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Life Cycle Assessment of Direct Air Carbon Capture and Storage with Low-Carbon Energy Sources

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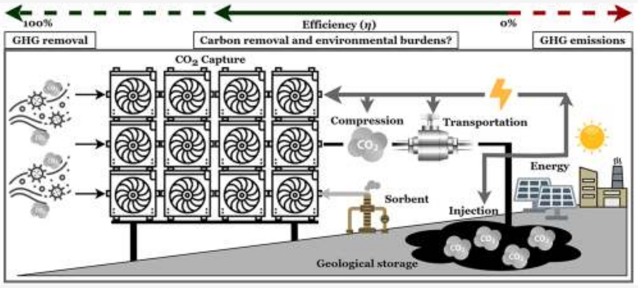
ABSTRACT: Direct air carbon capture and storage (DACCS) is an emerging carbon dioxide removal technology, which has the potential to remove large amounts of CO_2 from the atmosphere. We present a comprehensive life cycle assessment of different DACCS systems with low-carbon electricity and heat sources required for the CO_2 capture process, both stand-alone and grid-connected system configurations. The results demonstrate negative greenhouse gas (GHG) emissions for all eight selected locations and five system layouts, with the highest GHG removal potential in countries with low-carbon electricity supply and waste heat usage (up to 97%). Autonomous system layouts prove to be a promising alternative, with a GHG removal efficiency of 79−91%, at locations with high solar irradiation to avoid the consumption of fossil fuel-based grid electricity and heat. The analysis of environmental burdens other than GHG emissions shows some trade-offs associated with CO_2 removal, especially land transformation for system layouts with photovoltaics (PV) electricity supply. The sensitivity analysis reveals the importance of selecting appropriate locations for grid-coupled system layouts since the deployment of DACCS at geographic locations with CO_2-intensive grid electricity mixes leads to net GHG emissions instead of GHG removal today.



KEYWORDS: *life cycle assessment (LCA), direct air carbon capture and storage (DACCS), carbon dioxide removal (CDR), negative emission technologies (NETs)*

1. INTRODUCTION

Carbon dioxide removal (CDR) technologies, or negative emission technologies (NETs), are expected to play a crucial role in the decarbonization of the global energy system, with carbon removal projections of more than 20 gigatonnes year^{-1} by 2050.[1,2] The carbon removal projections of CDR technologies are, however, associated with high uncertainties and depend, among others, on the modeling assumptions used.[1,2] Prospective energy scenarios, generated by integrated assessment models (IAMs), demonstrate that the 1.5 °C target of the Paris Agreement is likely to be infeasible without the large-scale deployment of CDR technologies, and most IAM scenarios rely on the large-scale deployment of CDR technologies to have a chance of more than 50% to reach the 2.0 °C target.[3,4]

Further, CDR technologies are required to a greater extent when climate mitigation is postponed to compensate for an overshoot of greenhouse gas (GHG) emissions.[5] A wide portfolio of CDR technologies has been proposed, such as the application of biochar, enhanced weathering (EW), ocean fertilization (OF), bioenergy with carbon capture and storage (BECCS), and direct air carbon capture and storage (DACCS).[1,2,6] A CDR technology only removes GHGs from the atmosphere—*i.e.*, yields negative emissions—when the total permanent GHG removal is higher than the total GHG emissions emitted to the atmosphere through several processes required for GHG removal during the entire life cycle of that CDR technology.[6,7] Further, CDR technologies can have substantial environmental side effects, such as impacts on land, water, and/or soil.[4,6] For example, CDR technologies associated with biomass feedstock (*e.g.*, BECCS and biochar) typically result in intensive land use, soil quality changes, and water consumption, although these can be partially avoided by making use of waste-based or sustainably sourced biomass feedstocks.[8] Potential unintended side effects of other CDR technologies (*e.g.*, OF) still need to be investigated.[4,6,9] DACCS systems could largely avoid impacts on the water and food security nexus and can be considered as a technology ready for small-scale deployment.[4,6,10,11] DACCS systems aim to extract CO_2 from ambient air and permanently store the captured CO_2 in a geological storage medium.[6] One drawback of direct air capture (DAC) is the comparatively high-energy requirement

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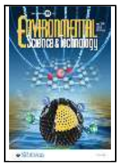

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needed for the capture of CO_2 from the atmosphere.[6,12,13] DACCS can be especially useful to capture GHG emissions and compensate for climate impacts, which are very hard to eliminate with conventional reduction measures, such as emissions from agriculture and indirect effects of aviation.[14,15] Further, DACCS enables to capture GHG emissions already emitted to the atmosphere to possibly reach net-negative global GHG emissions in the future. DACCS can therefore be a key element of a decarbonization technology portfolio. Importantly, DACCS should complement a decarbonization strategy and should not replace climate change mitigation—a transition toward renewable energy sources should be the first priority.[13,16]

DAC usually includes two steps: the adsorption (or absorption) step and the desorption (regeneration) step. During the former process, sorbents with strong absorption characteristics are used in a contacting area to bind CO_2, which is challenging due to the extreme dilute concentration of CO_2 in ambient air.[10] The regeneration process aims to regenerate the sorbents and to separate CO_2. The latter process is energy-intensive due to the requirement of heat at relatively high-temperature levels (~100 °C for low-temperature DAC).[10]

DACCS systems offer some flexibility compared to other CDR technologies since they can remove CO_2 independently from point sources in both time and space.[6,17] Consequently, optimal locations can be selected, considering CO_2 storage potential and local costs for energy supply.[13] DAC systems are usually based either on aqueous solutions with high-temperature regeneration (up to 900 °C) or on solid sorbents with a low-temperature regeneration (~100 °C).[10] This distinction is established based on the sorbent used in the CO_2 capture process and the temperature level required for its regeneration.[10] Few companies currently offer such DAC systems. Carbon Engineering (Canada) implemented high-temperature DAC systems on the North-American market.[10,18] Climeworks (Switzerland) has installed low-temperature DAC plants in Europe in Hinwil (Switzerland) and Troia (Italy), and realized the first DACCS project with negative CO_2 emissions in Hellisheiði (Iceland) in 2017.[10,19,20] Global Thermostat recently deployed pilot and demonstration plants based on low-temperature DAC in the United States.[10] Other DACCS initiatives and pilot plants usually apply the low-temperature DAC approach.[21] Generally, not all current DAC demonstration or pilot plants are designed as DACCS, *i.e.*, with permanent storage of captured CO_2, but rather connect to an industrial carbon utilization process, *i.e.*, an approach that does not allow for negative GHG emissions.

As an emerging technology with a potentially decisive role in future low-carbon energy systems, DACCS-based solutions must be thoroughly evaluated regarding their environmental performance in a transparent and consistent way over their entire lifetime to determine environmental trade-offs and to examine whether they can indeed achieve negative GHG emissions.[6] Life cycle assessment (LCA) is a suitable and flexible assessment tool to identify environmental hotspots (*i.e.*, the main contributors to environmental burdens) and to evaluate the total life cycle environmental performance of a product or service.[22,23] Only a few DACCS LCA studies, with mostly limited scopes, have been conducted so far.

de Jonge et al.[24] assessed the life cycle carbon removal efficiency of a high-temperature DAC system and determined the main environmental contributors to overall LCA scores. The contribution analysis revealed a high environmental impact due to the energy needed for the CO_2 capture process. A recent DACCS LCA study conducted by Deutz and Bardow[25] showed that two commercial low-temperature DAC(CS) plants, in Hinwil (Switzerland) and Hellisheiði (Iceland) produced and operated by Climeworks, achieved GHG removal efficiencies of 85 and 93%, respectively. Ref 25 also determined the environmental impacts of six different adsorbents and concluded that climate benefits are mainly influenced by energy sources used for CO_2 capture.

To the best of our knowledge, no comprehensive LCA of DACCS has been published in the scientific literature. We acknowledge that the study of Deutz and Bardow[25] is comprehensive for the DAC system and the associated supply chains but lacks a detailed assessment of the CO_2 storage stage since their work only considered electricity demand for CO_2 injection. However, the CO_2 storage stage includes, for example, environmental impacts from infrastructure (*e.g.*, pipelines, injection wells, and the compression station), the drilling of injection wells, and CO_2 leakage during the transportation of CO_2.[26,27] These additional processes need to be considered into the system boundaries of a comprehensive LCA to determine additional environmental impacts as well as trade-offs coming along with the transportation and storage of CO_2. Besides, the latter study excludes energy storage when intermittent (renewable) energy sources are integrated, while we believe that appropriate energy storage units need to be considered when (renewable) intermittent energy sources are used for the CO_2 capture process, which inevitably leads to additional environmental impacts. Further, the DACCS study of de Jonge et al.[24] only focused on the life cycle carbon efficiency and hence excluded other potentially important environmental impacts. In addition, the latter study reported limitations regarding the quality of their life cycle inventory (LCI) data for the DAC infrastructure.

Other available LCA studies mainly focus on DAC, thereby excluding the carbon storage stage required for permanent CO_2 removal from the atmosphere. In addition, these LCAs are simplified regarding LCA modeling choices, such as the exclusion of life cycle phases and of environmental impact categories besides climate change.[6] Further, some studies do not consider a certain amount of CO_2 equivalents removed from the atmosphere as a functional unit, which impedes the comparison of different CDR technologies.[6] Essentially, a comprehensive LCA on the entire DACCS supply chain, which assesses multiple environmental impact categories, uses an appropriate functional unit, is transparent in the methodology used, thoroughly assesses all life cycle stages, and examines a wide set of energy sources for CO_2 capture including autonomous system designs, is missing.

In this context, the contributions of this paper can be summarized as follows:

- We present a detailed and transparent LCA of a low-temperature potential DACCS system, based on Climeworks' technology and verified with the available data on low-temperature DAC, with different electricity (*i.e.*, grid and photovoltaics (PV) power) and heat sources (*i.e.*, electricity, waste, and solar heat) for CO_2 capture.

- Different innovative and autonomous system layouts are included, namely, the integration of high-temperature heat pumps (HTHPs), the integration of a Fresnel solar heat plant at locations with high solar irradiation and system layouts with electricity and heat storage.

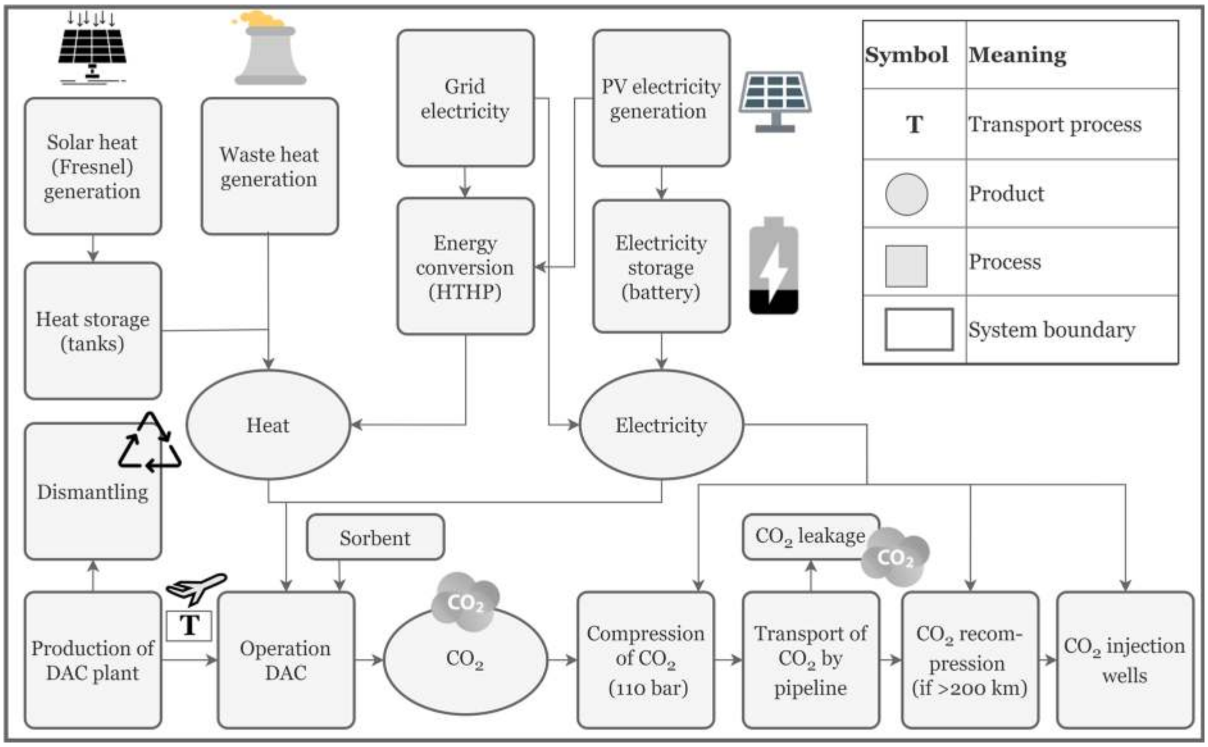

Figure 1. System boundaries of the potential DACCS product system. Note the different electricity and heat system layouts in the upper part of the figure—the different combinations/configurations of these energy sources are provided in Section 2.2. All upstream and downstream materials, services, and emission flows are included but are not shown in this figure to reduce complexity. Our functional unit is defined as the "gross removal of 1 ton CO_2 from the atmosphere *via* the use of a DAC plant combined with geological CO_2 storage". HTHP, high-temperature heat pump; DAC, direct air capture.

- We include several processes needed for CO_2 storage, parameterized on transportation distance and geographical storage location: energy needs for compression, infrastructure requirements (*e.g.*, pipelines and compression stations), drilling of boreholes, country-specific electricity for the injection of CO_2, and CO_2 leakage resulting from the transportation in pipelines.
- We address land transformation in detail: while grid-connected DACCS systems could exhibit low land requirements compared to other CDR technologies,[6] direct solar electricity and heat supply for the DAC unit might result in substantial land transformation.[4,21]
- A global analysis is included for all-electric grid-connected DACCS systems.
- We integrate two future IAM scenarios[28−30] and modify the ecoinvent background LCA database to determine the future global carbon removal potential of grid-connected DACCS systems.

The structure of the paper is as follows. Section 2 describes the methodology, where LCA modeling steps and different DACCS system layouts are discussed. Section 3 presents the LCA results, discussions, and limitations. And finally, the conclusions, implications, and recommendations of DACCS deployment are drawn in Section 3.4.9.

2. METHODOLOGY: LIFE CYCLE ASSESSMENT

LCA is a methodology that aims to quantify environmental impacts of a product or service over its entire life cycle.[22,23] LCA is standardized by the international organization for standardization (ISO), where ISO 14040 describes the general principles and framework of LCA[31] and ISO 14044 presents guidelines and general practices for LCA.[32]

2.1. Goal and Scope. Our study aims at quantifying the net carbon removal efficiency of DACCS under different boundary conditions and in various system layouts, as well as at identifying potential environmental trade-offs coming along with CO_2 removal. Elements and outcomes of our analysis should be incorporated into system-level assessments regarding the future role of DACCS in the transformation of the global economy toward net-zero GHG emissions. Our work focuses on innovative low-carbon energy sources and electrification of DACCS systems for the following reasons. Previous analysis of DAC(CS) demonstrated that impacts on climate change mainly depend on the carbon intensity of electricity and heat sources.[24,25,33] However, the overall environmental impact of the integration of low-carbon energy sources, using a wide set of environmental impact categories, has not been well examined until now. Further, we expect that the penetration of low-carbon energy sources and the electrification of energy systems will expand further in the future.[29] And finally, site-dependent boundary conditions, such as the lack of waste heat, geological CO_2 storage sites, or low-carbon electricity from the grid, might require novel system designs including heat and electricity storage allowing for (near-)autonomous DAC operation in proximity of geological CO_2 storage sites.

In our analysis, we consider the following countries, which exhibit different climates: Greece, Mexico, Jordan, Spain, and Chile are included as (semi)-arid countries with high annual solar irradiation. Besides, cooler and temperate climate regions are covered with Norway, Iceland, and Switzerland. We have selected these countries based on their geological storage

Table 1. Main Parameters per DACCS Configuration[a]

main parameters	Autonomous (Fresnel + PV)	Autonomous (HTHP + PV)	HTHP + Grid	Waste Heat + Grid	Waste Heat + PV + Battery	unit
storage medium(s)	heat + battery	battery			battery	
capture capacity	100	100	100	100	100	kt CO_2 year^{-1}
lifetime	20	20	20	20	20	years
electricity consumption (for CO_2 capture, HTHP, and compression)	690	1271	1132	614	690	kWh $t_{CO_2}^{-1}$ captured
waste or Fresnel heat consumption (for CO_2 capture)	1500	0	0	1500	1500	kWh $t_{CO_2}^{-1}$ captured
sorbent consumption	3.0	3.0	3.0	3.0	3.0	kg $t_{CO_2}^{-1}$ captured

[a] More information per system configuration is provided in Note S3, SI.

potentials,[34] difference in the grid electricity mix,[35] climate variations, and data availability for the Fresnel solar collector. A comprehensive argumentation for the selection of these countries is provided in Note S2, Supporting Information (SI).

Our functional unit is defined as the “gross removal of 1 ton CO_2 from the atmosphere *via* the use of a DAC plant combined with geological CO_2 storage”, with a reference flow of a DAC unit removing 100 kt CO_2 year^{-1} with varying system layouts and electricity and heat inputs as specified in the individual scenarios (see Section 2.2.1). Consequently, the total GHG emissions produced from all upstream and downstream DACCS activities must be less than 1 ton CO_2-eq. to result in net-negative GHG emissions, *i.e.*, a net CO_2 removal from the atmosphere.

We identify no multifunctionality of our DACCS system since the main purpose is to remove CO_2 from the atmosphere in a permanent way. Hence, allocation or system expansion is not required.[36]

We use the ILCD 2.0 (2018) life cycle impact assessment (LCIA) method[37] to assess the environmental performance of the proposed DACCS system layouts. We adopt 15 midpoint categories from ILCD in the protection areas of climate change, ecosystem quality, human health, and resources. Further, we add one additional impact category to capture water consumption with the water depletion impact category of the ReCiPe 2016 LCIA methodology (1.1 (20180117)).[38,39] Finally, we are interested in the total amount of land transformed associated with different DACCS configurations since we include different PV-coupled DACCS configurations, which could be land-intensive.[4,21] Therefore, we aggregate all life cycle inventory flows associated with land transformation (in m^2) since the considered environmental impact category “land use” of the ILCD 2.0 LCIA method represents land use impacts in terms of points and as such does not lead to meaningful land transformation surface areas. Results are shown for impacts on climate change in the main text, while the complete set of results is shown in Note S6, SI.

An attributional LCA perspective with the ecoinvent database (v3.6, system model “allocation, cut-off by classification”[40,41]) as a source of background inventories is applied. The open-source Python package Brightway2 is used to conduct our LCA.[42] Our LCIs and corresponding assumptions will be discussed in Sections 2.2 and 2.2.1. In addition to the analysis for the specific countries listed above, a sensitivity analysis shows GHG emissions of an all-electric system layout for 144 countries on a world map (see Section 3.3.2).

2.2. System Boundaries and Technical Description. We evaluate five different DAC configurations with the specific supply of heat and electricity for CO_2 capture:

(1) Electricity from the grid, heat from a waste heat source.
(2) Electricity from the grid, heat from a HTHP operated with electricity from the grid.
(3) Electricity from a PV installation, heat from a Fresnel solar-thermal heat collector.
(4) Electricity from a PV installation, heat from a HTHP operated with electricity from a PV installation.
(5) Electricity from a PV installation, heat from a waste heat source.

These configurations are detailed in subsequent sections. A simple representation of our system boundaries is shown in Figure 1. The DACCS product system includes, depending on the specific system configuration, the production and transportation of system components, energy generators, and storage units, such as the DAC plant, a Fresnel heat plant, PV systems, heat storage tanks, and batteries as well as transport and injection of CO_2 and business flights (the so-called “foreground processes”). Detailed assumptions regarding the infrastructure and dismantling of the DAC plant (based on Climeworks’ technology), sorbent used for CO_2 capture, business trips, and geological storage of CO_2 are provided in Note S5, SI.

We differentiate between two DAC units: a today’s state-of-the art unit representing Climeworks’ current technology (4 kt CO_2 captured year^{-1}) and a near-future design representing an upscale of their current standard DAC plant to capture 100 kt CO_2 year^{-1}. Energy requirements for CO_2 capture are non-confidential (obtained from Climeworks) and amount to 500 kWh $t_{CO_2}^{-1}$ for electricity (without electricity consumption for CO_2 compression) and 1500 kWh $t_{CO_2}^{-1}$ for heat (at around 100 °C).[25] The analysis in the main body of this article represents the upscaled near-future DAC unit since we expect this upscaling to take place before any large-scale roll-out. Furthermore, the DAC unit of Climeworks is a modular unit, which can therefore be easily scaled up (or scaled out).[13] To show the consequences of expected technology developments, LCA results for the current DAC unit are shown for comparison in Note S1, SI. Here, we analyze a DAC plant with an annual gross carbon capture capacity of 100 kt CO_2 and a system lifetime of 20 years.[25]

Earlier DAC(CS) LCAs showed that energy consumption for the CO_2 capture can be perceived as the crucial process in terms of environmental impacts.[24,25,33] Therefore, we evaluate system layouts with different energy sources for the CO_2 capture process. We focus on solar energy, waste heat, and all-electric system layouts for the following reasons. First, solar energy is one of the fastest-growing renewable energy sources with large potential for further expansion and comparatively low costs and can be used for both heat and electricity production.[43] Second, waste heat, if available, can be considered as the optimal heat source due to its low cost and the fact that it comes (almost) burden-free in terms of environmental impacts.[10,41] Third, further low-carbon electrification of future energy systems can be expected.[29]

Large-scale and economically attractive implementation of DACCS might, however, require remote installations close to suitable sites for geological CO_2 storage. Therefore, we introduce two autonomous off-grid system layouts entirely based on solar energy. Besides, three grid-coupled alternatives are considered. DAC operators generally aim for renewable-based energy sources and low-carbon DAC operation, so that exclusively low-carbon grid electricity is an option for them. An overview of the different system layouts (system components, capacities, lifetimes) is given in Note S3, SI. The common system lifetime is indicated as 20 years, which is the lifetime of the DACCS unit. In case of a longer lifetime of a system component, we assign a proportional fraction of the inventory of the system component to the DACCS system by dividing the common system lifetime with the lifetime of the system component. An overview of the main parameters used in the DACCS system configurations is provided in Table 1.

2.2.1. Autonomous (Fresnel + PV). The *Autonomous (Fresnel + PV)* system layout is supposed to allow for an autonomous off-grid DAC system operation entirely based on solar energy. However, solar energy is intermittent, resulting in fluctuations in power and heat output of the PV and Fresnel units.[44] These fluctuations are mitigated by two storage media: heat storage tanks and battery electricity storage. This system design enables an assessment based on the common functional unit with the same goal, *i.e.*, to capture 100 kt CO_2 annually from the ambient air. Less CO_2 would be captured without a storage medium when the same DAC capacity is installed since fluctuating electricity and heat supply would not allow for continuous DAC operation.

Solar heat can be generated with Fresnel solar collectors when sufficient solar irradiation is present.[45] Steam temperatures up to 400 °C can be achieved with Fresnel solar collectors, which makes Fresnel solar heat an appropriate heat source for industrial applications as well as for DAC systems.[46] For the desorption of CO_2, a temperature of 100 °C is required.[10] LCI of the Fresnel solar collector has been generated in collaboration with Industrial Solar (Freiburg, Germany), based on an existing Fresnel plant in Jordan. Detailed information and LCI of the Fresnel system are provided in Note S4, SI.

Site-specific annual solar irradiation is a key factor for the design and heat output of Fresnel units, and therefore, location-specific plant designs are required.[45] We use data received from Industrial Solar for eight locations in five countries with a direct normal irradiance of more than 2000 kWh m^{-2} $year^{-1}$, which were comprehensively modeled regarding their techno-economic performance. The sites considered are located in Chile (Antofagasta), Greece (Creta), Jordan (Amman), Mexico (San luis Potosi), and Spain (Tabernas).

Electricity is supplied by large-scale ground-mounted PV arrays. Therefore, country-specific LCI data sets are generated and used to represent multi-Si PV modules (see Note S5, SI). Further, we use a stationary battery system to store excess PV electricity during the daytime to be consumed during nighttime. We include a lithium nickel−manganese−cobalt oxide (NMC) battery, representing the mainstream technology for stationary electricity storage today.[47]

We assume that the NMC battery is able to store 12 h of the electricity load to provide sufficient electricity during the night for CO_2 capture and compression, in line with the heat storage sizing and the aim to capture 100 kt CO_2 $year^{-1}$. The minimum battery storage capacity requirement ($C_{bat,req}$) is calculated to capture 100 kt CO_2 $year^{-1}$ considering 12 h of storage. We oversize the NMC battery to consider battery degradation.[48] Hence, the battery capacity (C_{bat}) is determined by considering the depth of discharge (DoD), the discharge efficiency (η_{dis}), and the percentage of the original storage capacity left, required at the end of its lifetime (EoL). We use a DoD of 93%, a discharge efficiency of 94.3%, and an EoL capacity percentage of 80%.[44] Equation 1 is used to size the energy capacity of the NMC battery[44]

$$C_{bat} = \frac{C_{bat,req}}{\eta_{dis} \cdot \mathrm{DoD} \cdot \mathrm{EoL}} \tag{1}$$

Besides, we assume a C-rate of 0.5 C (*i.e.*, 2 h for battery charge or discharge) as most appropriate since the NMC battery is installed in (semi)-arid locations with high PV power peaks. Consequently, a high power capacity could be beneficial to charge during PV power peaks to avoid curtailment of PV electricity. Further, we consider additional electricity needed for system layouts with battery deployment to compensate for the roundtrip efficiency (RE)-related losses of the battery. Hence, the required PV electricity is divided by the RE (*i.e.*, 89%) of the NMC battery to function as a safety factor.[44] The latter assumption can be perceived as a worst-case scenario to be conservative, assuming that all produced PV electricity will go through the NMC battery to be used for electricity for the DAC plant.

2.2.2. Autonomous (HTHP + PV). The *Autonomous (HTHP + PV)* system layout is an all-electric off-grid system entirely supplied by PV electricity, including a HTHP to deliver high-temperature heat for the CO_2 capture process. Hence, the difference with our previous system layouts is the replacement of solar heat with heat produced by a HTHP. Further, an NMC battery is used to store PV electricity during the night; the storage capacity of the battery is calculated using eq 1. Note that the battery storage capacity is larger compared to *Autonomous (Fresnel + PV)* due to the larger electricity requirement for this all-electric system layout. We assume a coefficient of performance (CoP) of 2.9 for the HTHP, which is conservative and at the lower range of presented CoPs of HTHPs.[49] CO_2 capture *via* DAC requires heat at relatively high temperatures (100 °C), compared to heat temperatures provided by HTHP on the market today; hence, a CoP at the lower end of the range seems reasonable.[49] LCI of the HTHP has been generated by linearly scaling up a 10 kW heat pump (HP) from the ecoinvent database to the appropriate heat pump size (17 MW) to deliver sufficient instant heat for CO_2 capture. We modify the LCI of the heat pump and use CO_2 (R744) as a refrigerant, instead of R134, based on information from MAN Energy Solutions (Zürich, Switzerland), to represent current and future industrial practices of the HTHP industry.[50]

2.2.3. HTHP + Grid. The all-electric *HTHP + Grid* system layout contains a HTHP connected to the electricity grid. The same assumptions for the HTHP are used as in the previous system layout. Other energy sources are not required since grid electricity is available in all selected countries. Consequently, the environmental impact of energy consumption (predominantly) depends on the national grid electricity mix and the performance of the HTHP. Country-specific LCI datasets of the ecoinvent

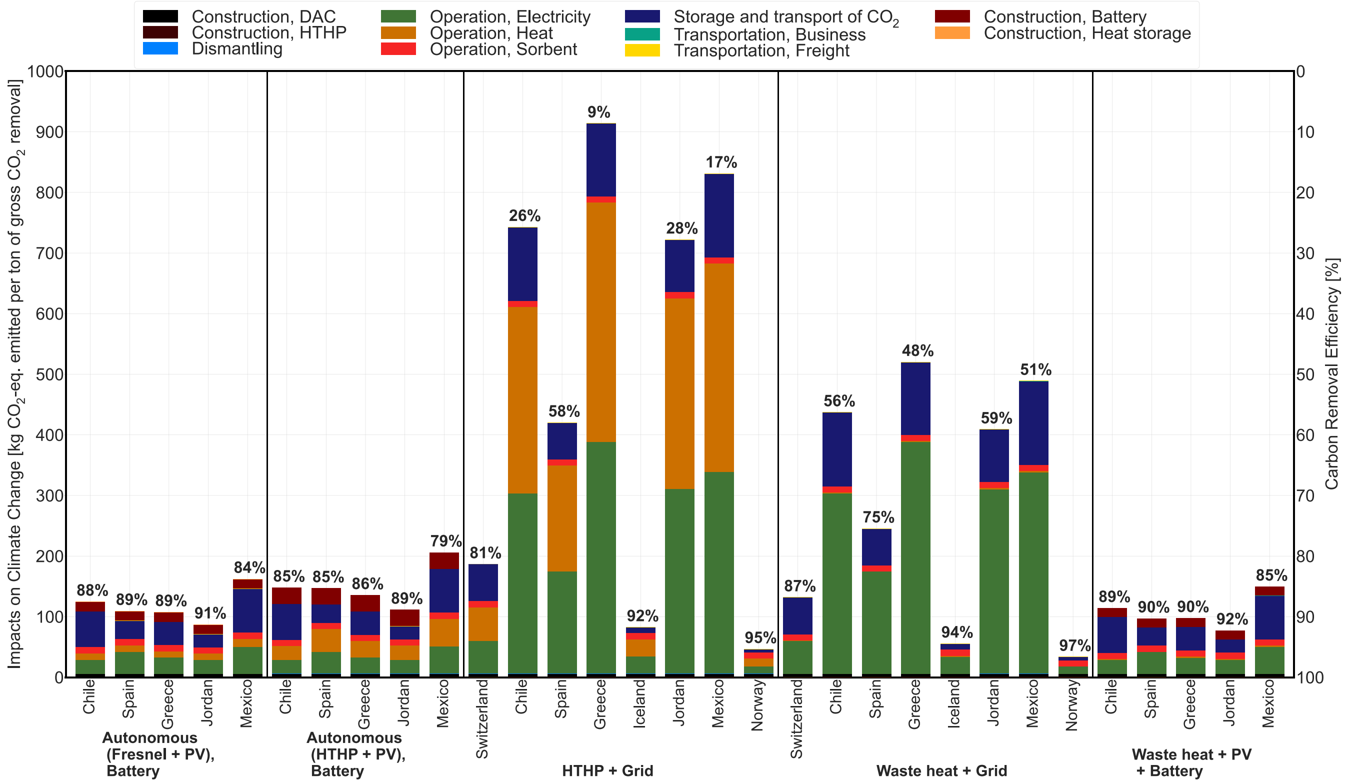

Figure 2. Life cycle GHG emissions in kg CO_2-eq. per ton of gross CO_2 removal with the DAC plant as well as carbon removal efficiencies [%] for different system layouts in selected countries. We define the carbon removal efficiency as "The share (in percentage) of net permanent GHG removal—"net" is the gross minus indirect (LCA related) emissions—of the initial gross GHG removal (100%) by the DAC unit". The size and colors of the bar segments correspond to the contributions of specific processes to the total life cycle GHG emissions. Note that "Storage and transport of CO_2" includes compression, transportation, recompression, injection, and the infrastructure requirements during the CO_2 storage stage.

database are used for grid electricity.[41] We conduct a sensitivity analysis for future electricity mixes (see Section 2.3.2).

2.2.4. Waste Heat + Grid. The *Waste Heat + Grid* system layout consumes (industrial) waste heat and is connected to the electricity grid. Note that waste heat comes (almost) burden-free in the "allocation, cut-off by classification" system model of ecoinvent.[41] The *Waste Heat + Grid* system layout is only applicable when waste heat at the correct temperature level is available, for example, generated from industrial processes. Therefore, a location-specific assessment is required to identify the potential of waste heat. For simplicity, we decided to include all eight countries with country-specific LCI from the ecoinvent database for waste heat and grid electricity.[41]

2.2.5. Waste Heat + PV + Battery. The *Waste Heat + PV + Battery* system layout consumes PV electricity and waste heat as energy sources for CO_2 capture. For waste heat, the same assumptions are used as in the previous system layout. Assumptions for the provision of PV electricity and battery storage are adopted from the *Autonomous (Fresnel + PV)* layout. Complete LCIs and additional information of all system layouts are provided in supplementary Excel files and in Note S5, SI.

2.3. Sensitivity Analysis. *2.3.1. Reduced Electricity Consumption.* DACCS is an emerging technology and will profit from technological improvements.[10] This could result in a reduction of energy consumption during the CO_2 capture process. Current figures used for energy consumption (500 kWh $t_{CO_2}^{-1}$ electricity and 1500 kWh $t_{CO_2}^{-1}$ heat[25]) are based on a very high CO_2 purity in the resulting CO_2 stream.[10] However, a lower CO_2 purity seems to be compatible with carbon capture and storage (contrary to carbon capture and utilization applications), which might result in lower energy requirements. Therefore, we examine the performance of the proposed system layouts with an electricity consumption of the CO_2 capture process reduced by 20%.

2.3.2. Electrification: HTHP + Grid Alternative. The all-electric *HTHP + Grid* system layout is further examined on a global scale to determine GHG emissions (*i.e.*, climate change) of DACCS in 144 countries. We specifically focus on this all-electric system layout in our geographical sensitivity analysis since energy system models predict an increase of low-carbon electrification in future energy systems.[29] Besides, this system layout is the simplest and (theoretically) could be implemented in all locations with a (low-carbon) grid connection providing sufficient electricity, as grid electricity is the only energy source needed. Climeworks has committed, and probably also other DAC suppliers, to only offer carbon dioxide removal services with grid-coupled DAC systems at geographical locations with low GHG-intensive grid electricity.

For simplicity, we exclude environmental impacts for transportation and business trips in this sensitivity analysis since GHG emissions from transportation processes are small according to our results. Further, we use average electricity supply (*i.e.*, "market group for electricity,...") for countries that are modeled with multiple regional electricity data sets in the ecoinvent database. In case there is no country-specific electricity data set available, we use the market (group) for electricity for the geographical area in which this country is located as approximation. Further, we assume a generic

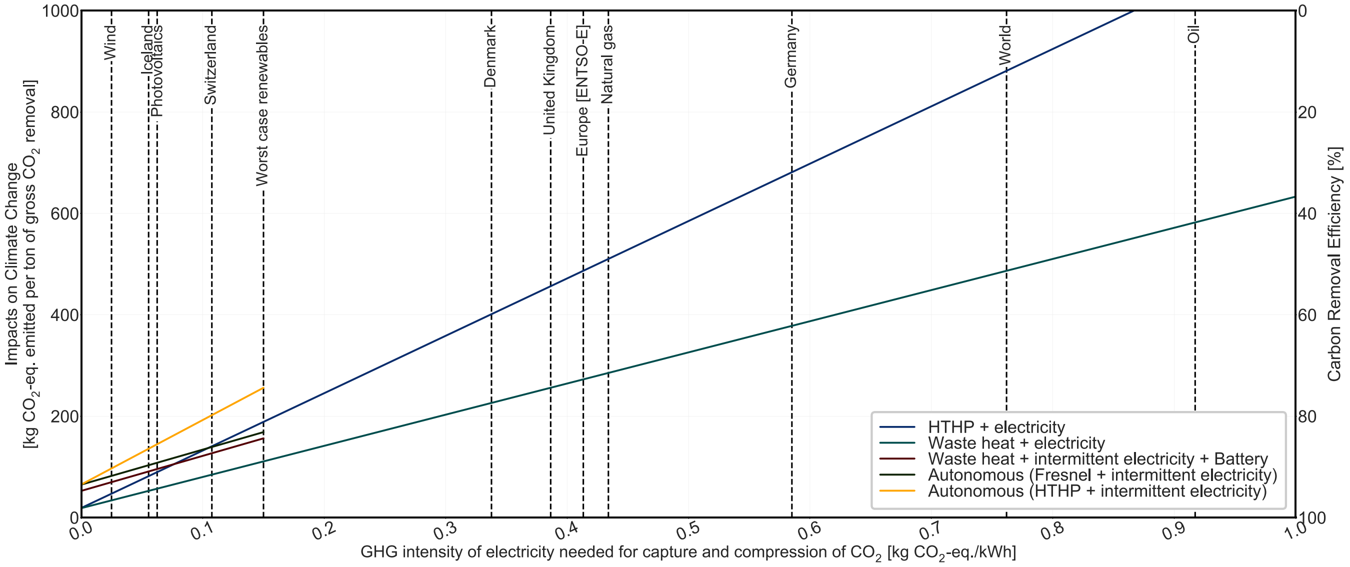

Figure 3. Effect of the GHG intensity of electricity used, needed for capture and compression of CO_2, for all alternatives on the overall GHG emissions of DACCS systems. We use the LCI of Norway as a proxy for *HTHP + electricity* and *Waste heat + electricity*. Further, LCI of Spain is used for the following configurations: *Waste heat + intermittent electricity + Battery, Autonomous (Fresnel + intermittent electricity)* and *Autonomous (HTHP + intermittent electricity)*. Note that the *Autonomous (Fresnel + intermittent electricity)* alternative can only be installed in (semi)-arid locations due to the requirement of high annual solar irradiation. Further, the waste heat alternatives can only be installed with a sufficient supply of waste heat. The DAC configurations operated with intermittent electricity are shown up to a GHG intensity of an electricity supply of 0.15 kg CO_2-eq. kWh^{-1}, which represents a worst case for renewables. The countries and specific power generation technologies on the *x*-axis represent the GHG intensities of national electricity grids and technologies according to ecoinvent (see Note S9, SI).[41]

transportation distance for CO_2 of 500 km to the injection wells and a generic storage depth of 2000 m.

2.3.3. Future Electrification: HTHP + Grid Alternative. Projections of IAMs show that electricity grid mixes will become less CO_2-intensive even in the most carbon-intensive energy scenarios.[28,29] Therefore, we examine GHG emissions of the *HTHP + Grid* system layout based on future grid electricity mixes for 2040. To achieve this, we use these future grid mixes for electricity supply for the DAC unit by modifying the ecoinvent background database with the rmnd-lca Python package,[51] adapting future electricity mixes in our background database, based on the output figures of the REMIND model scenarios.[28] Note that this results in geographically aggregated future electricity datasets since the REMIND model subdivides the world into only 11 regions.

First, we use the "SSP2-base" energy scenario to determine future GHG emissions of the *HTHP + Grid* system layout. The "SSP2-base" scenario is a scenario with no additional climate policy.[28,51] Second, we include a more ambitious future climate policy with the "SSP2-PkBudg1300" energy scenario, which corresponds to a maximum average temperature increase of 2 °C.[51]

3. RESULTS AND DISCUSSION

3.1. Assessment of Climate Change Impact. The outcome of the assessment of the "Impacts on Climate Change" of DACCS technologies for the different system layouts and countries considered is illustrated in Figures 2 and 3 (the reader will best appreciate the color figures). The former figure is a bar chart plotting the climate change impact in kilograms of CO_2 emitted per gross removal of 1 ton CO_2 from the atmosphere *via* the use of a DAC plant (left vertical axis) as well as the net carbon removal efficiency (right vertical axis, increasing from top to bottom, and the figure reported above each bar); each bar consists of the overall impact, whereby the life cycle contributions of 11 different upstream and downstream components of the value chain are identified with different colors. The latter figure plots the (linear) dependence of the climate change impact (and of the carbon removal efficiency) on the GHG intensity of electricity generation for the different system layouts; the different slopes of these lines reflect the electricity demand of each specific system layout (and its corresponding impact on climate change), whereas the vertical gray dashed lines indicate the GHG intensities of different electricity generators and national grid electricity mixes according to the ecoinvent 3.6 database.[41]

Note that for all cases, considered in Figure 2, the climate change impact is smaller than the amount of CO_2 removed and stored, *i.e.*, negative emissions are indeed generated in all cases. However, the range of carbon removal efficiencies is large, going from only 9% all the way to 97%. It is worth noting that carbon removal efficiency has a strong impact on unitary costs per net CO_2 removed. If we assumed that the two value chains leading to 9% and to 97% removal costed the same per ton CO_2 removal, say 100 monetary units, then the unitary cost per net CO_2 removed would be 1111 and 103 monetary units, respectively. Additionally, the carbon removal efficiency can be strongly dependent on the GHG footprint of electricity generation, as illustrated in Figure 3. The main findings can be summarized as follows.

Energy supply—heat and electricity used—for CO_2 capture are the key factors driving GHG emissions of DACCS configurations. Waste heat is environmentally attractive as a heat supplier when available, as waste heat comes (almost) burden-free in the "allocation, cut-off by classification" system model. Please note that this is a consequence of our choice of this system model and that the availability and environmental benefit of using waste heat should be determined on the actual case-specific scenario of waste heat provision (please refer to

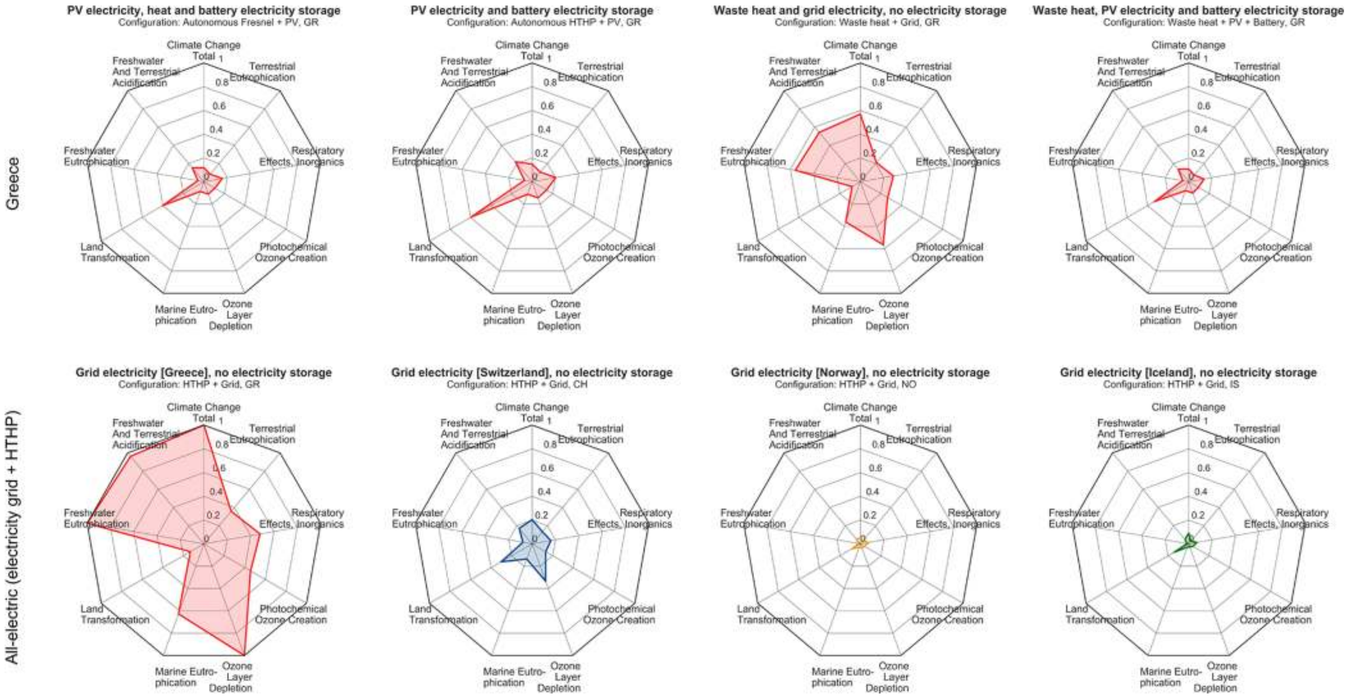

Figure 4. Spider graphs illustrating environmental trade-offs for a selection of DACCS configurations on nine environmental impact categories with a focus on Greece and all-electric DACCS configurations (please refer to the main body of the text for an explanation of this selection). Complete spider graphs considering all DACCS system configurations and environmental impact categories are provided in Note S6, SI. The absolute maximum impacts for each environmental impact category are also provided in Table S6 in Note S6, SI.

Section 3.4.6 for more discussion). GHG emissions generated from energy consumption can be reduced by using renewable energy: in our analysis, solar energy in countries with high annual solar irradiation.

The GHG intensity of the grid electricity mix is a crucial factor for grid-coupled DACCS configurations and is very country-specific. Consequently, countries with low GHG-intensive electricity mixes in combination with waste heat exhibit lowest GHG emissions, with GHG removal efficiencies of 97% and 94% for Norway and Iceland, respectively. On the contrary, high GHG emissions are generated, almost approaching net-positive GHG emissions in Greece with 0.91 ton CO_2-eq. per ton of gross CO_2 removal, for grid-coupled DACCS configurations when the national electricity grid relies on a large share of fossil fuels. Figure 3 shows that the break-even point, in terms of net-negative GHG emissions for the grid-coupled HTHP DACCS configuration (*HTHP + Grid*), is on ∼0.87 kg CO_2-eq. kWh^{-1} electricity used for the capture and compression of CO_2.

In absolute terms, minor GHG emissions are generated from the DAC construction (6 kg CO_2-eq. per ton of gross CO_2 removal) and sorbent consumption (10 kg CO_2-eq. per ton of gross CO_2 removal). However, these components can be substantial in relative terms if energy supply for CO_2 capture is clean in terms of GHG emissions. The relative climate change impact of transportation processes directly related to DAC (*i.e.*, business and freight transport for the collectors and process units) can be considered small, with contributions of less than 1%.

GHG emissions from the storage and transport of CO_2 are significant, and their relative contribution fluctuates between 11% and 52%, mainly driven by electricity requirements for the compression, recompression, and injection of CO_2 as well as CO_2 leakage during long-distance CO_2 transport, respectively. Note that we included the medium scenario presented in Holloway et al.[27] for CO_2 leakage during transportation. The contribution of CO_2 leakage during the transportation of CO_2 in pipelines would increase by a factor of 10 if we used their pessimistic scenario for CO_2 leakage, which emphasizes the need for a well-designed pipeline network for CO_2 transportation. A longer transportation distance of CO_2 logically results in a higher probability of CO_2 leakage.

DACCS-related GHG emissions in countries with a CO_2-intensive grid electricity mix can be substantially reduced by shifting to renewable energy sources as energy suppliers and can be installed as (nearly) autonomous DACCS configurations. The GHG removal efficiencies of autonomous system layouts, entirely supplied by solar energy, are between 79% and 91% and are therefore at a similar, or even higher, level as grid-connected DAC in countries with rather low-carbon electricity supply and available waste heat, such as Switzerland with a GHG removal efficiency of 87%.

Autonomous system layouts with a Fresnel solar collector exhibit slightly higher GHG removal efficiencies (84−91% for *Autonomous (Fresnel + PV)*) in comparison with autonomous DACCS configurations, entirely based on PV arrays in combination with a HTHP (79−89% for *Autonomous (HTHP + PV)*), mainly due to the provision of lower carbon heat with a Fresnel solar collector and the requirement of a smaller battery (∼125 *vs* ∼221 MWh). A bigger battery is required for autonomous configurations with a HTHP as electricity needs to be stored to provide sufficient electricity to the HTHP during night. Further, the production of heat storage tanks (made of low-alloyed steel) causes lower GHG emissions compared to that of battery energy storage systems, which means lower GHG emissions for DACCS configurations with Fresnel heat.

The production of electricity storage (NMC battery) media can, in relative terms, have a significant impact on GHG emissions, from 9% to 24%, while that of the production of the heat storage (steel tanks) units is small with a relative contribution of less than 1%. Consequently, GHG emissions from the infrastructure of autonomous systems are comparably higher (see the lower-left part of Figure 3) due to the impact of the production of energy storage units, such as battery energy storage systems. Figure 3 illustrates how alternatives with waste heat consumption, and the autonomous system layout coupled with Fresnel heat, are less sensitive when using high GHG-intensive electricity due to less electricity consumption required for CO_2 capture. For example, the autonomous DACCS configuration with Fresnel heat generates solar heat and therefore less electricity is required for CO_2 capture, although this alternative can only be installed at locations with sufficient annual solar irradiation.

3.2. Assessment of Other Environmental Impact Categories. Figure 4 illustrates the overall environmental burdens and trade-offs for a selection of DACCS configurations and environmental impact categories. The spider graphs show the normalized scores (scores from 0 to 1 visualized with the colored line) of DACCS configurations on nine environmental impact categories, normalized to the system configuration with the highest environmental impact among all configurations (*i.e.*, highest score = 1, presented in Note S6, SI) on a specific environmental impact category. A greater colored area therefore demonstrates an overall higher environmental burden for a specific DACCS configuration.

Environmental impact categories, presented in Figure 4, are selected based on recommendations by Fazio et al.[37] We only show impact categories of recommendation levels I and II in the main article and add land transformation since these are important for CDR technologies in general.[6] Greece is included as a reference and worst-case scenario regarding GHG emissions. All-electric DACCS system configurations, *i.e. HTHP + Grid* electricity, exhibit a large sensitivity regarding overall environmental impacts between countries. We therefore include three other countries with cleaner electricity mixes in terms of GHG emissions, Switzerland, Norway, and Iceland, since Climeworks has committed to only offer carbon dioxide removal services with grid-coupled DAC systems at geographical locations with low GHG-intensive grid electricity. The complete set of spider graphs and results for all DACCS configurations and environmental impact categories is provided in Note S6, SI.

In general, the lowest overall environmental burdens can be achieved with DACCS configurations with waste heat in combination with renewable electricity supply, for example, in Norway and Iceland. Fundamental differences between environmental impact categories and countries are found for grid-coupled DACCS configurations in combination with a HTHP. This is illustrated in the second row of Figure 4. Low overall environmental burdens can also be achieved with grid-coupled DACCS configurations, in combination with a HTHP *(HTHP + Grid)*, when the national grid electricity mix is based on a large share of renewables as, for example, in Norway and Iceland, which both rely on a high share of hydropower. Switzerland has slightly higher overall environmental burdens compared to Norway and Iceland, mainly due to a higher reliance on nuclear power and fossil fuels, respectively.

On the contrary, substantially higher environmental burdens are generated from grid-coupled DACCS configurations when the national grid electricity mix is based on a large share of fossil fuels as, for example, in Greece (see Figure 4). This generally results in high environmental burdens on almost all environmental impact categories.

Autonomous DACCS configurations generate low environmental burdens on almost all environmental impact categories, although several environmental trade-offs occur. The most important one concerns land use, which can be considered as the main "outlier" among all of the impact categories: autonomous DACCS layouts with solar energy supply perform, in general, worse than grid-connected systems on land transformation (see the first two spider graphs in the top row of Figure 4). Figure S5, in Note S7 (SI), presents a detailed contribution analysis on land transformation (in m^2) and reveals that autonomous energy systems exhibit large land transformations (up to 1.87 m^2 per ton of gross CO_2 removal), mainly due to large surface area requirements for ground-mounted PV panels and the Fresnel heat collector, while the DAC unit as such exhibits very low land transformation impacts (a life cycle land transformation of 0.01 per ton of gross CO_2 removal). In addition, autonomous energy systems exhibit comparatively high environmental scores on the mineral and metal impact categories (see Note S6, SI), mainly resulting from material requirements for the production of PV arrays and the NMC battery. Note that this impact category is associated with substantial uncertainties and also methodological shortcomings and is therefore assigned with recommendation level III.[37]

These examples demonstrate the importance to assess and compare system layouts on a wide set of environmental impact categories to examine environmental trade-offs. Additional figures regarding the contribution analysis of other environmental impact categories, of recommendation levels I and II, are provided in Note S10, SI.

3.3. Sensitivity Analysis. *3.3.1. Reduced Electricity Consumption.* Figure S6, in Note S8 (SI), demonstrates the absolute change in the climate change impact category when the electricity consumption for the CO_2 capture process is reduced by up to 20%. Such an efficiency improvement would result in substantial reductions of impacts on climate change for system layouts, which consume large amounts of CO_2-intensive electricity (*i.e., HTHP + Grid* in Greece and Mexico). In contrast, the environmental impact of such an electricity reduction is hardly visible for configurations using low-carbon energy sources. Nevertheless, reducing pressure on the electricity grid requires increased efficiency of any technology, including DACCS.

3.3.2. Electrification: HTHP + Grid Alternative. Figure 5 illustrates the performance on the climate change impact category for the *HTHP + Grid* system layout for 144 countries. Net-negative GHG emissions can be obtained when life cycle

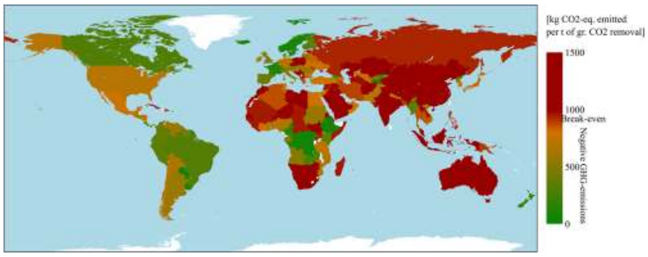

Figure 5. Country-specific results for the climate change impact category for the *HTHP + Grid* system configuration. Green and orange indicate net-negative GHG emissions, while dark red shows net-positive GHG emissions of DACCS.

GHG emissions are lower than 1000 kg CO_2-eq. per ton of gross CO_2 removal. Countries that fulfill this requirement are shaded in green and orange (*i.e.,* orange meaning higher GHG emissions than green). Net-positive GHG emissions are shown in dark red.

Significant variations between countries can (mainly) be explained by the difference in the CO_2 intensity of national grid electricity mixes. For the *HTHP* + *Grid* system layout, the GHG break-even point is reached with a grid electricity mix GHG intensity of ~0.87 kg CO_2-eq. kWh^{-1} electricity (see also Figure 3 for the break-even point), which means that grid electricity mixes with a lower GHG intensity than the GHG break-even point exhibit GHG removal from the atmosphere. It turns out that most countries in Europe, North America, South America, and central Africa already show large GHG removal potentials with DACCS *HTHP* + *Grid* system layouts today. However, few countries in these continents exhibit net-positive GHG emissions. In general, Australia and countries in Asia, Southern Africa, and Northern Africa are nowadays not (or less) suitable to install DACCS *HTHP* + *Grid* systems.

3.3.3. Future Electrification: HTHP + Grid Alternative. Figure 6 demonstrates the possible future impact (in the year 2040) regarding the climate change impact category for the *HTHP* + *Grid* system layout according to the "SSP2-base" scenario of the REMIND model,[28,30] which extrapolates observed trends but is not particularly ambitious in terms of climate policy. It turns out that the deployment of *HTHP* + *Grid* DACCS system layouts could result in net-negative GHG emissions in all world regions by 2040. North America, South America, Europe, Australia, and Africa exhibit a large net GHG removal potential, while India and Asia seem to be less suitable due to their still relatively high CO_2-intensive grid electricity mix.

When implementing a more ambitious climate scenario (*i.e.,* "SSP2-PkBudg1300"), DACCS exhibits GHG emissions of less than 230 kg CO_2-eq. per ton of gross CO_2 removal by 2040 in all regions. Hence, all regions would be green if the world map for this case were drawn using the same color codes as used for Figures 5 and 6. Therefore, effective climate policy would be very beneficial for all-electric DACCS system layouts connected to the grid. The suitability of grid-coupled DACCS configurations is highly dependent on the carbon intensity of the future electricity mix.

Note that in these two prospective evaluations we not only use future regional electricity mixes for electricity supply to the DACCS unit but also populate the scenario-specific development of the electricity sectors throughout the entire background LCA database. This procedure results, for example, in a reduction of GHG emissions due to PV electricity since the electricity used for PV wafer production exhibits a scenario-specific GHG intensity lower than today's GHG intensity.

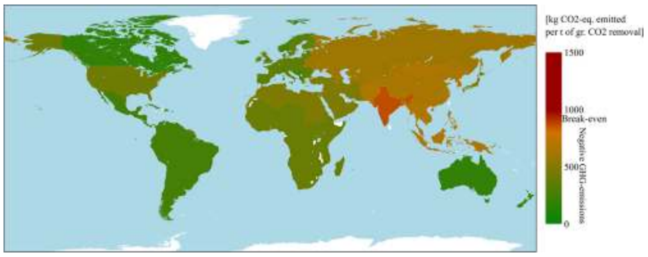

Figure 6. Results for the climate change impact category for the *HTHP* + *Grid* alternative according to the "SSP2-base" scenario. Green and orange colors indicate net-negative GHG emissions, while dark red shows net-positive GHG emissions.

3.4. Discussion: Limitations, Solutions, and Future Work. Our analysis shows that within the LCA of DACCS systems, there are few elements and key factors, which determine the results: first of all, the energy required for DAC operation and the sources of supply. While we consider data availability and quality regarding energy demand of and supply for the CO_2 capture process to be crucial, there are other issues that call for further analysis in the future. More sophisticated modeling integrating the operational experience of DAC units of different scales and at different locations to be gained in the future will allow for a more precise representation of the environmental burdens due to DACCS. We discuss the most important of these elements in the following subsections.

3.4.1. High-Temperature Heat Pumps (HTHPs). We included a HTHP in different DACCS system layouts. Currently, no complete LCA of HTHP has been conducted. We received information regarding potential future refrigerants used in HTHPs from MAN Energy Solutions (Zürich, Switzerland) but had to establish most of our LCI as an extrapolation of a low-temperature HP. Future analysis should consider the utilization of different types of refrigerants since refrigerants usually exhibit large contributions to environmental burdens caused by heat pumps.[49,52]

3.4.2. Assessment of Fresnel Heat Plant in Other Locations. Only five potential locations for the Fresnel solar collector were included in the analysis due to the site-specific performance of Fresnel solar collectors and limitations on data for further locations. The Fresnel performances at these locations in semi-arid climate regions were thoroughly modeled by Industrial Solar (Freiburg, Germany). Since the integration of Fresnel solar heat systems turned out to be beneficial from the environmental perspective, especially for autonomous DACCS systems, site-specific assessments and modeling should be expanded to determine the environmental merits in other geographical locations.

3.4.3. Competition between Electricity Sources. Future energy systems tend to show higher reliance on electricity than current energy systems. Additional electricity will be required by the mobility and heat sector, possibly on top of increased power demand due to population growth and higher electrification rates. Increasing the pressure on the power sector through the installation of grid-coupled DACCS systems competes with other demands of electricity. This was not a topic of our research but has to be considered.

Note that country-specific results for grid-connected DACCS represent single units within the current energy system "as of today". Further, the installation of DACCS should always come along with the installation of new low-carbon electricity generation capacities to avoid the installation of fossil-fuel-based electricity grid capacity.

3.4.4. Battery Storage in (Semi)-Arid Countries. We proposed to install NMC batteries in (semi)-arid countries (*i.e.,* Chile, Spain, Greece, Jordan, and Mexico) with high ambient air temperatures. Temperature levels within these countries could easily reach more than 40 °C during warm periods. The acceptable temperature range for lithium-ion batteries is between −20 and 60 °C, with an optimal operation temperature between 15 and 35 °C.[53] Therefore, location-specific measures should be considered when outside temperatures approach critical temperature levels to avoid battery damage. These measures could result in additional energy and/or material requirements, *i.e.*, higher environmental impacts.

3.4.5. Autonomous System Layouts. Two autonomous system layouts, entirely based on solar energy supply, were included: *Autonomous (Fresnel + PV)* and *Autonomous (HTHP + PV)*. Twelve hours of energy storage capacity was assumed for storage media for these two system layouts. This led to a large storage capacity needed for both the battery system (~125 and ~221 MWh, respectively) and the heat storage tanks.

Alternatively, a doubling of the DAC capacity (to comply with our functional unit) with discontinued operation overnight could be installed to reduce the need for energy storage in the proposed two autonomous system layouts. However, we emphasize that such a system with doubled DAC capacity would still require energy storage for intermittent (renewable) electricity generation, which would result in additional environmental impacts from the production of storage media. Further, a doubling of the capacity of the DAC plant would incur large additional capital expenditures for the DAC plant since capital expenditures of DAC systems are still high.[10] Therefore, we argue that such a DAC system is currently unrealistic from an economic and technological point of view, and further research is required for a complete assessment of the operation of such an autonomous DAC system.

Further, more sophisticated research is needed to confirm the self-sufficiency, *e.g.*, the 12 h of energy storage, of the proposed autonomous system layouts. For the *Autonomous HTHP + PV*, the environmental and economic performance can possibly be (slightly) improved with the installation of a heat storage medium to reduce the size of the battery. In this way, heat supply, needed for the CO_2 capture process, can be stored in a heat storage medium instead of electricity storage in a battery.

In addition, autonomous system designs with DAC units entirely powered by solar energy require site-specific assessments in terms of land use. The area covered by PV arrays required for electricity supply for a DAC unit removing 100 kt CO_2 year^{-1} in our most land-intensive case and location (*i.e.*, *Autonomous HTHP + PV* in Mexico) amounts to a direct land transformation (for the DAC unit and PV panels considering their lifetimes) of almost 4.7 km^2. Such a land surface area will not always be available. The scale-up of solar PV-coupled DACCS to the gigatonne scale, for example, removing 1 Gt CO_2 year^{-1} from the atmosphere with a GHG removal efficiency of 79% (representing worst-case *Autonomous HTHP + PV* for Mexico), would therefore require almost 12 700 DACCS plants (of 100 kt CO_2 year^{-1}) in combination with a direct land transformation area as large as ~59 000 km^2, which is equal to ~1.5 times the total land surface area of Switzerland. Such land area, material requirements, and availability of potential sites for the DAC infrastructure can limit the large-scale implementation of DACCS and needs to be further investigated on a global level.

Land surface area requirements can be significantly reduced by, for example, using grid electricity sources, although this would result in indirect land use transformations mainly generated from electricity generators. Note that the land transformation for PV panels includes area requirements for the open-ground power plant including the space between the modules and some surrounding areas[41] and can therefore be considered as conservative. Further, the land between and underneath the PV modules may be used for grazing of small livestock such as sheep or poultry, bee-keeping, or other innovative agricultural uses. Such land use options should already be considered during the design phase of the solar field.

3.4.6. Location-Specific Limitations: Waste Heat and Geological Storage Formations. Our analysis demonstrated the best environmental performance for DACCS configurations coupled to waste heat usage. Location-specific assessments are required to identify the availability of waste heat from industrial applications since the availability of waste heat is constrained for the following reasons. First, waste heat is usually a constrained co-product from fuel combustion. Second, the future decarbonization of the energy system with a reduced combustion of fossil energy carriers will likely result in a further reduction of waste heat sources, in general, and/or at the required temperature level. Third, waste heat could be inaccessible in (for example) remote locations. And fourth, there are competing uses. Therefore, we provide different options to replace waste heat; both grid-coupled and autonomous DACCS energy systems provide a good alternative for waste heat in certain case-specific situations and should be considered in the early phase of deploying DACCS systems. It is worth noting that waste heat comes burden-free in the attributional system model “allocation, cut-off by classification” of the ecoinvent database and that alternative LCA approaches (such as an economic allocation) would result in higher environmental burdens. Our DACCS configurations with waste heat usage should therefore be considered as the best option regarding overall environmental impacts.

Another site-specific boundary condition of DACCS is the availability of geological storage sites. We therefore examined different transportation distances from CO_2 capture to the injection wells, although the transportation distance of CO_2 can be minimized, to avoid CO_2 leakages from the transportation of CO_2, by installing the DAC facility close to geological storage sites. However, the large-scale deployment of DACCS requires large geological storage potentials and therefore CO_2 transportation might be required. This could result in competition for geological storage sites between DACCS and, for example, carbon capture and storage from conventional power plants, which could limit the large-scale implementation of DACCS.

3.4.7. Future Electrification: REMIND Regions. We used the outputs of the IAM REMIND[28] for quantification of future GHG intensity of electricity supply and to modify our background LCA database to assess the future performance of all-electric DACCS systems. However, the geographical resolution of the REMIND model is limited to 11 world regions. Therefore, the future environmental potential of the electrified system layouts had to be aggregated to those regions, while our results for current systems showed that regional differences can be significant in terms of GHG emissions (see Figure 5). Further geographical disaggregation would be beneficial for prospective LCA of DACCS, especially for all-electric system layouts. However, our findings demonstrate very well how effective climate policy could improve the environmental performance of all-electric DACCS systems. On the other hand, the decarbonization of the power supply system could lead to additional environmental impacts on other life cycle environmental impact categories. Luderer et al.[54] investigated the benefits and side effects of the decarbonization of power supply.

3.4.8. Comparison with Literature on DACCS. The single purpose of DACCS systems is to remove CO_2 from the atmosphere in a permanent way. Our analysis focused on low-temperature DACCS systems and must not be considered as a representative for high-temperature DACCS systems, due to the need of different processes for CO_2 capture and their associated energy requirements.[10,12]

One recent DACCS LCA study of Deutz and Bardow[25] examined the same low-temperature DAC technology of Climeworks although focused on a smaller DAC plant (with a capture capacity of 4 kt CO_2 year^{-1}) and therefore provided less detailed results for the larger DAC plant (with a capture capacity of 100 kt CO_2 year^{-1}). Their GHG removal efficiencies are generally slightly higher compared to the reported GHG removal efficiencies in this work. For example, Deutz and Bardow[25] found a carbon removal efficiency of 88.8% for Switzerland, consuming the same amount of grid electricity and waste heat as energy sources for CO_2 capture as our analysis, for the 4 kt CO_2 year^{-1} DAC facility. The CO_2 removal efficiency can be expected to be higher for the 100 kt CO_2 year^{-1} DAC facility due to reductions of sorbent consumption as well as the optimization of the DAC infrastructure (see Section 2.2 and Note S1, SI). Reported GHG emissions of the carbon storage phase turned out to have a minor contribution with less than 2% on total GHG removal efficiencies for DACCS configurations with GHG removal efficiencies higher than 86%.

On the contrary, our results show a lower GHG removal efficiency of 86.8% for the waste heat and grid electricity configuration (*Waste Heat + Grid*) in Switzerland, (even) for a DAC plant with a capacity of 100 kt CO_2 year^{-1}. First, this can be partly explained by the application of different background LCA data: ecoinvent 3.6 in our analysis *vs* both Gabi and ecoinvent 3.5 in ref 25. Second, our analysis includes a more thorough (and conservative) analysis regarding the transportation and storage of CO_2, while ref 25 expected sufficient CO_2 storage sites close to the DAC facility as well as the utilization of existent infrastructure for CO_2 storage; they therefore excluded CO_2 transportation and infrastructure requirements for geological storage sites. However, the availability of geological storage sites for DACCS can be limited due to geographical availability as well as the competition with conventional CCS and other CDR technologies, such as BECCS (see Section 3.4.6). For Switzerland, CO_2 leakage during transportation (1500 km) reduced the GHG removal efficiency already by more than 2%, while the compression, recompression, and injection of CO_2, as well as infrastructure requirements, further reduced the GHG removal efficiency to a total of 6% in our analysis.

In addition, the work of Deutz and Bardow[25] reported a generic land use requirement of 4450 km^2 (3683 DAC plants with a capture capacity of 100 kt CO_2 year^{-1}, with 1.21 km^2 per DAC plant) for PV-coupled DACCS to capture 1% of global CO_2 emissions in the year 2019 (0.368 Gt CO_2[25]). Our main environmental trade-off was found for land transformation, manifested by the big range of both direct and life cycle land transformations between DACCS configurations and countries. The worst-case land transformation scenario in our analysis demonstrated much higher land use requirements for autonomous PV-coupled DACCS systems, almost 4.7 km^2 per DAC facility (with a capacity of 100 kt CO_2 year^{-1}), mainly due to the lower annual solar irradiation for this specific location (and for the reason given in Section 3.4.5).

Since our analysis and this comparison show that specific DACCS system configurations at different locations exhibit very different environmental performances and that transport and storage of CO_2 can cause substantial burdens, we argue to perform site-specific assessments of DACCS configurations during the design phase, including a thorough analysis of the transportation and storage of CO_2 as well as a location-specific assessment of land use requirements.

3.4.9. Comparison with CDR Technologies. We demonstrated that the comparison of DAC(CS) LCA results with other CDR technologies should consider the variability of the life cycle GHG removal efficiency. This is, however, not always the case, and the presentation of the GHG removal performance of DAC(CS) as one single number can be misleading (*e.g.*, by Müller et al.[36] for DAC).

Further, DACCS systems should be compared with other CDR technologies on the same functional unit, to evaluate benefits and potential trade-offs of various CDR technology options.[6] We propose to compare CDR technologies per unit of CO_2 removal from the atmosphere. Unfortunately, we are currently not able to present such a comparison between CDR technologies, as demonstrated in Terlouw et al.,[6] due to the immature research state and several limitations of current LCA research on CDR technologies, in general.[6]

3.5. Implications and Recommendations for DACCS Deployment. To conclude, the results demonstrated that energy supply—heat and electricity—used for CO_2 capture are the key factors driving GHG emissions of DACCS configurations. All eight selected countries exhibited net-negative GHG emissions on all proposed system layouts, meaning that GHG emissions associated with energy supply and material demand for DACCS are below the amount of gross CO_2 removal, equivalent to a net removal of CO_2 from the atmosphere. However, the level of "net-negativity" showed a substantial variation between DACCS layouts and countries of application: while in our best case, 97% of the captured CO_2 is permanently removed from the atmosphere, our worst case resulted in a life cycle GHG removal rate of 9%. The best climate change-related performances were achieved by system layouts using waste heat, and in countries with low CO_2 intensities of the national electricity grid. The CO_2 intensity of the national grid electricity mix turned out to be the crucial factor for grid-coupled system layouts.

Autonomous DACCS layouts, which consume solar energy, are from an environmental perspective (except for land use considerations) promising alternatives in regions where the grid electricity mix relies on a high share of fossil fuels and at remote locations without grid access (potentially close to CO_2 storage sites). Therefore, we recommend solar-based autonomous DACCS systems for countries with (semi)-arid climates, which have a CO_2-intensive grid electricity mix and the required land area available. All-electric DACCS system layouts are recommended when new low-carbon electricity generation capacity can be installed and when the national grid electricity mix relies on low-carbon electricity sources. Further contributions to life cycle GHG emissions of DACCS, associated with, for example, DAC infrastructure, CO_2 transport and storage, as well as energy storage units, can be substantial in relative terms if energy supply for CO_2 capture is clean in terms of GHG emissions, but absolute GHG emissions due to these contributions are rather small. The assessment of a wide variety of environmental impact categories, in addition to impacts on climate change, showed different trade-offs on a few environmental impact categories, especially regarding land transformation associated with PV-coupled DACCS configurations. This observation confirms the importance of a comprehensive LCA approach not only focusing on climate change.

The sensitivity analysis demonstrated the large variation of GHG emissions between countries for all-electric DACCS systems. Hence, selecting inappropriate locations in countries with CO_2-intensive grid electricity mixes could lead to net GHG emissions instead of GHG removal. Consequently, the operation of all-electric DACCS systems with fossil-fuel based grid electricity mixes should be avoided, and we recommend to assess the suitability of DACCS systems based on site-specific conditions, such as the availability of (renewable) energy sources as well as land, waste heat, and the potential of carbon storage sites. Further, our prospective analysis demonstrated that more ambitious climate policy will have beneficial effects on GHG emissions for all-electric DACCS systems.

Finally, we demonstrated significant carbon removal efficiencies of DACCS, although with a large dependency on the energy sources used for CO_2 capture. We therefore argue to utilize low-carbon energy sources for CO_2 capture and to compare alternative potential DACCS configurations with a comprehensive and transparent LCA approach. Such an approach enables policy-makers to select the most environmentally friendly DACCS configuration under given local boundary conditions. Besides environmental aspects, effective policy is required to facilitate the development of the DAC industry to improve the economic viability of DACCS. With sufficient geological storage potentials and an economically viable DAC system, we foresee DACCS as a key component in the pathway toward net-zero GHG emissions.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.1c03263.

> Verification of information obtained from Climeworks and corresponding contribution analysis (regarding impacts on climate change) of the DAC infrastructure (plant) (Note S1); selection of locations for DACCS system configurations (Note S2); overview table of energy system layouts (Note S3); life cycle inventory: additional information Fresnel solar collector (Note S4); LCI - DACCS alternatives and additional information (Note S5); LCA results: all environmental impact categories (Note S6); LCA results for land transformation (Note S7); sensitivity analysis: reduced electricity consumption for CO_2 capture (Note S8); ecoinvent 3.6 data sets used for Figure 3 (Note S9); and contribution analysis: other environmental impact categories (Note S10) (PDF)
>
> Life cycle inventories compatible with Brightway2 (ZIP)

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Notes

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