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Techno-economic insights and deployment prospects of permanent carbon dioxide sequestration in solid carbonates†

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While a rapid defossilisation of the energy-industry system is at the highest priority for climate change mitigation, additional post-fossil carbon dioxide removal (CDR) for net-negative emissions will likely be necessary to ensure a safe future. An in-depth techno-economic analysis of differentiated sequestration options for carbon dioxide (CO_2) in solid carbonates is not yet available, as direct air capture-based mineralisation is usually aggregated in direct air capture and carbon sequestration. This research gap is closed by studying mineralisation as a key CDR option to sequester atmospheric CO_2 permanently, based on available literature. The most frequently discussed routes for mineralisation, *i.e.*, *in situ*, *ex situ* mineralisation, and enhanced rock weathering, are examined. The deployment potentials of these options are determined globally for nine major regions. Results indicate that costs for all mineralisation options can be kept below 100 € per tCO_2 from 2050. From 2030 onwards, *in situ* mineralisation, with low energy-intensity, can be realised at cost of $\leq$131 € per tCO_2, *ex situ* mineralisation at $\leq$189 € per tCO_2, and enhanced weathering at $\leq$88 € per tCO_2. Final energy demand for CO_2 sequestration *via in situ* mineralisation is $\leq$1.8 MWh per tCO_2, *via ex situ* mineralisation $\leq$3.7 MWh per tCO_2, and *via* enhanced weathering $\leq$1.1 MWh per tCO_2 from 2030. Large-scale deployment of mineralisation options supporting 60% of projected CDR demand is assessed to require up to 0.06% and 0.21% in global gross domestic product and up to 2.5% and 8.6% additional primary energy demand in 2070 for a 1.5 °C and 1.0 °C climate target, respectively. Implications, permanence of sequestration, and limitations are discussed, and a research outlook is provided.

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Broader context

The fight against climate change requires different actions. Beneath the adaption to climate change, mitigating climate change through the deployment of renewable energy is of upmost importance. However, even for the 1.5 °C climate target, an entirely renewable energy system is not sufficient anymore. An expected 500 $GtCO_2$ must be removed to balance delayed action in climate change mitigation. In addition, climate restoration may be required to reach safe planetary boundaries at 350 ppm CO_2 in the atmosphere, or a 1.0 °C climate target. This requires an expected 1750 $GtCO_2$ to be removed from the atmosphere within the 21st century. For such large amounts of CO_2 to be sequestered, many factors play a role in assessing suitable options for carbon sequestration, such as energy demand, costs, area demand, technology readiness level, or permanence. The latter is an important point of discussion for gaseous or geological sequestration of CO_2 commonly associated with direct air carbon capture and sequestration. However, possible leakage, earthquakes, or well failures increase the risk of large-scale geological sequestration. This draws light on carbon mineralisation, where CO_2 is fixed in solid carbonates not to be released for at least thousands of years.

1. Introduction

The rising atmospheric greenhouse gas concentration leads to increasing global warming, putting human civilisation at risk.[1] The trade-offs of intensive fossil fuel combustion enabling the industrial revolution and further economic growth[2] are becoming increasingly severe with rising mean air temperatures and more frequent extreme weather events.[3] To re-balance the climate system and to limit global warming to a sustainable level, rapid defossilisation and electrification of all industry sectors are necessary.[4,5] However, the latest findings of the Intergovernmental Panel on Climate Change (IPCC) imply the requirement for negative carbon dioxide (CO_2) emissions through carbon dioxide removal (CDR) employing negative emission technologies (NET) and natural climate solutions

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(NCS).[6] While CDR can reduce CO_2 levels in the atmosphere, the management of other greenhouse gases such as methane or nitrogen dioxide is subject to current research.[7] CDR is partially required to offset residual emissions; however, the definition of residual emissions is subject to vested interests and often debatable.[8] Post-fossil CDR is also essential to achieve a net-negative energy-industry system.[4,9] To limit global warming to 1.5 °C by 2100, total negative emissions (TNE) of about 500 $GtCO_2$ from the atmosphere must be realised.[10,11] To reach even more ambitious yet safe and just targets of limiting global warming to 1.0 °C, TNE of up to 1750 $GtCO_2$ may be required.[10–12] The 1.5 °C and 1.0 °C climate targets will require annual CO_2 removal rates of up to 10 $GtCO_2$ per a and 40 $GtCO_2$ per a, respectively, when fully ramped if the climate shall be rebalanced in a timely manner.[10–12] The ambitious climate target of limiting global warming to 1.0 °C might be significantly more energy- and cost-intensive; however, it can help to achieve a sustainable and just future for civilisation[12,13] by avoiding major tipping points and potential cascades thereof.[14–16] An unprecedented reduction in cost of renewable electricity, which is currently observed, can enable such ambitious climate targets to return to the Holocene.[17] It is expected that the mineralisation of CO_2 can play a major role, with an expected potential of up to 10 $GtCO_2$ per a by 2050, sequestering atmospheric CO_2 safely for geological time scales.[18,19] Sandalow *et al.*[19] projected that, given adequate political incentives and measures, about 1 $GtCO_2$ per a and 10 $GtCO_2$ per a can be mineralised by 2035 and 2050, respectively. However, most integrated assessment models (IAM) that are considered as scientific basis for IPCC reports include either no mineralisation option or only enhanced weathering (EW) in their studies, making these options underrepresented in current climate change mitigation research.[11]

NCS can sequester atmospheric CO_2 and have co-benefits on the environment, but the long-term effectiveness and storage duration as well as sustainable scalability are potential bottlenecks.[20,21] The CDR potential of NCS has been discussed intensively in literature.[9,22] NETs including direct air carbon capture and sequestration (DACCS),[23–25] bioenergy with carbon capture and sequestration (BECCS),[26–29] EW,[30–33] or biochar production[34] must be considered and potentially deployed on large-scale,[11,35] though research has indicated that diverse portfolios of CDR are preferable.[11,36,37] Bio-geo-chemical options such as EW, biochar production, or afforestation combine the CO_2 capture and storage step by sequestering atmospheric CO_2 in carbonates or biogenic materials.[11,31,38] In contrast, DACCS and BECCS are realised *via* concentrated CO_2 that can either be used as feedstock for e-fuels and e-chemicals to defossilise hard-to-abate sectors[39–43] or can be safely sequestered as negative emissions.[11,44] While most IAMs as of today model DACCS and BECCS without further specifying CO_2 sequestration modalities,[11] the production of carbon-

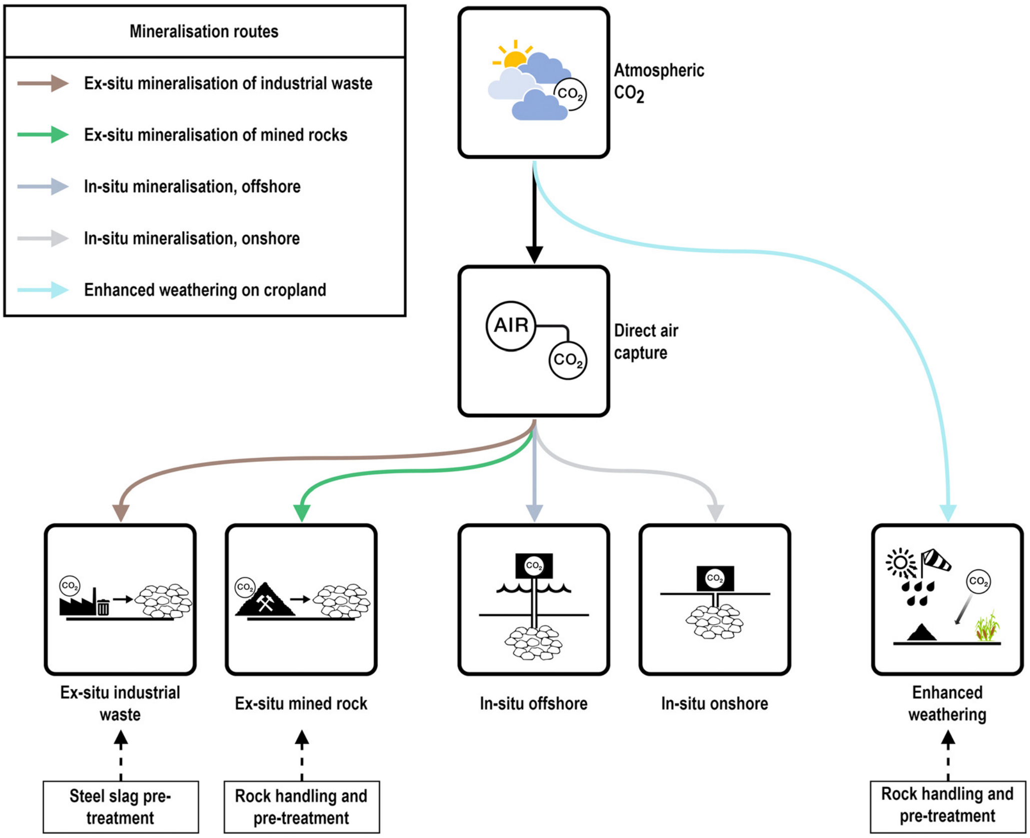

Fig. 1 Schematic overview on CO_2 mineralisation options adapted from Mühlbauer *et al.*[11]

bearing and electricity-based solid materials is introduced as alternative option.[45–49] The anticipated low-cost renewable electricity[5,50] can be utilised to enable novel NETs with relatively high energy demand following the overall trend towards a Power-to-X Economy[51] and higher sequestration security compared to geological underground CO_2 sequestration.[11,52] Solid carbon-bearing carbonates can be produced using *ex situ* mineralisation (MIN_{EX})[53,54] while simultaneously acting as valuable building material.[55] Also, CO_2 can be sequestered safely in suitable underground formations allowing for the *in situ* mineralisation (MIN_{IN}) of CO_2 to form solid carbonates within a few years.[44,56,57] MIN_{IN} can help mitigate concerns and potential flaws of other underground sequestration options such as leakage through previously impermeable caprocks[58,59] or in deep ocean storage options.[60] The sequestration of atmospheric CO_2 in stable solid materials can enable permanent CDR[21] and ensure effective negative emissions to aim for more ambitious climate targets.[20] CO_2 mineralisation may be a key technology to achieve such effective long-term stable atmospheric CO_2 sequestration.

An overview of the most frequently discussed CO_2 mineralisation options, *i.e.*, EW, MIN_{EX}, and MIN_{IN} is depicted in Fig. 1. While MIN_{EX} and EW can both use suitable mafic and ultramafic rock from open pit mines to produce solid carbonates as a main product, MIN_{IN} dissolves CO_2 in water to inject it into suitable deep formations of basaltic rocks or peridotites.[44] MIN_{EX} can also use industrial solid wastes bearing magnesium (Mg) or calcium (Ca).[61] While this is also reported for EW,[62] this option is not considered within this work due to possible sustainability bottlenecks when applying waste material to large open areas.

The aim of this study is to provide a techno-economic assessment of options to sequester atmospheric CO_2 in minerals by different processes, closing research gaps of lacking literature on comprehensive CO_2 mineralisation options and a dedicated techno-economic assessment of respective NETs. A novelty of this study is the assessment of global-local potentials as well as economics of CO_2 mineralisation to provide the basis for future research in the energy-industry-CDR nexus.[4] Therefore, the novelties of this study include:

- Global-local potentials for CO_2 mineralisation enabling dedicated energy-industry-CDR system transition studies considering different CO_2 mineralisation options.
- Techno-economic parameters for CO_2 mineralisation options for further use in future energy-industry-CDR studies, including final energy demand and primary energy demand per tonne of sequestered CO_2.
- Technology readiness level (TRL) of all mineralisation options to assess maturity.
- Global implications for the cost and primary energy demand of large-scale CO_2 sequestration with major shares of mineralisation.

By providing a novel basis for further research in implementing CO_2 mineralisation in energy-industry-CDR system transition simulations, this study aims to support the discussion about CO_2 mineralisation options for permanent CDR. Early investigations of future cost, energy demand, and global regional potential are required to pave a way to a safe future. This study aims to provide a step towards in-depth understanding of CO_2 mineralisation in the context of energy-industry-CDR systems to simulate pathways to such respective safe futures.

2. Literature review

Geochemical NETs have been comprehensively reviewed[19,63–67] and current research gaps, *i.e.*, the identification of regional potentials or advanced understanding of biological influences on CO_2 mineralisation rates and other performance factors, have been identified.[57,66] Geochemical NETs have also recently been studied on a regional level as for the case of Spain,[68] South Africa,[69] the United Kingdom,[70–72] Austria,[73] or Japan.[74] Also, field trials have been carried out[75] to verify modelled CDR rates.[76]

Minerals containing alkaline-earth metal oxides such as calcium oxide (CaO) or magnesium oxide (MgO) react with CO_2 in an exothermic mineralisation reaction.[77] The natural weathering of rocks, induced by intense tectonism, is a significant part of the Earth's carbon cycle,[78] removing about 300 MtC per a from the atmosphere.[19,79] Rock types containing significant shares of suitable metal oxides include olivine, brucite, pyroxene, serpentine, wollastonite, and dunite among others.[64] Also, certain industrial wastes such as steelmaking slag can be used in MIN_{EX} to obtain useful products.[80,81] Such dual use enables CO_2 capture, utilisation, and sequestration (CCUS) *via* mineralisation[82–84] or the production of renewable electricity-based and carbon-bearing solid materials.[45–47] As an example, Pan *et al.*[85] elaborate on the cases of electric arc furnace steelmaking, which also occur in a defossilised industry, and Portland cement manufacturing for a waste-to-resource supply chain by producing useful construction material from steelmaking slags.

The subsurface reaction of CO_2 with suitable rock to carbonates is usually referred to as *in situ* mineralisation (MIN_{IN}) of CO_2[19,44,63,64,86] and is one out of four possible trapping mechanisms for CO_2 underground sequestration.[60] Suitable sequestration sites are basaltic rock formations or peridotites.[57] Rapid mineralisation removes the need for a long-term stable caprock.[44,87,88] MIN_{EX} refers to engineered processes to mineralise CO_2 with suitable feedstock in reactors as first proposed by Lackner *et al.*[53] as early as 1995.[86,89] The reverse reaction of exothermal mineralisation, *i.e.*, calcination, is favoured at high temperatures (>900 °C for $CaCO_3$ and >300 °C for $MgCO_3$ at 1 bar CO_2 partial pressure), whereas mineralisation is favoured at relatively low temperatures.[90] Various reactor setups and the impact of different process parameters such as retention time have been studied[54,91] and lab-scale demonstration reactors are being operated.[92] MIN_{EX} can be characterised either as direct or indirect mineralisation.[77,90] Both direct and indirect routes can be conducted in an aqueous or gas–solid environment,[77,90] whereas, in indirect mineralisation, the reactive oxide (of Mg or Ca) or hydroxide is extracted from the feedstock.[93–95]

Enhancing the natural surface weathering of rocks is commonly referred to as EW.[31–33] Strefler *et al.*[32] investigated in detail the effect of the weathering efficiency of three different rock types, the influence of temperature and soil pH, and the optimal grain size of rocks for a study of the global CDR potential and the cost of EW in high spatial resolution. Best suited locations are therein characterised as warm and humid,[32] which was also confirmed in field experiments.[96] Beerling *et al.*[31] employ a one-dimensional vertical reactive transport model for basalt weathering with a steady-state flow to conduct their techno-economic and potential assessment for EW on a global scale in 2050. In contrast to Strefler *et al.*,[32] Beerling *et al.*[31] account for soil pH and varying grain size by considering a log-normal grain size distribution. Also, Beerling *et al.*[31] apply the fractal dimension to account for uncertainties in grain topography and porosity and consider annual rock application over a 10-year time horizon. Goll *et al.*[33] studied EW and the implications of fertility enhancement by basalt application to global hinterland. Enhancing the soil fertility by basalt application can improve the ecosystem's carbon uptake, further increasing the CDR potential.[33] Cipolla *et al.*[97] study the impact of rainfall, vegetation, and soil type on the efficacy of EW of olivine, calculated with a formerly introduced model[98,99] for three case studies. Results indicate a major impact from annual rainfall distribution and the authors concluded that irrigation can substantially increase weathering rates.[97] The grain size of the applied rock also significantly impacts the EW rate.[73] Eufrasio *et al.*[100] conducted a thorough life cycle assessment (LCA) of EW and found that, in order to maintain a high carbon efficiency, renewable electricity is required to satisfy the comminution electricity demand. Eufrasio *et al.*[100] also build on the results provided by Beerling *et al.*[31] and compare EW's impact in terms of energy demand, land requirement, and water requirement to other NET options. Vakilifard *et al.*[101] studied the impact of EW, modelled by the approach introduced by Beerling *et al.*,[31] co-deployment on the Earth system. The authors conclude that the additional CDR provided can increase of the likelihood of limiting global warming in 2100 to 1.5 °C and the ocean alkalinity to benefit marine ecosystems.[101] EW using different suitable industrial waste as feedstock is studied frequently.[31,62,102,103] Further literature findings on the cost and energy demand for EW are aggregated in Note 4 in the Supplementary material 1 (ESI†).

As stated by Sandalow *et al.*,[19] the global potential for different CO_2 mineralisation options in high spatial resolution is a major research gap that must be addressed. Additionally, Wei *et al.*[104] find that current assessments should be harmonised using a hierarchical framework that they proposed. The global potential of mine tailings for CDR was studied by Bullock *et al.*[105] on a global to regional scale. Kremer *et al.*[106] mapped and categorised potential input material for CO_2 mineralisation in Europe; however, comprehensive data in high spatial resolution on availability and accessibility of sequestration sites is still largely missing.[19,57] This study aims to close the abovementioned research gaps by providing a comprehensive techno-economic overview on CO_2 mineralisation options and respective global-local sequestration potentials.

3. Methods and data

Within this study, techno-economic parameters for different CO_2 mineralisation options are elaborated to provide the basis for future energy-industry-CDR system analyses. The following sections are structured as follows. In subsection 3.1, assumptions for process configurations are described. In subsection 3.2, the assessed processes of this study are further specified, followed by a global-local potential investigation for these processes in subsection 3.3. Techno-economic data are aggregated from various sources in literature and presented in Table 1 for traceability. Considering the state of deployment of mineralisation options, those techno-economic assumptions are subject to uncertainty. All costs are corrected for inflation to the year 2020 and cost that are given in USD are adjusted to € with a long-term exchange rate of 1.2 USD per € applied.

3.1. Mineralisation routes

Carbonates, the final products of CO_2 mineralisation, are highly stable solid products showing low Gibb's free enthalpy compared to CO_2 and is, therefore, expected to remain stable and sequestering atmospheric CO_2 safely.[110] The low Gibb's free enthalpy of carbonates compared to gaseous CO_2 is reflected in the high thermal inertia and stability of carbonates and results in reported CO_2 sequestration times of geological timescales.[44] Further fundamentals of the mineralisation reaction are elaborated in Note 1 in the Supplementary material 1 (ESI†).

3.1.1. *In situ* mineralisation. At the CarbFix and CarbFix2 site in Iceland, MIN_{IN} is operated successfully for one decade already[111] in combination with a geothermal powerplant.[112] Kali *et al.*[87] studied the underground sequestration of CO_2 in different sites such as in subsurface basalt formations suitable for MIN_{IN}. While an abundant global theoretical sequestration potential in basalt is expected, a major bottleneck of MIN_{IN} is the extensive water demand, making MIN_{IN} about twice as expensive as other geological sequestration options.[87] Dissolving CO_2 in water prior to injection is crucial to enhance the reaction kinetics to rapidly mineralise the CO_2 for safe sequestration.[44] Injected water can partially be circulated,[113] as shown at the CarbFix site's geothermal powerplant.[114] The substitution of freshwater with seawater for underground injection of CO_2 is currently studied[115] and mineralisation of CO_2 in underground basaltic rock formations at mineralisation rates of up to 80% and 95% within one and two years, respectively,[44] ensures safe long-term sequestration of atmospheric CO_2 in solid carbonates underground.[113] For MIN_{IN}, supercritical CO_2 or CO_2 dissolved in water is injected to suitable sequestration sites with highly permeable and porose rocks at depths of 1000–2500 m.[87] The average lifetime of injection projects is about 20 years.[87] The overview of large igneous provinces (LIP) and

Table 1 Initial techno-economic parameters for technologies and processes for CO_2 mineralisation. All numbers taken from references were recalculated to 2020 values in €. Electricity input represents the electricity demand for driving processes involved and heat input is thermal energy used in reaction reactors. Further information can be found in the Notes 2–4 of Supplementary material 1 and in the Supplementary material 2 (ESI)

Technology	Parameter	Unit	Value	Ref.
Open-pit mining	CAPEX	€ per (tOre/a)	6.0	31
	OPEXfix	€ per tOre	4.6	
	OPEXvar	€ per tOre	0.0	
	Lifetime	years	10	
	Electricity input	kWh_{el} per tOre	5.2	32,89
	Heat input	kWh_{th} per tOre	0.0	
	Heat output	kWh_{th} per tOre	0.0	
Rock transportation	Cost	€ per (tRock·100 km)	4.4	32
	Lifetime	Years	—	
	Electricity input	kWh_{el} per (tOre·100 km)	1.4	
	Heat input	kWh_{th} per tOre	0.0	
	Heat output	kWh_{th} per tOre	0.0	
Rock comminution	CAPEX	€ per (tOre/a)	—	
	OPEXfix	€ per tOre	—	
	OPEXvar	€ per tOre	—	
	Lifetime	Years	—	
	Electricity input	kWh_{el} per tOre	127.8	
	Heat input	kWh_{th} per tOre	0.0	
	Heat output	kWh_{th} per tOre	0.0	
Rock spreading	CAPEX	€ per (tCO_2/a)	208.4	
	OPEXfix	€ per tCO_2	—	
	OPEXvar[a]	€ per tCO_2	13.8	
	Lifetime	Years	50	Own assumption
	Electricity input[b]	kWh_{el} per tCO_2	0.0	
	Heat input	kWh_{th} per tCO_2	0.0	
	Heat output	kWh_{th} per tCO_2	0.0	
Direct aqueous mineralization – serpentine	CAPEX	€ per (tCO_2/a)	208.4	55,107
	OPEXfix	€ per tCO_2	6.3	
	OPEXvar[a]	€ per tCO_2	13.8	
	Lifetime	Years	50	89
	Electricity input	kWh_{el} per tCO_2	455.0	108
	Heat input	kWh_{th} per tCO_2	452.0	
	Heat output	kWh_{th} per tCO_2	0.0	
Direct aqueous mineralization – olivine	CAPEX	€ per (tCO_2/a)	208.4	55,107
	OPEXfix	€ per tCO_2	6.3	
	OPEXvar[a]	€ per tCO_2	13.8	
	Lifetime	years	50	89
	Electricity input	kWh_{el} per tCO_2	689.0	108
	Heat input	kWh_{th} per tCO_2	103.0	
	Heat output	kWh_{th} per tCO_2	0.0	
Direct aqueous mineralization – steel slag	CAPEX	€ per (tCO_2/a)	208.4	55,107
	OPEXfix	€ per tCO_2	6.3	
	OPEXvar	€ per tCO_2	13.8	
	Lifetime	Years	50	89
	Electricity input	kWh_{el} per tCO_2	592.0	108
	Heat input	kWh_{th} per tCO_2	407.0	
	Heat output	kWh_{th} per tCO_2	0.0	
CO_2 underground injection – onshore	CAPEX	€ per (tCO_2/a)	35.0	109
	OPEXfix	€ per tCO_2	1.5	
	OPEXvar	€ per tCO_2	0.0	
	Lifetime	Years	40	
	Electricity input	kWh_{el} per tCO_2	70.0	44
	Heat input	kWh_{th} per tCO_2	0.0	
	Heat output	kWh_{th} per tCO_2	0.0	
CO_2 underground injection – offshore	CAPEX	€ per (tCO_2/a)	99.5	109
	OPEXfix	€ per tCO_2	3.5	
	OPEXvar	€ per tCO_2	0.0	
	Lifetime	Years	40	
	Electricity input	kWh_{el} per tCO_2	70.0	44
	Heat input	kWh_{th} per tCO_2	0.0	
	Heat output	kWh_{th} per tCO_2	0.0	

[a] $Opex_{var}$ represents the cost including energy of feedstock preparation normalised to 1 tonne CO_2 sequestered. [b] The electricity demand for rock spreading is assumed to be already included in rock transportation.

flood basalt shown in Fig. 2 implies that potential for MIN_{IN} is available over the globe with potentially accessible sequestration sites characterised by mafic or ultramafic basalt located on all continents.[44] LIP and flood basalt were built by

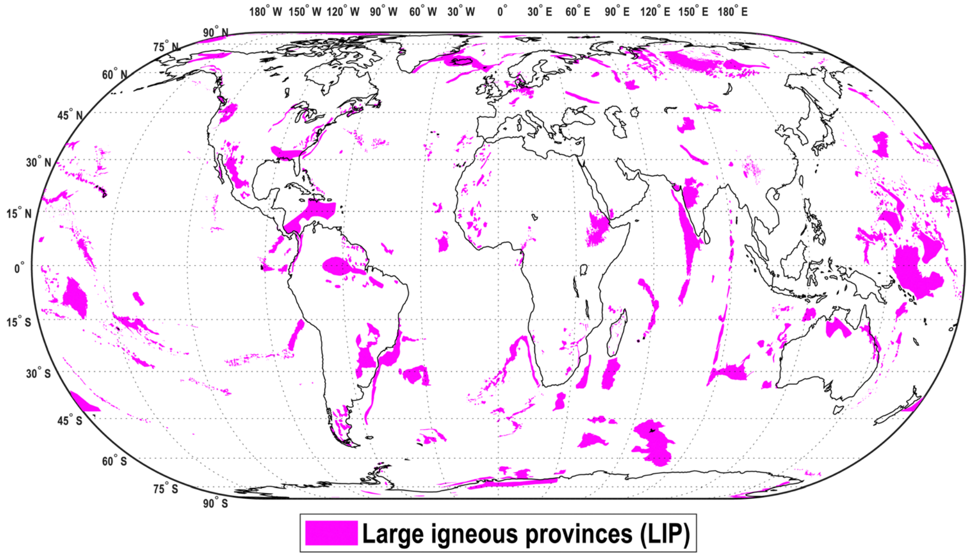

Fig. 2 Global overview on large igneous provinces (LIP) and flood basalt showing potential MIN_{IN} sequestration sites[44] which are characterised by mafic and ultramafic basalt.[44,57] Data source.[117]

volcanic magma in a rather short period of time for geologic time scales and are rich in required minerals for CO_2 mineralisation.[116] This study does not consider MIN_{IN} of the CO_2 brine layer.

The CO_2 mineralisation reaction of Wollastonite (calcium silicate, $CaSiO_3$) or Enstatite (magnesium silicate, $MgSiO_3$) present in ultramafic rocks to magnesium carbonate ($MgCO_3$) or calcium carbonate ($CaCO_3$) and silicon dioxide (SiO_2) can be summarised by eqn (1):[67]

$$(\mathrm{Mg,Ca})\mathrm{SiO_3(s)} + \mathrm{CO_2} \rightarrow (\mathrm{Mg,Ca})\mathrm{CO_3(s)} + \mathrm{SiO_2(s)} \quad (1)$$

For the mineralisation reaction, gaseous CO_2 is generally dissolved in water. Further information on the MIN_{IN} process is available in Supplementary material 1, Note 1 (ESI†).

3.1.2. *Ex situ* mineralisation.

MIN_{EX} can be conducted in direct or indirect ways. Indirect MIN_{EX} pathways extract reactive material from feedstock in a separate step prior to carbonation. In this study, the direct aqueous carbonation reactor design as parametrised by Ostovari *et al.*[89] is considered. No indirect mineralisation options are considered in this study due to the lower carbon efficiency compared to direct routes when cement substitution is not accredited.[89] Ostovari *et al.*[89] consider a continuously stirred tank reactor for olivine, serpentine, and steel slag. The pre-treatment differs between feedstocks: olivine is grinded and milled; serpentine is grinded before magnetite is magnetically separated, with the remaining feedstock being thermally treated;[89] and steel slag only needs grinding.[89] All feedstock is mixed with water and additives before the carbonation in a rotary packed bed reactor and is de-watered in a subsequent post-processing step.[89] Dri *et al.*[118] noted that even though gross freshwater demand for aqueous MIN_{EX} is significant, about 99.96% can be reused, limiting the net freshwater demand of MIN_{EX} to about 0.08 tH_2O per tCO_2. Rock pretreatment processes can be mechanical (crushing, grinding, milling), thermal or mechanochemical.[119] Main process parameters for MIN_{EX} are the feedstock's cation content, the pretreatment (grain diameter, temperature), operating pressure, temperature, potential additives, and feedstock residence time in the reactor.[93] Process temperatures generally range from up to 200 °C for aqueous processes to up to 700 °C for rock pretreatment.[119] All heat demand for MIN_{EX} in this study is considered as high-temperature heat that cannot be provided through heat pumps. However, aqueous MIN_{EX} using pretreated feedstock is feasible at process parameters of 100–180 °C and 100–160 bar, possibly enabling the use of high-temperature heat pumps.[120] Optimal carbonation conditions as proposed by Gerdemann *et al.*[121] are 185 °C and 150 bar CO_2 partial pressure for olivine, 100 °C and 40 bar CO_2 partial pressure for wollastonite, and 155 °C and 115 bar CO_2 partial pressure for serpentine. Such as conducted by Ostovari *et al.*,[108] the life cycle inventory data in this study for the above mentioned processes are adapted with operational pressures of 100 bar, 115 bar, and 20 bar for MIN_{EX} of olivine, serpentine, and steel slag, respectively.

The reaction of mined rocks is similar to eqn (1). The mineralisation of industrial waste, more specifically the containing minerals magnesium oxide (MgO), calcium oxide (CaO), magnesium hydroxide ($Mg(OH)_2$), or calcium hydroxide ($Ca(OH)_2$) to $MgCO_3$ or $CaCO_3$ and water (H_2O) is described by eqn (2) and (3):[67]

$$(\mathrm{Mg,Ca})\mathrm{O(s)} + \mathrm{CO_2} \rightarrow (\mathrm{Mg,Ca})\mathrm{CO_3(s)} \quad (2)$$

$$(\mathrm{Mg,Ca})(\mathrm{OH})_2\mathrm{(s)} + \mathrm{CO_2} \rightarrow (\mathrm{Mg,Ca})\mathrm{CO_3(s)} + \mathrm{H_2O}(l) \quad (3)$$

Gaseous CO_2 may be in aqueous solution when reacting to solid carbonates, however, dry reactions are also possible, thus the state is not further specified in eqn (2) and (3).The ranges of metal oxide share in different rock types and industrial waste vary by a multitude of parameters and influence both the total weathering potential (WP) and reaction kinetics.[63] Metal oxides contributing to mineralisation of industrial waste also encompass Al, Fe, Na, and K.[122] The WPs of different rock types have been reviewed in several studies and elaborated in experiments.[123] The dissolution rate of rock feedstock is crucial to understand reaction kinetics and research in creating comprehensive databases is advancing.[124] The energy and material demand for the respective $\mathrm{MIN_{EX}}$ configurations are also adapted from Ostovari *et al.*[89,108] (*cf.* Note 3 in the Supplementary material 1 and in the Supplementary material 2, ESI†).

3.1.3. Enhanced weathering. Within this study, EW is considered with rock handling, which includes mining, comminution, and transportation as well as the spreading of rock on suitable land. In this study, potentials are only assessed for cropland as it is actively managed and rock can be applied with existing machinery[32] but other types of land such as hinterland were previously assessed to show promising potentials.[33] Section 3.1.4 further elaborates on comminution. The crushed rock is applied mainly to agricultural land to enhance the natural CO_2 uptake of calcium- and magnesium-rich silicate rocks.[31] Rock spreading in this study is largely parametrised according to Strefler *et al.*,[32] while Beerling *et al.*[31] also provided an effective approach to capture the cost of EW.

For EW, as Strefler *et al.*[32] describe, the annual CDR rate for a specific land area $r_{\mathrm{CO2}}^{\mathrm{CDR}}$ is determined by the total amount of rock spread m, the dissolution rate d, and the CO_2 sequestration potential p of the applied rock type. The dissolution rate d is depending on the specific surface area a, the weathering rate w, and the molar mass M. The constant ϑ is used to investigate annual weathering using a conversion rate of 3.155×10^7 s a^{-1}. The grain size x is applied in μm. Therefore, the amount of CO_2 removed annually is calculated as described in eqn (4)–(6) as proposed by Strefler *et al.*[32] All input parameters assumed for further calculations can be found in Supplementary material 2 (ESI†).

$$r_{\mathrm{CO2}}^{\mathrm{CDR}} = m \cdot d(x) \cdot p \tag{4}$$

$$d(x) = a(x) \cdot w \cdot M \cdot \vartheta \tag{5}$$

$$a(x) = 69.18 \cdot x^{-1.24} \tag{6}$$

This study relies on geochemical modelling shown in previous studies[31,32] and does not largely focus on physical processes of EW. Instead, this study focuses on the overall energy demand and economic parameters derived from previous studies.

3.1.4. Mined rock feedstock preparation. Derived from the formulas given by Strefler *et al.*[32] the electricity demand per tonne of CO_2 removed *via* EW through basalt annually is plotted in Fig. 3. Based on the share of rock weathered per year at 20 °C in relation to the grain size, the electricity demand for grinding rock to the respective grain size from an initial grain size of 100 μm is calculated. Strefler *et al.*[32] assume that 1 $\mathrm{kgCO_2}$ can be captured and sequestered by 0.8 kg dunite or 3.3 kg basalt.

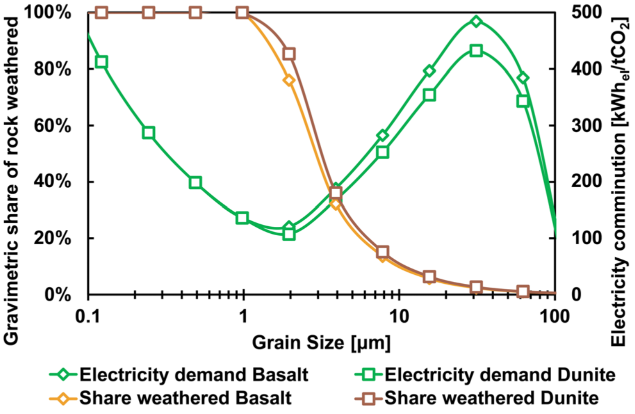

Fig. 3 The share of rock weathered (left vertical axis) after one year at 20 °C based on formulas given by Strefler *et al.*[32] and the resulting electricity demand for comminution per tonne of CO_2 removed (right vertical axis) based on different grain size for Basalt and Dunite.

As can be seen in Fig. 3, the electricity demand for comminution is optimal at a grain size of about 2 μm from a feed size of 100 μm at an electricity demand of about 120 $\mathrm{kWh_{el}}$ per $\mathrm{tCO_2}$ (*cf.* Supplementary material 2 for details, ESI†). At this grain size, about 80% of the rock particles are weathered within one year minimising the specific electricity demand for rock comminution relative to the mass of CO_2 mineralised. However, a grain size below 10 μm is anticipated to potentially bear health risks for humans.[32] A grain size of ground feedstock material of 10 μm is assumed in this study corresponding to an electricity demand of 127.8 $\mathrm{kWh_{el}}$ per tRock (*cf.* Note 3 in the Supplementary material 1, ESI†) which would correspond to an electricity demand of about 322 kWh per $\mathrm{tCO_2}$ and 288 kWh per $\mathrm{tCO_2}$ when using basalt and dunite, respectively. This assumption is made for EW as well as for $\mathrm{MIN_{EX}}$ feedstock preparation. For rock mining and crushing, an electricity demand of 50 $\mathrm{kWh_{el}}$ per tRock is assumed (*cf.* Note 3 in the Supplementary material 1, ESI†). As rock mining and crushing does not affect the final grain size and, therefore, the rate of weathering, the electricity demand is equivalent to 165 kWh per $\mathrm{tCO_2}$ and 40 kWh per $\mathrm{tCO_2}$ when using basalt and dunite, respectively, when assuming the weathering efficiencies provided by Strefler *et al.*[32]

3.2. Techno-economics and process specifications

In this subsection, all techno-economic input data and assumed process configurations, which are also displayed in schematics shown in Fig. 1, are presented. Within this work all energy- and mass balances and cost are normalised to the functional unit of 1 $\mathrm{tCO_2}$ permanently removed and sequestered.

In this study, it is assumed that industrial solid waste suitable for mineralisation can be acquired for no additional cost and without additional energy demand, *i.e.*, it is available as waste product that is otherwise disposed. For DAC, techno-economic specifications from Fasihi *et al.*[23] and as adapted in Mühlbauer *et al.*[11] are assumed. Details are elaborated in the

Supplementary material 1 (ESI†). Rock crushing, grinding, and comminution is estimated to be 8.3 € per t (10 USD per t) for full life-cycle cost including energy.[125] Within this study, the estimation by Strefler *et al.*[32] for electricity demand of 127.8 kWh t^{-1} is applied which leads to cost of 5.3 € per t in 2020. Technology learning is a well-established approach to quantify the future cost reduction of technologies.[107,126,127] While rock comminution, rock mining and handling, as well as CO_2 injection are assumed to have no further substantial learning due to the widespread maturity and high historic installed capacity, capital expenditure (CAPEX) learning of MIN_{EX} is assumed based on estimates by Faber *et al.*[107] Thus, CAPEX reduction calculated with a learning rate of 10.55% and a deployment projection reaching 3.46 $GtCO_2$ per a cumulative installed capacity for MIN_{EX} is assumed (*cf.* Supplementary material 2, ESI†). Cost reductions for DAC are adapted from Mühlbauer *et al.*[11] which are based on Fasihi *et al.*[23]

The initial techno-economic input parameters used in this study are listed in Table 1. Assumptions regarding the future cost development are elaborated in more detail in the Notes 2–4 of Supplementary material 1 and in the Supplementary material 2 (ESI†).

CO_2 mineralisation generally produces solid carbonates and other by-products. The potential of these by-products in the cement industry has been investigated in several studies.[55,128–130] The energy- and CO_2-intensive cement production is challenging to decarbonise,[131] therefore, synergies with CO_2 mineralisation can be a valuable option. For example, substituting up to 5% of Portland cement in mortars with feed and by-products of mineral carbonation was found to maintain compressive strength while reducing CO_2 emissions and cost of waste disposal.[128] Also, other industries such as paper or rubber production have been identified as potential customers for by-products of CO_2 mineralisation.[83]

All process chains presented within this study are evaluated regarding their final energy (FE) demand and levelised cost of CDR. The FE demand of each process chain for CO_2 mineralisation $E_{FE,NET}$ is calculated according to eqn (7).

$$E_{\mathrm{FE,NET}} = \frac{\sum_p E_{\mathrm{FE},p} \cdot m_{\mathrm{out},p}}{1\mathrm{tCO_2\ removed}} \tag{7}$$

Wherein $E_{FE,p}$ is the final energy demand in MWh per tCO_2 or MWh per tRock for rock comminution and transportation of each process step p and $m_{out,p}$ is the output mass of CO_2 or rock of each process p that is reacted for sequestering 1 tCO_2 in carbonates. Future improvements in the energy demand of CO_2 mineralisation processes are omitted. Low temperature heat demand is converted to final energy in the form of electricity assuming a coefficient of performance of the heat pump of 2.16, 2.48, and 2.80 in 2030, 2040, and 2050, respectively.[11] Heat at higher temperature levels is assumed to be provided by direct electric heating with an efficiency of 100%. The primary energy (PE) demand of each process chain $E_{PE,NET}$ is further calculated using eqn (8):

$$E_{\mathrm{PE,NET}} = \frac{E_{\mathrm{FE,NET}}}{f_{\mathrm{PE}}} \tag{8}$$

wherein f_{PE} is a PE factor that includes curtailment, intermediate battery storage with associated losses, and transmission losses and is set to 90.41%, 93.57%, and 93.58% in 2030, 2040, and 2050, respectively, as applied by Mühlbauer *et al.*[11] based on LUT-DEMAND.[10,132]

For each CO_2 mineralisation process chain, the levelised cost of CDR LCOCDR is calculated according to eqn (9).

$$\mathrm{LCOCDR_{NET}} = \sum_p \left[\mathrm{LCOP}_{p,\mathrm{co}} + H_{\mathrm{LT},p} \cdot \mathrm{LCOH_{LT}} + \left(E_{\mathrm{FE},p} + H_{\mathrm{HT},p}\right) \cdot \mathrm{LCOE}\right] \cdot \frac{m_{\mathrm{out},p}}{1\mathrm{tCO_2\ removed}} \tag{9}$$

wherein $LCOP_{p,co}$ is the levelised cost of product based on capex and opex only of each process in the process chain, $H_{LT,p}$ is the low temperature energy demand of each process and $LCOH_{LT}$ is the levelised cost of low temperature heat provided by heat pumps that is calculated in eqn (13). High temperature heat demand $H_{HT,p}$ is assumed to be provided with direct electric heating, hence is treated as electricity demand. The sum of the final energy demand of each process $E_{FE,p}$ and the high temperature heat demand $H_{HT,p}$ of each process is multiplied by the levelised cost of electricity LCOE. The summand of each process is normalised to the mass output $m_{out,p}$ for removing 1 tCO_2.

The $LCOP_{p,co}$ is calculated according to eqn (10)–(12) wherein the CAPEX, fixed operational expenditures OPEXfix, the variable operational expenditures OPEXvar, the weighted average cost of capital WACC and lifetime N, and the availability τ of each process p are used.

$$\mathrm{LCOP}_{p,\mathrm{co}} = \frac{\left(\mathrm{CAPEX}_p \cdot \mathrm{crf}_p + \mathrm{OPEXfix}_p\right) \cdot \mathrm{cap}_p}{\mathrm{out}_p} + \mathrm{OPEXvar}_p \tag{10}$$

$$\mathrm{crf}_p = \frac{\mathrm{WACC} \cdot (1 + \mathrm{WACC})^{N_p}}{(1 + \mathrm{WACC})^{N_p} - 1} \tag{11}$$

$$\mathrm{out}_p = \mathrm{cap}_p \cdot \tau_p \tag{12}$$

where out_p is the output of a process, and cap_p the capacity of a process. The units depend on the process. The levelised cost of low temperature heat $LCOH_{LT}$ provided *via* heat pump HP is calculated using eqn (13).

$$\mathrm{LCOH_{LT}} = \frac{\mathrm{CAPEX_{HP}} \cdot \mathrm{crf_{HP}} + \mathrm{OPEXfix_{HP}}}{8760 \cdot \tau} + \mathrm{OPEXvar_{HP}} + \frac{\mathrm{LCOE}}{\mathrm{COP}} \tag{13}$$

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

3.3. Applied scenarios for total cost and primary energy demand estimation

The primary energy demand and cost of large-scale mineralisation is assessed for the exemplary years 2050 and 2070. The analysis is conducted by assuming that 60% of the CDR demand for the 1.5 °C and 1.0 °C climate targets[10,12] is covered with mineralisation options. The basic primary energy demand is estimated using LUT-DEMAND, applying the LUT-delayed economic equality scenario (LUT-DEES) for GDP per capita and medium population projection of the United Nations (UN) as macro-economic basis.[10] Of these 60%, the contribution is assumed to be 20% each onshore and offshore MIN_{IN}, 10% EW, and 3.3% for each MIN_{EX} using serpentine, olivine, and industrial waste. Details on the calculation can be found in the Supplementary material 2 (ESI†), where alternative assumptions can be applied.

3.4. Assessment of sequestration potentials

Literature findings on the cumulative and annual CO_2 mineralisation potential on a regional spatial resolution are allocated to the nine major regions as of the LUT Energy System Transition Model (LUT-ESTM), which are Europe, Eurasia, Middle East and North Africa (MENA), sub-Saharan Africa (SSA), South Asian Association for Regional Cooperation (SAARC), Northeast Asia, Southeast Asia, North America, and South America.[50]

For MIN_{IN}, a summary on global-regional total potential by Oelkers *et al.*[57] is used. To identify economically feasible sequestration capacities, 10% of the capacity reported by Oelkers *et al.*[57] is assumed to be economic potential and sequestration sites are assigned to the above-mentioned major regions. This approach bears significant uncertainty and should be further challenged as a major point on the research agenda for MIN_{IN} proposed by Oelkers *et al.*[57] To derive the annual injection and sequestration potential of MIN_{IN} locations in $MtCO_2$ per a, the surface area, as communicated by Oelkers *et al.*,[57] is multiplied by the area specific injection rate as proposed by Wijaya *et al.*[133] Several methods for sequestration capacity estimation of CO_2 underground sequestration sites have been established.[60] Nevertheless, thorough MIN_{IN} potential analysis in high spatial resolution is a current research gap.[57] Vishal *et al.*[134] estimate a total MIN_{IN} potential for India of 97–316 $GtCO_2$ as a conservative theoretical estimate. They apply methods proposed by McGrail *et al.*[135] and Snæbjörnsdóttir *et al.*[136] to assess the sequestration potential of Indian basaltic formations.

Myers and Nakagaki[74] conducted a regional study on MIN_{EX} and concluded that Japan alone can achieve CDR at the rate of up to 7.6 $GtCO_2$ per a. Slag-based MIN_{EX} alone is expected to enable cumulative MIN_{EX} of 26.4–41.9 $GtCO_2$ between 2020 and 2100.[137] Steel slag's high CaO and MgO content of about $37\%_{wt}$ and $9.1\%_{wt}$, respectively, and the resulting WP of around 384.7 $kgCO_2$ per t of slag, make it a valuable feedstock for MIN_{EX} with an expected global potential of 320–870 $MtCO_2$ per a in 2100.[122] Renforth[122] notes that about 185 t of blast furnace slag and 117 t of steel slag are produced per tonne of crude steel produced. Through the decarbonisation of steel production, blast furnaces will be phased out and blast furnace slag is therefore not further considered in this work. Production of one tonne of aluminium produces 3.45 t of bauxite residues, that can neutralise 44–66 $kgCO_2$ per t of bauxite residues.[122] About 115 kg of cement kiln dust are produced per tonne of cement clinker.[122] All these industrial solid wastes may be used for CO_2 mineralisation. In this study, only industrial solid waste is assumed as feedstock to MIN_{EX} and additional potential of mined rock, *e.g.*, serpentine or olivine is neglected. Pan *et al.*[61] also emphasise the potential for additional indirectly avoided CO_2 emissions by utilising carbonates as filler material in concrete blocks or cement mortars.

EW potential on agricultural land for the nine major regions considered in this work is determined as follows. The available agricultural land of each country[138] is multiplied by a basalt application rate of 40 t per (ha·a) and a lower as well as higher estimate for the EW efficiency of 0.5 tCO_2 per tRock and 0.67 tCO_2 per tRock, respectively.

4. Results

In subsection 4.1, the global potential for MIN_{IN}, MIN_{EX}, and EW is presented for the nine major regions of LUT-ESTM.[10] Subsection 4.2 elaborates on the techno-economic findings for CO_2 mineralisation, and subsection 4.3 condenses key findings on the technology readiness levels of the mineralisation options. Results are calculated in five-year steps from 2020–2100, though presented for 2030, 2050, and 2070, as the key years. These years provide concise information for times steps decisive for ramping CDR to reach a safe future.[12] Results for all years can be found in Supplementary material 2 (ESI†) where all calculations can be traced.

4.1. Global-regional mineralisation potential

The global-regional potential for MIN_{IN} is depicted in Fig. 4. The global calculated annual injection potential equals about 0.7% per a of the total sequestration capacity. This stems from a combination of area and sequestration potential estimates in Oelkers *et al.*[57] and the assumed area demand for injection of 28 km^2 per ($MtCO_2$/a).[133] Some MIN_{IN} sequestration locations can only achieve 0.2% per a injection rates due to different geologic characteristics. In Fig. 4, the secondary (right) vertical axis presenting the annual MIN_{IN} potential is therefore normalised to 0.7% per a of the maximum total MIN_{IN} potential presented on the primary (left) vertical axis. Hence, discrepancies in length of the coloured and black bar, shows discrepancies in the ratio of annual to total sequestration potential.

As can be seen in Fig. 4, all major regions except for Europe, MENA and North America are assumed to have Gt-scale annual injection and MIN_{IN} potential. South America shows the highest total and annual potential for MIN_{IN} at 543.0 $GtCO_2$ and 3.9 $GtCO_2$ per a, respectively. Only Europe and the MENA region show annual potential considerably below 1 $GtCO_2$ per a. The SAARC region can sequester about 2 $GtCO_2$ per a. The

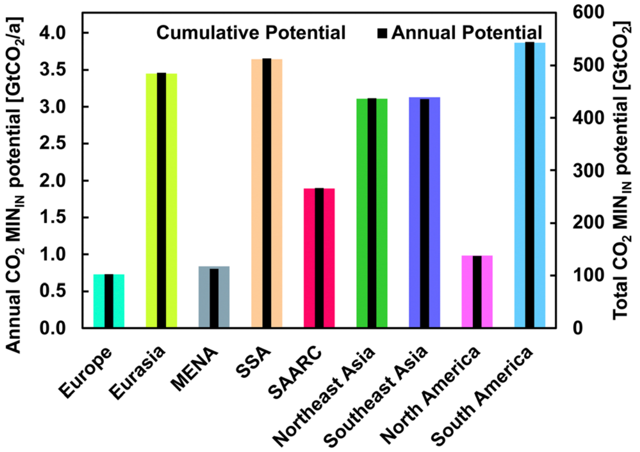

Fig. 4 Global-regional CO$_2$ *in situ* mineralisation potential. Coloured bars indicate the total sequestration potential and the black bars indicate the annual injection and sequestration potential in each region. The left vertical axis is normalised to 0.7% per a of total potential as derived from the global total and annual sequestration potential. The total sequestration potential is represented by the right vertical axis. Divergences of this average value can be seen in the figure. Further information can be found in the Supplementary material 2 (ESI†).

secondary vertical axis in Fig. 4 indicates that some potential MIN$_{IN}$ sites have different estimated height (volume-to-area ratio) which results in a slight divergence in the annual injection rate calculated when using the area demand approximation for CO$_2$ underground injection provided by Wijaya *et al.*[133]

The annual and cumulative potential for MIN$_{EX}$ using industrial solid wastes, *i.e.*, steel slag, cement kiln dust, and red mud, are depicted in Fig. 5. Steelmaking slag occurs also in defossilised processes utilising hydrogen and electric arc furnaces[139] and is, therefore, expected to be available despite the transition to green steel.

SAARC is expected to produce most of the global industrial waste output that can be utilised for CO$_2$ MIN$_{EX}$ by the mid of the 21st century. The global MIN$_{EX}$ potential using industrial waste is projected to peak in 2045 at about 584 MtCO$_2$ per a before declining to 394 MtCO$_2$ per a by 2100. These results confirm findings of Pan *et al.*[61] who found a global total direct MIN$_{EX}$ potential using alkaline solid wastes of about 310 MtCO$_2$ per a without considering some countries, especially in South America and Africa. Northeast Asia, especially China, is a major contributor both in results provided by Pan *et al.*[61] and in this study; however results in this study indicate that SAARC and SSA will overtake Northeast Asia in terms of CO$_2$ mineralisation using alkaline solid wastes. By the end of the 21st century, SSA is projected to be the major producer of industrial waste enabling substantial MIN$_{EX}$ at an annual rate of 98 MtCO$_2$ per a. A total of 37.4 GtCO$_2$ can be permanently sequestered in carbonates using industrial solid waste as input for MIN$_{EX}$, which confirms the findings by Myers *et al.*[137] It can be seen in Fig. 5 that the potential for MIN$_{EX,IW}$ is lowest in Europe, Eurasia, MENA, North America, and South America. The potential projected for Southeast Asia is significantly lower compared to Northeast Asia, SAARC, and SAA where significant economic growth is projected in the LUT-DEES macroeconomic scenario.[10]

The technical potential for EW in the nine major regions of LUT-ESTM is depicted in Fig. 6. The EW potential is directly related to the available cropland in each region. Whether the full technical potential of EW on croplands can be realised is uncertain and must be assessed in future studies.

SSA has the highest theoretical EW potential with about 5.5 GtCO$_2$ per a. North America and SAARC follow with a theoretical EW potential of about 5.3 GtCO$_2$ per a and 5.2 GtCO$_2$ per a, respectively. MENA has a theoretical EW potential of about 1.5 GtCO$_2$ per a, and South America, Southeast Asia, Northeast Asia, Eurasia, and Europe all have average theoretical EW potential in the range of 3.4–4.3 GtCO$_2$ per a. As EW performance is linked to precipitation, arid regions will show lower sequestration kinetics than humid regions, potentially constraining the annual CDR potential.

4.2. Techno-economic process parameters and cost of carbon sequestration

Subsection 4.2.1 presents findings on the PE demand of different CO$_2$ mineralisation options. Subsection 4.2.2 then shows the resulting future projected cost of CO$_2$ mineralisation

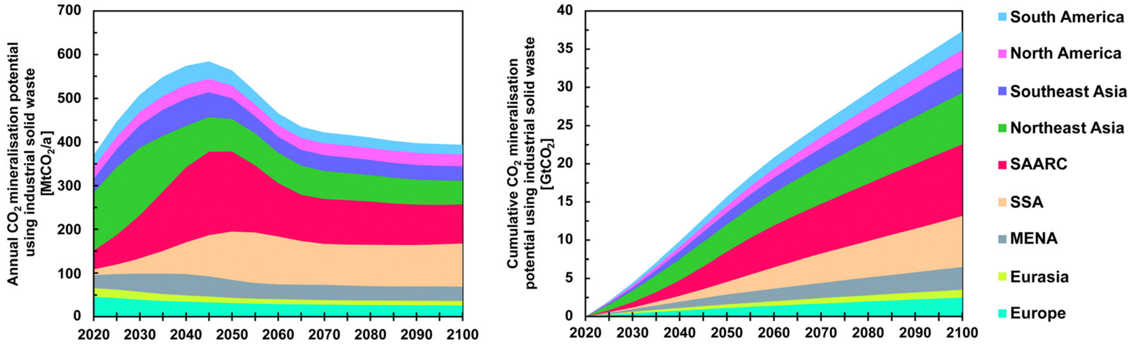

Fig. 5 Global-regional CO$_2$ *ex situ* mineralisation potential using steel slag, cement kiln dust, and red mud for the nine major regions used in LUT-ESTM.[10] The left panel shows the annual CO$_2$ mineralisation potential, and the right panel shows the cumulative CO$_2$ mineralisation potential in the 21st century.

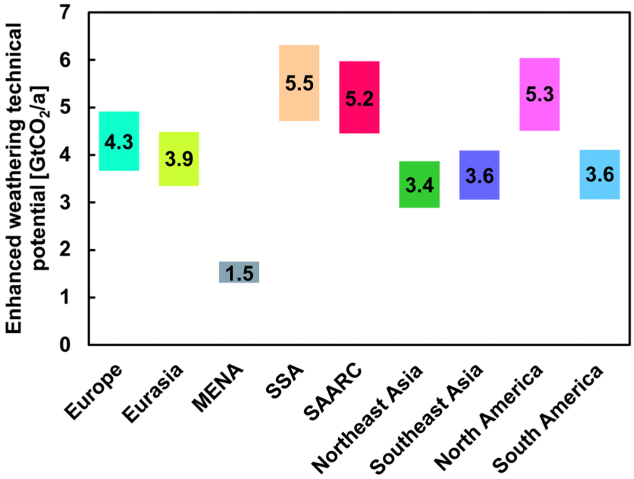

Fig. 6 Global-regional potential for EW using basalt on cropland for the nine LUT-ESTM major regions.[50] The position of the numeric values indicates the mean of the available technical potential range. There is uncertainty in the weathering efficiency when considering soil acidity[140] as weathering rates differ with soil pH. As the average soil pH across the major regions ranges from about 5.7–7.5 (*cf.* Supplementary material 1 note 3, ESI†), this effect is not further considered.

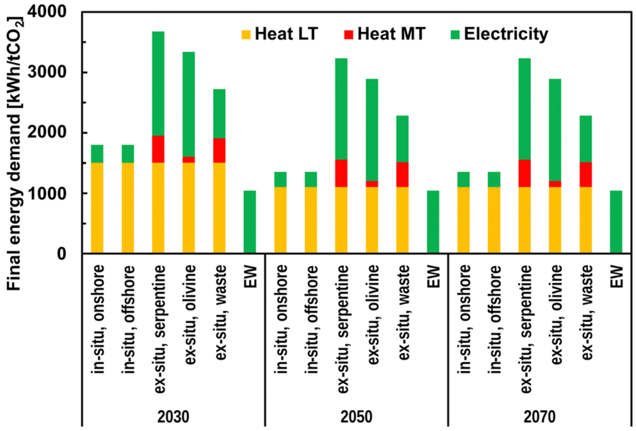

Fig. 7 Final energy demand for mineralisation options for CDR. Final energy demand is clustered in low-temperature heat (Heat LT) for DAC, medium-temperature heat (Heat MT) for the mineralisation process and electricity for feedstock handling, mineralisation process, and DAC. Rock transportation distance for EW and MIN_{EX} is assumed to be 200 km.

to make these CO_2 mineralisation options suitable for energy-industry-CDR system integration.

4.2.1. Energy demand of mineralisation options. Energy and mass balances of processes are crucial for consideration in energy system modelling. The process chains considered for EW, MIN_{IN}, and MIN_{EX} are depicted in Fig. 1. MIN_{IN} requires DAC units close to underground formations suitable for CO_2 injection, assuming rather limited suitability of large-scale CO_2 pipelines. EW requires rock handling with a subsequent spreading step for CDR. MIN_{EX}, however, involves both a capture and sequestration step requiring rock handling. The FE demand for mineralisation options for CDR is presented in Fig. 7.

EW is the only mineralisation option that solely relies on electricity. All other options require also heat for DAC and the mineralisation process. Mature technologies for rock mining, transportation, and comminution are assumed to show no reduction in final energy. EW requires the least total FE of about 1.0 MWh per tCO_2. The total FE demand for MIN_{EX} is 3.7 MWh per tCO_2, 3.3 MWh per tCO_2, and 2.7 MWh per tCO_2 if serpentine, olivine, and industrial solid waste is used as a feedstock in 2030. The total FE is expected to decrease in the future mainly due to advances in DAC technologies. In 2070, the total FE demand using MIN_{EX} is expected to decrease to 3.2 MWh per tCO_2, 2.9 MWh per tCO_2, and 2.3 MWh per tCO_2 when using serpentine, olivine, and industrial solid waste, respectively. Using olivine for MIN_{EX} results in the lowest mid-temperature heat demand but shows the highest total electricity demand. MIN_{IN} requires a total FE supply of about 1.8 MWh per tCO_2 and 1.4 MWh per tCO_2 in 2030 and from 2050 onwards, respectively. While EW shows the lowest total FE demand of all mineralisation options examined in this study, the electricity demand is sensitive to the rock transportation distance. Increasing the transportation distance from the base assumption of 200 km to 400 km can increase the total FE demand from 1.0 MWh per tCO_2 to 1.9 MWh per tCO_2. For MIN_{EX} options, the longer transportation distance increases the total FE to 4.4 MWh per tCO_2 and 3.9 MWh per tCO_2 when using serpentine and olivine, respectively, in 2030.

The PE demand for CDR with CO_2 mineralisation for long-term stable sequestration is presented in Fig. 8.

The PE demand of MIN_{EX} using industrial solid waste is the highest at about 2.1 MWh per tCO_2 in 2030 which declines to 1.8 MWh per tCO_2 and 1.7 MWh per tCO_2, in 2050 and 2070, respectively. Using serpentine and olivine as feedstock for MIN_{EX}, increases the PE demand of mineralisation to 3.2 MWh per tCO_2 and 2.8 MWh per tCO_2, respectively, in 2030. In 2070, MIN_{EX} using mined rocks requires about 2.7 MWh per tCO_2 and 2.4 MWh per tCO_2 for serpentine and olivine, respectively. MIN_{IN} does not require feedstock handling and has significantly lower PE demand related to the sequestration process, which results in comparably lower total PE demand of 1.1 MWh per tCO_2, 0.8 MWh per tCO_2, and 0.7 MWh per tCO_2 in 2030, 2050, and 2070, respectively. EW has the lowest PE demand at about 1.2 MWh per tCO_2 in 2030; however, the demand increases with the distance of rock transportation. Increased transportation distance from the base assumption of 200 km to 400 km results in a PE demand of 1.9 MWh per tCO_2 in 2030.

4.2.2. Estimation of future cost for carbon dioxide mineralisation. The impact of technology learning on the cost of MIN_{EX} in a scenario where MIN_{EX} is widely deployed for secure CO_2 sequestration (*cf.* Supplementary material 2, ESI†) is depicted in Fig. 9.

The high initial cost related to DAC makes MIN_{IN} and MIN_{EX} options relatively expensive compared to EW in the short term. MIN_{IN} options cost about 123 € per tCO_2 and 131 € per tCO_2 if CO_2 is injected onshore and offshore, respectively, in 2030. The cost is expected to decline to 40 € per tCO_2 and 47 € per tCO_2 for

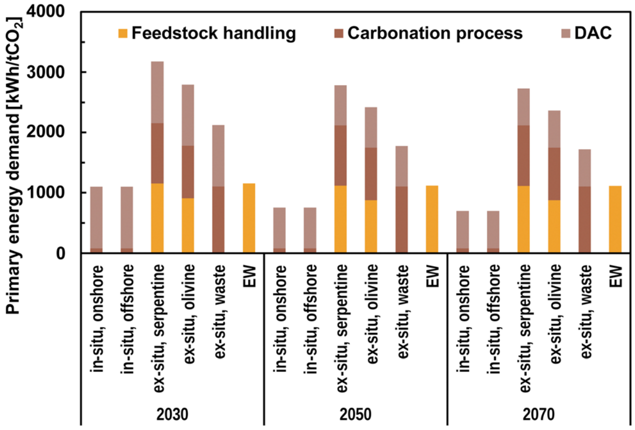

Fig. 8 Primary energy demand for CO_2 mineralisation options in 2030, 2050, and 2070. The energy demand is distinguished between feedstock handling, which includes open-pit mining, transportation, and comminution, energy demand directly related to the sequestration process, such as reactor operation for MIN_{EX}, and additional energy demand for DAC. DAC assumptions are taken from Fasihi *et al.*[23] and adapted as in Mühlbauer *et al.*[11] The rock transportation distance is assumed to be 200 km for EW and MIN_{EX}.

onshore and offshore MIN_{IN}, respectively, until 2070. EW costs about 88 € per tCO_2, 68 € per tCO_2, and 66 € per tCO_2 in 2030, 2050, and 2070, respectively. Therefore, anticipated future cost decline for DAC makes MIN_{IN} cost-competitive with EW starting in 2050. For MIN_{EX}, using industrial waste is economically preferable over mined rocks due to the avoided cost for feedstock mining and handling. The cost of MIN_{EX} using serpentine, olivine, and industrial solid waste for mineralisation is estimated to be 188 € per tCO_2, 189 € per tCO_2, and 182 € per tCO_2, respectively, in 2030. Until 2070, the costs for those MIN_{EX} options are expected to decrease to 74 € per tCO_2, 73 € per tCO_2, and 61 € per tCO_2, respectively. Generally, the LCOCRs of all mineralisation options' are in the range of 50–100 € per tCO_2 from 2050 onwards, making mineralisation an economically viable option for safe and long-term CO_2 sequestration. Future cost projections are uncertain, and those findings will need re-evaluation once technologies such as DAC become more mature and robust cost estimates are publicly available. Technology learning is only assumed for MIN_{EX}, as both MIN_{IN} and EW can build fully on well-established and mature technologies used in mining and fossil fuel extraction.

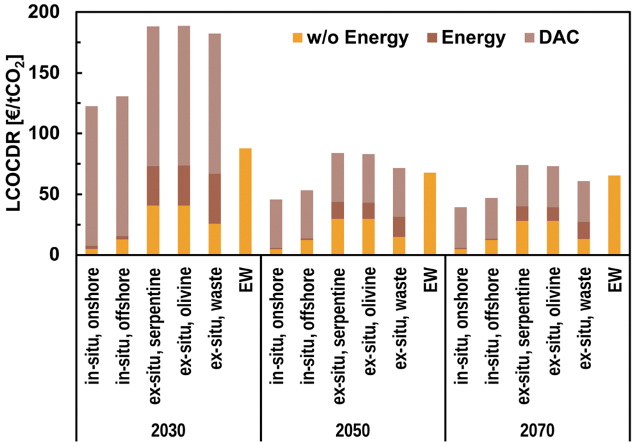

Fig. 9 Future estimations of the levelised cost of CDR (LCOCDR) with secure CO_2 sequestration *via* mineralisation. Projected DAC cost are adapted from Mühlbauer *et al.*[11]

4.3. Technology readiness

TRL is an important measure for energy system modelling to project future technology development. As Young *et al.*[141] find, low-TRL technologies often experience cost escalations before becoming mature (high TRL) technologies, which show technology learning reducing the cost.[127] Rock mining and handling is a well-established industry with mature technology. Furthermore, EW relies solely on relatively simple technology requiring feedstock mining, handling, transportation, and spreading.[11,31,32] The TRL of EW can therefore be assessed at high level of 8–9. MIN_{EX}, in contrast, requires a specialised reactor, and no large-scale plants have been reported yet. Consequently, the TRL is lower compared to EW at a level of 5–6. MIN_{IN} generally builds on well-established technologies such as deep drilling and fluid injection that are used for geothermal energy, fossil fuel extraction or enhanced oil and gas recovery. While the subsurface reservoir is different, there are already several relatively large-scale operations proofing the concept on a TRL of 8–9.[142]

4.4. Cost and primary energy demand of large-scale mineralisation

The marginal energy demand and cost of CDR options with long-term CO_2 sequestration *via* mineralisation are presented in subsections 4.2.1 and 4.2.2, respectively. Also, mineralisation is evaluated to be ready to satisfy a major share of future CDR demand to achieve ambitious climate targets (*cf.* subsection 4.3). The results for the assumptions mentioned above are presented in Table 2.

The cost of large-scale mineralisation deployment for long-term CO_2 sequestration is about 69.1 b€ in 2050 and increases to 333.4 b€ in 2070 due to a fully ramped CDR sector in a trajectory compliant with limiting long-term global warming to 1.5 °C. For more ambitious targets, *i.e.*, limiting global warming to 1.0 °C, the cost increase to 241.7 b€ and 1166.9 b€ in 2050 and 2070, respectively. If these results are put into perspective with the projected total GDP, the cost of sequestering 60% of projected CDR demand long-term in stable materials is estimated at 0.06% and 0.21% of the GDP for the 1.5 °C and 1.0 °C trajectories, respectively.

Primary energy demand for CO_2 mineralisation in the discussed case amounts to about 1.3 PWh and 6.7 PWh in 2050 and 2070, respectively, for the 1.5 °C climate target. For the 1.0 °C target, the primary energy demand increases to about 4.5 PWh in 2050 and 23.4 PWh in 2070. This increase implies additional primary energy demand compared to the basic energy-industry system of 2.46% and 8.6% in 2070 for a 1.5 °C and 1.0 °C climate target, respectively.

Table 2 Total energy demand and cost for large-scale mineralisation for long-term CO_2 sequestration. MIN_{IN} offshore and MIN_{IN} onshore are assigned 20% of CDR contribution each. MIN_{EX} with serpentine, olivine, and industrial solid waste are assigned 3.3% each and EW is assigned 10% of total CDR demand. Therefore, mineralisation would sequester 60% of the total CDR demand. Further information can be found in the Supplementary material 2 (ESI)

			1.5 °C target[a]		1.0 °C target[a]	
		Unit	2050	2070	2050	2070
Cost	Total GDP (LUT-DEES)	b€	340,827	566,658	340,827	566,658
	Total annualised cost mineralisation	b€	69.1	333.4	241.7	1166.9
	Share in projected total GDP	%	0.02	0.06	0.07	0.21
Energy	Primary energy demand energy-industry system	PWh	192.4	271.7	192.4	271.7
	Primary energy demand mineralisation	PWh	1.3	6.7	4.5	23.4
	Share in projected total primary energy demand	%	0.67%	2.46%	2.34%	8.60%

[a] The CDR demand is adapted from Keiner *et al.*[10] and Breyer *et al.*[12] for a climate target at the end of this century, limiting global warming to 1.5 °C and 1.0 °C. The 1.5 °C target assumes a total CDR demand of 500 $GtCO_2$, the 1.0 °C target 1750 $GtCO_2$.

5. Discussion

The discussion section focuses on the implications drawn from the presented results (subsection 5.1), the key issue of permanence of CDR (subsection 5.2), and the limitations and the research outlook (subsection 5.3) of this study.

5.1. Implications from the results

As concluded in a recent study,[11] CO_2 mineralisation shows the highest permanence and safety, *i.e.*, permanent CO_2 sequestration, at a cost-competitive level compared to other CDR options. While there is still significant uncertainty in the global potentials of the options, the presented results indicate that all CDR from NETs based on captured dilute atmospheric CO_2 and concentrated gaseous CO_2 as intermediate step can be realised with the safe sequestration of CO_2 in carbonates *via* mineralisation.

The required energy for CO_2 mineralisation would lead to gross emissions in the fossil fuel-dominated energy system.[63] Resulting low carbon efficiencies would make net CDR significantly more expensive and emphasises the priority of a rapid defossilisation of the current energy-industry system and future energy-industry-CDR system before large-scale CDR with DAC and CO_2 mineralisation can be implemented effectively.[12,17] The need for raw material seems negligible for MIN_{IN} but is significant for MIN_{EX}, with mined rocks, and EW. Both technologies can at least partially build on existing infrastructures; however, the satisfaction of raw material demand should be questioned carefully on the background of substantial pressure on material availability.[143]

As shown in previous work, CO_2-to-solid processes can play a major role in future CDR endeavours if the security of CO_2 sequestration is prioritised.[11] Safe and long-term sequestration of atmospheric CO_2 will be key to achieve ambitious climate targets to enable a safe future within planetary boundaries.[12,13]

CO_2 mineralisation options in general, and MIN_{EX} in particular, show higher cost and energy demand compared to geological underground sequestration of CO_2 or afforestation.[11] However, non-permanent CDR such as afforestation and potentially geological underground CO_2 sequestration come with several challenges including complicated and challenging accounting and subsequent monetary compensation and the risk of potential leakage, respectively.[21,144] As stated by Vielstädte *et al.*,[52] former hydrocarbon extraction sites with multiple injection wells may not always be suitable for geologic CO_2 sequestration due to potential leakage.

Alternative pathways to produce solid materials from atmospheric CO_2 have been presented in previous works with electricity-based silicon carbide and carbon fibres as promising options.[45,46] The business-case of selling by-products from MIN_{EX} and potential synergies with the cement industry to reduce the cost of CO_2 sequestration has been elaborated;[55,130] however, MIN_{EX} is less dependent on a market and demand for by-products compared to electricity-based silicon carbide and carbon fibres, which are considerably more energy- and cost-intensive.[45,46] Nevertheless, MIN_{EX} is largely constrained by the availability of industrial waste because mined rocks for carbonation make the approach significantly more expensive.

Further technologies relying on CO_2 mineralisation for removal and sequestration of atmospheric CO_2 have been proposed. Repeated ambient weathering of MgO for DAC was studied[63,141,145] and is found to be a cost-competitive alternative to other DAC technologies.

Due to the high potential demand for minerals, concerns such as mineral poverty should be considered throughout the discussion of geochemical measures for CDR.[143] However, in most literature, a large potential for CO_2 mineralisation is presented (*cf.* section 2) and even a fraction of that could be sufficient to enable the safe sequestration of CO_2 at the scale required for a 1.5 °C or 1.0 °C climate target. Future LCAs will be needed to thoroughly determine the impacts of different mineralisation options on the environment. The insights of such studies can be utilised to enhance the implications on future CDR portfolios for different societal preferences.[11]

While the additional primary energy demand is significant, renewable and clean energy sources, *i.e.*, wind and especially solar photovoltaic, are on a promising trajectory to supply abundant low-cost electricity. It is not expected that large-scale CDR for ambitious climate targets is adversely affected by a possible limitation due to the availability of renewable electricity.[146] As the additional demand in 2050 for electricity is limited to about 0.67% and 2.34% of projected total PE demand for a 1.5 °C target and a 1.0 °C target, respectively, the deployment of CDR, especially mineralisation, is not expected to significantly hamper the energy transition of other sectors such as power, transport, or industry through competition for renewable electricity.

CO_2 mineralisation faces several current challenges. MIN_{IN} or direct aqueous MIN_{EX} are currently operated with potable freshwater.[44,119] This may severely compromise the sustainable potential of these operation considering the rising challenges in the global freshwater scarcity. MIN_{IN} may be operated with seawater instead of freshwater in the foreseeable future[44] and MIN_{EX} shows a high water recovery rate[119] which should further be improved to solve water demand issues. EW has some sustainability issues considering potential health threats through dispersion of ultrafine particles[32] or particles which are potentially contaminated, which calls for stringent management of operations and use of non-hazardous waste only[102] in the future.

5.2. Permanence and timing of mineralisation

Mineralisation produces solid materials from gaseous atmospheric CO_2. The permanence of CO_2 sequestration in carbonates is well documented.[20,65] While EW and MIN_{EX} can deliver permanent CDR as well as desirable co-benefits, MIN_{IN} shows relatively low cost and energy demand and can be an attractive alternative to geologic sequestration as commonly proposed and often 'over used' in IAM scenarios.[147] Geologic sequestration comes with several challenges. Fossil fuel producers that now own depleted reservoirs will benefit from underground sequestration of CO_2 in their sites. This starkly contradicts the polluters-pay approach that is often proposed in the context of CO_2 emissions[148] by providing an attractive business case for fossil fuel producers for both polluting and restoring the atmosphere.

One public concern regarding geologic CO_2 sequestration is the risk of potential leakage[87,149] that imposes significant risk to the environment and of morbidity.[150,151] This risk requires significant efforts to actively monitor sequestration sites.[152,153] For example, similar to nuclear power plants in Germany, there are currently no options for insuring geologic sequestration projects.[154] Although there are no major leakage events reported so far, the risk is acknowledged and more investigation and research on the topic are needed.[155] Non-permanent CDR options are difficult to properly account[21,144] and, in light of the above-mentioned uncertainties regarding geologic sequestration, CO_2 mineralisation can be a key technology in sustainable large-scale CDR.

5.3. Limitations and research outlook

Even though the CO_2 mineralisation potentials are elaborated on a global and regional basis, further improving the spatial resolution is necessary to provide effective guidance to decision makers. However, since the required data is still not sufficient,[57] providing potential estimations for all CO_2 mineralisation options that are currently discussed in a comprehensive manner will advance the discussion about CO_2 mineralisation for permanent CDR. Precisely mapping technical, economic, and sustainable potentials is yet to be conducted in regional level assessments. Missing data indicates the requirement of field trials and further endeavours to precisely estimate the potential of underground MIN_{IN} sites globally in high spatial resolution. The CO_2 mineralisation potential projection for the 21st century in conjunction with techno-economic parameters will enable future energy-industry-CDR system transition analyses.

A static techno-economic framework is chosen to evaluate the cost and energy demand for large-scale CO_2 mineralisation enabling permanent CDR on the large scale. Therefore, results of this study are intended to advance the consideration of CO_2 mineralisation in energy-industry-CDR systems by providing the required data, but the results should be validated in comprehensive system transition optimisation studies and LCAs. Further developments of CO_2 mineralisation options in energy demand are omitted and should be further studied in specialised assessments. This also relates to the consideration of local parameters such as soil pH on the weathering rate for EW mineralisation. Future research with a high spatial resolution will be required to assess mineralisation options in full global-local detail.

6. Conclusions

This work provides a techno-economic assessment as well as estimations for the global-regional potential of the CO_2 mineralisation options with high probability to shape the future energy-industry-carbon dioxide removal system if a safe and long-term sequestration of atmospheric CO_2 is desired. Three main pillars of CO_2 mineralisation can play a significant role in future energy-industry-carbon dioxide removal systems: enhanced weathering, *in situ* mineralisation, and *ex situ* mineralisation. Enhanced weathering can be implemented on agricultural crop land or marginal hinterland as a potentially no-regret option for carbon dioxide removal. *in situ* mineralisation is similar to geologic underground sequestration, but it requires different sequestration sites that enable the rapid mineralisation and therefore safe long-term sequestration at elevated temperature and pressure of CO_2 in aqueous solution. These suitable sequestration sites are composed of basaltic rocks or peridotite, are distributed globally, and show sufficient CO_2 mineralisation potential. At similar cost and energy demand to geological underground sequestration, *in situ* mineralisation of CO_2 provides an attractive alternative. *ex situ* mineralisation is the option investigated with highest cost and energy demand. Mined rocks must be transported, crushed, grinded, eventually pre-treated, and then mineralised in special reactors. Alternatively, industrial wastes of steelmaking, among others, can be used for *ex situ* mineralisation.

The *in situ* mineralisation adds a moderate extra energy demand and cost required to direct air capture of CO_2 enabling safe and long-term atmospheric CO_2 sequestration. The cost of all mineralisation options for permanent CDR is expected to fall below 100 € per tCO_2 by 2050 while enhanced weathering will likely reach that threshold by 2030. The primary energy demand for CDR *via* CO_2 mineralisation is lowest for enhanced weathering, which does not require any direct air CO_2 capture operations or powering a reactor as it only requires feedstock mining and spreading. At about 1150 kWh per tCO_2, enhanced weathering would add moderate additional primary energy demand on the future projected demand. *in situ* mineralisation requires similar primary energy at about 1100 kWh per tCO_2 in 2030, which is expected to decrease mainly due to improvements in direct air CO_2 capture in the long term. While direct air CO_2 capture constitutes about 50% of the total primary energy demand, *ex situ* mineralisation for carbon dioxide removal requires about 2100–3200 kWh per tCO_2 in 2030, which reduces to about 1700–2700 kWh per tCO_2 in 2070. The primary energy demand of mineralisation options must be considered for the determination of future carbon dioxide removal portfolios, but the sharply declining cost of renewable energy makes other indicators, which must be studied in dedicated life cycle assessments, potentially more important. *in situ* mineralisation is already conducted commercially on industrial scale and is considered to be on an advanced technology readiness level of 8 or 9. While enhanced weathering and *ex situ* mineralisation are not yet deployed on significant scale, both options rely on established technology with the exception of aqueous carbonation reactors.

Large-scale deployment of mineralisation to contribute a 60% share of total carbon dioxide removal endeavours necessary to achieve ambitious climate targets is shown to be manageable at a share of 0.06% and 0.21% in projected gross domestic product until 2070 for a 1.5 °C and 1.0 °C temperature target, respectively. The total energy demand for mineralisation in this case requires about 2.5% and 8.6% additional primary energy demand compared to the total primary energy demand for all other sectors. While this demand is substantial, current development of cost and deployment of clean, renewable sources such as wind power and solar photovoltaics can support additional primary energy demand for safe CO_2 sequestration. Also, due to the limited additional demand in 2050, *i.e.*, 0.67% and 2.34% for the 1.5 °C and 1.0 °C temperature target, respectively, additional primary energy demand for CO_2 mineralisation is not expected to substantially hinder the defossilisation of all other sectors until 2050. Possible synergies of by-products are discussed.

With this work, a first step for implementation of CO_2 mineralisation for permanent carbon dioxide removal in modelling frameworks for investigating the characteristics of future energy-industry-carbon dioxide removal systems is taken. Future iterations of techno-economic and potential assumptions for the presented technology options will further reduce uncertainty that exists at this early stage of development in CO_2 mineralisation and carbon dioxide removal in general. Permanent carbon dioxide removal *via* CO_2 mineralisation can be a most valuable endeavour to reach ambitious climate targets that should be considered for the transition phase, which include large-scale carbon dioxide removal in a 100% renewable post-fossil energy-industry-carbon dioxide removal system.

Nomenclature

BECCS	Bioenergy with carbon capture and sequestration
Ca	Calcium
CAPEX	Capital expenditures
CDR	Carbon dioxide removal
CO_2	Carbon dioxide
DAC	Direct air capture
DACCS	Direct air carbon capture and sequestration
EW	Enhanced weathering
FE	Final energy
IAM	Integrated assessment model
IPCC	Intergovernmental Panel on Climate Change
LCA	Life cycle assessment
LCOCDR	Levelised cost of carbon dioxide removal
MENA	Middle East and North Africa
Mg	Magnesium
MIN_{EX}	*Ex situ* mineralisation
MIN_{IN}	*In situ* mineralisation
NCS	Natural climate solutions
NET	Negative emission technology
OPEX	Operational expenditures
OPEXfix	Fixed operational expenditures
OPEXvar	Variable operational expenditures
PE	Primary energy demand
SAARC	South Asian Association for Regional Cooperation
SSA	Sub-Saharan Africa
TNE	Total negative emissions
TRL	Technology readiness level
UN	United Nations
WP	Weathering potential

Data availability

The data supporting this article have been included as part of the ESI.†

Conflicts of 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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