



ACT Project Number:
271498

Project name:
ELEGANCY

Project full title:
Enabling a Low-Carbon Economy via Hydrogen and CCS

ERA-Net ACT project

Starting date: 2017-08-31
Duration: 36 months

D5.3.4
Scientific paper on Life Cycle Analysis (LCA) of
Direct Air Carbon Capture and Storage (DACCS)

Date: 2020-08-31

Organization name of lead participant for this deliverable:
Climeworks

ACT ELEGANCY, Project No 271498, has received funding from DETEC (CH), BMWi (DE), RVO (NL), Gassnova (NO), BEIS (UK), Gassco, Equinor and Total, and is cofunded by the European Commission under the Horizon 2020 programme, ACT Grant Agreement No 691712.		
Dissemination Level		
PU	Public	
CO	Confidential, only for members of the consortium (including the Commission Services)	X (until finalization of the paper)

Deliverable number:	D5.3.4
Deliverable title:	Scientific paper on Life Cycle Analysis (LCA) of Direct Air Carbon Capture and Storage (DACCS)
Work package:	WP 5 – case studies
Lead participant:	Climeworks

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Keywords
Direct Air Carbon Capture and Storage (DACCS); Life Cycle Assessment (LCA); Carbon dioxide removal (CDR)

Abstract
<p>Direct Air Carbon Capture and Storage (DACCS) is an emerging Carbon Dioxide Removal (CDR) technology, which has the potential to remove large amounts of CO₂ from the atmosphere. However, DACCS systems have hardly been evaluated regarding their environmental life-cycle performance. Therefore, we present a comprehensive Life Cycle Assessment (LCA) of different DACCS systems with low-carbon electricity and heat sources required for the CO₂ capture process – both stand-alone and grid-connected system configurations. The results demonstrate negative Greenhouse Gas (GHG) emissions – i.e. a net removal of CO₂ from the atmosphere – for all eight selected locations and system layouts. Highest GHG-removal potential is found for Norway with a grid-coupled system layout and waste heat usage – with a GHG removal efficiency of ca. 96% per ton of gross captured CO₂ with the Direct Air Capture (DAC) plant. Autonomous system layouts – entirely supplied by solar energy – prove to be a promising alternative at locations with high annual solar irradiation to avoid the consumption of fossil fuel based grid electricity and heat: Their GHG removal efficiency is at a similar – or even higher – level as grid-connected DAC in countries with rather low-carbon electricity supply, such as Switzerland. The analysis of environmental burdens other than GHG emissions shows some trade-offs associated with CO₂ removal and confirms the need for a comprehensive LCA approach. Further, the sensitivity analysis reveals the importance of selecting appropriate locations for grid-coupled all-electric system layouts, since the deployment of DACCS at geographic locations with CO₂-intensive grid electricity mixes leads to net GHG emissions instead of GHG removal today. However, a prospective global analysis with the integration of two future energy scenarios shows net-negative GHG emissions for the all-electric system layouts on all continents in 2040.</p>

The following represents the final draft of the scientific paper, as of 18th of November 2020.

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 (CDR) technology which has the potential to remove large amounts of CO₂ from the atmosphere. However, DACCS systems have hardly been evaluated regarding their environmental life-cycle performance. Therefore, we present a comprehensive Life Cycle Assessment (LCA) of different DACCS systems with low-carbon electricity and heat sources required for the CO₂ capture process - both stand-alone and grid-connected system configurations. The results demonstrate negative Greenhouse Gas (GHG) emissions - *i.e.* a net removal of CO₂ from the atmosphere - for all eight selected locations and system layouts. Highest GHG-removal potential is found for Norway with a grid-coupled system layout and waste heat usage - with a GHG removal efficiency of ~96% per tonne of gross captured CO₂ with the Direct Air Capture (DAC) plant. Autonomous system layouts - entirely supplied by solar energy - prove to be a promising alternative at locations with high annual solar irradiation to avoid the consumption of fossil fuel based grid electricity and heat: Their GHG removal efficiency is at a similar - or even higher - level as grid-connected DAC in countries with rather low-carbon electricity supply, such as Switzerland. The analysis of environmental burdens other than GHG emissions shows some trade-offs associated with CO₂ removal and confirms the need for a comprehensive LCA approach. Further, the sensitivity analysis reveals the importance of selecting appropriate locations for grid-coupled all-electric system layouts, since the deployment of DACCS at geographic locations with CO₂-intensive grid electricity mixes leads to net GHG emissions instead of GHG removal today. However, a prospective global analysis with the integration of two future energy scenarios shows net-negative GHG emissions for the all-electric system layouts on all continents in 2040.

Keywords: Life Cycle Assessment (LCA), Direct Air Carbon Capture and Storage (DACCS), Carbon Dioxide Removal (CDR), Negative Emission Technology (NET), Energy Storage.

1. Introduction

Carbon Dioxide Removal (CDR) technologies, or Negative Emission Technologies (NETs), are expected to fulfill a crucial role in the decarbonization of the global energy system, with average carbon removal estimations ranging from 10-15 Gt of captured CO₂ year⁻¹ at the end of the 21st century [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 more than 50% of all projected IAM energy scenarios require the deployment of CDR technologies to reach the 2.0° C target [3, 4]. Further, CDR technologies are required to a greater extent when climate mitigation measures are postponed, to compensate for an overshoot of GHG emissions [5]. A wide portfolio of CDR technologies have been proposed, such as the application of biochar,

Enhanced Weathering (EW), Ocean Fertilisation (OF), Bioenergy with Carbon Capture and Storage (BECCS) and Direct Air Carbon Capture and Storage (DACCS) [1, 2, 6].

However, 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, while potential unintended side-effects of other CDR technologies (*e.g.* OF) still need to be investigated [4, 6]. DACCS systems could largely avoid impacts on the water and food security nexus and can be considered as technology ready for small-scale deployment [4, 6, 7]. DACCS systems aim to extract CO₂ from ambient air and permanently store the captured CO₂ in a geological storage medium [6]. Direct Air Capture (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 CO₂, which is challenging due the extreme dilute concentration of CO₂ in ambient air [7]. The regeneration process aims to regenerate the sorbents and to separate CO₂. The latter process is energy-intensive due to the require-

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Nomenclature

Abbreviations

BECCS	Bioenergy with Carbon Capture and Storage
CCS	Carbon Capture and Storage
CCU	Carbon Capture and Utilization
CDR	Carbon Dioxide Removal
CoP	Coefficient of Performance
DAC	Direct Air Capture
DACCS	Direct Air Carbon Capture and Storage
DoD	Depth of Discharge
EW	Enhanced Weathering
GHG	Greenhouse Gas
GWP	Global Warming Potential
HT DAC	High Temperature aqueous solutions DAC
HTHP	High Temperature Heat Pump
IAM	Integrated Assessment Model
ISO	International Organization for Standardization

LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LCIA	Life Cycle Impact Assessment
LT DAC	Low Temperature solid sorbents DAC
NET	Negative Emission Technology
NMC	Nickel-Manganese-Cobalt oxide
OF	Ocean Fertilisation
PV	Photovoltaic
RE	Roundtrip Efficiency

Parameters

η_{dis}	Battery discharging efficiency [-]
C_{bat}	Battery energy capacity [kWh]
$C_{\text{bat,req}}$	Required battery energy capacity [kWh]
DoD	Depth of Discharge [-]
EOl	End of Life [-]

ment of heat at high temperature levels [7]. DACCS systems offer some flexibility compared to other CDR technologies, since they can remove CO₂ independently from the point source, both in time and space [6, 8]. Consequently, optimal locations can be selected, considering CO₂ storage potential and local costs for energy supply [9]. DAC systems are usually based on High Temperature aqueous solutions (HT DAC) or Low Temperature solid sorbents (LT DAC) [7]. This distinction is established on the temperature level and the sorbent used in the CO₂ capture process [7]. Few companies currently offer such DAC systems. For example, Carbon Engineering (Canada) implemented HT DAC systems on the North-American market [7, 10]. Clime-works (Switzerland) installed pilot LT DAC plants in Europe and realized the first DACCS project with negative CO₂ emissions in Hellisheiði (Iceland) in 2017 [7, 11]. Global Thermostat recently deployed pilot and demonstration plants based on LT DAC in the United States [7].

As emerging technology with a supposedly decisive role in future low-carbon energy systems, DACCS systems must be thoroughly evaluated regarding their environmental performance in a transparent and consistent way over their entire lifetime [6]. Life Cycle Assessment (LCA) is a suitable and flexible assessment tool to identify environmental hotspots (*i.e.* the main contributors) and to evaluate the total life-cycle environmental performance of a product or service [12, 13]. Only few DACCS LCA studies - with mostly limited scopes - have been conducted so far.

De Jonge et al. [14] assessed the life-cycle carbon efficiency of a HT DAC system and determined the main environmental contributors to overall LCA scores. The contribution analysis revealed a high environmental impact due to energy needed for the CO₂ capture process. Lastly, a recent DACCS LCA study of Deutz and Bardow [15] showed that two commercial LT DAC(CS) plants - in Hinwil (Switzerland) and Hellisheiði (Iceland) produced and operated by Clime-works - achieved GHG removal efficiencies of 85% and 93%, respectively. The latter study also determined the environmental impacts of six differ-

ent adsorbents and concluded that climate benefits are mainly influenced by energy sources used for CO₂ capture.

To the best of our knowledge, no comprehensive LCA of DACCS has been published in the scientific literature. We believe that the study of Deutz and Bardow [15] is comprehensive for the DAC system and the associated supply chains, but lacks a detailed assessment of the CO₂ storage stage, since their work only considered electricity for CO₂ injection. However, the CO₂ storage stage includes for example environmental impacts from infrastructure (*e.g.* pipelines, injection wells and the compression station), the drilling of injection wells and CO₂ leakage during the transportation of CO₂ [16, 17]. Besides, the latter study excludes energy storage when intermittent (renewable) energy sources are integrated and claim that their energy system layouts can be installed at remote locations, while we believe that appropriate energy storage mediums need to be considered when (renewable) intermittent energy sources are used for the CO₂ capture process - which inevitably leads to additional environmental impacts. Further, the other DACCS study of de Jonge et al. [14] only focused on the carbon capture efficiency, hence excluded other potentially important environmental impacts. In addition, the latter study reported data limitations regarding the quality of their Life Cycle Inventory (LCI) data for the DAC infrastructure.

Other available studies mainly focus on DAC, thereby excluding the carbon storage stage required for permanent CO₂ removal from the atmosphere. In addition, these LCAs are simplified regarding LCA modelling choices, such as the exclusion of life-cycle phases and environmental impact categories besides climate change [6]. Further, some studies do not consider a certain amount of CO₂ equivalents removed from the atmosphere as 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 FU, is transparent in the methodology used, assesses all life-cycle stages (including a thorough assessment of the CO₂ stor-

age stage) and examines a wide set of energy sources for CO₂ capture, is missing. The contribution of our paper can be summarized as follows:

- We present a detailed and transparent LCA of a LT DACCS system, based on Climeworks' technology, with different electricity (*i.e.* grid and Photovoltaic (PV) power) and heat sources (*i.e.* electricity, waste and solar heat) for CO₂ 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 (batteries) and heat storage.
- We include several processes needed for CO₂ storage - parameterized on transportation distance, geographical storage location and storage depth: energy needs for compression, infrastructure requirements (*e.g.* pipelines and compression stations), drilling of boreholes, country-specific electricity for the injection of CO₂, and CO₂ leakage resulting from the transportation in pipelines.
- A global analysis is included for all-electric grid connected DACCS systems.
- We integrate two future IAMs scenarios [18, 19] and modify the ecoinvent background LCA database to determine the future global potential of grid connected DACCS systems.

The structure of our paper is as follows. Section 2 presents the methodology, where LCA modelling steps and different DACCS system layouts are discussed. Section 3 shows the LCA results, discussions and limitations. Finally, the conclusion is presented in Section 4.

2. Methodology: Life Cycle Assessment

LCA is a methodology which aims to quantify environmental impacts of a product or service over its entire life-cycle [12, 13]. LCA is standardized by the International Organization for Standardisation (ISO), where ISO 14040 describes the general principles and framework of LCA [20] and ISO 14044 presents guidelines and general practices for LCA [21].

2.1. Goal and Scope

Our goal is to quantify the environmental impacts and to determine environmental hotspots along the DACCS life-cycle considering low-carbon energy sources for CO₂ capture in different DACCS system layouts. 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 [14, 15, 22]. However, the overall environmental impact of the integration of low carbon energy sources - using a wide set of environmental

impact categories - is not 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 future [19]. And finally, site dependent boundary conditions - such as the lack of waste heat or low-carbon electricity from the grid - might require novel system designs including heat and electricity storage allowing for autonomous DAC operation.

We consider the following countries - with different climates - in the main analysis: Greece, Mexico, Jordan, Spain and Chile are included as (semi)-arid and 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 potentials [23], difference in grid electricity mix [24], climate variations and data availability for the Fresnel solar collector.

Our functional unit is defined as '*Gross removal of 1 tonne CO₂ from the atmosphere via the use of a DAC plant combined with geological CO₂ storage*', with a reference flow of a DAC unit removing 100kt CO₂ year⁻¹ with varying system layouts and electricity and heat inputs as specified in the individual scenarios (see Section 2.2.1). Consequently, total GHG emissions produced from all upstream and downstream DACCS activities should be less than 1 tonne CO₂-eq. to result in net negative GHG emissions, *i.e.* CO₂ removal from the atmosphere.

We identify no multi-functionality of our DACCS system, since the main purpose is to remove CO₂ from the atmosphere in a permanent way. Hence, allocation or system expansion is not required.

We use the ILCD 2.0 (2018) Life Cycle Impact Assessment (LCIA) method [25] to assess the environmental performance of the proposed DACCS system layouts. We adopt 16 midpoint categories from ILCD in the protection areas 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)) [26, 27]. Results are shown for impacts on climate change in the main text, while the complete set of results is shown in the Supplementary Information (Appendix E).

An attributional LCA perspective with the ecoinvent database (v3.6, system model *Allocation, cut-off by classification* [28]) as source of background inventories is applied. Besides, open-source Python package Brightway2 is used to conduct our LCA [29]. Our LCIs and corresponding assumptions will be discussed in Section 2.2 and Section 2.2.1. Besides, the 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

A simple representation of our system boundaries is visualized in Figure 1. The DACCS product system includes 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 CO₂ and business flights (the so-called "foreground processes").

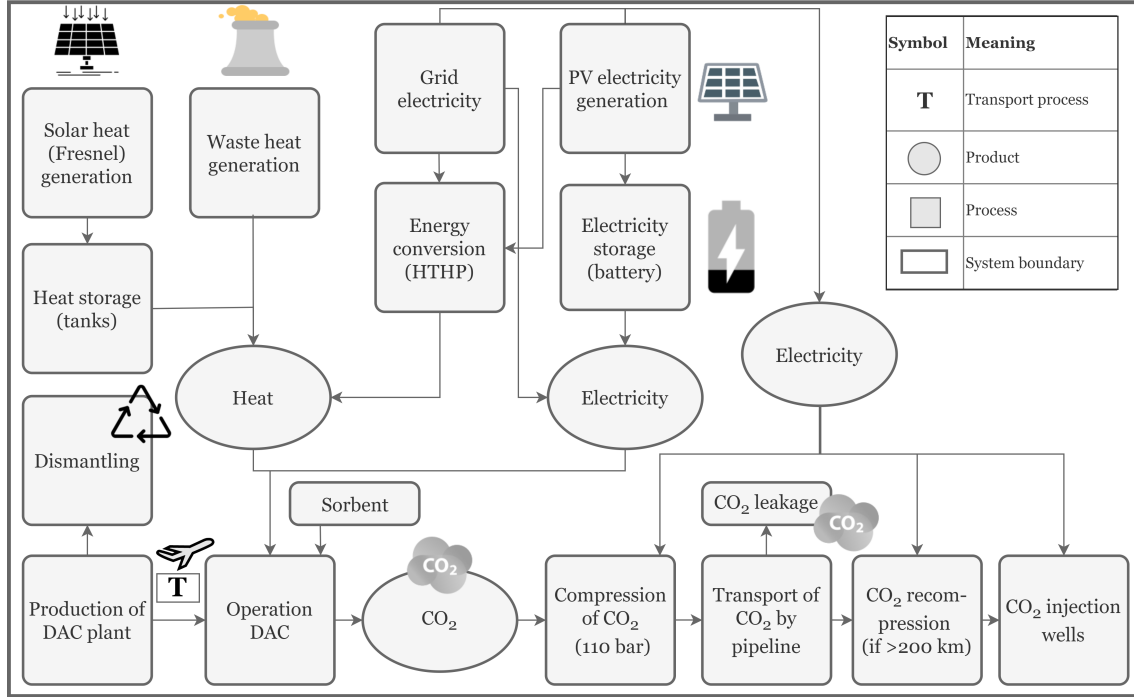


Figure 1: System boundaries of the DACCS product system. Note the different electricity and heat system layouts in the upper part of the figure. Further, all upstream and downstream materials, services and emission flows are included, but are not shown in this figure to reduce complexity.

DAC plant. Specific production and operation data of the DAC plant is based on industrial information provided by Climeworks (Zürich, Switzerland). A contribution analysis of the main components and materials used for the DAC construction for two scenarios (year 2020 and year 2025) is presented in Appendix A of the Supplementary Information. The LT DAC technology of Climeworks embraces a cellulose-based filter complemented with amine-rich materials [7]. Deutz and Bardow [15] present an overview and environmental assessment of different sorbents potentially used for the CO₂ capture process of Climeworks. A current goal of Climeworks is to upscale their standard plant DAC size to capture 100 kt CO₂ per year within the next few years. Hence, we assume a DAC plant with an annual gross carbon capture capacity of 100 kt CO₂ and a system lifetime of 20 years [15]. Note 'gross', since GHG emissions from all upstream and downstream activities, generated from the entire DACCS life-cycle, are not included in this figure which inevitably leads to less than 100 kt annual net CO₂ removal from the atmosphere.

Further, we assume that the production of DAC components and related engineering work is conducted in Switzerland. LCI of the DAC is obtained from Climeworks (2025 scenario) and the work of Deutz and Bardow [15]. The latter study generally showed small environmental impacts in absolute terms due to adsorbent consumption. Besides, a near future scenario (year 2025) of Climeworks shows that adsorbent consumption is expected to decrease from 7.5 to 3.0 kg adsorbent t⁻¹_{CO₂} captured. Therefore, we consider a generic proxy for the sorbent; 'market for chemical, organic', and use a sorbent consumption of 3.0 kg t⁻¹_{CO₂} captured. Near-future (2025) energy requirements for CO₂

capture are estimated as 500 kWh t⁻¹_{CO₂} and 1500 kWh t⁻¹_{CO₂} captured for electricity (without electricity consumption for CO₂ compression) and heat (at around 100° C), respectively [15]. We believe that the latter assumptions on energy consumption could have a big influence on LCA results. Hence, we consider an improvement in efficiency for electricity consumption to examine the impact on the Climate Change impact category (see Section 2.3).

Business trips. We consider environmental impacts of business trips for acquisition, negotiation, installation, trouble shooting and maintenance of the DAC plant by Climeworks engineers. We estimate a conservative total number of 86 trips from Zürich to the location of the DAC installation (and back).

Dismantling. Dismantling of the DAC plant is included. All main materials (e.g. steel, plastics, copper and aluminium) are assumed to be treated after the system lifetime of 20 years, partially recycled, partially disposed of.

Geological storage of CO₂. After the CO₂ is captured, the CO₂ needs to be compressed (from 1 atm to 110 bar) by consuming locally available electricity sources, in our alternatives provided with grid or PV electricity. We assume that CO₂ is transported with pipelines (110 bar pressure) to the injection wells (80 bar assumed as pressure at pipeline end), due to the high capacity needed for large-scale CO₂ transportation [16]. Further, additional compression of CO₂ is included when the transportation distance is larger than 200 kilometer [16]. We consider CO₂ leakage from CO₂ transmission pipelines using baseline (i.e. Medium) emission factors from an IPCC report [17], hence we update the LCI of [16] appropriately. After that, the CO₂ is injected into wells - using the country-specific elec-

tricity mix - to store the CO₂ in geological layers. CO₂ storage in suitable geological layers is considered to exhibit the highest CO₂ storage potential, hence we focus on CO₂ storage in geological layers [16]. We use the LCI from Volkart et al. [16] for the infrastructure requirements for transportation, (re)compression and injection of CO₂ into wells. We parameterize the latter inventory to generate location specific environmental impacts of CO₂ storage, based on the specific transportation distance and storage depth needed for CO₂ storage in a country. We assess the feasibility of geological CO₂ storage based on a geological storage map developed by the Global CCS Institute [23]. Based on this map, we approximately estimate transportation distances to potential CO₂ injection wells in the same or other countries. The following transportation distances to CO₂ injection wells are assumed: Chile (1250 km to Brazil), Greece (500 km), Jordan (100 km), Mexico (1000 km to USA), Norway (500 km), Iceland (500 km to Norway), Spain (500 km) and Switzerland (1000 km to Norway). For simplicity, we assume a generic CO₂ storage depth of 2000 meters for each country, since [16] have shown that this depth hardly affects LCA results. CO₂ leakage from injection wells is assumed to be negligible [30].

Energy supply for CO₂ capture Earlier DAC(CS) LCAs showed that energy consumption for the CO₂ capture can be perceived as the crucial process in terms of environmental impacts [14, 15, 22]. Therefore, we evaluate system layouts with different energy sources for the CO₂ 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 [31]. 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 [7, 28]. Third, further electrification of future energy systems can be expected [19]. Large-scale and economically attractive implementation of DACCS might, however, require remote installations close to proper sites for geological CO₂ 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 renewables-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 the Supplementary Information (B). 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.

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 intermit-

tent, resulting in fluctuations in power and heat output of the PV and fresnel units [32]. 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 CO₂ annually from the ambient air. Less CO₂ 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 [33]. 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 DAC systems [34]. For the desorption of CO₂ heat at a temperature of 100° C is required [7].

LCI of the fresnel solar collector has been generated in collaboration with Industrial Solar (Freiburg, Germany). The fresnel construction is largely made of low-alloyed steel and (for a smaller part) of stainless steel and aluminium. Industrial Solar offers commercial solar heat systems, such as the fresnel solar collector LF-11 [34]. Fresnel solar plants use reflective mirrors (made of glass) to concentrate solar irradiation on a solar collector. Water is pumped through the solar collector and is partly evaporated due to the concentration of solar irradiation.

Next, the resulting steam is stored as latent heat in a steam drum reservoir [34]. We scale the heat storage tank, made of low-alloyed steel, to be able to store the amount of steam generated within 12 hours, since the fresnel plant only produces solar-based heat during the day. The fresnel plant is produced in Germany by Industrial Solar. Hence, transportation distances to other countries use Freiburg (Germany) as reference point and include freight transportation by lorries and ships. The latter transportation mode is only used when it is more efficient to reach a destination by ship. Further, we include business trips needed for the acquisition, negotiation, installation, trouble shooting and maintenance of the fresnel plant. The total business trips are estimated on 51 trips. Dismantling of the fresnel plant after the system lifetime is considered, with generic recycling, incineration or disposal activities from theecoinvent database. A system lifetime of 25 years has been assumed. The efficiency of the fresnel plant is obtained from modelling work of Industrial Solar. It varies between 40-47%, mainly influenced by - but not linearly linked to - the incoming direct normal irradiance. The functional unit used in the fresnel LCA is 1 MJ of heat delivered, to be subsequently consumed in the CO₂ capture process of the DAC plant. The LCI of the fresnel system is presented in the Supplementary Information (Appendix C).

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 [33]. We received data for 11 potential locations in 8 countries with a direct normal irradiance of more than 2000 kWh/m²/year from Industrial Solar, which were comprehensively modelled regarding their techno-economic performance. Chile (Antofagasta), Greece (Crete), Jordan (Amman), Mexico (San Luis Potosí) and Spain (Taber-

nas) are included in our analysis to represent 5 countries with sufficient solar irradiation.

Electricity is supplied by PV arrays. Therefore, country-specific LCI datasets are used to represent multi-Si PV modules. Further, we use a stationary battery system to store excess PV electricity during day-time in order to be consumed during night-time. We include a lithium Nickel-Manganese-Cobalt oxide (NMC) battery, representing mainstream technology for stationary electricity storage today [35].

We assume that the NMC battery should be able to store 12 hours of the electricity load to provide sufficient electricity during night for CO₂ capture and compression, in line with the heat storage sizing and the aim to capture 100kt CO₂/year. The minimum battery storage capacity requirement ($C_{\text{bat,req}}$) is calculated to capture 100 kt CO₂ per year considering 12 hours of storage. We oversize the NMC battery to consider battery degradation [36]. 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% [32]. Equation 1 is used to size the energy capacity of the NMC battery [32].

$$C_{\text{bat}} = \frac{C_{\text{bat,req}}}{\eta_{\text{dis}} \cdot \text{DoD} \cdot \text{EoL}} \quad (1)$$

Besides, we assume a power to energy ratio of 1:2 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 in order 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 [32]. The latter assumption can be perceived as a worst case scenario, 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 CO₂ 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 equation 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 8500 load hours per year and a Coefficient of Performance (CoP) of 2.9 for the HTHP, which is conservative and at the lower range of presented CoPs of HTHPs [37]. CO₂ 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 [37]. LCI of the HTHP has been generated by linearly scaling up a 10 kW heat pump from the ecoinvent database, to the appropriate heat pump size (19 MW) to deliver sufficient instant heat for CO₂ capture. We modify the LCI of the heat pump and use CO₂ (R744) as refrigerant - instead of R134 - based on information from MAN Energy Solutions (Zürich, Switzerland), to represent current and future industrial practices of the HTHP industry [38].

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 database are used for grid electricity [28]. 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 [28]. The *Waste heat + Grid* system layout is only applicable when waste heat at the correct temperature level is available. Therefore, a location specific assessment is required to identify the potential of waste heat. For simplicity, we decided to include all 8 countries in the waste heat system layouts. Country-specific LCI of the ecoinvent database is used for waste heat and grid electricity [28].

2.2.5. Waste heat + PV + Battery

The *Waste heat + PV + Battery* system layout consumes PV electricity and waste heat as energy sources for CO₂ 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 LCI of all system layouts are provided in the Supplementary Information (Appendix D).

2.3. Sensitivity Analysis

2.3.1. Reduced electricity consumption

DACCS is an emerging technology and will profit from technological improvements [7]. This could result in a reduction of energy consumption during the CO₂ capture process. Current figures used for energy consumption (500 kWh t_{CO₂}⁻¹ electricity and 1500 kWh t_{CO₂}⁻¹ heat, respectively [15]), are based on a very high CO₂ purity in the resulting CO₂ stream [7]. However, a lower CO₂ purity seems to be feasible with a CCS instead of Carbon Capture and Utilization (CCU) application, which might result in lower energy requirements. Therefore, we examine the performance of the proposed system layouts with an electricity consumption of the CO₂ 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 scope to determine GHG emissions (*i.e.* Climate Change) for 144 countries. We specifically focus on this all-electric system layout in our geographical sensitivity analysis, since energy system models predict an increase of electrification in future energy systems [19]. Besides, this system layout is the simplest and could be implemented in all locations with a (low-carbon) grid connection, 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 which are modelled with multiple regional electricity datasets in the ecoinvent database. In case there is no country-specific electricity dataset available, we use the market (group) for electricity for the geographical area in which this country is located as approximation. For example, we assume the market group datasets 'RLA' (*i.e.* composed of all available Latin American and the Caribbean electricity mixes according to their corresponding electricity production volumes) as approximative electricity dataset for Puerto Rico, since Puerto Rico has no specific electricity dataset in the ecoinvent database. Further, we assume a generic transportation distance for CO₂ of 500 kilometers to the injection wells and a generic storage depth of 2000 meters.

2.3.3. Future electrification: HTHP + Grid alternative

Projections of IAMs show that electricity grid mixes will become less CO₂-intensive, even in the most carbon intensive energy scenarios [18, 19]. 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 [39], adapting future electricity mixes in our background database, based on the output figures of the REMIND model scenarios [18]. Note that this results in geographically aggregated future electricity datasets, since the REMIND model subdivides the world into only 11 regions.

Firstly, 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 [18, 39]. Secondly, 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 as stated in the Paris Agreement [5, 39].

3. Results and discussions

3.1. Climate Change impact per system layout

Figure 2 shows the results for the impact category "Impacts on Climate Change" including the contribution of the various

system components to the total score. The red dotted line visualizes the GHG "break-even", and all bars below the red dotted line represent net negative GHG emissions. That means, CO₂-capture removes more CO₂ than the GHG emissions generated from all upstream and downstream activities during the DACCS life-cycle. Figure 3 shows the sensitivity of the results on "Impacts on Climate Change" for all alternatives, when using electricity sources (average country supply mixes and specific generation technologies) with different GHG-intensities for CO₂-capture.

In general, Figure 2 and Figure 3 show that system layouts with waste heat and DAC operated in countries with a high share of renewables in the electricity mix result in the best performance regarding GHG emissions. Waste heat comes (almost) burden-free in the *Allocation, cut-off by classification* system model of ecoinvent. Further, national grid electricity mixes with a large share of renewables have a low CO₂-intensity. Consequently, lowest GHG emissions (40 kg CO₂-eq. per FU, *i.e.* net-negative GHG emissions of 0.96 t CO₂-eq.) are obtained from the *Waste heat + Grid* system layout in Norway. Norway has the grid electricity mix with the lowest CO₂-intensity among the selected countries. It turns out that the grid electricity mix is a crucial factor for grid-coupled alternatives, especially for alternatives which require large amounts of electricity (*i.e.* HTHP + Grid). Consequently, largest GHG emissions are obtained in Greece with 0.91 t CO₂-eq. per ton of CO₂ captured for the HTHP + Grid system layout. Grid electricity in Greece has a relatively high CO₂-intensity due to the large share of lignite and natural gas. Hence, GHG emissions for the HTHP + Grid system layout are very country-specific.

DACCS-related GHG emissions in countries with a CO₂-intensive grid electricity mix can be substantially reduced by shifting to renewable energy sources as energy suppliers for the DAC operation (see Figure 3). For Greece (*i.e.* *Waste heat + Grid*), GHG emissions can be reduced by 80% when grid electricity is replaced by PV electricity (*i.e.* *Waste heat + PV + Battery*). For autonomous system layouts, the *Autonomous (Fresnel + PV)* (between 94-123 kg CO₂-eq.) configuration demonstrates slightly lower GHG-emissions compared to the *Autonomous (HTHP + PV)* system layout (between 123-168 kg CO₂-eq.), due to the provision of low carbon heat with the fresnel solar collector.

Figure 3 demonstrates that alternatives with waste heat consumption and the *Autonomous (Fresnel + electricity)* system layout are less sensitive when using high GHG-intensive electricity, due to less electricity consumption for CO₂ capture and compression. For example, the *Autonomous (Fresnel + electricity)* generates solar heat and therefore less electricity is required for CO₂ capture, although this alternative can only be installed at locations with sufficient annual solar irradiation. Besides, the zoom into the lower-left corner of Figure 3 shows that GHG-emissions from the production of autonomous systems are comparably higher due to the production of energy storage units, such as battery energy storage systems. Further, lower GHG emissions are obtained with *Autonomous (Fresnel + electricity)* compared to the *Autonomous (HTHP + electricity)* system layout when using higher GHG-intensive electricity

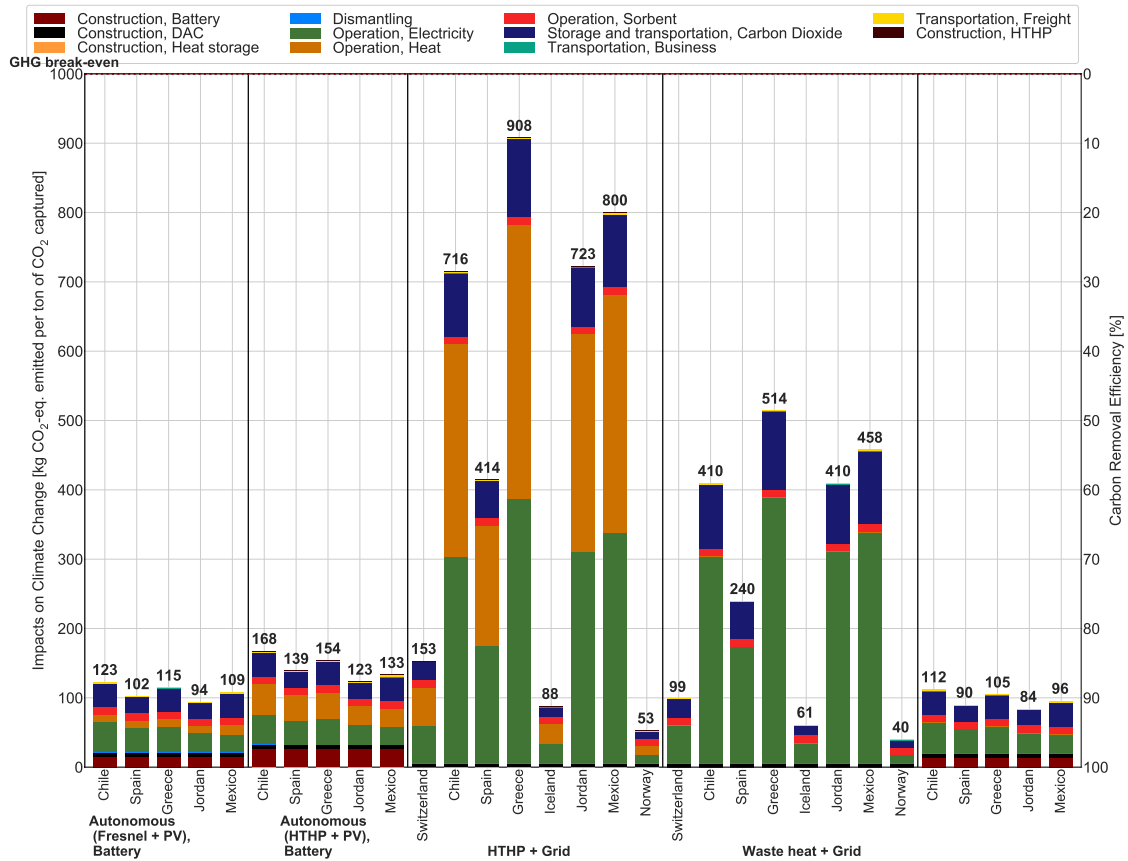


Figure 2: Life-cycle GHG emissions in kg CO₂-eq. per tonne of gross CO₂ captured with the DAC plant, for different system layouts in selected countries. The size and colors of the bar segments correspond to the contributions of specific processes to the total life-cycle GHG emissions. Results below the "break-even level" indicate a net-negative effect (*i.e.* net removal of GHG from the atmosphere), while results above this level would indicate a net-positive effect of DACCS. Note that 'Storage and transportation, Carbon Dioxide' includes compression, transportation, re-compression, injection and the infrastructure requirements for CO₂ during the CO₂ storage stage.

for CO₂ capture and compression, due to the provision of low carbon heat with the fresnel collector. Besides, a bigger battery (~221 MWh vs. ~125 MWh) is needed for *Autonomous (HTHP + electricity)*, since more electricity needs to be stored to provide sufficient electricity to the HTHP during night. The production of heat storage tanks (made of low-alloyed steel) results in smaller GHG-emissions compared to the production of bigger battery energy storage systems, which means lower GHG-emissions for the *Autonomous (Fresnel + PV)* system layout.

The contribution of the various system components to the total score is also presented in Figure 2. It reveals the absolute contribution of processes to the total GHG emissions of a system layout in a specific country. In general, energy consumption is a key factor for the Climate Change impact category, which can be reduced by using solar energy in countries with a high annual solar irradiation. Besides, the absolute impact on Climate Change of the DAC construction (6 kg CO₂-eq. t_{CO₂}⁻¹ captured), sorbent consumption (10 kg CO₂-eq. t_{CO₂}⁻¹ captured) and dismantling (0.2 kg CO₂-eq. t_{CO₂}⁻¹ captured) is identical for

all alternatives.

The relative Climate Change impact of transportation processes directly related to DAC (*i.e.* business and freight) can be considered as small with contributions of less than 4%. Relative GHG emissions from the carbon dioxide storage stage are significant - and fluctuate between 12-36% - mainly driven by electricity requirements for the compression and injection of CO₂, and CO₂ leakage during CO₂ transportation, respectively. Note that we included the Medium scenario presented in [17] for CO₂ leakage during transportation. The contribution of CO₂ storage would further increase, if we used the pessimistic scenario (factor of 10 increase) for CO₂ leakage during the transportation of CO₂ in pipelines, which emphasizes the need for a well-designed pipeline system for CO₂ transportation.

Further, the contribution analysis reveals that the production of electricity storage (NMC battery) units can - in relative terms - have a significant impact on GHG emissions with 12-22%, while the production of the heat storage (steel tanks) mediums are small with less than 1%. Further, the production of the HTHP has a negligible impact on total GHG emissions with a

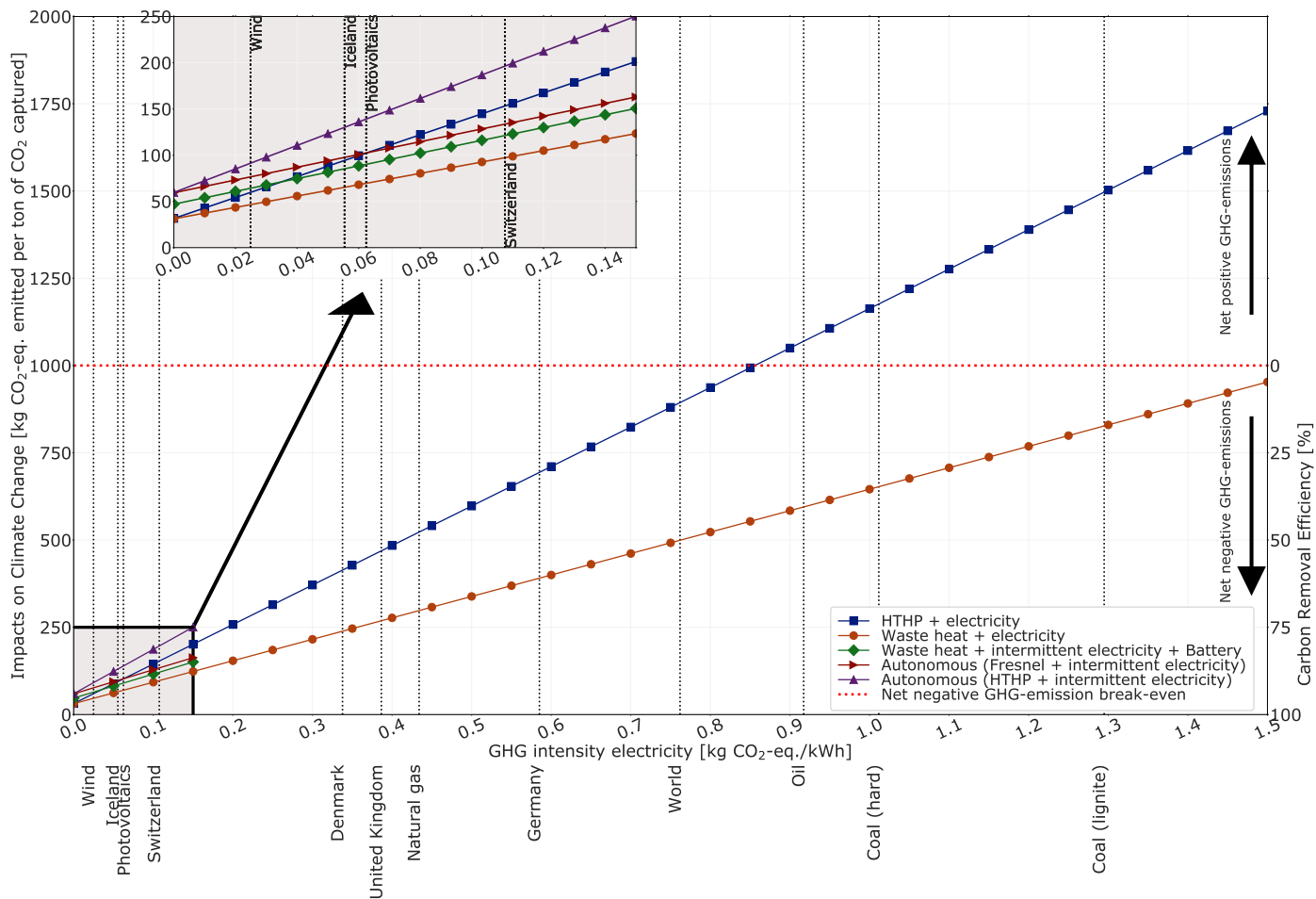


Figure 3: Sensitivity on the GHG-intensity of the electricity source used for CO₂ capture and CO₂ compression for all alternatives. We use the LCI of Switzerland as proxy for *HTHP + electricity* and *Waste heat + electricity* with a CO₂ transportation distance of 1000 km to Norway. Further, LCI of Spain is used for *HTHP + electricity*, *Autonomous (Fresnel + electricity)* and *Autonomous (HTHP + electricity)*. Note that the *Autonomous (Fresnel + electricity)* alternative can only be installed in (semi)-arid locations due to the requirement of high annual solar irradiation. Besides, the waste heat alternatives can only be installed with sufficient supply of waste heat.

contribution of less than 1%, which can be (partly) explained by the replacement of R134a (Global Warming Potential (GWP) of ~1300) with R744 (GWP of 1, *i.e.* CO₂) as refrigerant.

3.2. Other environmental impact categories

Appendix E in the Supplementary Information shows the results of all system layouts on all selected environmental impact categories. Low environmental burdens in almost all impact categories can be achieved with the *Waste heat + PV + Battery* and the *Autonomous Fresnel + PV* system layouts. For the *Waste heat + PV + Battery*, the low environmental impacts can be explained due to consumption of PV electricity and the use of waste heat as (almost) burden-free heat source. However, this system layout has a moderate score in the Minerals and Metals impact category resulting from material requirements for the production of PV arrays and the NMC battery. For the *Autonomous Fresnel + PV*, the low environmental impacts can be explained due to consumption of PV electricity and the use of low-carbon heat production with the Fresnel solar collector. Further, the *Waste heat + Grid* layout causes low burdens in

countries with a large share of renewables in their grid electricity mix (*e.g.* Norway and Iceland).

Fundamental differences between environmental impact categories and countries are found for the *HTHP + Grid* system layout. For example, DACCS in Greece and Chile causes high environmental burdens in different impact categories, such as Fossils, Freshwater and Terrestrial Acidification, Freshwater Eutrophication, Terrestrial Eutrophication and Photochemical Ozone Creation, mainly due to the consumption of fossil fuel based grid electricity. DACCS in Norway causes a high environmental impact on Water Depletion, predominantly due to the reliance on hydropower. Switzerland and Spain show high environmental burdens for the Ionising Radiation impact category, explained by the relatively high share of nuclear power in their grid electricity mixes.

In general, the *Autonomous HTHP + PV* system layout shows low burdens in most impact categories. However, the environmental impacts on Minerals and Metals is high, due to the use of raw materials for the production of PV arrays and the

NMC battery. These examples show the importance to assess and compare system layouts on a wide set of environmental impact categories.

3.3. Sensitivity Analysis

3.3.1. Reduced electricity consumption

Figure C (Appendix F in the Supplementary Information) demonstrates the absolute change in the Climate Change impact category when the electricity consumption for the CO₂ capture process is reduced by up to 20%. An efficiency improvement has a bigger (beneficial) influence on the Climate Change impact category - especially on system layouts which consume large amounts of CO₂-intensive electricity (*i.e.* *HThP + Grid* in Greece and Mexico).

3.3.2. Electrification: *HThP + Grid* alternative

Figure 4 shows 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 the impact on the Climate Change impact category is lower than 1000 kg CO₂-equivalent per ton of gross CO₂ captured. Countries which fulfill this requirement are indicated in green and orange (*i.e.* orange meaning higher GHG emissions than green). Net positive GHG emissions are indicated in dark red.

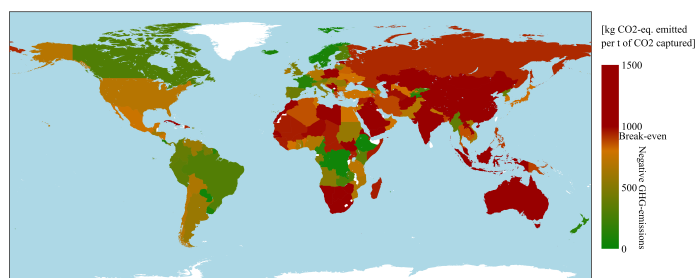


Figure 4: Country-specific results for the Climate Change impact category for the *HThP + Grid* system configuration. Green and orange indicate net-negative GHG emissions, while dark red shows net-positive GHG emissions of DACCS.

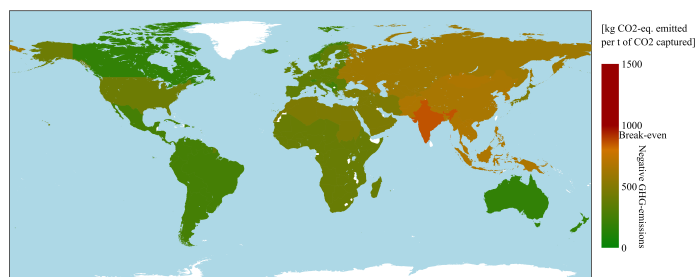


Figure 5: Results for the Climate Change impact category for the *HThP + Grid* alternative, according to the *SSP2-Base* scenario. Green and orange colors indicate net-negative GHG emissions, while dark red shows net-positive GHG emissions.

Significant variations between countries are found for the Climate Change impact category, which can (mainly) be explained by the difference of the CO₂-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 ~0.86 kg CO₂-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 results in GHG removal from the atmosphere. It turns out that most countries in Europe, North-America, South-America and middle Africa already show large GHG removal potentials with DACCS *HThP + Grid* system layouts. However, few countries in these continents show 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 5 demonstrates the possible future impact (2040) regarding the Climate Change impact category for the *HThP + Grid* system layout according to the *SSP2-Base* scenario of the REMIND model. It turns out that the deployment of *HThP + Grid* DACCS system layouts could result in net-negative GHG emissions in all world regions in 2040. North-America, South America, Europe, Australia and Africa show a large net GHG removal potential, while India and Asia seem to be less suitable due to their still relatively high CO₂-intensive grid electricity mix.

Figure 6 shows the results after implementing a more ambitious climate scenario (*i.e.* *SSP2-PkBudg1300*), which aims to curb the global temperature increase to 2°C. It turns out that DACCS in all regions shows GHG emissions of less than 230 kg CO₂-eq. per ton of CO₂ captured in 2040. Therefore, effective climate policy would be very beneficial for all-electric DACCS system layouts connected to the grid.

3.4. Discussion - limitations and future work

Our analysis shows that within the LCA of DACCS systems, there are few elements and key factors, which determine the results. While we consider data availability and quality regarding

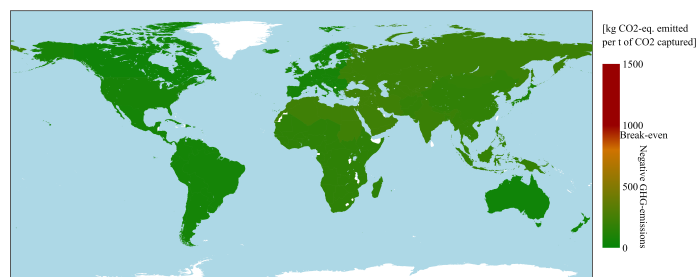


Figure 6: Results for the Climate Change impact category for the *HThP + Grid* alternative according to the *SSP2-PkBudg1300* scenario. Green and orange colors indicate net-negative GHG emissions, while dark red shows net positive GHG emissions.

energy demand of and supply for, respectively, the CO₂ capture process - the most crucial factor by far driving LCA results - to be high, there are other issues, which call for further analysis in the future. More sophisticated modeling integrating operational experience of DAC units of different scale 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:

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 show large contributions to environmental burdens caused by heat pumps [37, 40].

3.4.2. Assessment of fresnel heat plant in other locations

Only five potential locations for the fresnel solar collector were included 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 modelled 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 modelling should be expanded to determine the environmental merits in other geographical locations.

3.4.3. Optimization of system layouts

Energy storage systems were sized with a static sizing methodology to reduce complexity. However, despite of this simplification, our LCA results should be accurate, since they show that variation in DACCS system component sizes would not change the outcomes in a substantial way. Still, optimization models could be useful to determine the optimal system layouts based on investment and operation costs or environmental impacts of storage units [32, 41]. Further, those models could be helpful to define the optimal PV array size as well as heat storage size and they could give first indication of the impact of grid-coupled DACCS system layouts on the electricity grid [32].

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 °C and 60 °C, with an optimal operation temperature between 15 °C and 35 °C [42]. Therefore, location-specific measures should be considered when outside temperatures approach the latter 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)*, *Autonomous (HTHP + PV)*. Twelve hours of energy storage capacity was assumed for storage mediums for these two system layouts. This led to a large storage capacity needed for both the battery system (~125 MWh and ~221 MWh, respectively) and the heat storage tanks.

Alternatively, a doubling of the DAC capacity (in order to comply with our functional unit) with discontinued operation over night could be installed to reduce the need for energy storage in the proposed two system layouts. However, we emphasize that such a system with doubled DAC capacity still requires energy storage for intermittent (renewable) electricity generation, which would result in additional environmental impacts from the production of storage mediums. Further, a doubling of the capacity of the DAC plant results in (large additional) capital expenditures for the DAC plant, since capital expenditures of DAC systems are still high [7]. 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 for the operation of such an autonomous DAC system.

Further, more sophisticated research is needed to confirm the self-sufficiency of the proposed autonomous system layouts. Finally, real demonstration projects are needed to test the results from optimization models and inputs to LCA in general.

3.4.6. Future electrification: REMIND regions

We used the outputs of the IAM REMIND 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 4). 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. These benefits and side-effects of the decarbonization of power supply have been investigated by Luderer et al. [43].

3.4.7. Comparison with DACCS and CDR technologies

The single purpose of DACCS systems is to remove CO₂ from the atmosphere in a permanent way. Our analysis focused on LT DACCS systems and can hardly be compared with HT

DACCS systems, especially due to the need of different processes used for CO₂ capture and their associated energy requirements [7]. 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 CO₂-removal from the atmosphere [6]. Unfortunately, we are currently not able to present such a comparison between CDR technologies due to the immature research state of CDR technology LCAs in general [6].

4. Conclusion

Our paper aimed to determine the life-cycle environmental performance and environmental hotspots of LT DACCS systems with low carbon energy sources for CO₂ capture. The results were presented per tonne of gross captured CO₂ with a DAC plant of an annual capture capacity of 100 kt CO₂ based on Climeworks' technology. We included different energy sources - solar heat, waste heat and electricity by means of a HTHP - to deliver high temperature heat to the DAC plant. In addition, different electricity sources - PV and grid electricity - were considered to supply electricity to the CO₂ capture process. The results were presented for different system layouts for eight geographic locations with specific assumptions regarding CO₂ transport and storage after capture.

The results revealed net-negative GHG emissions for all eight selected countries and all proposed system layouts, meaning that GHG emissions associated with energy supply and material demand for DACCS are below the amount of CO₂ captured, *i.e.* removed from the atmosphere. However, the level of "net-negativity" showed substantial variation between DACCS layouts and countries of application: while in our best case 96% of the captured CO₂ 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 CO₂-intensities of the grid electricity due to high shares of renewables in their national grid electricity mix. The CO₂-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 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 CO₂ storage sites). Therefore, we recommend solar-based autonomous DACCS systems for countries with (semi)-arid climates which have a CO₂-intensive grid electricity mix. All-electric DACCS system layouts are recommended when the national grid electricity mix relies on low-carbon electricity sources. Further contributions to life-cycle GHG emissions of DACCS - associated to for example DAC infrastructure, CO₂ transport and storage, and energy storage units - can be substantial in relative terms, if energy supply for CO₂ capture is clean in terms of GHG emissions, but absolute GHG emissions due to these contributions are significant but small.

The assessment of a wide variety of environmental impact categories - in addition to impacts on climate change - showed a different ranking of DACCS systems on a few environmental impact categories, which confirmed the importance for 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 CO₂-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, waste heat and the potential of carbon storage sites. Further, our prospective analysis demonstrates that more ambitious climate policy will have beneficial effects on GHG emissions for all-electric DACCS systems.

Based on our findings, we recommend to compare alternative DACCS system layouts and technologies with a comprehensive and transparent approach based on location-specific parameters to identify and select the most environmentally friendly DACCS system layout under given boundary conditions. With DACCS implementation performed in this way, we foresee a promising CO₂-removal potential of DACCS systems.

Supplementary Information

Supplementary information available in a Word document: A. Contribution analysis of the DAC infrastructure (plant) B. Overview table of energy system layouts. C. LCI of the fresnel system. D. LCI of all system layouts. E. Results on all environmental impact categories. F. Figure C: Reduced electricity consumption.

Acknowledgements

This work has mainly been funded by ACT ELEGANCY, Project No 271498, which has received funding from DETEC (CH), BMWi (DE), RVO (NL), Gassnova (NO), BEIS (UK), Gassco, Equinor and Total, and is co-funded by the European Commission under the Horizon 2020 program, ACT Grant Agreement No 691712. This project is supported by the pilot and demonstration program of the Swiss Federal Office of Energy (SFOE). In addition, this work was partially funded by innosuisse via the Swiss Competence Centres for Energy Research "Efficient Technologies and Systems for Mobility" and "Heat and Electricity Storage". Further financial support was provided by the Kopernikus Project Ariadne (FKZ 03SFK5A), funded by the German Federal Ministry of Education and Research.

The authors thank D. Sutter and colleagues from Climeworks (Zürich, Switzerland), C. Zahler and S. Scherer from Industrial Solar (Freiburg, Germany) and E. Jacquemoud and R. Decorvet from MAN Energy Solutions (Zürich, Switzerland) for the fruitful discussions, feedback and data provision.

Conflicts of interest

There are no conflicts to declare.

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