,OFF}$	0.0	4.0	1.6
PSC•$MIN_{EX,MR}$	0.0	16.9	6.6
PSC•$MIN_{EX,IW}$	0.0	14.4	5.6
PSC•BAWL	0.0	19.8	7.7
PSC•SiC	0.0	96.9	37.7
PSC•CF	0.0	539.3	209.7
EW	0.0	3.8	1.5
BC_{CP}	433.0	433.2	433.1
AF_{RF}	3483.5	3483.5	3483.5
AF_{DB}	619.2	627.7	626.6

The total estimated annual PED for the CDR sector correlates with the CDR deployment and rises to about 36.1 PWh/a (129.9 EJ/a) for the LEP. Interestingly, the BP and LEP show the same structure of NETs. This structure can be explained by the fact that energy and cost strongly correlate; therefore, low-energy options are prioritised in the BP for their low energy demand as well as for their resulting low LCOCDR. In case of the average PED for CDR normalised to 1 tCO_2 removed, all portfolios except the LCP and the HTP show the same average PED in the initial years due to low CDR demand and the portfolios' deployment characteristics elaborated in Section 4.5.1. In the long term, the LCP has the highest average PED of about 3.87 MWh/tCO_2 and the LEP has the lowest average PED of 0.88 MWh/tCO_2. The enormous PED of LCP is due to the full deployment of e-CF production which is further discussed in Section 5.1.

To put the results in perspective, the annual PED for CDR is put into relation to the estimated TPED projection of the LUT-DEES as provided by Keiner et al. (Keiner et al., 2023) in Fig. 10. The BP, LCP, LEP, HSP, LAP, and HTP peak in additional PED in the projected TPED at about 12.0%, 37.6%, 12.0%, 12.6%, 27.4%, and 11.9% respectively. Notably, no portfolio requires more than 13.3% additional PED in the context of the LUT-DEES (Keiner et al., 2023), except the LCP and the LAP due to low-cost e-CF utilisation and low area NETs such as SiC deployment, as discussed in Section 5.1.

4.5.3. *Cost for carbon dioxide removal*

Cost for CDR is decisive for enabling ambitious climate mitigation pathways as economic means must be negotiated in societies and finally agreed in decision-making processes and linked to concrete policies for the respective deployment. Fig. 11 shows the annualised cost including investments, operations and energy for the investigated CDR portfolios, the specific cost for the CDR portfolios normalised to 1 tCO_2 removed and the total cost in ratio to the global GDP in the context of the LUT-DEES (Keiner et al., 2023).

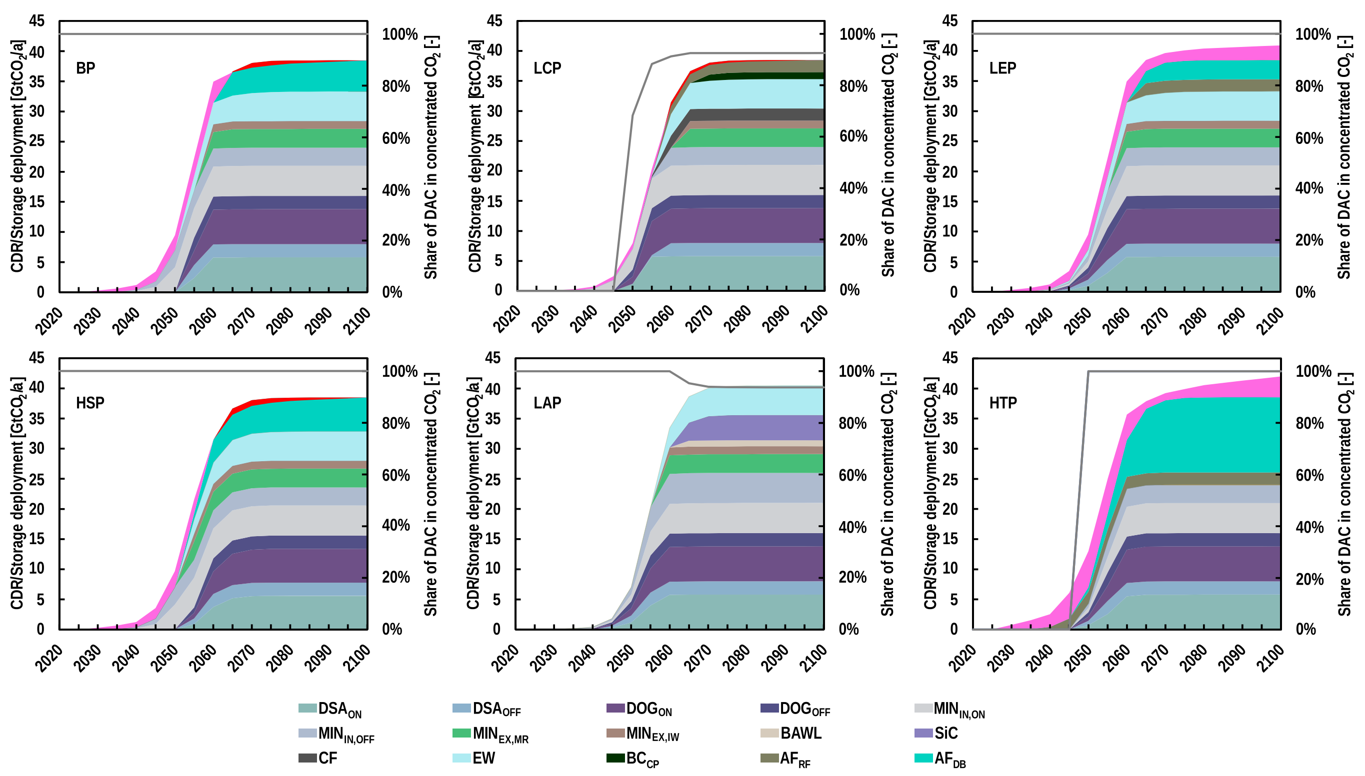

Fig. 9. CDR deployment scenarios limiting global warming to 1.0°C for a BP (top left), LCP (top centre), LEP (top right), HSP (bottom left), LAP (bottom centre), and HTP (bottom right). For visualisation purposes only storage options are displayed for NETs storing concentrated CO_2 from either DAC or PSC. Please note that storage options are not ordered according to their deployment but in a regular order.

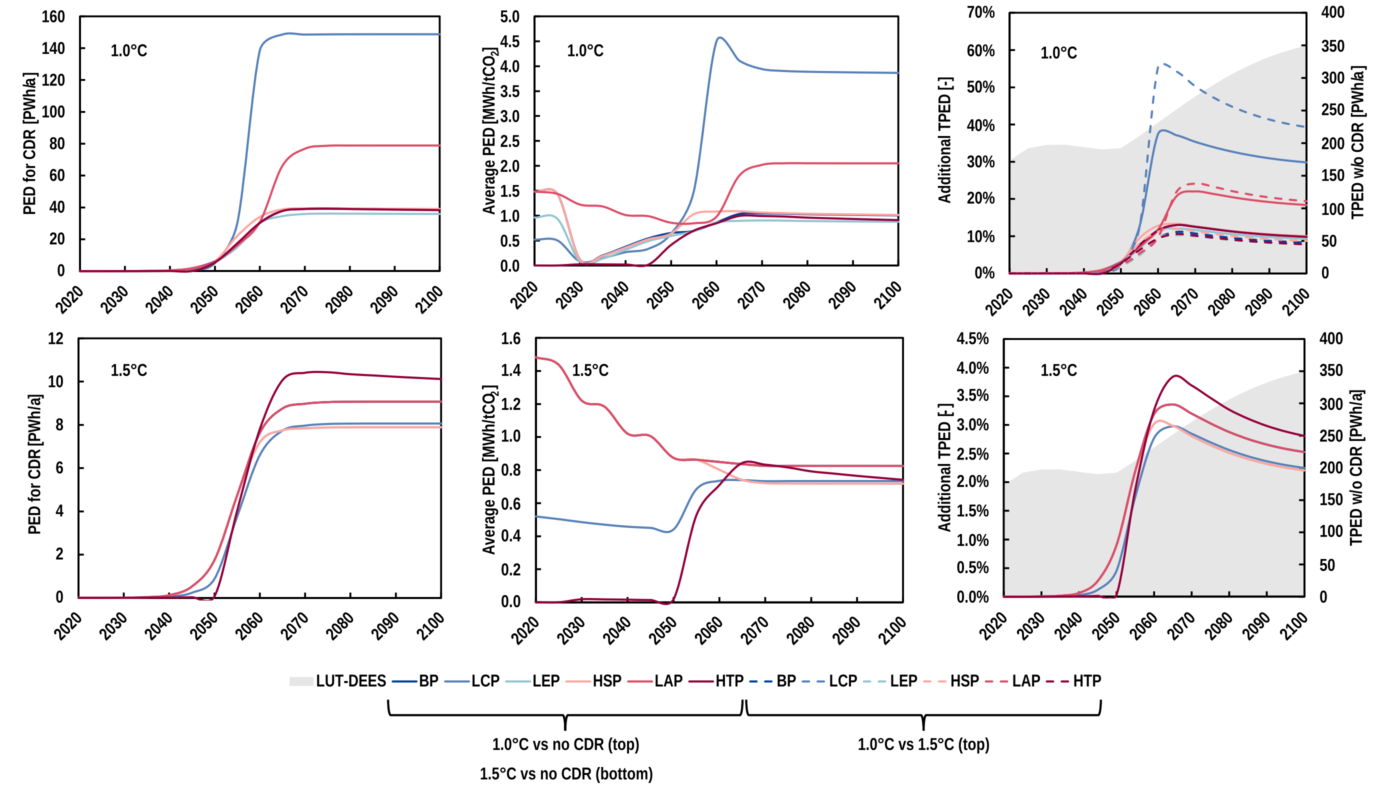

Fig. 10. PED for CDR portfolios compliant with a 1.0°C pathway (top) based on 1750 $GtCO_2$ total negative CO_2 emissions within this century and a 1.5°C pathway (bottom) based on 500 $GtCO_2$ total negative emissions. Total annual PED for CDR (left), average PED of NETs (centre), PED for limiting global warming to 1.0°C (solid lines top right) and 1.5°C (solid lines bottom right) compared to LUT-DEES TPED projection without any CDR demand, and with CDR in alignment with limiting global warming to 1.5°C considered (dashed lines top right) (Keiner et al., 2023). Additional TPED is calculated as the share of PED for CDR in TPED including the PED for CDR. The portfolio evaluation year is set to 2050. Please note that the BP, LEP and HTP lead to similar results in some years. Please note different y-axis scaling.

The BP, LEP, HSP, HTP, and especially LAP show higher annualised costs compared to the LCP, due to the cost-optimisation of the LCP. Interestingly, the LEP is equivalent to the BP which may be explained by the strong correlation of energy and cost, i.e., low-energy options being prioritised due to the low energy demand and the implied low cost. The cost for electricity steadily decreases until 2070, so energy becomes less cost-intensive until then. The annualised cost for the LCP and LEP peak in 2080 and 2075 at 2296 b€/a and 2807 b€/a, respectively. The HSP reaches its maximum total annual cost of 2789 b€/a in 2075.

While the average cost declines steeply from almost 200 €/tCO_2 and up to 300 €/tCO_2 for the HTP in 2020 to about 68 - 72 €/tCO_2 for all portfolios except the LCP which remains relatively constant at 52 - 56 €/tCO_2 except for the scale-up phase around 2030.

It should be noted that, as shown in Fig. 11, less than 0.42% of the projected global GDP in 2100, except for the LAP, would be sufficient to finance large-scale CDR that might limit global warming to 1.0°C if ambitious short-term emission mitigations are implemented and a low-cost CDR portfolio, i.e., the LCP, would be realised. The specific cost for large-scale CDR peaks at 0.55%, 0.42%, 0.55%, 0.56%, 0.65%, and 0.60% of the global GDP for the BP, LCP, LEP, HSP, LAP, and HTP, respectively. As the global GDP projection shows a steadily increasing development in the applied macro-economic scenario until 2100 and CDR cost decrease as shown in Fig. 11, the specific cost of CDR decreases to below 0.5% for all portfolios in 2100, reaching the 1.0°C target assumed in this research. The cost of CDR to meet the TNE for the 1.5°C target, measured in additional cost as a ratio to the projected GDP, remains below 0.19% throughout the century for all portfolios.

5. Discussion

In this section, several aspects of the results and the context of this study are discussed. This includes a classification of the results in comparison to results from literature in Section 5.1, sustainability challenges of large-scale CDR with focus on DACCS, BECCS and CO_2 leakage in Section 5.2, limitation of the modelling done in this study in Section 5.3, aspects of how to finance large-scale CDR with focus on a post emission trading framework in Section 5.4, general purpose of CDR under the light of tipping points 5.5, and an outlook on possible research based on this study in Section 5.6.

5.1. Comparison and classification of results

A general issue when assessing literature is non-conforming terminology. It is important to be clear what exact sub-processes are included when discussing a specific NET to produce comparable results. For example, DACCS and BECCS are discussed frequently in literature (Minx et al., 2018, Fuss et al., 2018, Nemet et al., 2018), though they are treated as single processes rather than the process chains that comprise the technologies. For the case of DACCS, there are several DAC technologies with different techno-economic specifications, as well as additional CO_2 transport and sequestration options. Therefore, to make comparable results and assumptions these specifications and open questions must be considered (Terlouw et al., 2021).

Since, in this study, practically no constraints were set for DAC, the deployment of DAC of 6.6 $GtCO_2$/a in the BP, LEP, HSP, and LAP, for the TNE compliant with 1.0°C global warming, exceeds the upper limits proposed by Minx et al. (Minx et al., 2018) or Fuss et al. (Fuss et al., 2018) for DACCS in 2050 by about 1 - 2 $GtCO_2$/a. However, a recent

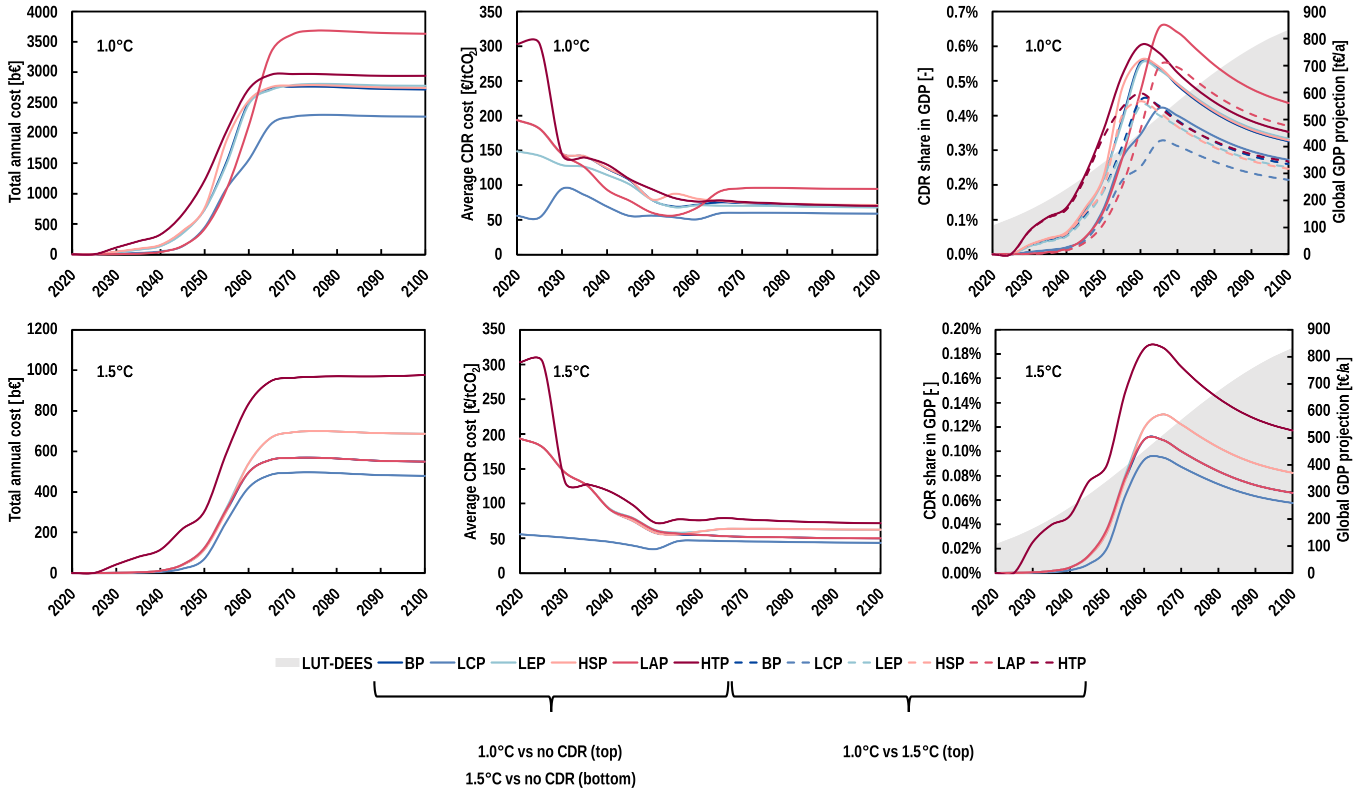

Fig. 11. Cost results of different CDR portfolios for a 1.0°C pathway (top) and a 1.5°C pathway (bottom). Total annualised cost of CDR for different CDR portfolios (left). Average cost for CDR in different portfolios (centre). Share of global GDP projection for CDR limiting global warming to 1.0°C (solid lines top right) and 1.5°C (solid lines bottom right) in LUT-DEES GDP projection with CDR for limiting global warming to 1.5°C considered (dashed lines top right) and without CDR considered (Keiner et al., 2023) (bottom right). The year of portfolio evaluation, impacting parameters for optimisation such as cost, energy demand, security, efficiency, etc., is set to 2050. Please note that the BP and LEP produce similar results. Please note different y-axis scaling.

report on CDR finds this scale feasible for DAC in 2050 (Smith et al., 2023). The early phase-in for DAC found in this study contrasts with previous findings but affirms the sensitivity of deployment to the potential of available alternatives and the maximum deployment found in a previous study (Chen and Tavoni, 2013). The DAC technology is expected to experience substantial growth rates similar to other modular technologies (Breyer et al., 2019, Galimova et al., 2022). Thus, a respective market ramping for DAC units for DACCU and DACCS applications should not be a limitation. The compound annual growth rate for DAC units remains below 35% from 2030 to 2050, except for one 5-years period with 80%, and it declines to below 10% beyond 2060. This growth rate can be compared to solar PV with compound annual growth rates of 56% and 24% for the strong growth periods between 2000 to 2010 and 2010 to 2020, respectively ([IEA] 2022).

Deutz et al. (Deutz and Bardow, 2021) examined components of DAC units that might be scarce in the future after a large-scale deployment. In the study, only adsorbent production was found to be challenging for future large-scale DAC deployment. In the LCP, the DAC deployment is even below 5 $GtCO_2$/a in 2050, which can be explained by the higher DAC LCOCDR compared to PSC that is prioritised in that portfolio. The LCOCDR of DACCS, i.e., DAC with subsequent CO_2 transportation and underground sequestration, as calculated in this research, ranges from about 56 - 66 €/tCO_2 in 2050. This levelised cost is at the rather lower range of estimations or even blow estimations made by Minx et al. (Minx et al., 2018), Fuss et al. (Fuss et al., 2018), Chauvy et al. (Chauvy and Dubois, 2022), Möllersten and Naqvi (Möllersten and Naqvi, 2022), or Bui et al. (Bui et al., 2018). Therefore, the projected cost can be seen ambitious, however, feasible as discussed by Fasihi et al. (Fasihi et al., 2019) and Breyer et al. (Breyer et al., 2019).

BECCS, AF_{RF}, and EW are NETs frequently discussed in the literature (Smith et al., 2023, Rueda et al., 2021, Strefler et al., 2021) and regularly deployed in IAMs (Table 1). EW's significant role in future CDR portfolios for TNE compliant with a 1.0°C target can be confirmed by the results derived from this research, i.e., it is fully deployed in the BP, LCP, LEP and partially deployed in the HSP, whereas AF_{RF} is only deployed in the LCP due to its low cost comparable to findings on other low-cost scenarios provided in literature (Rueda et al., 2021). In other portfolios calculated, AF_{RF} is also deployed at a rather small scale as also found by Migo-Sumagang et al. (Migo-Sumagang et al., 2022). All other portfolios in this study deploy AF_{DB} instead, implying advantages over AF_{RF} such as lower land occupation through more efficient tree growth and in particular higher security due to continuous irrigation (Chiquier et al., 2022).

BECCS, i.e., bioenergy-based PSC, often plays a key role in IAMs for 1.5°C compliant mitigation scenarios ([IPCC] 2022). However, in this study, BECCS is only deployed in LCP due to its perceived lower cost compared to DACCS and a low weighting of other factors such as security, which includes carbon efficiency and area demand. This finding is in line with the results by Rueda et al. (Rueda et al., 2021) where BECCS was not deployed in portfolios with a low CDR demand. BECCS is also not deployed in portfolios calculated by Migo-Sumagang et al. (Migo-Sumagang et al., 2022).

Most reviews on NETs list total cost numbers for many compound technologies or process chains including DACCS or BECCS (Minx et al., 2018, Fuss et al., 2018). This issue is curbed in this study (cf. Section 2). A major aim of this research is to reduce unclarities about what process steps exactly are employed for a specific NET but some simplification is still necessary. Thorough assessments investigating different plant configurations are yet to be done and further systematic CDR assessments in the portfolio context with clear techno-economic specifications are possible based on this study. The role of plant location on different technologies' cost and energy demand should be subject to future research on regional level.

Table 4 summarises the key findings of this research. Large-scale CDR deployment with the possibility of limiting global warming to 1.0°C in synthesis with a rapid defossilisation of the energy-industry system requires investments that are a minor share of the global GDP. Even for a highly secure CDR portfolio, no more than 0.56% of the global GDP must be invested annually to curb severe effects of global warming, which may lead to significantly higher cost for compensating the effects of climate change. For a cost-optimal CDR portfolio, no more than 0.42% of global GDP must be invested; for an energy-optimal portfolio no more than 12.0% additional TPED is required annually. It should be noted that the LCP for 1.0°C shows enormous PED, which may make it infeasible. However, the PED is related to large-scale e-CF production with product sales from 2050 onwards and can also substitute steel, which shows substantial energy demand as well (Lopez et al., 2022). Therefore, this result requires further investigation to see the impact in a full energy-industry-CDR system transition analysis. All portfolios for TNE being compliant with a 1.5°C global warming target seem much feasible with additional cost in projected GDP of up to 0.19% and additional PED in projected TPED of up to 3.8%.

As displayed in Table 4 the maximum total annual cost of LCP for a 1.0°C pathway is close to today's global annual military expenses of 2100 bUSD/a ([SIPRI] 2022), while all portfolios compliant with TNE for 1.5°C cost up to 24% of today's global annual military expenses. It should be noted that results relative to a projected GDP scenario imply significant economic growth throughout the century. In addition, portfolios for a TNE of 500 $GtCO_2$ by 2100 require significantly less versatile NETs with mainly three storage options being deployed in the different portfolios (c.f. supplementary material 1, note 10).

5.2. Sustainability challenges

Removing CO_2 from the atmosphere can have beneficial health effects on humans by mitigating climate change (Cobo et al., 2022). This may, however, also bring negative impacts on health compared to even more ambitious emission mitigation (Jacobson, 2019), if fossil fuels combustion is still in use for CDR as suggested for the non-electric high-temperature DACCS approach. That approach is not considered in this research. According to Cobo et al. (Cobo et al., 2022), monetising the possible health benefits of CDR to mitigate global warming can offset the cost for CDR as they conclude for a case study for high-temperature liquid sorbent DACCS. Major environmental effects of large-scale NETs must be examined thoroughly and could generally be minimised by transformational change at adequate rates, lowering the overall demand of CDR by avoiding emissions. The aim should be minimising negative side effects on the environment and energy demand. As results indicate, the LEP can enable large-scale CDR compliant with limiting global warming to 1.0°C and 1.5°C at a maximum energy demand of 36.1 PWh and 7.9 PWh, respectively.

Table 4
Key findings on total cost and energy demand for large-scale CDR to limit global warming to 1.0°C (1750 $GtCO_2$ TNE) and 1.5°C (500 $GtCO_2$ TNE).

TNE		Maximum Cost		Maximum Energy Demand	
		Total annual	Additional cost in GDP*	Total annual	Additional PED in TPED*
$GtCO_2$	Unit	b€	-	PWh	-
1750	BP	2764.6	0.55%	39.2	13.0%
	LCP	2295.5	0.42%	148.8	37.5%
	LEP	2807.3	0.55%	36.1	12.0%
	HSP	2789.4	0.56%	39.5	13.3%
	LAP	3685.0	0.65%	78.8	22.0%
	HTP	2971.4	0.60%	39.2	13.0%
500	BP	568.8	0.11%	9.1	3.4%
	LCP	496.2	0.09%	8.1	3.0%
	LEP	699.5	0.13%	7.9	3.0%
	HSP	699.5	0.13%	7.9	3.0%
	LAP	568.8	0.11%	9.1	3.4%
	HTP	976.5	0.19%	10.4	3.8%

* compared to the macro-economic projection of LUT-DEES without CDR as presented by Keiner et al. (Keiner et al., 2023)

Even though there are detailed LCA of NETs such as DACCS (Madhu et al., 2021, Deutz and Bardow, 2021), they are not yet always comparable (Terlouw et al., 2021) or are lacking important NETs and should be investigated further. In the end, shifting the responsibility for the climate crisis to other sectors that may not be present today must be avoided in any case (Jeswani et al., 2022). Günther and Ekardt (Günther and Ekardt, 2022) examined the compatibility of BECCS and DACCS with different human rights. They find that BECCS can cause major infringements on human rights, e.g., the right to food caused by water and land stress, while they see DACCS mainly constrained by the right to energy (Günther and Ekardt, 2022). However, this study finds that projections employing large-scale DAC deployment have a manageable additional TPED, which is in line with findings by Keiner et al. (Keiner et al., 2023).

EW can have several positive side effects on soil such as fertilising and therefore increasing the productivity or buffering soil erosion (Smith et al., 2019). Since the carbonation rate of spread minerals is affected by moisture, further synergies with AF_{DB} could arise maximising the benefits of constant irrigation and the resulting soil moisture and the fertilising effect of EW. Side effects of AF vary widely depending on the modalities and scale-up. Biodiversity can be comprised by large-scale AF, which may also compete with land available for food production (Smith et al., 2019).

Freshwater demand can be a significant limitation to large-scale CDR deployment. With several regions in the world suffering from droughts, freshwater is already a valuable resource today and the situation might worsen in the future (Sovacool et al., 2023). Mineralisation is usually supported by freshwater to increase reaction kinetics; however, the water can be reused (Snæbjörnsdóttir et al., 2020). The share of freshwater that is ultimately recyclable is key and is not accounted for in this study. Further research in this field will be required to gather more knowledge. Voigt et al. (Voigt et al., 2021) study the impact of seawater utilisation for mineralisation as proposed in previous studies (Snæbjörnsdóttir et al., 2020). Rosa et al. (Rosa et al., 2021) project the global green water (root-zone soil moisture available to plants) and blue water (freshwater in surface and groundwater bodies available to humans) demand for different scenarios and find that large-scale BECCS deployment has most severe impact to future water demand and conclude that a portfolio approach is desirable. Another limiting factor for CDR deployment is area occupation as presented in subSection 4.4. However, these results should be further investigated in future studies employing a thorough LCA. It is also questionable whether the area occupied through afforestation should be compared to the area occupied by power generators. Within this study, this topic is out of scope.

Storing CO_2 in its gaseous form underground is the cheapest storage option identified. However, this storage option bears the risk of CO_2 leakage (Alcalde et al., 2018). While mineral trapping, so called in-situ mineralisation, stores CO_2 for geological timespans, there are concerns about other more vulnerable trapping mechanisms (Ajayi et al., 2019). Alcalde et al. (Alcalde et al., 2018) present a numerical model to investigate storage security of different geological underground sequestration sites. The study concludes that carbon efficiency of storage after 10,000 years is about 98% in well-regulated and 78% in poorly regulated storage sites (Alcalde et al., 2018). However, Alcalde et al. (Alcalde et al., 2018) point out the high uncertainty regarding development of subsurfacially stored CO_2 in 10,000 years. Lyngfelt et al. (Lyngfelt et al., 2019) conclude that leakage rates higher than 1% per year can result in significant increase in CDR demand to achieve substantial net CDR rates sufficient for climate change mitigation (Lyngfelt et al., 2019). For the assumptions of this research, these leakage rates may mean up to 17.5 $GtCO_2/a$ additional CDR demand in the very long term to balance the annual leakage. Risk assessment, monitoring, and detection of leakage at the injection site is crucial for geological CO_2 storage and subject to multiple studies (Connelly et al., 2022, Gholami et al., 2021). To avoid these risks in the first place, sites suitable for in-situ mineralisation in basaltic rock formations should be prioritised. The respective cumulative potential seems to be no limitation for large-scale storage operations (Oelkers et al., 2023). The risk of CO_2 leakage and therefore the durability of storage was included for this study with a risk score calculated as described in subSection 3.5 that is considered for all portfolios presented.

5.3. Modelling limitations

The LUT-CDR spreadsheet tool used for the portfolio creation limits the options for optimisation of CDR portfolios. For example, as described in Section 3.7, portfolio creation and optimisation can only be conducted after a single year of choice and the deployment requires several steps to calculate viable portfolios that cover the demand for negative CO_2 emissions in every year. Also, non-linearities, i.e., afforestation-based CDR trajectories can only be considered with some limitations and corrections. In this research, the initially calculated deployment is corrected since the logarithmic growth rate of trees (Caldera and Breyer, 2023) is not able to match the logistic curve shaped CDR demand. The method is described in more detail in the supplementary material 1, note 7–8. The deployment of afforestation-based CDR as modelled in this study creates the necessity of significant investment starting as early as 2030, while the return of this investment, including total cost decrease, follows about 25 - 45 years after the first investment. It can be questioned whether such NETs will be preferred by society or decisionmaker (Dietz et al., 2020). However, AF is a much practiced and recommended NET. The timing of NETs is regarded to be critical (Chiquier et al., 2022). However, AF_{DB} might bring significant positive co-benefits, such as cooling the surrounding, fight desertification, or food production (Caldera and Breyer, 2023). While climate change is expected to exceed the threshold for human adaptability in some regions, as for instance in parts of the Middle East (Pal and Eltahir, 2016), such side effects might become the main reason for AF_{DB} deployment and can, therefore, make afforestation-based CDR a viable option if immediate economics without considering positive side effects is not the most important factor.

In this study, no income related positive side effects or product utilisation are considered, except for e-SiC and e-CF according to their material value. The cases of e-CF and e-SiC represent the rare class of a combined CCS and CCU application, so that further income can be generated that may lead to a profitable future business (Mühlbauer et al., 2024, Keiner et al., 2023). The used tool, LUT-CDR, shows the sensitivity to the CF product value that may be generated in the future, indicating an enormous CDR potential in combination with an attractive business case if CF product applications were enabled in respective volumes, e.g., for vehicle structures, buildings, etc. (Keiner et al., 2023, Choi et al., 2019, Böhm et al., 2018, Backes et al., 2023). Also, carbonates have several possible applications (Strunge et al., 2022), potentially lowering cost and boosting deployment. These effects are not considered within this study and offer the potential for further cost decrease.

With this study, options for CDR and a tool for creating portfolios, which are optimised with respect to five societal preferences, are presented. However, indicators of NETs' performance may differ substantially depending on the location of deployment (Fridahl et al., 2020). Regional constraints to NETs must be considered to guide future CDR development (Fajardy et al., 2019). Therefore, CDR indicators must be assessed on a rather regional basis in the future to enable high resolution modelling of NETs such as that done by Sendi et al. (Sendi et al., 2022). However, regional data are often insufficient with some exceptions (Beerling et al., 2020, Goll et al., 2021). With these limitations in mind, it should be noted that not all dimensions for CDR evaluation can be considered within this framework. Most prominently, environmental impacts of land-based CDR options are subject to recent LCAs (Terlouw et al., 2021) but cannot be generalised without considering scale, local conditions, and management strategies. The impact on the local environment can be either positive or negative depending on multiple factors ([IPCC] 2022). A thorough assessment of such factors is considered out of scope of this study. The lack of spatial resolution means that no further inferences for the interregional transfer of CDR can be made [240].

The methodology used to consider learning effects applied for calculating future capex and fixed opex estimations can be further improved adapting literature recommendations, such as including learning-by-doing and learning-by-searching effects among others, as proposed by Thomassen et al. (Thomassen et al., 2020). This approach can be used in future work to further verify results presented in this study and to improve results qualitatively. While learning effects for NETs are considered, industrial cost curves for NETs are neglected in this study. These would lead to higher cost of technologies with higher deployment, but the presented portfolio approach would somewhat avoid such issues similar to least-cost renewable energy portfolios. The effect of industrial cost curves on the presented results should be studied in future work.

In this study, a specific CDR deployment was assumed to examine the future cost of different NETs and portfolios thereof for societal preferences. However, since the learning rate approach used in this study is sensitive to the historic cumulative installed capacity of each technology and, therefore, to the deployment scenario assumed initially, the results presented are directly related to the initial deployment scenario. This issue can be addressed in follow-up work, by assessing the cost of CDR portfolios in energy-industry-CDR system (Breyer et al., 2022) scenarios variations. Therefore, this work provides the basis for diverse energy-industry-CDR studies in the future. Also, the actual deployment of NETs is compared to the initial deployment scenario in the supplementary material 2.

Alternative deployment scenarios derived from data provided in the IPCC AR6 WGIII ([IPCC] 2022) (Table S20 in the supplementary 1, note 11) and results thereof are shown in Figure S10-S11 in the supplementary 1, note 11. The comparison with the IPCC literature scenario shows that NETs present in leading IAMs show relatively low diversity, low security, but also lower cost and energy demand compared to the BP and LCP presented in this study. As shown in Table 1 leading IAMs only use consistently BECCS and AF, while DACCS is not yet standard and EW is hardly used, and CO_2 obtained from point sources or DAC units is assumed to be geologically stored. The broad set of NETs presented in this study enables stakeholders to overcome these gaps in literature.

The field of CDR is highly diverse and attracts increasing attention in science and industry. Therefore, the field of NETs will further diversify, and further NETs as mentioned in sub-section 3.2.10 will emerge. These will widen the portfolio options presented in this study and can reduce the TNE demand for engineered NETs. For NCS, however, since the average cost for a 11.3 $GtCO_2/a$ scale is projected to be around 83 €/tCO_2 (Griscom et al., 2017), these would theoretically not be phased-in in an LCP. Options not considered within this work shall be investigated further to enable results that make them viable for inclusion in energy-industry-CDR portfolio analyses. The synergies between NETs can bring further benefits such as lower land occupation and overall CDR cost and should, therefore, be considered for simulating CDR portfolio systems (Abraham et al., 2022, Migo-Sumagang et al., 2022). Additional synergies enabled by the CDR sector with the energy-industry system can be investigated in high temporal and spatial resolution as already initiated (Breyer et al., 2020, ElSayed et al., 2023) for the case of DACCS. There is also further potential for industrial flows, such as coupling the steel industry and CDR sector for steel slags use. The PED for NETs is largely based on electricity, which adds another application to the diverse power-to-X field (Sterner and Specht, 2021), in this case, power-to-CDR, as a further almost fully electricity-based sector in the Power-to-X Economy (Breyer et al., 2023).

5.4. Financing of large-scale carbon dioxide removal portfolios

The biggest hurdle for application of CDR portfolios might be unsolved financing. As displayed in Table 4, the maximum total annual cost of the LCP for a 1.0°C pathway is close to today's global annual military expenses of 2100 bUSD/a ([SIPRI] 2022). The annual CDR cost can be estimated to about 0.4 - 0.6% of the projected global GDP (cf. Fig. 11). This cost could exceed the capabilities of a voluntary carbon market, as already practiced by leading stakeholders (Joppa et al., 2021). These are substantial financial means, though it can be expected that there is willingness-to-pay for overcoming a real existential threat. Evidence is growing that the adaptation costs are much higher than those for CDR (Sanderson and O'Neill, 2020). The cheapest option would be to keep fossil fuels in the ground. To minimise lagged social external cost, a moderate CDR deployment is preferable over extreme ramp-up at the end of the 21st century (Obersteiner et al., 2018).

Some CDR approaches create co-benefits, such as afforestation with a positive impact on local climate (Caldera and Breyer, 2023, Pal and Eltahir, 2016), soil enrichment with higher agricultural yields (Laihonen et al., 2022) and rewetting of wet land fosters recovery from biodiversity loss. Therefore, some CDR approaches might be easier to finance than others. A kind of global CDR tax may be required to collect the means for the about 0.42–0.65% of GDP in respective CDR cost (Table 4). The Organisation on Economic Co-operation and Development (OECD) average tax-to-GDP ratio was 34.1% in 2021 ([OECD] 2022), which indicates that a 0.42–0.65% global tax for CDR to rebalance to 1.0°C is not out of reach, as this may represent about 1.2–1.9% of the globally generated taxes. A more detailed discussion on the topic of CDR financing is available in the supplementary material 1, note 14.

5.5. Carbon dioxide removal and tipping points

The IPCC defines CTP to be the "critical threshold beyond which a system reorganizes, often abruptly and/or irreversibly" ([IPCC] 2022). CTP differ greatly in terms of global and regional impact, time scale at which they are triggered and act, and the global surface air temperature (GSAT) by which they can be triggered (Armstrong McKay et al., 1979). There is high confidence in the existence of CTP but still uncertainties remain about their parameters including thresholds and responding time to climate forcing (Wang et al., 2023). CTP events may imply disastrous consequences (Lenton et al., 2008, Zhang et al., 1979) and are partly interconnected with the risk of cascades (Lenton et al., 2019, Liu et al., 2023, Wunderling et al., 2023), which further increase the necessity of risk assessment and mitigation, in particular in the form of CDR. However, CDR technologies still require major development pushes to be available cost-effectively on a large scale by mid-century. A legitimate question is: Is CDR impacting quickly enough to avoid CTP in the Earth system, or is this development coming too late?

In this work, a level of atmospheric CO_2 concentration of about 350 ppm in 2100 is targeted in the scenario assumptions, which corresponds to about 1.0°C (Hansen et al., 2017, Azar and Rodhe, 1979, Hansen et al., 2008) to provide scenarios for ambitious temperature targets that can enable a safe and just climate (Rockström et al., 2023, Breyer et al., 2023), while a mid-century increase in concentration to about 450 ppm can be estimated. Accordingly, only CTP are considered that are triggered with high probability at 450 ppm or less than 2°C. These are four CTP in the cryosphere (collapse of the West Antarctic and Greenland ice sheet with a global impact and melting of Barents sea ice and Alpine glaciers with an regional impact), two CTP in the biosphere (collapse of coral reefs and thawing of boreal permafrost, both with direct regional but indirect global impacts), and one CTP as an ocean-atmosphere element, the North Atlantic subpolar gyre Labrador Sea convection with a global impact (Armstrong McKay et al., 1979).

The coral reefs are considered lost with high probability, i.e., even with CDR it is too late. However, all melting processes occur over decades to centuries, and in this long timescale lies the opportunity of CDR for reducing the probability of CTP in the cryosphere. At 350 ppm in 2100, glaciation resumes, so recovery can be expected, as has been observed in paleorecords (Foster and Rohling, 2013). The same applies to the melting of the boreal permafrost, that may release about 1035 GtC as CO_2 or methane (Schuur et al., 2015). However, there is medium or even low confidence in an abrupt thaw or collapse, rather than high confidence in a gradual thaw (Armstrong McKay et al., 1979). This thawing could be slowed down or even stopped by CDR.

Less clear is the impact of CDR on CTP in ocean-atmosphere interactions. The oceanic convection of the subpolar gyre in the North Atlantic is an important process in regulating the heat exchange between the ocean and the atmosphere. Oceanic convection of the subpolar gyre has been included in past years as a new CTP with high probability. However, the implications are not yet clear. A collapse is likely to lead to global cooling by 0.5 K but at the same time to a shift of jet streams and weather extremes in Europe (Swingedouw et al., 2021).

It is also uncertain whether the absolute GSAT or the total cumulative CO_2 emissions trigger the CTP. The bottom line is that the probability of CTP is reduced by CDR. Hence it makes sense to take a closer look at these technologies or scenarios in more depth. Further energy-climate modelling with a higher spatial resolution can underline the importance of CDR for preventing or managing CTP.

5.6. Research outlook

Within this study, most NCS were not considered to contribute to the TNE. This limitation is mainly due to the lack of suitable techno-economic input parameters as required for the implementation in this study. However, it is expected that NCS can effectively contribute to the TNE in the order of up to 11.3 $GtCO_2$/a at a cost of below 83 €/tCO_2 (Griscom et al., 2017). Therefore, NCS can possibly reduce the CDR demand of NETs employed in this study while simultaneously enabling positive side effects on biodiversity etc. (Griscom et al., 2017). These co-benefits are not considered within this study due to a lack of spatial resolution that is crucial for adequately projecting the effects of land-based NETs such as AR_{RF} ([IPCC] 2022). Since in this study only CO_2 is considered, the effects of other GHG such as methane might further increase the TNE that must be removed to reach a specific temperature level. Therefore, the effects of NCS, i.e., lowering the TNE and non-CO_2 GHG, i.e., increasing the TNE, are not considered. Literature suggests that non-CO_2 mitigation and possible removal options might ultimately be required to find cost-effective emission reduction pathways (Iyer et al., 2022, Jackson et al., 2021). Allen et al. (Allen et al., 2022) investigate the idea of net-zero climate with several GHG in detail. However, literature data on techno-economic parameters for removal of diverse GHG is still vague. The topic will become increasingly important in the future. Besides CO_2, there is a rise in research insights on methane removal (Ming et al., 2022, Wang et al., 2022), which may allow future research to combine CDR with methane removal for a more effective rebalancing to climate safety levels.

The trajectory of CDR demand, i.e., the annual demand for CDR, is simplified by approximating a logistic curve covering the historic CO_2 emissions projected for 2050 (Breyer et al., 2021). The total CDR demand in the 21st century is approximated by Breyer et al. (Breyer et al., 2021), and Keiner et al. (Keiner et al., 2023). There are other methods proposed to approximate CDR trajectories, e.g., Terhaar et al. (Terhaar et al., 2022) propose a method to create individual CO_2 emission trajectories in a simplified way by determining emission reductions in an iterative adaptive manner. For cross checking purposes, 97 emission trajectory scenarios with the respective average GSAT, that fulfil the C1 level were examined (Byers et al., 2022). C1 scenarios limit peak warming to 1.5°C or below with a chance of at least 33% and limit the global warming to 1.5°C or below with a chance of at least 50% (Kikstra et al., 2022). The findings as well as a linear approximation of the GSAT as a function of net CO_2 emissions in 2015–2100 is depicted in Fig. 12. The linear approximation shows the strong proportionality between

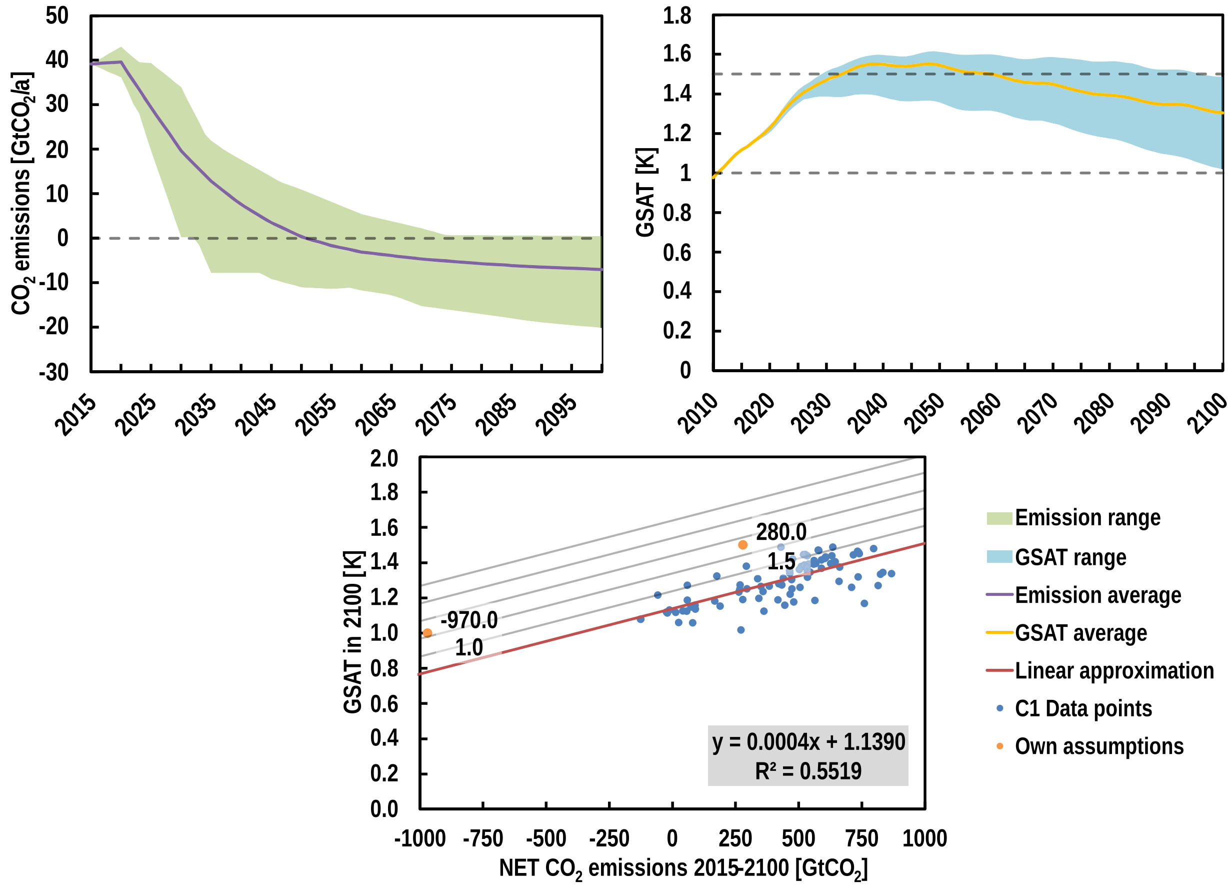

Fig. 12. Linear approximation of the relation between the GSAT in 2100 and the net CO_2 emissions in 2015 - 2100 (bottom) based on available data for CO_2 emission trajectories (top left) and resulting GSAT (top right) of 97 C1 scenarios (Byers et al., 2022). In this study for limiting GSAT to 1.0°C a CDR demand of 1750 $GtCO_2$ and for limiting GSAT to 1.5°C a CDR demand of 500 $GtCO_2$ is assumed. With estimated remaining emissions of about 780 $GtCO_2$ until CO_2 neutrality, net CO_2 emissions in 2015 - 2100 of 280 $GtCO_2$ and -970 $GtCO_2$ for 1.5°C and 1.0°C, respectively, are assumed (see Section 3.6).

global warming and atmospheric CO_2 concentration, i.e., the net CO_2 emitted, that is also suggested in literature (Rogelj et al., 2019).

As can be seen in Fig. 12, the CDR demand assumed in this study is rather conservative compared to the linear approximation of investigated C1 scenarios. For this study, higher TNE compared to the linear approximation of IAM scenarios are assumed, so that uncertainties, in form of offsets in the CO_2-temperature relation, regarding non-CO_2 emissions or the effect of aerosols are covered to some extent. The relationship between global warming and atmospheric CO_2 concentration shows a hysteresis effect as simulation results by Jeltsch-Thömmes et al. (Jeltsch-Thömmes et al., 2020) or Vakilifard et al. (Vakilifard et al., 2022) suggest. Also, high temperature overshoots must be avoided to lower the risk of long-term climate change effects that may be driven by meeting tipping points (Lenton et al., 2008, Lenton et al., 2019, Liu et al., 2023, Armstrong McKay et al., 1979) (cf. subSection 5.5) or by the climate systems characteristic response to addition and subsequent removal of CO_2 as studied by Koven et al. (Koven et al., 2023).

Moreover, the effects of aerosols reduction through deep electrification and thus avoided combustion and respective aerosols can increase peak warming further by approximately 0.5 K (Westervelt et al., 2015) or even more (Samset et al., 2018, Brasseur and Roeckner, 2005). Thus, the rather conservative CDR demand assumptions seem reasonable given the high uncertainties, in particular since most of the C1 level trajectories as shown in Fig. 12 still assume substantial combustion shares in the energy-industry-CDR system across all sections including BECCS. An additional CO_2-temperature effect of 0.1 K would induce a further CDR demand of about 250 $GtCO_2$. Therefore, it should be noted that CDR demand assumptions are sensitive to the assumption for defossilisation, electrification, and emission reduction.

The ambitious goals and CDR deployment require verification of comprehensive IAM analyses also applying highest shares of electrification such as in Luderer et al. (Luderer et al., 2021), so that ambitious CO_2 reduction scenarios can verify the actual effect of large-scale CDR combined with rapid defossilisation and the consequent phase-out of combustion processes as for instance projected in Bogdanov et al. (Bogdanov et al., 2021). This phase-out will also lead to a reduction of aerosols in the atmosphere eliminating a cooling effect (Brasseur and Roeckner, 2005). In addition, this study and the IAM scenarios can be extended by the resource demand for all applied technologies to verify possible further planetary boundaries important for a safe and just future (Rockström et al., 2023). The general uncertainty of projecting the future deployment potential of specific NETs should be emphasised, and further research is necessary to refine respective assumptions. The provided LUT-CDR spreadsheet tool can easily employ alternative assumptions on CDR measures, e.g., as given in the IPCC AR6 WGIII ([IPCC] 2022) (cf. supplementary material 1, note 11) or in the recent State of CDR report (Smith et al., 2023), and should encourage stakeholders as well as academia to create their own CDR portfolios.

In future research, the long-term durability, expressed in the present study via the security aspect of fixed CO_2 must be assessed more thoroughly for respective portfolios. For example, Chiquier et al. (Chiquier et al., 2022) and Dees et al. (Dees et al., 2023) already provide such an assessment for single NETs; however, a portfolio view will have to be provided in the future for a more thorough basis for decision-making. The same applies to a thorough assessment of environmental impacts of NETs in LCA, which were conducted for several options, especially

DAC (Madhu et al., 2021, Deutz and Bardow, 2021, Terlouw et al., 2021, Chauvy and Dubois, 2022, Terlouw et al., 2021). However, LCA analysis must be elaborated from the portfolio perspective and for the entire value chain in future works to understand the implications of large-scale CDR deployment for the environment.

The results of this study can be used to provide techno-economic parameters for NETs for CDR studies embedded in energy-industry-CDR system analyses. The parameters are provided in Table S21 and verified in Table S22-S23 in the supplementary material 1, note 12 and 13.

6. Conclusion

With the results presented in this work, the basis for further energy-industry-carbon dioxide removal system analyses is provided. The results can furthermore aid decision makers in paving the way to a safe climate aligning with the sustainable development goals. Based on a literature review, carbon dioxide removal technologies were investigated in detail, to examine techno-economic parameters, global potential, area demand, technology readiness level, and security of each carbon dioxide removal approach. Technologies were then combined to account for complete carbon dioxide removal technology chains, consisting of capture, transport, and sequestration steps. For these, the global potential, energy demand, area demand, technology readiness level, security, and the cost were projected until 2100. After examining the distinct carbon dioxide removal options, these were investigated and compared. A novel approach to develop carbon dioxide removal portfolios with a deployment projection considering different hypothetical societal preferences was employed.

The portfolios were created respecting technologies' maximum potential derived from literature. The portfolios were analysed for their future cost, energy demand, and land occupation until 2100. The results are then compared with the projected trajectory of the macro-economic LUT-DEES as a case study to put them into a broader perspective. Other scenarios can be chosen for comparison in the LUT-CDR spreadsheet tool provided. The structural results show a high sensitivity to the individual annual deployment potential of negative emission technologies considered. This sensitivity implies both the uncertainty and potential for further improvements. With input data in higher resolution for regional studies, multi-objective optimised portfolios can be created to be implemented in energy-industry-carbon dioxide removal studies. Increased spatial resolution would also enable the inclusion of further dimensions for portfolio creation. Most notably, various environmental impacts other than area demand are most prominent in today's discussion about carbon dioxide removal and should be considered for future studies to respect the Sustainable Development Goals.

The results indicate the enormous potential of direct air capture to contribute substantially to lowering the atmospheric CO_2 concentration to a safer level, including reducing the risk of triggering climate tipping points and respective management. While this result is enabled by virtually putting no constraints on the future installed capacity of this highly scalable technology, direct air capture generally performs well in many criteria considered. All portfolios examined (balanced, low-cost, low-energy, high-security, low-area, high technology readiness level) deploy direct air capture in combination with a variety of storage options that are well suited to the respective portfolio. In all portfolios, mineralisation (in-situ and ex-situ) is among the most attractive sequestration options for concentrated CO_2. This appeal is due to the great overall performance in a combination of key parameters such as cost, energy demand, and security. Desalination-based afforestation also covers major shares in all portfolios examined, indicating a future key role in the carbon dioxide removal sector. Enhanced weathering accounts for a major share mainly in a high-security CDR portfolio. Rainfall-based afforestation and bioenergy with carbon capture and sequestration, highly deployed options in many scenarios calculated with integrated assessment models, are not deployed in the balanced, high-security and low-area portfolio, indicating severe drawbacks in key parameters such as area demand and security.

With this work, the basis for further carbon dioxide removal portfolio studies is provided by investigating economic parameters as well as the energy demand of carbon dioxide removal technologies, the cost and energy demand of distinct and clearly described options, and the wider perspective of a diverse carbon dioxide removal portfolio deployment until 2100. The results of this research show, that the ambitious large-scale deployment of technologies to remove atmospheric CO_2 can be feasible. Removing 1750 $GtCO_2$ by 2100, which may limit global warming to 1.0°C if ambitious fossil phase-out can be achieved, may cost up to 2296 b€ annually if a low-cost portfolio is deployed. This is in the order of today's global military expenses and accounts for up to 0.42% of the projected global gross domestic product in a medium economic projection scenario, and represents 1.2% of the taxation ratio in the OECD. Portfolios prioritising security cost up to 2789 b€ annually or 0.56% of the global gross domestic product. Financing through carbon dioxide removal taxes seem feasible. The sensitivity of the cost results to the weighted average cost of capital shows a possible reduction of the integral total cost for CDR for the 1.0°C scenario from 2020 to 2100 of 7.0 - 14.5% for different portfolios if a weighted average cost of capital of 4% instead of 7% is assumed. Also, the additional energy demand seems manageable with the low-energy portfolio increasing the total primary energy demand by a maximum of 36.1 PWh/a (129.9 EJ/a). This results in additional primary energy demand of up to 12% compared to the projected total primary energy demand without any carbon dioxide removal. For a highly secure portfolio, 39.5 PWh/a (142.2 EJ/a) additional primary energy demand, or additional 13.3% total primary energy demand are required.

The results are discussed and compared with literature findings and further research options are identified. This study advances the discussion of carbon dioxide removal and negative emission technologies while pointing out its limitations, notably a strong impact of potential assumptions on the results and the incomprehension of important factors including the environmental impact of negative emission technologies. By investigating future pathways of energy-industry-carbon dioxide removal systems using the results of this research, carbon dioxide removal can become an integral part of future energy system discussions. While the future cost and energy demand as well as general impacts on the environment projected are substantial, past incompetence in adapting adequate transformational change create today's necessity of large-scale carbon dioxide removal deployment to enable a safe climate and just future. Carbon dioxide removal may be proposed to the wider public and policy makers as a required evolutionary step and improvement of 100% renewable energy transition for a sustainable future enabling society to curb the most severe effects of global warming and to maintain a safe, just, and liveable planet.

CRediT authorship contribution statement

Andreas Mühlbauer: Writing – review & editing, Writing – original draft, Visualization, Software, Methodology, Investigation. **Dominik Keiner:** Writing – review & editing, Validation, Methodology, Investigation, Conceptualization. **Christoph Gerhards:** Writing – review & editing, Methodology. **Upeksha Caldera:** Writing – review & editing, Methodology. **Michael Sterner:** Writing – review & editing, Methodology. **Christian Breyer:** Writing – review & editing, Validation, Supervision, Methodology, Conceptualization.

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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Supplementary materials

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Data availability

Data are available in the supplementary files and in the online repository.

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