COMPREHENSIVE LIFECYCLE ASSESSMENT OF DIRECT AIR CAPTURE SYSTEMS FOR CARBON-DIOXIDE REMOVAL FROM THE ATMOSPHERE

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Abstract

This thesis evaluates the environmental performance of Direct Air Capture (DAC) technologies through a review of life cycle assessments (LCA), focusing on low-temperature systems integrated with solar energy for adsorbent regeneration.

The methodology combines ISO-standardized LCA modeling with experimental validation using silica-supported polyethyleneimine adsorbents under photothermal regeneration conditions. Three heating configurations (natural gas, solar-thermal, and hybrid solar-electric) were evaluated across diverse geographical regions.

Results show solar-based systems significantly outperform conventional systems, with potential emissions reductions up to 290 kg CO_2-eq per ton of CO_2 captured. Laboratory experiments demonstrated thermal system desorption required 3.0 GJ/kg CO_2 (0.07% efficiency), while the photothermal system required 707 MJ/kg CO_2 (0.29% efficiency), confirming photothermal regeneration's feasibility for low-energy DAC operation.

While regeneration energy dominates environmental impact, infrastructure, transport, and storage collectively contribute significantly. DAC deployment should prioritize regions with abundant solar resources and clean electricity grids, with policy frameworks incentivizing these optimal configurations.

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The most heartfelt acknowledgment is reserved for my parents. It is their unconditional love and support that have enabled me to pursue and achieve my academic goals. With immense gratitude and a sense of fulfillment, I dedicate this thesis to my mother, Patromina, whose sacrifices and inspiration have been the foundation of my academic pursuits.

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List of Abbreviations

AR	Afforestation/Reforestation
BECCS	Bioenergy with Carbon Capture and Storage
BCR	Blue Carbon Restoration
CDR	Carbon Dioxide Removal
CCUS	Carbon Capture, Utilization, and Storage
CM	Carbon Mineralization
CO_2	Carbon Dioxide
DAC	Direct Air Capture
DACCS	Direct Air Carbon Capture and Storage
DACCU	Direct Air Carbon Capture and Utilization
ESA-DAC	Electro-Swing Adsorption Direct Air Capture
EW	Enhanced Weathering
GWP	Global Warming Potential
HOMER	Hybrid Optimization of Multiple Energy Resources
HT-DAC	High-Temperature Direct Air Capture
IEA	International Energy Agency
IPCC	Intergovernmental Panel on Climate Change
ISO	International Organization for Standardization
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LCIA	Life Cycle Impact Assessment
LM	Land Management
LT	Low Temperature
LT-DAC	Low-Temperature Direct Air Capture
m-DAC	Membrane Direct Air Capture
MOF	Metal-Organic Framework
NETs	Negative Emission Technologies
NOx	Nitrogen Oxides
OF	Ocean Fertilization
PEI	Polyethylenimine
POC	Proof of Concept
POPs	Porous Organic Polymers
SCS	Soil Carbon Sequestration
SI	Silica Gel
SI-PEI	Silica-supported Polyethyleneimine
TSA	Temperature Swing Adsorption

Chapter 1

Introduction

1.1 Thesis Motivation

Climate change represents one of the most pressing challenges of our time, demanding innovative solutions to mitigate its effects and stabilize atmospheric greenhouse gas concentrations. The global average temperature has risen by approximately 1.1°C above pre-industrial levels and continues to climb at an alarming rate, threatening to exceed the critical 1.5°C threshold established by the Paris Agreement (IPCC, 2023). To meet the goals outlined in this landmark accord—limiting warming to well below 2°C, preferably to 1.5°C—greenhouse gas emissions must be reduced by 45% below 2010 levels by 2030 and reach net-zero by 2050 (IPCC, 2018).

Carbon dioxide (CO_2) remains the primary driver of anthropogenic climate change, with global emissions reaching 36.6 $GtCO_2$ in 2022, continuing a long-term upward trend from approximately 22 $GtCO_2$ in 1990 and 2 $GtCO_2$ in 1900 (Friedlingstein et al., 2022; Ritchie & Roser, 2024). Given emissions in 2023 were approximately 37 $GtCO_2$ (Ritchie & Roser, 2024) and the 2030 target is around 18 $GtCO_2$, 19 $GtCO_2$ per year must be avoided or removed by 2030 through a combination of rapid emission reductions and carbon dioxide removal (CDR) technologies. This immense challenge necessitates not only rapid decarbonization across all sectors but also the development and deployment of negative emissions technologies (NETs).

Figure 1.1 shows the global average temperature compared to the 1961-1990 baseline, highlighting the concerning upward trend. Figure 1.2 provides estimates of annual CO_2 emissions in billions of tonnes from fossil fuels and industry, demonstrating the exponential increase in emissions since the industrial revolution, with only temporary plateaus during major global events.

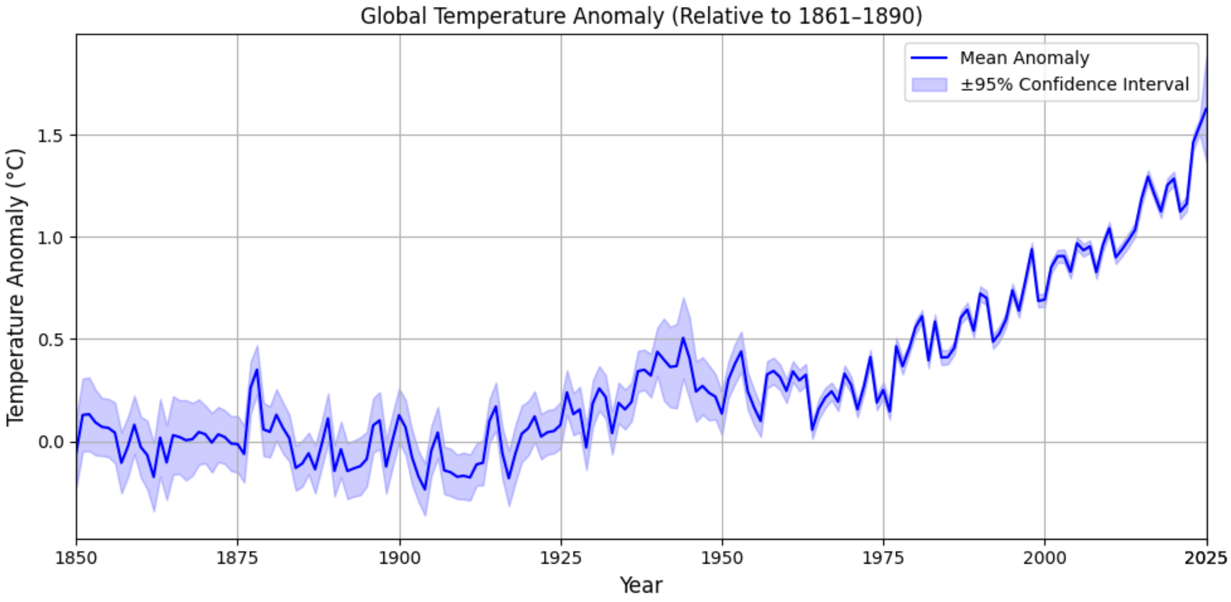

Figure 1.1: Global average temperature anomaly showing the acceleration of global warming in recent decades. Adapted from Met Office Hadley Centre (2023)

The urgent need to mitigate climate change has driven considerable attention towards carbon capture technologies. Direct Air Capture (DAC) technology has emerged as a promising approach to extract CO_2 directly from ambient air, offering potential for both carbon removal and utilization in various applications. This technology holds significant promise as a tool for achieving global climate goals, such as those outlined in the Paris Agreement, which seeks to limit global warming to 1.5°C above pre-industrial levels.

This research is motivated by the need to address the climate crisis through technological innovation and critical assessment of DAC technology. However, despite DAC's potential, significant challenges remain. High energy consumption and substantial environmental impacts associated with traditional DAC systems have hindered their widespread deployment. Furthermore, the scalability of DAC remains uncertain, with many uncertainties surrounding the long-term feasibility of large-scale implementation.

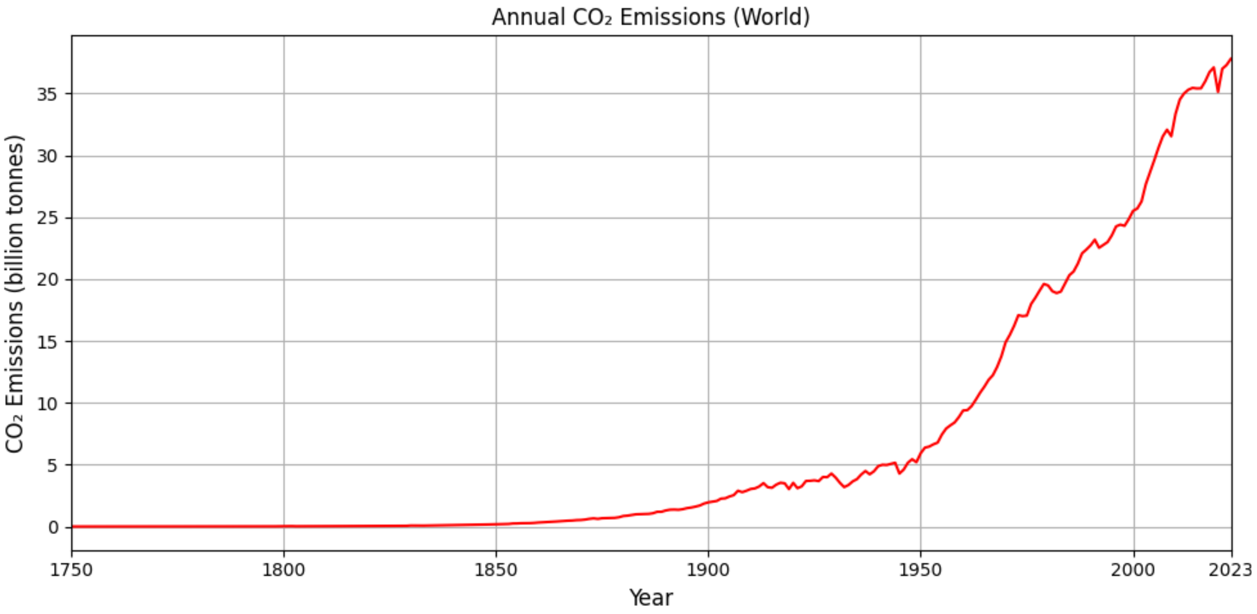

Figure 1.2: Global annual CO_2 emissions from fossil fuels and industry (1750-2022).
Adapted from Ritchie & Roser (2024).

This research seeks to address these critical challenges by conducting an extensive review of DAC technology and modifying an existing Life Cycle Assessment (LCA) from the literature. Through this modified LCA, this thesis aims to evaluate the environmental performance, life cycle efficiency of DAC, identify the most environmentally impactful stages in the process, and identify areas that require optimization. The motivation for this study lies in the desire to contribute to the scientific understanding of DAC's potential, enhance its efficiency, and provide insights into improving its environmental sustainability. By exploring innovative approaches, such as integrating solar thermal energy for adsorbent regeneration and utilizing renewable energy sources, the research aims to make DAC a more viable solution for large-scale carbon removal and climate change mitigation.

The outcomes of this research have significant relevance in Canada, where the Federal Government continues to focus on climate change mitigation strategies to achieve Net Zero emissions by 2050. These plans include a target of reducing emissions by 40 to 45 percent below 2005 levels by 2030, which is a significant step toward the net-zero goal (Environment and Climate Change, 2022). Furthermore, through the systematic analysis and experimental-based recommendations, this thesis strives to contribute meaningful knowledge that can guide policy decisions, technological development, and implementation strategies for DAC as part of the broader climate mitigation portfolio.

1.2 Background and Context

The growth of clean energy technologies holds promise for achieving net zero greenhouse gas emissions and limiting global warming to 1.5°C (IEA, 2023). The Intergovernmental Panel on Climate Change (IPCC) proposed Carbon Capture, Utilization, and Storage (CCUS) as a technique to limit increasing atmospheric CO_2 concentrations (Metz et al., 2005). Within this broader category, Direct Air Capture (DAC) represents an approach that targets CO_2 already dispersed in the atmosphere rather than capturing emissions at point sources such as power plants or industrial facilities. Negative Emission Technologies (NETs), also known as Carbon Dioxide Removal (CDR) technologies such as DAC, play a crucial role in potential climate scenarios to curb global warming below 2°C.

DAC technologies extract CO_2 from ambient air through chemical processes, typically using either liquid solvent systems (absorption - primarily aqueous alkali solutions) or solid sorbent materials (adsorption - such as amine-functionalized adsorbents) (Fasihi et al., 2019; Sanz-Pérez et al., 2016). The captured CO_2 can then be permanently sequestered underground, referred to as Direct Air Carbon Capture and Storage (DACCS), or utilized as a feedstock for various products, ranging from synthetic fuels to building materials referred to as Direct Air Carbon Capture and Utilization (DACCU). Figure 1.3 illustrates a schematic representation of DAC technology pathways, from sorption process to desorption process to storage and or utilization stages.

Studies by Keith et al. (2018) and Breyer et al. (2019) emphasize the potential in reducing atmospheric CO_2 concentrations. Keith et al. (2018) provided a design and cost basis for a comprehensive DAC system, focusing on aqueous KOH sorbents, enabling continuous operation with durable, low-cost cooling-tower hardware. The system's design ensures long contactor lifetimes despite environmental exposure, while centralized CO_2 regeneration enhances efficiency and scalability. Breyer et al. (2019) stressed DAC's importance, highlighting the need for research on CO_2 sorbents, waste heat integration, and a global CO_2 inventory which will accelerate the learning rate and cost projections of DAC.

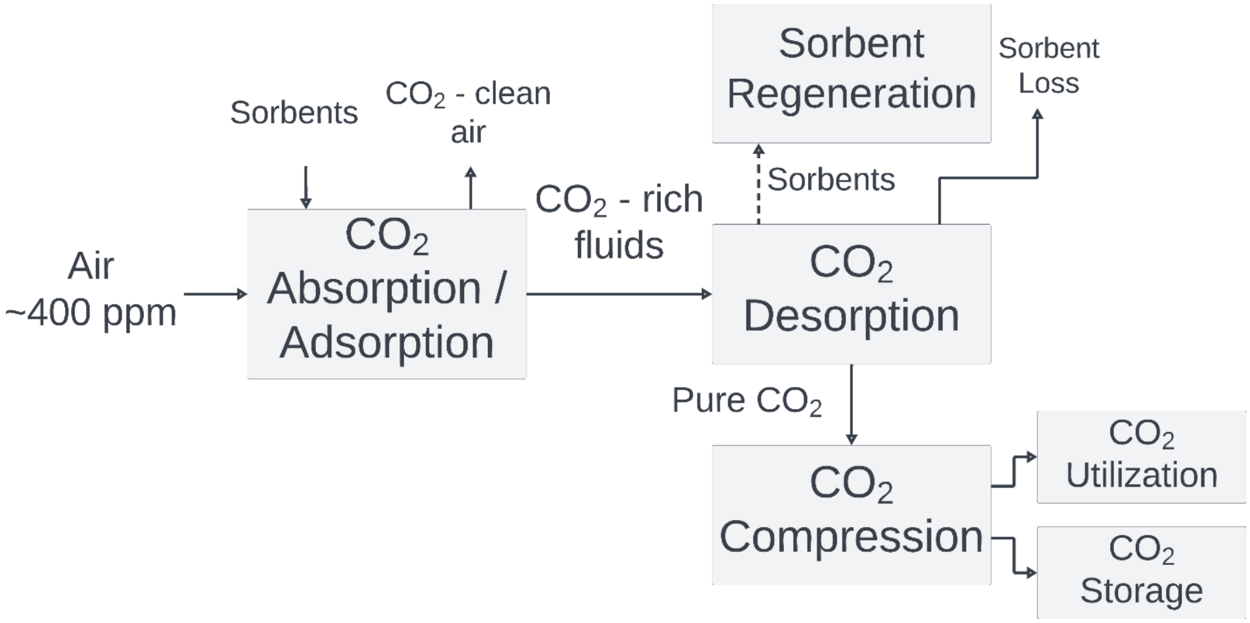

Figure 1.3: Schematic representation of DAC technology pathways—liquid solvent and solid sorbent systems—showing key process steps from atmospheric capture through regeneration and CO_2 recovery.

Unlike conventional point-source carbon capture, DAC offers several distinct advantages. First, DAC facilities can be situated strategically near renewable energy sources, geological storage sites, or utilization facilities, minimizing transportation requirements (Realmonte et al., 2019). Second, DAC can target emissions from difficult-to-decarbonize sectors such as aviation, agriculture, and existing building stocks, which together account for approximately 27–30% of global greenhouse gas emissions (Smith et al., 2016), IPCC, 2022). Third, when powered by low-carbon energy sources and combined with permanent storage, DAC can achieve net-negative emissions of up to 1,000 kg CO_2 per ton captured, effectively reversing some accumulated atmospheric carbon burden (Breyer et al., 2020). Finally, compared to biological carbon removal approaches like afforestation or bioenergy with carbon capture and storage (BECCS), DAC systems generally require less land area and pose fewer potential conflicts with food production (Fuss et al., 2018; Smith et al., 2016).

DAC technology has progressed substantially in recent years, moving from laboratory demonstrations to commercial-scale pilot plants. Notable developments include Climeworks' Orca plant in Iceland, which began operation in 2021 as the world's first commercial DAC facility dedicated to permanent geological storage, capturing approximately 4,000 tons of CO_2 annually (Climeworks, 2022). At this rate, Orca would need to operate for over 9 million years to offset just one year of global emissions, highlighting the challenge of scaling up DAC technology. Carbon

Engineering has demonstrated large-scale DAC technology in British Columbia, employing liquid solvent systems capable of capturing 1 million tons of CO_2 per year in future commercial implementations (Keith et al., 2018). While a step forward, this still offsets less than 0.003% of annual global emissions, which reinforces the urgent need to expand DAC capacity and reduce costs.

Despite these advances, DAC faces significant challenges that limit widespread adoption. Current DAC technologies consume large amounts of energy in the form of both electricity and heat. The heat demand for sorbent regeneration in DAC systems vary substantially based on the sorbent type. For liquid sorbents, the regeneration process with temperature of (>800 °C) demands approximately 6 to 10 GJ of energy per ton of CO_2 captured. In contrast, solid sorbents are relatively more energy efficient with regeneration temperatures ranging from 80- 100 °C, requiring only about 4 to 6 GJ per ton of CO_2 captured. The electricity consumed to operate fans, pumps and control system in DAC plants can range from 300 to 900 kWh per ton of CO_2 captured. This represents a significant portion of the total DAC system energy usage, often between 20 and 40% (Fasihi et al., 2019; Ozkan, 2021).

The cost estimates for DAC vary widely, with estimates in the literature ranging from 100-1000 USD per ton of CO_2 captured, which is significantly higher than many alternative mitigation strategies (Broehm et al., 2015). Fasihi et al. (2019) outlined levelized capture costs of CO_2, ranging from 100 to 250 USD, projecting reductions post-2050, while Breyer et al. (2020) estimated the cost of capturing CO_2 will be approximately 125 USD/tCO_2 in the year 2030 and predicted that the cost of DAC will decline after 2040 due to technological advancements and growing acceptance. For comparison, the combustion of 1 ton of natural gas emits approximately 2.75 tons of CO_2. At a natural gas price of 201 USD per ton (based on CNBC, 2025), capturing these emissions would raise the effective cost to 545–889 USD per ton of natural gas — 2.7 to 4.4 times higher than the cost of fossil fuel use without capture. These figures highlight the urgency of reducing DAC costs, ideally toward 100 USD per ton or lower, to make large-scale deployment both technically and economically feasible (Ozkan et al. 2022). For this reason, various academic and startup initiatives continue to explore novel materials, regeneration techniques, and energy integration strategies aimed at dramatically improving efficiency and reducing cost (An et al., 2023; Bouaboula et al., 2024; Sanz-Pérez et al., 2016).

The scientific and policy communities remain divided regarding DAC's ultimate role in climate mitigation strategies. An inter-model analysis by Realmonte et al. (2019) suggested DAC could substantially reduce mitigation costs if rapidly scaled to remove approximately 1.5 Gt CO_2 annually by 2100. However, Chatterjee & Huang (2020) argued that such projections underestimate practical challenges related to adsorbent production, energy supply chains, and financing requirements. Concerns have also been raised regarding DAC's potential burden on electricity grids and power sectors. Bistline & Blanford (2021) projected that even under optimistic deployment scenarios, DAC would consume less than 5% of global electricity, while complementing electrification and energy storage efforts in deep decarbonization pathways. Conversely, critics have questioned whether resources directed toward DAC development might be more effectively allocated to preventive mitigation measures and renewable energy expansion (Smith et al., 2016).

Despite these ongoing debates, DAC offers distinct advantages that maintain research momentum and policy interest. Unlike biological carbon removal approaches, DAC has minimal land and water requirements, reducing potential conflicts with food security and biodiversity conservation (Fuss et al., 2018). Additionally, DAC's flexibility regarding location and operational timing provides unique benefits for addressing distributed emissions sources (Smith et al., 2016). The National Research Council, USA, emphasized that successful DAC implementation requires addressing risks associated with long-term CO_2 storage, including potential leakage and induced seismicity (National Research Council, 2015). Recent lifecycle analyses estimate energy requirements of up to 45 GJ per ton of CO_2 removed for certain DAC configurations, highlighting the need for efficiency improvements to achieve economic viability (van der Giesen et al., 2017). By comparison, energy requirements must generally fall below 6 GJ/tCO_2 to achieve capture costs under 100 USD per ton, a widely cited benchmark for large-scale deployment (Fasihi et al., 2019). Advanced systems targeting 2–4 GJ/tCO_2 are considered to be more aligned with long-term climate and economic targets, whereas current experimental setups often exceed these targets by a wide margin, highlighting the need for further energy optimization.

Clearly, questions about the final role of DACCS technology to climate mitigation continue. Outstanding problems exist about realistic scaling potentials, lifecycle impacts monitoring and understanding holistic trade-offs like resource consumption. It is for this reason that LCA is required.

LCA is a widely acknowledged tool for environmental evaluation in academia and industry (Müller et al., 2020). During every stage of a process or product's life cycle, it can be used as an appropriate and flexible assessment method to pinpoint the main causes of environmental effect and gauge the complete environmental performance. This research aims to unlock new insights and strategies for enhancing the efficiency, sustainability, and scalability of DAC technology. Through rigorous experimentation, analysis, and modeling, we seek to elucidate the potential benefits and drawbacks of utilizing solar thermal energy for CO_2 desorption and adsorbent regeneration in DAC systems, thereby paving the way for the development of more effective and environmentally sustainable climate mitigation solutions.

1.3 Scientific Question and Technological Gap

Despite the growing interest in DAC technology, several scientific and technological gaps persist. While numerous studies have investigated DAC's potential as a CO_2 mitigation tool, there is still limited understanding regarding the full life cycle environmental impacts of DAC systems. While traditional DAC systems operate using temperature-swing adsorption methods, they tend to be energy-intensive, requiring large amounts of heat for the desorption of CO_2 and regeneration of adsorbents. This thermal energy requirement constitutes the largest energy demand within DAC operations. This energy consumption limits the scalability of DAC and raises concerns about its overall sustainability.

To address these gaps, this research seeks to explore the potential benefits of integrating solar heat into the adsorbent regeneration process to reduce emissions associated with the energy consumed by DAC systems. Utilizing solar radiation to directly generate the heat required for CO_2 desorption from capture materials (referred to as photothermal desorption) has the potential to significantly reduce the conventional energy demand of the CO_2 capture process. This integration could greatly enhance the energy efficiency and sustainability of DAC systems, making them more viable for large-scale implementation. This study will investigate whether photothermal desorption can reduce energy consumption and environmental impact compared to traditional temperature-swing desorption methods.

Additionally, this research will address another key gap: the lack of comprehensive assessments of DAC's feasibility for large-scale deployment. While DAC systems have demonstrated their potential in laboratory and pilot-scale settings, there are still uncertainties regarding their economic viability, scalability, and the associated environmental impacts at a global scale. Most existing LCA studies typically focus on specific deployment scenarios without adequately addressing how performance varies across different climatic conditions, energy mixes, and infrastructure contexts. This study aims to assess the environmental "hotspots" in DAC processes and identify areas that require optimization to improve the efficiency and sustainability of the technology.

The fundamental scientific questions that this research aims to answer are:

I. Can the integration of solar thermal energy for CO_2 desorption and adsorbent regeneration significantly reduce energy consumption and environmental impact in DAC systems compared to traditional temperature-swing adsorption methods?
II. What are the primary environmental hotspots in DAC systems that require improvement to make DAC a viable solution for large-scale CO_2 mitigation?

By addressing these questions and knowledge gaps, this research aims to provide a more comprehensive understanding of DAC's environmental implications, identify promising pathways for technology optimization, and contribute to evidence-based decision-making regarding climate mitigation strategies.

1.4 Goals and Objectives

The primary goal of this research is to evaluate the current state of the art pertaining to LCA of DAC as reported in the literature. Building upon established LCA methodologies, this research adapts and extends a model from Madhu et al. (2021) to identify environmental impact "hot-spots" and conduct sensitivity analysis to determine the effects of different parameters such as regeneration energy on the LCA results Through this approach, the research identifies areas for optimization to improve the energy efficiency of DAC systems under different scenarios. The specific objectives of this research are as follows:

i. To provide a state-of-the-art review of existing LCA studies on DAC technologies, to establish a solid foundation by understanding the current landscape, methodologies, identifying gaps and areas for improvement.

ii. To develop a model to comprehensively assess the environmental impacts of DAC technology, focusing on parameters such as the Global Warming Potential (GWP) and life cycle efficiency.

iii. To experimentally demonstrate and compare thermal and photothermal regeneration of CO_2-loaded adsorbents, assessing energy input and efficiency under lab-scale solar-simulated conditions to establish proof-of-concept for light-driven desorption.

1.5 Outline of the Thesis

This thesis is structured to guide the reader through a comprehensive exploration of DAC technology, its environmental impacts, and potential for large-scale deployment: To facilitate the investigation of the research questions, the structure is as follows:

- Chapter 1: Introduction – The current chapter establishes the motivation, background, research questions, and objectives of this study. It contextualizes DAC technology within broader climate mitigation efforts and outlines the knowledge gaps this research aims to address.
- Chapter 2: Literature Review – This chapter provides a comprehensive overview of the current DAC landscape, including evolution and current state of DAC technologies, with a focus on LCAs, energy requirements, environmental impacts, and potential innovations. The literature review will also explore the knowledge gaps, challenges and opportunities in DAC, including the integration of renewable energy sources such as solar energy.
- Chapter 3: Methods – This chapter describes the LCA methodology employed in this study. It outlines the assumptions, inputs, and data sources used in the analysis and explains the model's structure. Specific methodological elements include LCA framework and standards employed, system boundaries, functional units, and impact assessment methods, data sources and software employed, key assumptions regarding technology parameters, energy systems, and geographical scenarios. It also explains the methods used to incorporate experimental results obtained using a photothermal approach for CO_2 adsorbent regeneration into the LCA analysis. This chapter will also discuss how the model is used to assess the environmental impacts of DAC, including GWP, regeneration energy consumption, and life cycle efficiency.

- Chapter 4: Results and Discussion – This chapter presents the findings of the LCA, including the environmental performance in terms of GWP and life cycle efficiency across different geographical locations and system configurations and a discussion of the implications of the results. The chapter will also provide a detailed analysis of the regeneration energy sensitivity analysis with respect to the GWP of DAC systems and comparison with experimental results suggesting process improvements.
- Chapter 5: Conclusions and Future work – The final chapter summarizes the main findings of the research and offers conclusions and recommendations for policymakers, DAC developers, and researchers. The chapter also highlights areas for future research to improve DAC's environmental performance and scalability.

Chapter 2

Literature Review

Note on Previously Published Content

Portions of this chapter have been adapted from a previously published peer-reviewed article:

Victor Eke, Tanay Sahu, Kulbir Kaur Ghuman, Marina Freire-Gormaly, Paul G. O'Brien, "A comprehensive review of life cycle assessments of direct air capture and carbon dioxide storage," *Sustainable Production and Consumption,* Volume 55, 2025, Pages 217–241, ISSN 2352-5509. DOI: 10.1016/j.spc.2025.02.017.

My Contribution (Victor Eke): Writing – original draft, Writing – review & editing, Methodology, Investigation, Formal analysis, Data curation, Conceptualization.

Co-authors Contributions:

Tanay Sahu: Writing – review & editing, Methodology, Investigation.

Kulbir Kaur Ghuman: Writing – review & editing, Formal analysis.

Marina Freire-Gormaly: Supervision, Writing – review & editing, Conceptualization.

Paul G. O'Brien: Supervision, Writing – review & editing, Conceptualization.

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2.0 Chapter Introduction

This chapter presents a comprehensive review of the current literature on DAC technologies, with a focus on technological developments, LCA, environmental impacts, and integration with renewable energy sources. The review synthesizes findings from peer-reviewed publications, industry reports, and governmental assessments to establish the state of knowledge regarding DAC as negative emissions technology (NET). By examining both the experimental investigations and practical implementations of DAC systems, this chapter aims to identify knowledge gaps and research opportunities that inform the subsequent chapters of this thesis. The literature review begins with an overview of NETs, DAC technology evolution and current systems, proceeds through environmental assessment methodologies with particular attention to life cycle analysis and concludes with a discussion of energy requirements and integration possibilities.

2.1 Negative Emission Technologies

As we move further into the 21st century, the need to reduce CO_2 emissions into the atmosphere will become increasingly important if 1.5°C or 2°C targets are to be met (IPCC, 2022). The primary method used to reduce emissions is decarbonizing the energy sector, with the focus on transitioning away from fossil fuels to renewable energy sources. In addition to reducing carbon emissions by phasing out fossil fuels and enhancing energy efficiency, NETs have become a key component of net-zero strategies (IEA, 2021).

The technologies associated with NETs are often referred to as CDR technologies. A NET is a technology which permanently removes CO_2 from the atmosphere, either by physically extracting it through chemical processes or by increasing the size of natural carbon sinks which absorb it, such as vegetation (NASEM, 2019). While emissions reduction remains the primary focus of climate mitigation efforts, it is becoming increasingly clear that there may be a need to physically remove CO_2 from the atmosphere to achieve climate goals. NETs will undoubtedly play an important role in tackling climate change, but it remains uncertain which specific NETs will be widely deployed at scale.

This uncertainty stems from the fact that, apart from afforestation/reforestation, most NETs are not mature technologies and have not been deployed on large scales (NASEM, 2019). Within the field of NETs, there is considerable overlap in their categorization. NETs which increase the size of carbon sinks can be referred to as natural climate solutions, soil carbon sequestration, or terrestrial carbon removal and sequestration. This can make categorizing NETs challenging but broadly speaking there are seven main types (EASAC, 2018; NASEM, 2019): afforestation/reforestation (AR), bioenergy with carbon capture and storage (BECCS), blue carbon restoration (BC), direct air capture (DAC), carbon mineralization (CM), land management (LM), and ocean fertilization (OF).

The range of NETs being pursued varies from more conventional techniques like afforestation and reforestation to novel approaches like BECCS and Direct Air Carbon Capture and Storage

(DACCS) as shown in Figure 2.1. With respect to NETs, Fuss et al. (2018) and Minx et al. (2018) presented an in-depth techno-economic review of a portfolio of CDR technologies. Their CDR technology portfolio included Afforestation and Reforestation (AR), biochar, Soil Carbon Sequestration (SCS), Enhanced Weathering (EW), Ocean Fertilization (OF), Bioenergy with Carbon Capture and Storage (BECCS), and Direct Air Carbon Capture and Storage (DACCS).

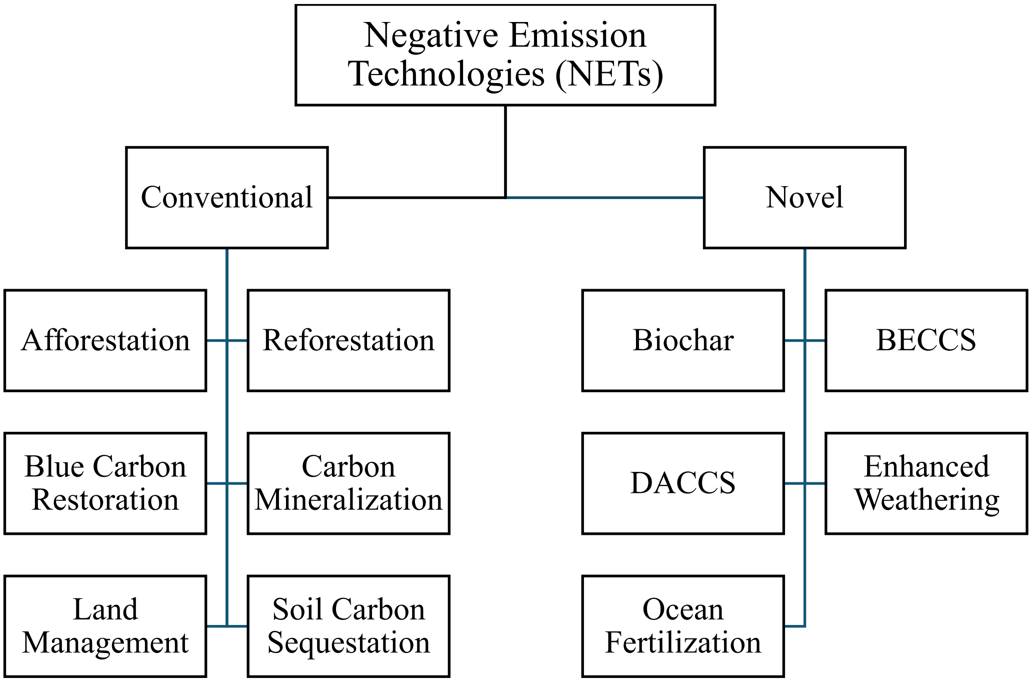


Figure 2.1: Examples of negative emission technologies

Each of these approaches offers distinct advantages and limitations regarding carbon removal potential, permanence of storage, land and resource requirements, environmental impacts, and cost-effectiveness, all of which must be carefully evaluated in the context of comprehensive climate mitigation strategies.

Among these NETs, DAC offers several distinct advantages that make it particularly promising for large-scale CDR. Unlike nature-based approaches, DAC is highly scalable and offers significant flexibility in deployment due to fewer ecological constraints. However, DAC deployment is practically influenced by factors such as the proximity to renewable energy sources, the availability of geological storage sites or CO_2-utilizing industries, and land-use considerations. Ideally, DAC systems are suited to regions with abundant renewable energy resources, minimal land-use conflicts, and proximity to suitable CO_2 storage or utilization infrastructure. DAC systems can thus effectively be powered by clean energy sources, aligning with broader decarbonization goals. By capturing CO_2 directly from ambient air, DAC reduces the need for long-distance CO_2 transport, lowering associated costs and environmental impacts. Furthermore, the high-purity CO_2 captured through DAC can be utilized by various industries or permanently sequestered underground. This thesis focuses on DAC technology due to its flexibility, scalability potential, and ability to be integrated with renewable energy sources—specifically exploring innovations that could significantly reduce its energy requirements, the primary barrier to widespread implementation.

2.2 Evolution and Current State of Direct Air Capture Technologies

2.2.1 Historical Development of Direct Air Capture

DAC as a concept emerged in the late 1990s, although the fundamental chemistry of CO_2 sorption has been understood since the early 20th century. The earliest conceptual proposals for extracting CO_2 directly from ambient air were published by Lackner et al. (1999), who proposed using strong alkali solutions to capture atmospheric CO_2. This pioneering work demonstrated that while thermodynamically challenging due to the dilute concentration of CO_2 in the atmosphere (approximately 420 ppm compared to 10-15% in power plant flue gas), DAC remained technically viable.

Throughout the 2000's, research momentum accelerated with significant contributions from Zeman & Lackner (2004), who explored various sorbent materials and process configurations to reduce energy penalties. The transition from theoretical concept to practical demonstration accelerated in the late 2000's and early 2010's. Holmes & Keith (2012) published influential work detailing a liquid solvent-based absorption system using hydroxide solutions, which later became the foundation for Carbon Engineering's approach. Simultaneously, researchers at ETH Zürich developed early prototypes of solid sorbent systems using amine-functionalized materials, which eventually led to the founding of Climeworks (Gebald et al., 2011). These parallel development paths established the two main technological approaches that continue to dominate the DAC landscape today.

The past decade has witnessed rapid advancement in DAC technology, moving from laboratory-scale demonstrations to pilot plants and the first commercial facilities. This progression reflects growing recognition of DAC's potential role in climate mitigation scenarios and has been supported by increased public and private funding (Fasihi et al., 2019; Realmonte et al., 2019). This evolution mirrors the development trajectory of other environmental technologies, with initial concepts requiring several decades to reach commercial implementation.

2.2.2 Current Direct Air Capture System Classifications

Current DAC technologies fall primarily into two categories based on their sorbent material and process configuration: liquid solvent systems and solid sorbent systems. Each approach offers distinct advantages and challenges regarding energy requirements, scalability, and environmental impacts.

2.2.2.1 High temperature direct air capture systems

The high temperature direct air capture (HT-DAC) systems often referred to as liquid solvent DAC typically employ aqueous alkaline solutions, most commonly potassium hydroxide (KOH) or sodium hydroxide (NaOH), to chemically absorb CO_2 from ambient air. In these systems, air is contacted with the alkaline solution, where CO_2 undergoes multiple intermediate reactions before ultimately forming carbonate ions. Initially, CO_2 reacts with water to form carbonic acid (H_2CO_3). The hydroxide ions (OH^-) then sequentially neutralize the carbonic acid, eventually yielding carbonate ions (CO_3^{2-}) and water. The overall net reaction is summarized as follows (Keith et al., 2018):

$$CO_2 + 2OH^- \rightarrow CO_3^{2-} + H_2O \tag{2.1}$$

The resulting potassium carbonate solution is then processed through a series of steps to regenerate the original hydroxide solution and produce a concentrated CO_2 stream. This regeneration typically involves precipitation of calcium carbonate through addition of calcium hydroxide, followed by calcination at high temperatures (> 800°C) to release CO_2 and regenerate the calcium hydroxide (Keith et al., 2018):

$$K_2CO_3 + Ca(OH)_2 \rightarrow 2KOH + CaCO_3 \tag{2.2}$$

$$Na_2CO_3 + Ca(OH)_2 \rightarrow 2NaOH + CaCO_3 \tag{2.3}$$

$$CaCO_3 \rightarrow CaO + CO_2 \tag{2.4}$$

$$CaO + H_2O \rightarrow Ca(OH)_2 \tag{2.5}$$

Liquid solvent systems benefit from fast reaction kinetics and high CO_2 absorption rates, allowing for efficient capture even at low atmospheric concentrations. The use of non-volatile solvents minimizes material losses, and the systems can operate continuously with relatively simple air contactors (Keith et al., 2018). However, they typically require high-temperature heat for sorbent regeneration (800-900°C), necessitating natural gas, biomass or hydrogen thermal inputs. Additionally, these systems often involve multiple chemical processing steps and may face corrosion challenges due to the caustic nature of the solutions (Realmonte et al., 2019).

2.2.2.2 Low temperature direct air capture systems

Low temperature direct air capture (LT-DAC) systems, often referred to as solid sorbent DAC, utilize porous materials functionalized with CO_2-reactive groups, most commonly amine-based compounds, to selectively capture CO_2 through chemical adsorption. In a typical configuration, ambient air is passed through modules containing the solid sorbent, where CO_2 is chemically bound to the surface. Once the sorbent approaches saturation, the modules are isolated and subjected to a temperature swing (typically 80-120°C), vacuum swing, or a combination of both to release concentrated CO_2 and regenerate the sorbent material (Fasihi et al., 2019).

The chemical mechanism for amine-based solid sorbents involves the formation of carbamates or carbamic acid under dry conditions, or bicarbonate in the presence of water (Drechsler & Agar, 2020):

$$2RNH_2 + CO_2 \rightarrow RNHCOO^- + RNH_3^+ \text{ (carbamate formation)} \qquad (2.6)$$

$$RNH_2 + CO_2 + H_2O \rightarrow RNH_3^+ + HCO_3^- \text{ (bicarbonate formation)} \qquad (2.7)$$

LT-DAC systems offer several advantages, including lower regeneration temperatures (80-120°C vs. 800-900°C for HT-DAC), which enables the use of low-grade waste heat or direct electric heating. Their modular architecture facilitates mass production and iterative deployment, and they typically involve fewer process steps compared to liquid systems (Fasihi et al., 2019). However, solid sorbent systems face challenges related to sorbent stability over repeated cycling, potential degradation from atmospheric contaminants, and mass transfer limitations within the solid material (Shi et al., 2020). Figure 2.2 illustrates the process flow diagram for both HT-DAC and LT-DAC systems, highlighting the key process steps, from the ambient air drawn in by the air contactor to the adsorbent regeneration process and CO_2 recovery.

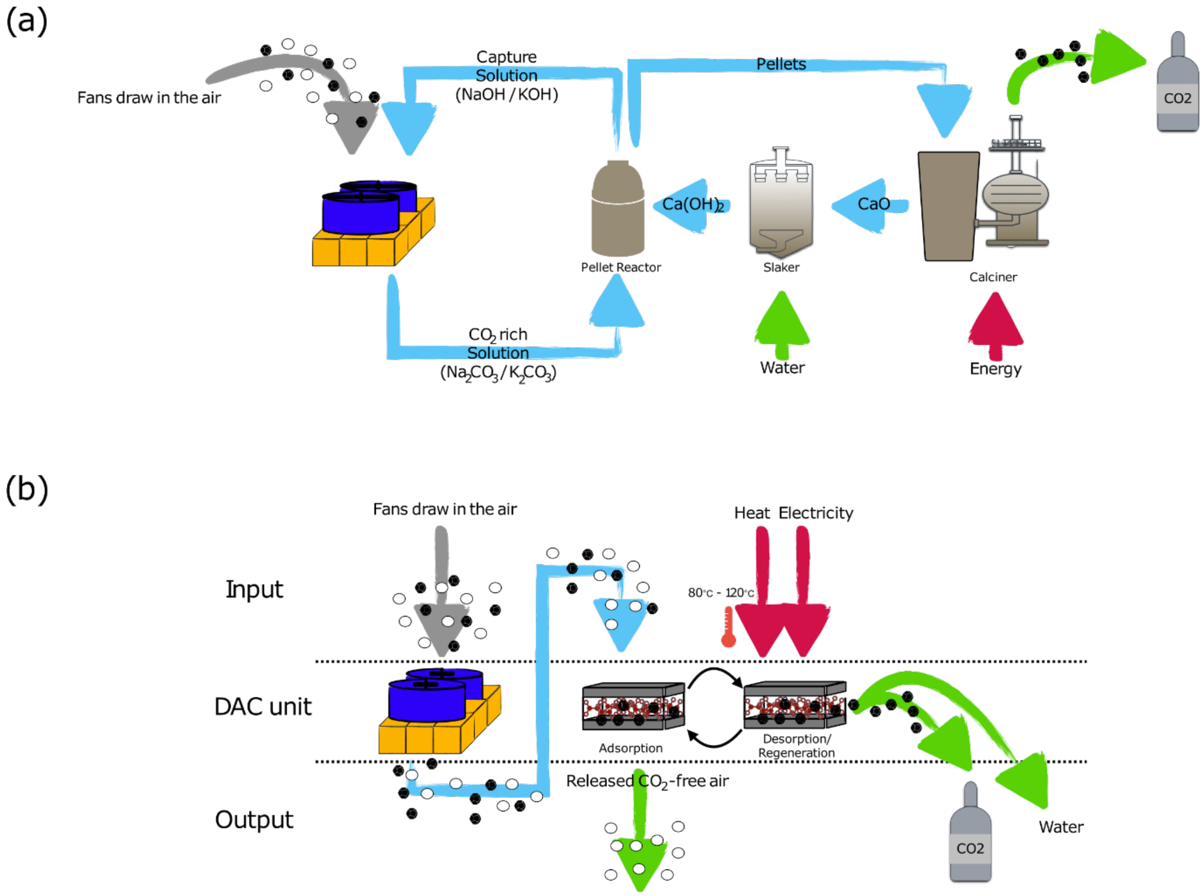


Figure 2.2: Process flow diagram of the (a) HT-DAC and (b) LT-DAC processes.

2.2.2.3 Emerging direct air capture technologies

Two alternate DAC methods that are being investigated include Electro-swing adsorption (ESA) DAC and membrane (m)-DAC.

ESA-DAC includes electrochemical approaches, which utilize electricity directly to drive CO_2 separation from air. These systems employ electrochemical cells with specialized membranes or electrodes to concentrate and capture CO_2 through electrically driven processes (Sanz-Pérez et al., 2018). The m-DAC involves the separation of CO_2 using multiple stages of separation via membranes (Fujikawa & Selyanchyn, 2022).

While these technologies are still predominantly at the research stage, they offer potential advantages in terms of operational flexibility and integration with renewable electricity sources.

2.2.3 Current Commercial and Pilot Scale Operations

The DAC industry has progressed substantially from conceptual designs to operational facilities in the past decade. Several companies have established pilot and commercial-scale plants, demonstrating technological viability while highlighting areas that require further development and optimization.

Climeworks has emerged as a leader in LT-DAC technology deployment, with multiple operational facilities in Europe. Their Orca plant in Iceland, commissioned in 2021, represents the world's first commercial-scale DAC facility dedicated to permanent CO_2 storage, with a nominal capacity of 4,000 tons of CO_2 removal annually (Climeworks, 2022). The plant utilizes geothermal energy for both electricity and heat requirements and injects captured CO_2 into basaltic formations for permanent mineralization in partnership with Carbfix. Climeworks has also operated smaller facilities in Switzerland and Italy, some of which supply captured CO_2 for utilization applications such as carbonated beverages and greenhouse fertilization (Deutz & Bardow, 2021).

Carbon Engineering has demonstrated HT-DAC technology at their pilot facility in Squamish, British Columbia, since 2015. This plant captures approximately 1 ton of CO_2 daily while serving as a development platform for their technology (Keith et al., 2018). The company has announced plans for commercial-scale facilities in partnership with Occidental Petroleum in the Permian Basin (Texas) and with 1PointFive in the United Kingdom, with projected capacities of up to 1 million tons of CO_2 annually (Carbon Engineering, 2023).

Global Thermostat operates a pilot plant in Huntsville, Alabama, using amine-functionalized monolithic ceramic sorbents. Their technology focuses on integration with existing industrial facilities to utilize waste heat for sorbent regeneration (Global Thermostat, 2023). Other emerging companies, including Skytree, Soletair Power, and Heirloom, are developing variations on these approaches or novel capture methods, though most remain at laboratory or early pilot scale. Table 2.1 presents a list of currently active DAC companies, their locations, and the technologies they are utilizing.

Table 2.1: Examples of currently active DAC companies.

Company	Location	Founding Year	Technology	Reference
Aircapture	USA	Founded 2019	LT-DAC	Aircapture (2025)
AspiraDAC	Australia	Founded 2022	LT-DAC	AspiraDAC(2025)
Climeworks	Switzerland	Multiple operating pilots & commercial plants	LT-DAC	Climeworks (2025)
Zero Carbon Systems (Global Thermostat)	USA	Multiple pilots (not operating) and two commercial construction projects	LT-DAC	Zero Carbon System (2025)
Heirloom Carbon	USA	Founded 2021; successful demonstration	LT-DAC	Heirloom (2025)
Infinitree	USA	Operating pilot	LT-DAC	Infinitree LLC (2025)
Noya	USA	Founded 2020	LT-DAC	Noya (2025)
Removr	Norway	Founded 2022	LT-DAC	Removr (2025)
Skytree	Netherlands	Operating pilot	LT-DAC	Skytree (2025)
Soletair Power	Finland	Operating pilot	LT-DAC	Soletair Power (2025)
Sustaera	USA	Founded 2021	LT-DAC	Sustaera (2025)
Capture6	USA	Founded 2021	HT-DAC	Capture6 (2025)
Carbon Engineering	Canada	Pilots operating; Constructing commercial facility	HT-DAC	Carbon Engineering (2023)
Mission Zero Technologies	UK	Founded 2020	ESA-DAC	Mission Zero Technologies (2025)
RepAir Carbon Capture	Israel	Founded 2020	ESA-DAC	Repair-Carbon (2025)
Verdox	USA	POC/Pilot in progress	ESA-DAC	Verdox (2025)
Carbyon	Netherlands	Founded 2019; demo	m-DAC	Carbyon (2025)

These deployments provide crucial real-world validation of DAC technologies while revealing practical challenges not evident in laboratory testing. Operational experience has highlighted issues such as sorbent degradation rates, contactor fouling from particulate matter, and mechanical system reliability under continuous operation (McQueen et al., 2021). Importantly, these facilities also generate empirical data regarding energy consumption, material requirements, and environmental impacts that can inform improved system designs and more accurate LCA studies.

2.2.4 Capture Materials and Technological Innovations

The development of improved capture materials represents one of the most active areas of DAC research, with significant implications for energy requirements, operational costs, and environmental impacts. Novel materials and system designs offer potential pathways to address current limitations.

For HT-DAC systems, research focuses on alternative hydroxide and amine solutions and process configurations to reduce the regeneration energy. Innovations include the exploration of amino acid salt solutions, which may offer lower volatility and regeneration energy requirements while maintaining favorable CO_2 absorption kinetics (Song et al., 2012). Process innovations such as bipolar membrane electrodialysis for hydroxide regeneration could potentially reduce the high-temperature calcination step in conventional liquid systems (Eisaman et al., 2011). Although solvents containing strong bases can absorb CO_2 rapidly, these solvents need high-grade heat demand for regeneration because of the strong bonding between the strong bases and CO_2.

In LT-DAC the solid sorbent materials used have seen particularly rapid development, with numerous studies investigating improved amine functionalization approaches and novel substrate materials. Polyethylenimine (PEI) remains widely used for functionalization due to its high amine content and strong CO_2 affinity, though degradation concerns have prompted research into more stable alternatives (Darunte et al., 2016). Zhang et al. (2014) presented their laboratory-scale experimental findings on the performance of PEI-silica adsorbent for capturing CO_2 from ambient air. Amine-based solid adsorbents have also been reported for the DAC process. These solid adsorbents are fabricated by impregnating polyamines into the porous materials or covalently bonding amines to the walls of them, resulting in less volatilization of amine than aqueous amine solvents. Solid physical and chemical adsorbents (Samanta et al., 2012) including activated carbon

(Ogungbenro et al., 2017), mesoporous silica (Sanz-Pérez et al., 2018), zeolites (Chen et al., 2017), metal organic frameworks (MOFs) (Belmabkhout et al., 2016), aerogel (Kong et al., 2012), covalent organic polymers (Xu et al., 2020; Yaqub et al., 2016), metal oxides (Bhatti et al., 2018), mesoporous alumina (Chen & Ahn, 2011), and porous organic polymers (POPs) (Masoumi et al., 2022) are regarded as the most recent methods for atmospheric CO_2 capture that have been explored to maximize surface area and optimize CO_2 diffusion properties.

Furthermore, the amine-based solid adsorbents can be regenerated at a lower temperature than the solid alkali carbonates and the aqueous amine solvents (Bhatti et al., 2018; Yaqub et al., 2016).

DAC efficiency depends on both sorbent materials and system architecture. Innovative contactor designs enhance air-sorbent contact efficiency, complementing material advancements in direct air capture. While traditional packed beds face inherent trade-offs between pressure drop and mass transfer, newer configurations like monolithic structures with parallel channels, hollow fiber modules, and radial flow contactors optimize this balance (Holmes & Keith, 2012; Sinha et al., 2017). Recently, Eke et al. (2024) proposed a multi-disciplinary optimization framework for air contactor design using gradient-based and heuristic methods. Their study achieved a CO_2 capture flux of 10,440 kg/m^2/yr at 0.068 USD per kg CO_2 captured, highlighting the potential for integrated design strategies to reduce DAC costs and improve mass transfer efficiency.

Beyond the core capture process, system-level innovations include heat integration strategies, advanced control systems, and hybrid configurations. Particularly promising are approaches that integrate DAC with renewable energy sources or industrial facilities to utilize waste heat and electricity (Cooper et al., 2022; Deutz & Bardow, 2021; Eke et al., 2025; Terlouw et al., 2021b). Strategic placement of DAC facilities near geological carbon storage sites or carbon utilization facilities can reduce transportation costs and energy requirements for the captured CO_2, further improving overall system economics and environmental performance (McQueen et al., 2021).

These technological improvements address key DAC deployment barriers including high energy consumption, material costs, and operational complexities. However, most innovations remain at laboratory or small pilot scale, requiring significant development before commercial implementation. Bridging the gap between theoretical performance and practical operation under real-world conditions remains a critical challenge for the field.

2.3 Life Cycle Assessment of Direct Air Capture Systems

LCA is crucial for evaluating the environmental impact of emerging technologies. Existing LCAs of DAC(CS) follow the International Organization for Standardisation (ISO) standards 14040 and 14044, ensuring a structured and standardized approach. These studies typically comprises of four essential phases: goal and scope definition, life cycle inventory (LCI), life cycle impact assessment (LCIA), and interpretation (ISO, 2014a, 2014b).

DAC systems, designed to reduce CO2 concentrations and combat climate change, pose unique challenges for LCA due to the need for reliable and complete LCI, methodological choices, and the potential variability of results. Factors such as functional units, system boundaries, and inventory data sources can influence LCA results, impacting their utility for decision-making (Arvidsson & Ciroth, 2021).

The results of DAC LCA studies can vary substantially depending on assumptions, scope boundaries, and other methodological choices. For instance, some assessments simplify the modeling by limiting life cycle phases (such as excluding production and disposal of sorbents, or the carbon storage stage) and different LCA studies require more considerations regarding the differences between many factors including location, energy sources, and system boundaries. These factors highlight the need for future development of LCA analyses for DAC which likely focuses on standardizing functional units, system boundaries, and impact assessment methods to facilitate technology comparison and policy decision-making.

2.3.1 Environmental Impacts of Current Direct Air Capture Systems from a Life Cycle Assessment Perspective

In LCA studies, environmental impacts are considered, enabling an exploration of how various indicators can provide valuable guidance in identifying real environmental issues.

LCA studies of current DAC technologies have revealed complex environmental profiles with impacts varying substantially based on system design, energy sources, and geographical location. The most extensively studied impact category is GWP, given DAC's primary purpose of climate change mitigation, though several studies have addressed broader environmental implications.

The GWP impact category can also be expressed in terms of net greenhouse gas (GHG) emissions, with a characterization factor measured in kilograms of CO_2-equivalents (kg CO_2-eq). Expressing emissions as CO_2-eq is necessary because it combines emissions from different greenhouse gases, such as methane (CH_4) and nitrous oxide (N_2O), into a single metric, accounting for their varying capacities to trap heat over a standard time horizon (typically 100 years). These net GHG emissions thus play a critical role in indicating the overall impact of a product system's life cycle on global warming and climate change, providing clear insights into tangible environmental challenges.

Net GHG Emissions refer to the overall impact of a process on greenhouse gas levels, is typically represented by Equation 2.8. Net GHG Emissions are calculated as the difference between the amount of CO_2 captured (in kgCO_2) and the CO_2 emitted (in kg$CO_{2\text{-eq}}$) during the entire process. A negative value indicates that the process results in a net removal of CO_2 from the atmosphere, while a positive value indicates net emissions.

$$\text{Net GHG Emission} = \text{CO}_2\ \text{Emitted} - \text{CO}_2\ \text{Captured} \tag{2.8}$$

In the context of LCA studies, a negative net GHG emission signifies a beneficial outcome (credit), where more CO_2 is captured and/or stored than emitted.

2.3.1.1 Net Greenhouse Gas Emissions

This section presents a comparison of the net GHGs derived from LCAs of DAC processes collected through literature review. Specifically, studies by Cooper et al. (2022), De Jonge et al. (2019), Deutz & Bardow (2021), Madhu et al. (2021), and Terlouw et al. (2021b), performed comparative analysis by evaluating pessimistic, baseline, and optimistic scenarios to assess the effectiveness of DAC(CS) systems. These scenarios represent a range of potential outcomes, shaped by different assumptions and conditions. While not all studies provided all three scenarios, the baseline scenario was consistently reported across most of the literature.

The pessimistic scenarios reflect the worst-case conditions under which DACCS technologies operate. These scenarios often assume higher energy consumption, lower efficiency of capture materials, reliance on non-renewable energy sources, and less favorable economic and regulatory conditions. As a result, they tend to show poorer performance, sometimes even net positive emissions, as seen in Cooper et al. (2022) study when grid electricity is used. These scenarios highlight the challenges and inefficiencies that need to be addressed for successful DACCS implementation.

The baseline scenario serves as a standard reference point, allowing for the comparison of different DACCS systems and offering insight into typical performance metrics. It represents the current state of the technology, with moderate energy consumption, average efficiency of capture materials, and a balanced mix of energy sources, reflecting realistic outcomes under prevailing conditions.

The optimistic scenarios illustrate the best-case conditions, assuming minimal energy consumption, high efficiency of adsorbent materials, and extensive use of renewable energy sources. These scenarios often show the highest life cycle carbon efficiency and the lowest net GHG emissions. Optimistic scenarios are vital for demonstrating the potential of DACCS technology under ideal conditions, providing aspirational targets for future research, development, and policy decisions by outlining the maximum benefits that could be achieved.

While some studies focused primarily on the baseline scenario, there were instances where certain papers did not provide any data on greenhouse gas emissions across the lifecycle of the systems they examined. For those studies that did not report quantitative greenhouse gas emissions values, they were excluded from the comparative analysis of net emissions and life cycle carbon efficiency in the results section.

Liquid sorbent and solid sorbent technologies exhibit substantial promise, each with its strengths and variations. The bar chart in Figure 2.3 represents the normalized net GHG emissions (in kg$CO_{2\text{-eq}}$/t CO_2) from various academic papers related to DAC systems. The chart positions the liquid sorbent technologies on the left and solid sorbent technologies on the right. The figure compares pessimistic, baseline, and optimistic scenarios, illustrating the variability in environmental performance among different DAC technologies. The studies included in Figure 2.3 were selected based on clear and explicit criteria: they had to provide comprehensive lifecycle data, clearly report their system boundaries, functional units, and assumptions, and employ a transparent LCA methodology consistent with ISO standards.

Within the realm of liquid sorbent technologies, several papers present noteworthy findings. De Jonge et al. (2019) stands out as particularly promising, with an optimistic baseline indicating a substantial reduction in emissions at -620 $kgCO_{2-eq}$/t CO_2. Liu et al. (2020) also demonstrates strong emissions reduction potential, specifically in the electric calciner technology, with a baseline of -840 $kgCO_2$-eq/t CO_2. Meanwhile, Madhu et al. (2021) explored various scenarios, showcasing significant emissions reduction potential from a baseline of -420 $kgCO_{2-eq}$/t CO_2 to an optimistic scenario of -454 $kgCO_{2-eq}$/t CO_2. Cooper et al. (2022), when considering renewable electricity, reveals substantial emissions reduction, with an optimistic scenario reaching -837 $kgCO_{2-eq}$/t CO_2. These findings collectively emphasize the effectiveness of liquid sorbent technologies in reducing carbon emissions.

Solid sorbent technologies, as presented in the papers by Ali et al. (2024), Deutz & Bardow (2021), Gutsch & Leker (2024), Ottenbros et al. (2024) and Terlouw et al. (2021b) also hold promise for emissions reduction strategies. Deutz & Bardow (2021) demonstrates the potential of solid sorbent technology in Hinwil (Switzerland), with a baseline emissions reduction of -854 $kgCO_{2-eq}$/t CO_2. Terlouw et al. (2021b), presents a range of outcomes, from a baseline of -737 $kgCO_{2-eq}$/t CO_2 to an optimistic scenario of -966 $kgCO_{2-eq}$/t CO_2, indicating the effectiveness of solid sorbent technologies in different contexts. Arjoon et al. (2022), further underscores the capacity of solid sorbent technology with a baseline emissions reduction of -799 $kgCO_{2-eq}$/t CO_2. In a study conducted by Ali et al. (2024) across ten European locations, most sites demonstrated a GWP lesser than or equal to -950 $kgCO_{2-eq}$/t CO_2. Similarly, Ottenbros et al. (2024) reported prospective GWP values ranging from -360 to -986 $kgCO_{2-eq}$/t CO_2, depending on the mix of electricity configurations used. These studies collectively suggest that solid sorbent technologies can be a valuable component of carbon capture plant strategies.

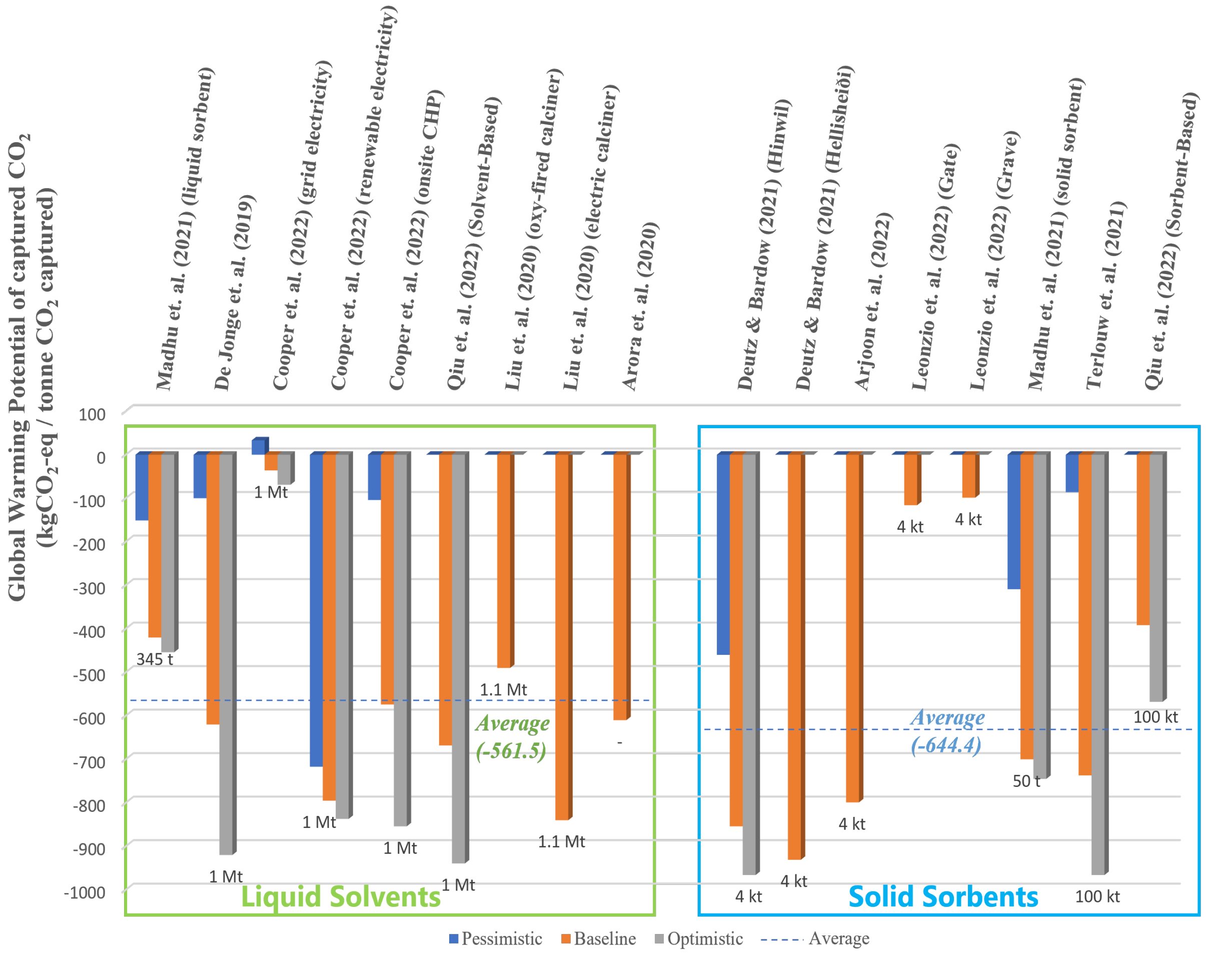


Figure 2.3: Net greenhouse gas emissions (CO2-eq) of DAC systems under pessimistic, baseline, and optimistic scenarios from various LCA studies. The horizontal dashed line and associated average value represent the average emissions from baseline scenarios.

Some papers, such as Madhu et al. (2021) and Qiu et al. (2022), provide insights into comparative analyses of sorbent technologies. Madhu et al. (2021), compares both solid and liquid sorbent technologies, highlighting emissions reduction potential with a baseline of -700 $kgCO_{2\text{-eq}}$/t CO_2 for solid sorbents and -151 $kgCO_{2\text{-eq}}$/t CO_2 for liquid sorbents in pessimistic scenarios. Qiu et al. (2022) offers a broader comparison across technologies, revealing a range of potential emissions reductions, from a baseline of -392 $kgCO_{2\text{-eq}}$/t CO_2 for solid sorbent to an optimistic scenario of -940 $kgCO_{2\text{-eq}}$/t CO_2 using liquid sorbent. These studies emphasize the importance of technology selection in emissions reduction strategies and suggest that different contexts may require different solutions.

The analysis showed that the average net GHG emissions of the baseline scenario for liquid sorbent DAC systems were -561.5 kg $CO_{2\text{-eq}}$ per ton of CO_2 captured. Solid sorbent technologies demonstrated a slightly lower average net GHG emission of -644.4 kg $CO_{2\text{-eq}}$ per ton of CO_2 captured, indicating a marginally better environmental performance.

These average values indicate that both liquid and solid sorbent DACCS holds promise as negative emission technologies. However, considering the baseline scenarios presented in Figure 2.3, the difference in the average emissions between the two remains quite small. This suggests that their environmental performance is similar. However, given the varying operating conditions and assumptions, choosing the right technology is crucial. It is important to consider the specific goals and limitations in which each technology will be used to fully assess its effectiveness.

2.3.1.2 Life Cycle Carbon Efficiency

Life cycle efficiency, defined as the ratio of net CO_2 removal to gross CO_2 captured, has emerged as a key metric for evaluating DAC performance. This efficiency accounts for all emissions associated with construction, operation, and decommissioning of the DAC system.

The bar chart in Figure 2.4 presents life cycle efficiencies as a percentage to gain insights into the DAC systems reported in the literature. Similarly, the studies presented in Figure 2.4 were chosen based on methodological transparency and data completeness. Inclusion required clearly defined system boundaries, ISO-compliant LCA frameworks, and consistent reporting of assumptions and functional units. The life cycle carbon efficiency of the liquid and solid DAC technologies was similarly differentiated on the same chart. Liquid sorbent systems, placed on the left, had an average efficiency of 56.2%, with a standard deviation of 23.7%. Solid sorbent systems, shown on the right, achieved an average efficiency of 64.4%, with a standard deviation of 29.5%.

Studies such as those reported by Arora et al. (2020), Cooper et al. (2022), De Jonge et al. (2019), and Liu et al. (2020), investigate the use of liquid sorbents for carbon capture. The data reveals significant variation in life cycle efficiency among these studies. For instance, De Jonge et al. (2019) present a pessimistic efficiency of 10%, while Liu et al. (2020) had ranging efficiency of 45% and 84% varying two different calciners and Cooper et al. (2022) achieve an optimistic efficiency of 85.4%. Arora et al. (2020) had a resulting efficiency of 61% for a CO_2 capture process (without considering storage). These variations highlight the sensitivity of liquid sorbent technologies to process design and operational parameters.

In contrast, studies carried out by Ali et al. (2024), Deutz & Bardow (2021), Gutsch & Leker (2024), Ottenbros et al. (2024) and Terlouw et al. (2021b) focus on solid sorbent technologies. These studies consistently demonstrate higher life cycle efficiencies across different scenarios, with baseline efficiencies ranging from 73.7% to 98.6%.

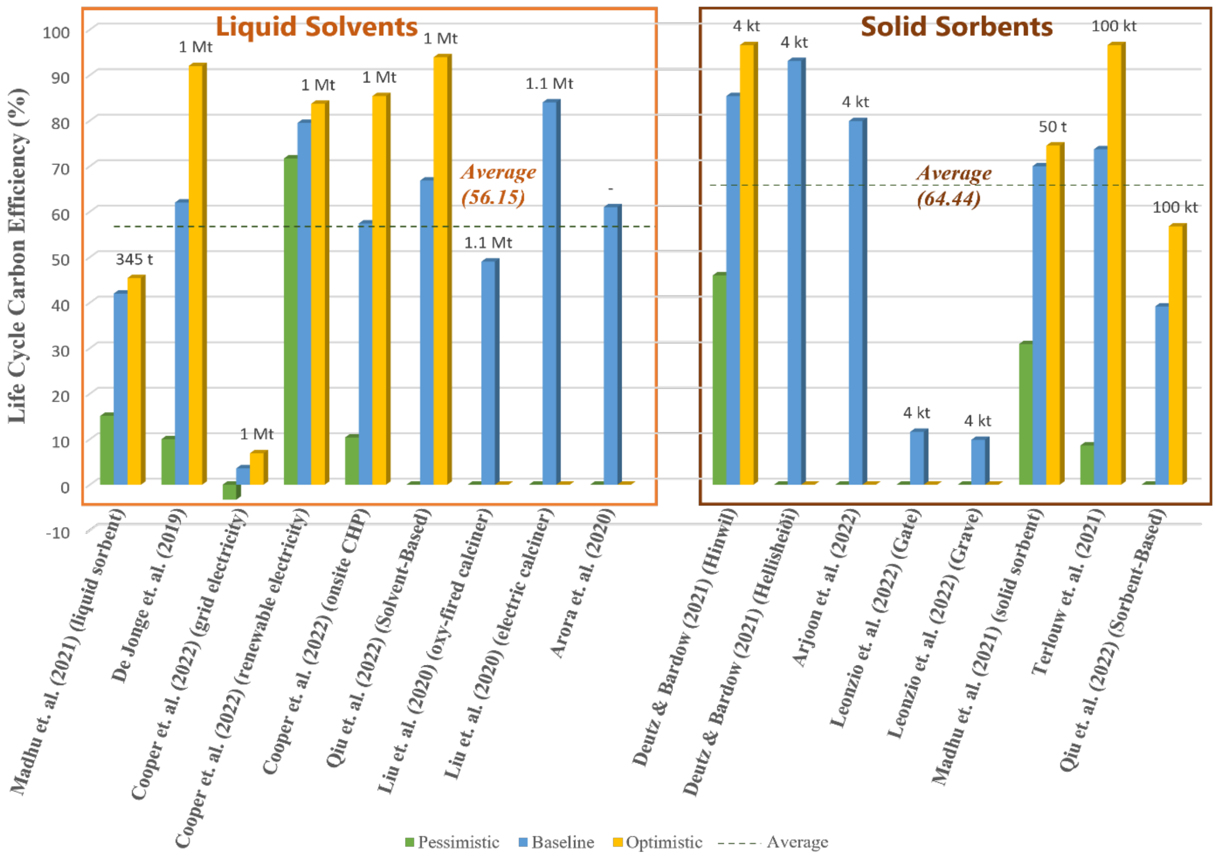


Figure 2.4: Life cycle carbon efficiency of DAC systems under pessimistic, baseline, and optimistic scenarios from various LCA studies. The horizontal dashed line and associated average value represent the average efficiency from baseline scenarios.

Some studies compare both solid and liquid sorbent technologies, such as Madhu et al. (2021), Qiu et al. (2022), and Simon (2023). These studies provide valuable insights into the trade-offs between the different technologies. For example, Qiu et al. (2022) reports a higher efficiency for solvent-based (66.81%) compared to sorbent-based (39.17%) technologies, while Madhu et al. (2021)

reported higher efficiency for solid sorbent-based (70%) compared to liquid solvent-based (42%) technologies, revealing the trade-offs between both technologies, with the choice of technology depending on project-specific goals and constraints.

The variations in life cycle efficiency among these studies underscore the importance of selecting the most suitable technology for DAC projects. While liquid sorbent technologies offer a wide range of efficiency potential, their effectiveness is highly contingent on precise process optimization. Additionally, the scale at which these technologies are assessed (kiloton-scale vs. megaton-scale per year) significantly impacts their overall practicality and environmental benefit. Ideally, fewer DAC plants operating at larger scales (megaton-scale capacities) would be preferred over numerous smaller kiloton-scale facilities, emphasizing the importance of both achieving high efficiencies and substantial deployment scale to rapidly approach net-zero targets.

In contrast, solid sorbent technologies consistently provide higher efficiencies, making them a preferable choice for projects prioritizing stable and efficient carbon capture. Notably, the efficiencies for the liquid and solid sorbent systems are comparable considering the large variations in study design parameters and assumptions made for different LCAs reported in the literature. Furthermore, the comparative studies reveal that mixed or hybrid approaches can also be viable, leveraging the strengths of both liquid and solid sorbent technologies. The bar chart and associated academic papers provide an overview of the different DAC plant technologies and their impact on reducing emissions.

2.3.2 Energy Consumption in Direct Air Capture Processes

Energy requirements represent one of the most significant determinants of DAC system performance, both economically and environmentally. Different DAC technologies exhibit distinct energy demand profiles regarding quantity, quality (temperature), and form (electricity versus heat), with important implications for system design and integration.

The energy sources used for DAC can be divided into two categories: electricity and thermal energy. Electricity is largely required to operate the fan to provide air to the CO_2 collector, which is usually an air contactor through which the air flows. In contrast, thermal energy is required for CO_2 desorption and regeneration of sorbent material. Since energy consumption has a huge impact on the environment, studies show that the use of low-carbon energy sources, namely renewable energies, can significantly reduce these negative impacts. Figure 2.5 presents the specific energy consumption provided in the literature, with Figure 2.5a showing electricity demand and Figure 2.5b showing heat demand per tonne of CO_2 captured and/or stored.

A comparative analysis of different DAC technologies reveals distinct energy consumption patterns across different DAC configurations. Electrical energy consumption in DAC systems varies widely across different technologies and configurations. Liquid solvent-based systems typically require 345–700 kWh/tCO_2, while solid sorbent-based systems tend to demonstrate lower electrical demands between 130–700 kWh/tCO_2, some systems report requirements up to 1,388 kWh/tCO_2 under specific operating conditions (Leonzio et al., 2022; Terlouw et al., 2021b). This variation primarily stems from differences in air handling requirements, process configurations, and system scale. Figure 2.5a illustrates the specific electricity consumption reported in LCA studies, highlighting the wide range of values for DAC systems across different configurations.

Thermal energy represents another critical component in the overall energy profile of DAC systems. HT-DAC systems utilizing liquid solvents typically demand 4–11.9 GJ/tCO_2. This higher thermal demand is primarily attributed to the calcination process required for carbonate decomposition, which operates at temperatures exceeding 800°C. In contrast, LT-DAC systems that use solid sorbents generally consume between 2.3 and 7 GJ/tCO_2, owing to their lower regeneration temperatures (60–120°C). Figure 2.5b presents the heat demand per tonne of CO_2 captured for various DAC configurations, where these variations arise primarily from differences in regeneration processes and types of sorbents or solvents used.

2.3.2.1 Energy demand profiles of different direct air capture technologies

The source and quality of energy significantly influence overall system efficiency and environmental impact. Recent studies have demonstrated substantial variations in performance based on energy source selection. Qiu et al. (2022) used electricity from the United States electricity system. The researchers explored the use of natural gas and renewable gas (biomethane) as heat sources for solvent-based DACCS and heat pumps, as well as biomethane for sorbent-based DACCS. Their facilities exhibit variations in electricity requirements ranging from 470–700 kWh/tCO_2 and thermal demands between 5.4–5.8 GJ/tCO_2. The findings revealed that for solvent-based DACCS, the use of biomethane as a heat source resulted in a lower global warming potential GWP of -668.1 kgCO_2-eq/t CO_2 and a high life cycle efficiency of 66.8%. Sorbent-based DACCS had a greater environmental impact, with a GWP of -391.7 kgCO_2-eq/t CO_2 and a life cycle efficiency of 39.2%. The study indicated that decarbonizing the power sector improves sequestration efficiency, implying that DACCS should be sited in areas with significant renewable energy potential.

De Jonge et al. (2019) described three scenarios in their investigation. In the baseline scenario, electricity accounted for 65% of energy consumption with demands ranging from 448–554 kWh/tCO_2, while natural gas combustion provided 32% of heat, with requirements of 5.1–8.4 GJ/tCO_2. In the optimistic scenario, solar power was used to generate electricity, and heat released during the condensation stage was recovered and reused, resulting in a 92% life cycle efficiency and an extremely low GWP of -920 kgCO_2-eq/t CO_2. The pessimistic scenario, which relied on an average US coal plant for power and did not include heat recovery, resulted in a 10% life cycle efficiency and a GWP of -100 kgCO_2-eq/t CO_2. Despite the lower efficiency, the DAC system still effectively captured CO_2, highlighting its potential as a mitigation option for climate change, especially when renewable energy is used.

Studies at commercial scale facilities provide valuable insights into real-world energy consumption patterns. The LCA study by Deutz & Bardow (2021) on Climeworks' two commercial LT-DACCS facilities in Hinwil (Switzerland) and Hellisheiði (Iceland) reported high GWP values of -854 and -931 kgCO_2-eq/t CO_2 and life cycle efficiencies of 85% and 93%, respectively. The Hellisheiði plant is powered by geothermal energy, while the Hinwil plant uses electricity and waste heat from an incineration plant. The study, while maintaining similar energy consumption profiles (approximately 700 kWh/tCO_2 electricity and 4.7–11.9 GJ/tCO_2 heat), confirmed that the energy sources used for the capture of CO_2 have a large effect on the GWP of the DAC processes.

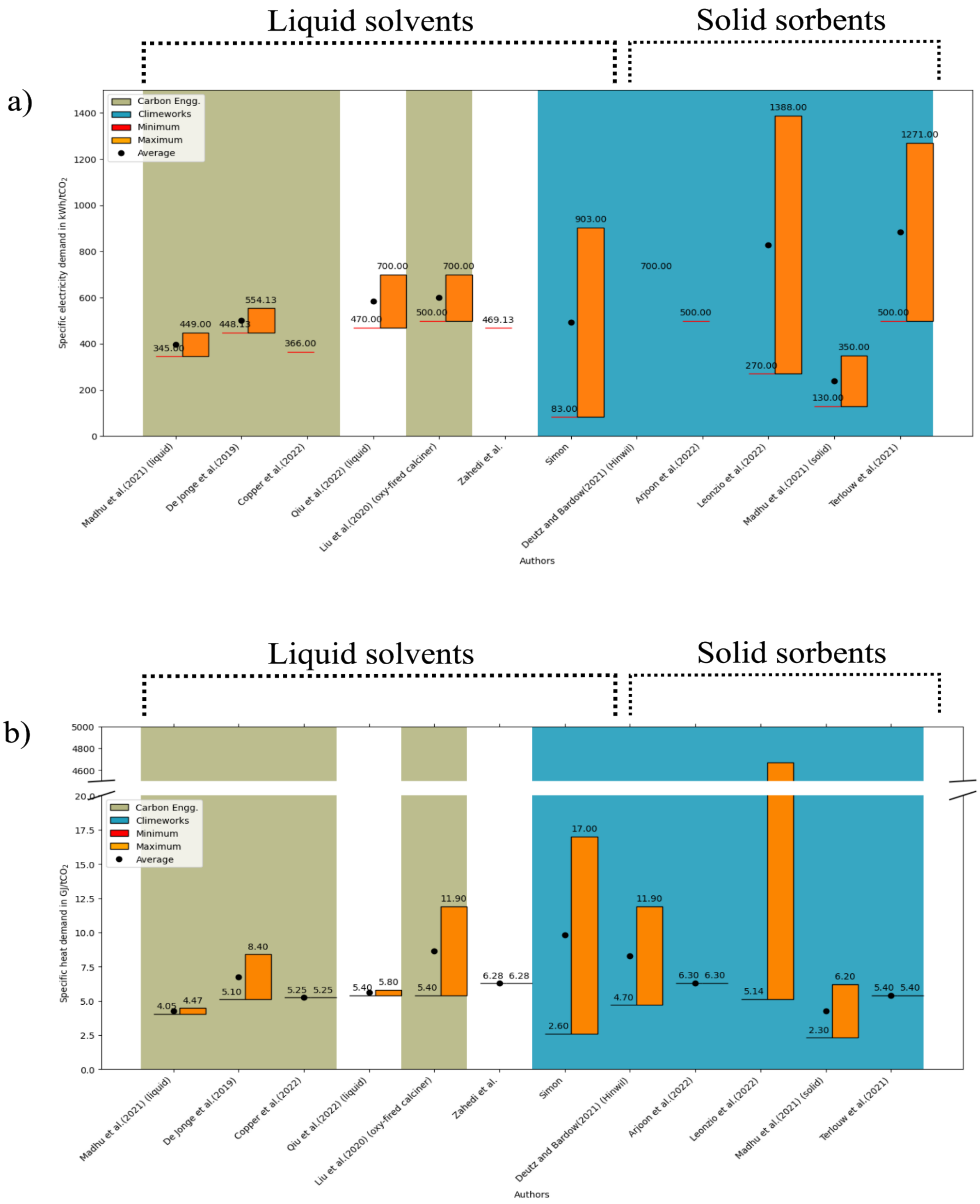


Figure 2.5: DACCS energy consumption from LCA studies: (a) electricity demand and (b) heat demand per tonne of CO_2 captured and/or stored

Terlouw et al. (2021b) recommended using low-carbon energy sources and avoiding fossil-fuel-based grid mixes. Their research stressed the importance of adequate energy storage units when utilizing renewable energy sources, as this leads to additional environmental impacts. Their facilities exhibit wider variations in electricity consumption (500–1,271 kWh/tCO_2) with thermal demands around 5.4 GJ/tCO_2. They explored different mixes of electricity (grid and photovoltaics) and heat sources (electricity, waste, and solar heat) for the capture of CO_2, finding that countries with low-carbon electricity mixes combined with waste heat exhibit the lowest GWPs, with Norway and Iceland achieving -966 and -940 kgCO_2-eq/t CO_2 and life cycle efficiencies of 97% and 94%, respectively. On the contrary, Greece, which relies heavily on fossil fuels, had a high GWP value that was close to net positive GHG emissions.

Cooper et al. (2022) compared grid electricity to renewable energy (wind) for DACCS consuming 366 kWh/tCO_2 electricity and 5.25 GJ/tCO_2 heat. The grid electricity's DACCS system had a high GWP of -36 kgCO_2-eq/t CO_2 and a low efficiency of 3.6%, whereas the wind-powered system had a GWP of -795 kgCO_2-eq/t CO_2 and an efficiency of 79.5%. Importantly, the study revealed that DACCS systems utilizing waste heat and renewable energy have the lowest environmental burdens, while grid-coupled designs based on fossil fuels exhibit significantly higher impacts. Further efficiency improvements could be achieved by leveraging surplus renewable energy, which would otherwise require storage in batteries or other storage systems, thus reducing storage-related environmental impacts. Additionally, integrating DACCS operations with smart-grid technologies, such as time-of-day demand management and adaptive energy usage strategies, could enhance overall system efficiency and reduce environmental burdens even further.

Ottenbros et al. (2024) conducted a similar study, comparing three different electricity configurations: grid, hybrid, and wind electricity, with heat provided by an integrated electric heat pump. The baseline GWP for wind-connected configurations with battery storage was -823 kgCO_2-eq/t CO_2, with a life cycle efficiency of 82%. The hybrid configuration outperformed the grid-connected system, with a GWP of -874 kgCO_2-eq/t CO_2 compared to -847 kgCO_2-eq/t CO_2. Additionally, the LCA study presented by Gutsch & Leker (2024) featured a refined model of an autonomous LT-DACCS system, where electricity is generated by a photovoltaic (PV) system, thermal energy is provided by heat pumps, and a battery storage system is incorporated to mitigate the intermittency issues of the PV system. This autonomous DAC system achieved a GWP of -884 kgCO_2-eq/t CO_2 and an efficiency of 88.4%, underscoring the importance of energy system selection from an LCA perspective.

Overall, the variation in energy consumption, GWP and life cycle efficiency values across DAC systems is influenced by several factors, including system configuration, sorbent or solvent selection, geographical location, and energy source. While both HT- and LT-DAC systems can achieve viable energy consumption profiles, the choice of energy source has a profound effect on the carbon footprint and operational efficiency of these technologies. As DAC systems continue to evolve, optimizing energy consumption, reducing reliance on fossil fuels, and integrating renewable energy sources will be key to improving the economic and environmental sustainability of these systems.

2.3.2.2 Heat sources and adsorbent regeneration

Given the substantial thermal energy demands of DAC systems, which range between 2.3 and 7 GJ/t CO_2 and account for approximately 60% to 80% of the total energy consumption (with the remaining 20% to 40% attributed to electricity consumption for fans, compressors, pumps and control systems, requiring 300 to 900 kWh/t CO_2), the selection of heat sources and thermal integration strategies plays a critical role in overall system performance. Thoughtful integration of heat sources can help reduce lifecycle environmental impacts while enhancing economic viability through synergistic operation. Various heat sources and integration approaches have been explored in the literature, each offering distinct advantages and limitations.

For HT-DAC systems, the elevated temperatures required for sorbent regeneration necessitate high-grade heat sources such as natural gas combustion, hydrogen, and biomass. Among these, natural gas combustion with carbon capture remains the most developed approach, as demonstrated in Carbon Engineering's design (Carbon Engineering, 2016; Keith et al., 2018). This configuration ensures that the required regeneration temperatures are met while maintaining reasonable carbon removal efficiency through the capture of combustion emissions.

In contrast, LT-DAC systems, which require lower regeneration temperatures, have been explored with several heat integration approaches. Biogas utilization, particularly biomethane, has been examined as a heat source (Qiu et al., 2022). Waste heat from industrial processes and power generation has also been leveraged, as demonstrated in Climeworks' Hinwil plant, which sources waste heat from an incineration facility. Similarly, Global Thermostat employs steam-based process heat (Deutz & Bardow, 2021; McQueen et al., 2021). Geothermal energy provides another renewable heat source, as seen in Climeworks' Icelandic facility, where it supplies continuous heat in geologically suitable locations (Deutz & Bardow, 2021). Additionally, heat pumps powered by renewable electricity, with coefficients of performance (COP) of 2.5 or higher, have been explored for generating the required temperatures efficiently (Deutz & Bardow, 2021; Madhu et al., 2021; McQueen et al., 2021).

An emerging research area involves the direct integration of solar energy into the regeneration process. Utilizing solar thermal energy presents a promising pathway to reducing conventional energy demands in DAC systems. This approach employs solar collectors that convert solar radiation into heat, which is then transferred to the sorbent to drive CO_2 desorption and sorbent regeneration. However, research on solar-driven DAC is still in its early stages, with most studies focusing on laboratory-scale photothermal methods rather than complete system designs that incorporate solar collectors and LCAs.

Several promising concepts have emerged, particularly in photo-thermal systems using aqueous amine solvents. Nguyen et al. (2014) demonstrated a suspension of carbon black nanoparticles dispersed in amine solvents that absorbed CO_2 and underwent regeneration via light irradiation. Similarly, Campbell et al. (2021) developed a transparent flow reactor containing aqueous amine solvents, where CO_2 desorption occurred using light-absorbing microparticles. Gao et al. (2021) introduced photo-triggered, mussel-inspired polymer solvents and nanogel particle adsorbents that released CO_2 when heated above 85°C through photoirradiation. While these systems have been tested for capturing CO_2 at higher concentrations than typically found in DAC applications, their direct application to DAC presents challenges, including amine volatility, corrosive properties, and the energy-intensive regeneration process (Sanz-Pérez et al., 2016).

Unlike liquid amine solvents, amine-based solid adsorbents offer several advantages for DAC, including low heat capacity, lower temperature requirements, compact unit size, scalability, and high CO_2 capacity (Elfving et al., 2021). As a result, these materials are being actively investigated for their compatibility with photo-thermal desorption methods. Kataoka et al. (2023) developed a novel adsorbent containing light-absorbing materials to enable energy-efficient DAC using light irradiation. This adsorbent was composed of polyethyleneimine (PEI) as the CO_2-absorbing component, fumed silica as a high-surface-area support, and carbon materials for photothermal heating. Similarly, Kar et al. (2025) reported an integrated gas-phase DAC and utilization (DACCU) reactor that captures CO_2 from ambient air, concentrates it, and converts it into renewable fuel using simulated sunlight. Their system employs a silica-polyamine CO_2 adsorber, desorbing CO_2 at 80–100°C via photothermal solar heating. A parabolic trough solar collector concentrates sunlight to heat the adsorbent, simultaneously enhancing the photoconversion efficiency of released CO_2. Amine-based adsorbents are sensitive to moisture, which can influence CO_2 capture capacity and long-term stability. These adsorbents can operate under humid gas streams, but their performance and degradation mechanisms are closely tied to water content. As such, many studies, including those on photothermal applications, focus on dry or controlled-humidity conditions to ensure material longevity and efficiency.

From an LCA perspective, Terlouw et al. (2021b) proposed a DAC system layout utilizing solar heat generated by Fresnel solar collectors, which can achieve high temperatures up to 400°C. For LT-DAC applications, where desorption occurs at 80–100°C, flat-plate solar collectors have been identified as an appropriate and efficient heat source. The integration of solar-driven regeneration into DAC systems presents several key advantages over conventional thermal regeneration methods. Notably, it can significantly reduce the operational energy costs associated with traditional DAC systems, which often rely on high-temperature (>100°C) thermal desorption. By utilizing renewable energy sources, such as solar radiation, solar-driven DAC also presents an opportunity for more environmentally sustainable carbon capture with a lower overall carbon footprint.

Despite promising developments, gaps remain in fully integrating solar-driven regeneration into operational DAC systems. This thesis aims to explore the feasibility and benefits of using solar energy for adsorbent regeneration in DAC processes. The research encompasses a comprehensive analysis of different system configurations through LCA methodology. Three distinct scenarios are evaluated: (1) a conventional system using grid electricity and natural gas from industrial furnaces for heating, (2) a system utilizing grid electricity with heat supplied by copper flat-plate solar-thermal collectors, and (3) a hybrid system employing grid electricity with solar-thermal collectors supplemented by auxiliary electrical heating. Additionally, experimental work investigates the performance of adsorbents under solar-simulated radiation, with LED light providing intensities of 2.3 kW/m^2 (twice the standard AM1.5 solar spectrum) to achieve regeneration temperature of 110°C for effective CO_2 desorption. While this method is not intended as a practical approach for real-world energy input because using electricity to generate light and then heat is inefficient, it serves as a controlled laboratory proxy for concentrated solar exposure. By integrating these experimental findings with data from literature into a modified process model, this research provides a comprehensive evaluation of solar-integrated DAC systems' technical viability, energy efficiency, and environmental impact—ultimately addressing a critical knowledge gap in sustainable carbon removal technologies.

Chapter 3

Methods

The LCA in this research follows the LCA methodology reported by Madhu et al. (2021) with modifications made to analyze the effects of using solar energy to regenerate the CO_2 adsorbents on the environmental impacts of the carbon capture process. This Methods section is divided into three sub-sections: (1) LCA methods, (2) experimental methods, and (3) the integration of solar energy in both calculations and experimental validation. Below, each section is outlined in detail.

3.1 Life Cycle Assessment Methods

3.1.1 Technical details for Life Cycle Assessment

The LCA framework used in this study is based on the approach developed by Madhu et al. (2021), which assesses the environmental impacts of TSA DACCS using a cradle-to-cradle perspective. The life cycle contains all activities from cradle-to-cradle: from the extraction of raw materials, transportation, production and product use to recycling and final disposal of waste. It also includes the construction phase, operational energy use (heat and electricity), operational material (sorbent) production and assumption of geological storage. All flows of energy and materials exchanged with the environment throughout the life cycle are collected and interpreted regarding their environmental impacts.

To conduct the LCA of my study in accordance with both the scientific and International Organization for Standardization (ISO) standards 14040 and 14044, OpenLCA software version 2.3 was used together with the Ecoinvent database version 3.10 (GreenDelta, 2025). The life-cycle impact assessment method used for this research study was ReCiPe Midpoint (H) without long term impacts, which is a standard in scientific LCA research that indicates how different environmental impact categories are affected by various anthropogenic emissions over a 100-year time horizon. The exclusion of long-term impacts means that the method does not account for highly uncertain or speculative long-term environmental effects, such as those arising from persistent pollutants (e.g., fluorinated compounds) whose full environmental or health consequences may extend beyond the modeled timeframe or are not yet fully understood.

3.1.2 Goal and Scope of the Life Cycle Assessment Study

This LCA study focuses on a TSA DAC pilot plant, with a CO_2 sequestration capacity of 50 t yr^{-1}. This study aims at quantifying the life cycle GHG emissions and life cycle efficiencies of DACCS under different boundary conditions and in various system layouts, as well as at identifying potential environmental trade-offs involved with CO_2 removal. In the analysis, the following countries and regions, which exhibit drastically different climates, are included: in Canada the provinces Alberta, New Brunswick, Ontario, and Quebec were investigated, as well as China and Greece, with Switzerland covering cooler and temperate climate regions. These regions were selected based on their geological storage potentials, difference in the grid electricity mix, climate variations and direct normal irradiance of greater than or equal to 2000 kWh m^{-2} $year^{-1}$.

3.1.3 Functional Unit

In alignment with Madhu et al. (2021), the functional unit is defined to be '1 ton CO_2 captured and stored.' To compare different scenarios, the environmental impacts of the LT-DAC plant are assessed based on the functional unit of 1 ton of CO_2 captured and stored. This evaluation considers plant lifetime, adsorbent consumption, and the total amount of CO_2 processed, ensuring consistency across scenarios.

3.1.4 System Boundaries and Technical Description

In line with the goal and scope of the LCA study, the system boundary includes all processes involved in capturing CO_2 from the atmosphere, from the construction and operation of the DAC plant to the end-of-life recycling and disposal of materials. For the purposes of this study, end-of-life recycling is modeled based on currently available technologies and typical industrial practices, constrained by economic feasibility and regional infrastructure. While the maximum theoretical recycling potential may be higher, only realistic, documented recycling pathways are considered.

Furthermore, the system boundaries are extended by incorporating additional considerations, such as the integration of solar energy as a heat source and expanded geographical analysis. The system boundary includes key components such as the DAC air contactor, sorbent materials, auxiliary infrastructure, and energy supply required to capture and permanently store CO_2 in geological formations, achieving CDR. Additionally, the system boundary considers the transportation of materials, components, and waste, as well as interactions with the natural environment, including resource consumption and emissions relevant to global warming potential impact categories.

This work evaluates three distinct DACCS system configurations, specifically focusing on modifications to the thermal energy supply for adsorbent regeneration while maintaining country specific grid electricity supply for CO_2 capture process and auxiliary operations. The key components of the system that differ from the work by Madhu et al. (2021) are indicated with colored borders in the schematic of the system boundaries shown in Figure 1. The grid electricity supply box is outlined in green, representing the consistent power source across all configurations. The heat source boxes are outlined in red, indicating the components that were modified in this study. The analyzed configurations are modeled using process data from the Ecoinvent database and include the following arrangements:

1. Electricity from the grid, heat from natural gas, at industrial furnace low-NOx >100kW
2. Electricity from the grid, heat from a Cu flat plate solar-thermal heat collector
3. Electricity from the grid, heat from a Cu flat plate solar-thermal heat collector with auxiliary heating operated with electricity from grid

These configurations are detailed in subsequent sections.

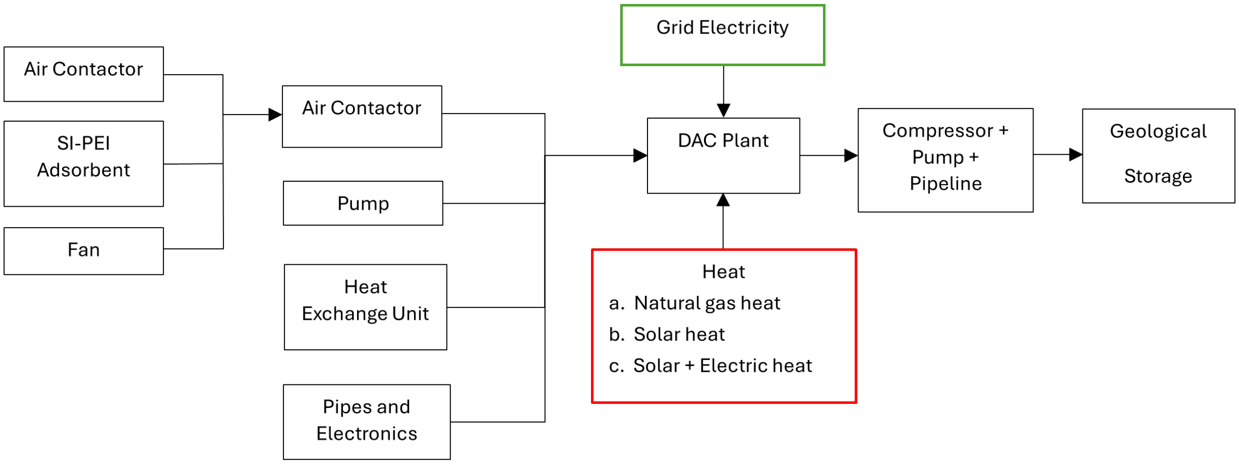


Figure 3.1: Product system flow diagram of a LT-DACCS LCA

3.1.5 Life-Cycle Inventory of Low Temperature Direct Air Capture System

In the life cycle inventory (LCI) phase, all mass and energy flows are collected and analysed for flows entering and leaving the system boundaries. The product system for LT-DAC is based on the commercialized DAC units manufactured by the Swiss company Climeworks. The analysed technology has an annual gross carbon capture capacity of 50 tCO_2, a carbon capture rate of 90% and an asset lifetime of 20 years. The adsorption and desorption of CO_2 take place at 25 °C and 100 °C, respectively. The main components modelled for the capture of CO_2 and regeneration of the sorbent material are the air contactor, the sorbent material and the heat exchange unit. The sorbent suggested by the Climeworks company for DAC systems is SI-AEATPMS (SI=silica gel, AEATPMS = [N-(2-aminoethyl)-3-aminopropyl]trimethoxysilane) (Leonzio et al., 2022), but no commercial production data was accessible. In the study by Madhu et al. (2021), SI-PEI (SI=silica gel, PEI= polyethyleneimine) is considered as amine proxy for the sorbent. This proxy was selected due to the availability of commercial-scale LCI data for PEI, including energy consumption, precursor chemicals, synthesis emissions, and transportation data. SI-PEI also exhibits comparable CO_2 adsorption capacity, regeneration temperatures, and stability under DAC conditions, making it a suitable alternative for modeling purposes. Approximately 340 kg of sorbent are needed every year, corresponding to an average sorbent loss of 7 kg per ton of CO_2 captured. Due to the high energy demand of sorbent regeneration, the reference case assumes that the plant incorporates a heat recovery system with 75% efficiency (Madhu et al., 2021). An overview of the main technical parameters used in the LT-DACCS system configurations is provided in Table 3.1.

Table 3.1: Technical and design parameters used in the LT-DACCS system configurations (Madhu et al., 2021).

Technical and design parameter for LT-DAC	Reference Case
CO_2 capture rate (%)	90
Single unit CO_2 capture capacity per year (t)	50
Number of air contactors	1
Lifetime of the air contactor (yr)	20
Carbon capture capacity of the sorbent (%)	10
Lifetime of sorbent (yr)	1
Sorbent required per t CO_2 captured (kg)	6.9
Air velocity (m s^{-1})	4
Fan efficiency (%)	68
Total electrical energy required (kWh)	180
Adsorption temperature (°C)	25
Desorption temperature (°C)	100
Heat recovery rate (%)	75
Heat required (GJ/ tCO_2 captured)	2.7
Transportation from site of production to site of installation to end-of-life treatment facility (km)	50
Energy required to compress 1 tCO_2 (kWh/ tCO_2)	111
Pipeline required to transport CO_2 (km)	300
Energy required to inject 1 tCO_2 in geological storage (kWh/ tCO_2 captured)	7

3.1.5.1. Compression, transfer and storage of captured CO_2

In the LT-DAC process, captured CO_2 is compressed on-site by a system requiring 111 kWh of electricity per ton. The CO_2 is then assumed to be transported 300 km through a pipeline before being injected into geological reservoirs, a step that demands 7 kWh per ton of CO_2 (Koornneef et al., 2008; Madhu et al., 2021).

Since energy consumption plays a crucial role in environmental impact, this work evaluates system layouts with different energy sources for the adsorbent regeneration process. Three layouts are evaluated, wherein the adsorbent is regenerated using either (1) solar energy, (2) solar energy and electric heating, and (3) heat from natural gas system layouts. This work investigates the benefits of using solar energy to regenerate the adsorbents because it is one of the fastest-growing renewable energy sources and has large potential for further expansion at comparatively low costs. The solar plus electric heat configuration addresses the intermittency of solar energy, with auxiliary heating helping the plant operate during periods without sunlight. Natural gas is used as a baseline for comparison based on traditional fossil fuel heat usage for the regeneration process.

3.1.5.2 Electricity consumption

The life cycle environmental impacts of DACCS were calculated using regionally explicit LCI data for electricity generation in Canada, China, Greece, and Switzerland. This approach aims to evaluate how country-specific electricity grid mixes influence the environmental profiles of DACCS. The electricity generation mix for each region highlights the share of different energy sources such as hydro, wind, solar, nuclear, fossil fuels, and other renewables. Notable differences include Quebec's predominantly hydro-based electricity (94%), Alberta's heavy reliance on fossil fuels (85%), Ontario’s predominant reliance on nuclear (55%), Switzerland's focus on hydro and nuclear power, and China's electricity generation dominated by coal (62%), with limited contributions from renewables. The diversity in grid mixes provides a nuanced understanding of how electricity sources can affect DACCS life cycle outcomes as shown in Table 3.2.

Table 3.2: Regional Electricity Generation Mix by Source (%) adapted from (Government of Canada, 2024; IEA, 2024).

Region	Hydro	Wind	Solar	Nuclear	Fossil Fuels	Other Renewables & Bioenergy
CA-QC	94	5	-	-	<1	<1
CA-NB	23	<10	-	40	27	<10
CA-ON	24	8	4	55	<10	<10
CA-AB	3	9	-	-	85	2
Greece	<10	23	17	-	49	<10
Switzerland	56	-	6	33	<5	<5
China	15	9	-	-	64	12

Note: Fossil fuel breakdowns are as follows—CA-AB: 22% coal, 63% natural gas; Greece: 15% coal & petroleum, 34% natural gas; China: 62% coal, 2% natural gas and petroleum.

3.1.5.3 Heat consumption

The LCA reported in this thesis extends Madhu et al.'s (2021) work by evaluating three distinct heat supply options for regenerating the adsorbent in the LT-DACCS system, each presenting unique operational characteristics and environmental implications. These heating approaches were selected to understand how different heat sources affect the overall environmental profile of the carbon capture process.

3.1.5.3.1 Natural gas heat

Natural gas heat production occurs through burning natural gas in co-generation units, boilers and industrial furnaces. This specific dataset represents heat production from natural gas at an industrial furnace (low-NOx >100kW) in a specific region/country. The activity starts with fuel combustion and ends with heat production, including emissions due to combustion, inputs needed for combustion, combustion by-products, and infrastructure requirements. While low-NO_x systems emit reduced nitrogen oxides, residual NO_x emissions are not a primary focus of this study, which centers on GWP and carbon efficiency. However, their contribution to environmental issues such as acidification and photochemical smog warrants further investigation in future LCA studies.

3.1.5.3.2 Solar heat

In this system, the solar heat is harnessed using a non-concentrating flat copper plate solar collector, which does not rely on semiconductor materials like photovoltaic cells. The system is composed of a pump, water storage, fluid circulation tubes, and the collector itself. The system relies predominantly on solar energy with minimal electricity input of about 0.49%, suggesting an extremely low carbon footprint and excellent operational cost-effectiveness. The system operates at approximately 87.9% efficiency, converting around 88% of total input energy into usable heat, with the remaining 12.1% accounting for collector and system losses. However, as a standalone system, its operation is constrained by solar availability, meaning heat supply may be interrupted during periods of low sunlight or at night.

The complete solar heating system incorporates solar system construction, circulation pumps for heat transfer fluid movement, water storage tanks for thermal energy retention, and an extensive distribution network including heat exchangers and control systems. This integration ensures optimal heat collection and distribution while maintaining system stability and performance. The operation excludes additional heating requirements, focusing solely on maximizing solar thermal conversion efficiency through careful system design and operation.

3.1.5.3.3 Solar and electric heat

The solar plus electric heat configuration produces heat through a combination of a solar collector system and an auxiliary electric heating unit. Approximately 25% of the heat is delivered through the collector system. This approach accounts for maintenance, operational electricity use, and the necessary auxiliary heating. This arrangement ensures uninterrupted heat supply regardless of solar conditions while maximizing the utilization of available solar resources.

3.1.6 Life Cycle Carbon Efficiency

To evaluate the carbon efficiency of the DAC system throughout its life cycle, from inception to production or across its entire lifespan, the methodology proposed by De Jonge et al. (2019) is used. The life cycle efficiency (E_c) is calculated using Equation 3.1:

$$E_c = \frac{CO_{2,cap} - GHG_{lc}}{CO_{2,cap}} \text{ x } 100 \qquad (3.1)$$

where $CO_{2,cap}$ represents the total amount of CO_2 (kg) captured over the system's lifetime, and GHG_{lc} denotes the total greenhouse gas emissions (kg CO_2-equivalent) produced throughout the entire life cycle. CO_2-equivalents (CO_2-eq) provide a standardized metric for comparing emissions from various greenhouse gases based on their GWP over a 100-year time horizon, in line with the ReCiPe Midpoint (H) method.

The GHG_{lc} of the LT-DACCS system, assessed from a cradle-to-grave perspective, includes emissions from sorbent production, system construction, operation, end-of-life processes, and CO_2 storage in geological reservoirs, all expressed in kg CO_2-eq.

If E_c is less than zero, it indicates that the system emitted more CO_2-eq than it removed during the capture and storage process. Conversely, a positive E_c value signifies that the amount of CO_2 stored exceeded the total CO_2-eq emissions. While any positive E_c indicates net-negative performance, values approaching 100% are generally considered favorable for large-scale deployment, as they reflect high carbon efficiency and minimal environmental trade-offs relative to the amount of CO_2

captured. Lower positive values may still be viable in specific contexts but would require optimization to reduce life cycle emissions further.

3.2 Experimental Methods

To validate key assumptions in the LT-DACCS LCA, experimental studies were conducted focusing on material synthesis, characterization, and CO_2 capture performance. The experimental methods were designed to provide empirical data for the LCA model while evaluating the system's practical feasibility. In these experiments, silica-supported polyethyleneimine (SI-PEI) adsorbents were synthesized and tested for CO_2 capture. The experimental apparatus used for CO_2 adsorption and desorption is shown in Figure 3.2. A LED light was used to provide solar-simulated light to regenerate the adsorbents, mimicking real-world solar energy conditions. The findings from these experiments serve as direct input into the LCA study, allowing for a comprehensive evaluation of solar-assisted DACCS performance.

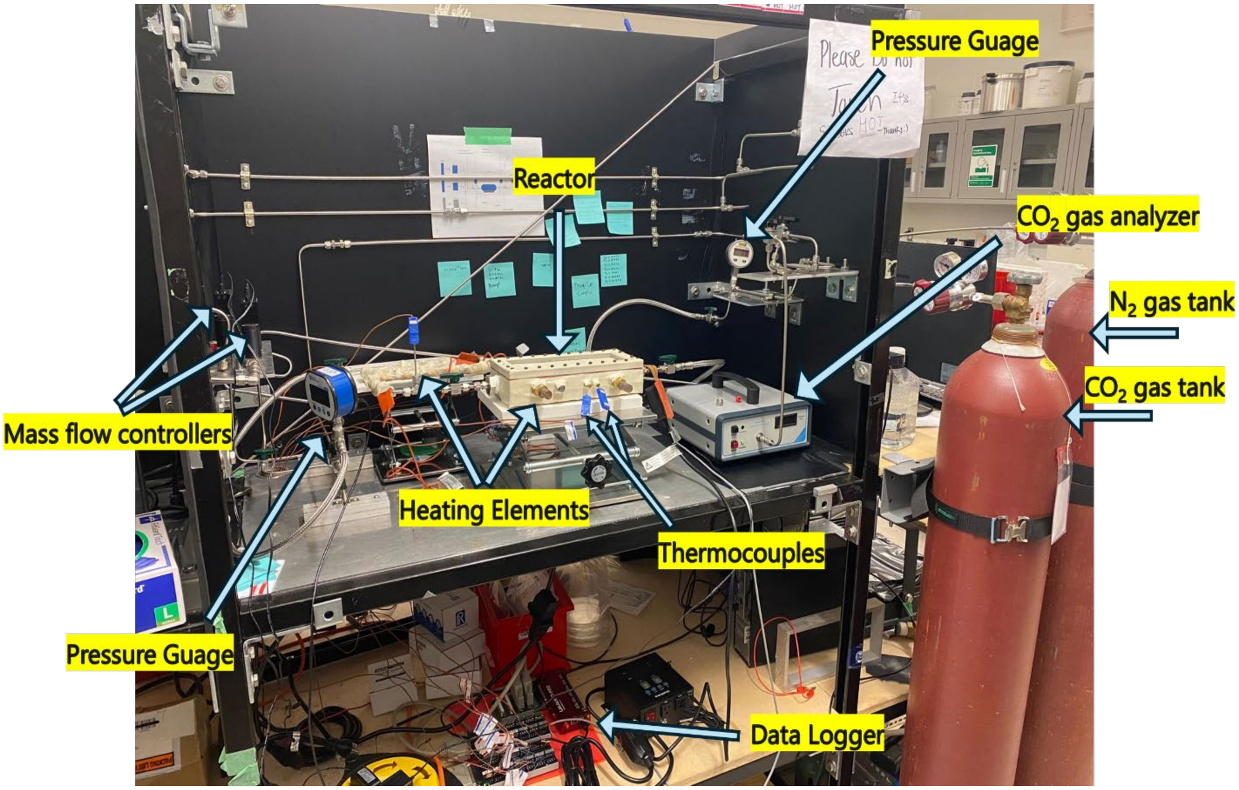


Figure 3.2: Experimental setup for CO_2 adsorption and desorption cycles. The system includes gas supply lines, mass flow controllers, fixed-bed reactor housing the adsorbent, thermocouples for temperature monitoring, and gas analyzers for real-time CO_2 concentration measurements.

3.2.1 Materials

All chemicals were of analytical grade and used without modification. High-purity CO_2 (>99.9%) and nitrogen (>99.95%) were supplied by Linde Canada Inc. Methanol (>99.5% purity), silica gel (40-200 mesh), and linear polyethyleneimine (PEI, average molecular weight (MW) 1200, boiling point (b.p.) 250ºC, density (ρ) = 1.05 g/mL) were obtained from MilliporeSigma Canada Ltd. (Oakville, Canada).

3.2.2 Sample Preparation

The SI-PEI adsorbents were synthesized via wet incorporation, following procedures reported by Sanz et al. (2010) and further refined based on the methodology described by Xu et al. (2019). The sample mass was optimized for a 250 cm^3 adsorbent-bed column to ensure appropriate gas–solid contact time. To prepare the adsorbent with 30 wt% PEI loading, 12 g of PEI was first dissolved in 224 g of methanol under continuous stirring at 400 revolutions per minute for 10 minutes at ambient temperature. This dissolution step was critical to achieving uniform polymer distribution within the porous support structure.

Subsequently, 28 g of silica gel was added to maintain a methanol-to-silica ratio of 8:1, providing sufficient solvent volume to facilitate complete pore filling without excessive residue. The resulting suspension was stirred continuously for 8 hours at room temperature, then dried at 70 °C for 12 hours to remove residual methanol. The process yielded 40 g of PEI-loaded silica adsorbent, consisting of silica gel embedded with amine-functional groups. The final material was stored in sealed glass containers to prevent moisture uptake and amine oxidation prior to characterization and CO_2 adsorption experiments.

3.2.3 Sample Characterization

Material characterization was performed using ultraviolet–visible (UV-Vis) diffuse reflectance spectroscopy to assess the optical properties of the synthesized adsorbents. The powdered samples were placed in a solid sample holder and analyzed using a Shimadzu UV-2600 spectrophotometer (Shimadzu Corporation, Japan) equipped with an integrating sphere attachment. Reflectance spectra were recorded over the wavelength range of 200 to 1400 nm with a spectral resolution of

1 nm. Barium sulfate ($BaSO_4$) was used as the reflectance reference standard for baseline correction.

3.2.4 CO_2 capture and Regeneration of Sorbents

The CO_2 capture experiments were conducted in a fixed-bed reactor under atmospheric pressure using simulated flue gas. In this work, 40 g of the dried adsorbent was packed into the bed in the middle of the reactor supported with quartz wool as shown in Figure 3.3.

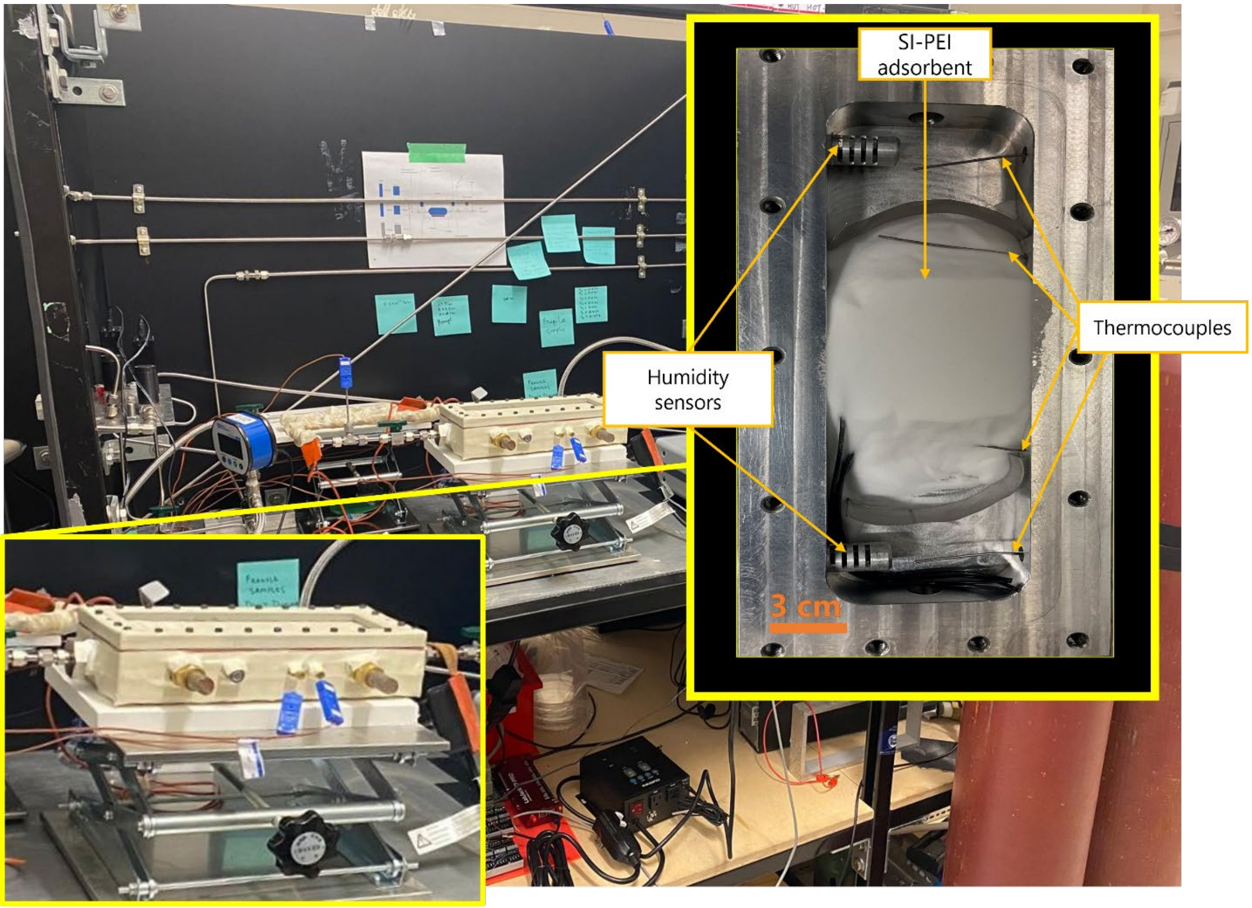


Figure 3.3: Exploded view of the adsorbent bed column with SI-PEI adsorbent. The humidity sensor for moisture monitoring.

Before adsorption, the adsorbent was treated with N_2 gas at 110ºC for 1 h, where the flow rate of N_2 gas was 100 SCCM. After cooling to the desired adsorption temperature, a CO_2/N_2 (10:90 v/v) gas mixture was introduced and passed through the adsorbent bed at a flow rate of 111 SCCM using a Masterflex mass flow controller (MFC) (Model 32907-59; Cole-Parmer, USA). The CO_2 concentrations in the effluent gas were monitored with an online CO_2 gas analyzer (Model 906;

Quantek Instruments Inc., MA, USA). The CO_2 adsorption capacity was calculated by integrating the area between the inert gas and CO_2 breakthrough curves. After the adsorption step, the adsorbents were regenerated by subjecting them to solar-simulated radiation from the LED light as shown in Figure 3.4.

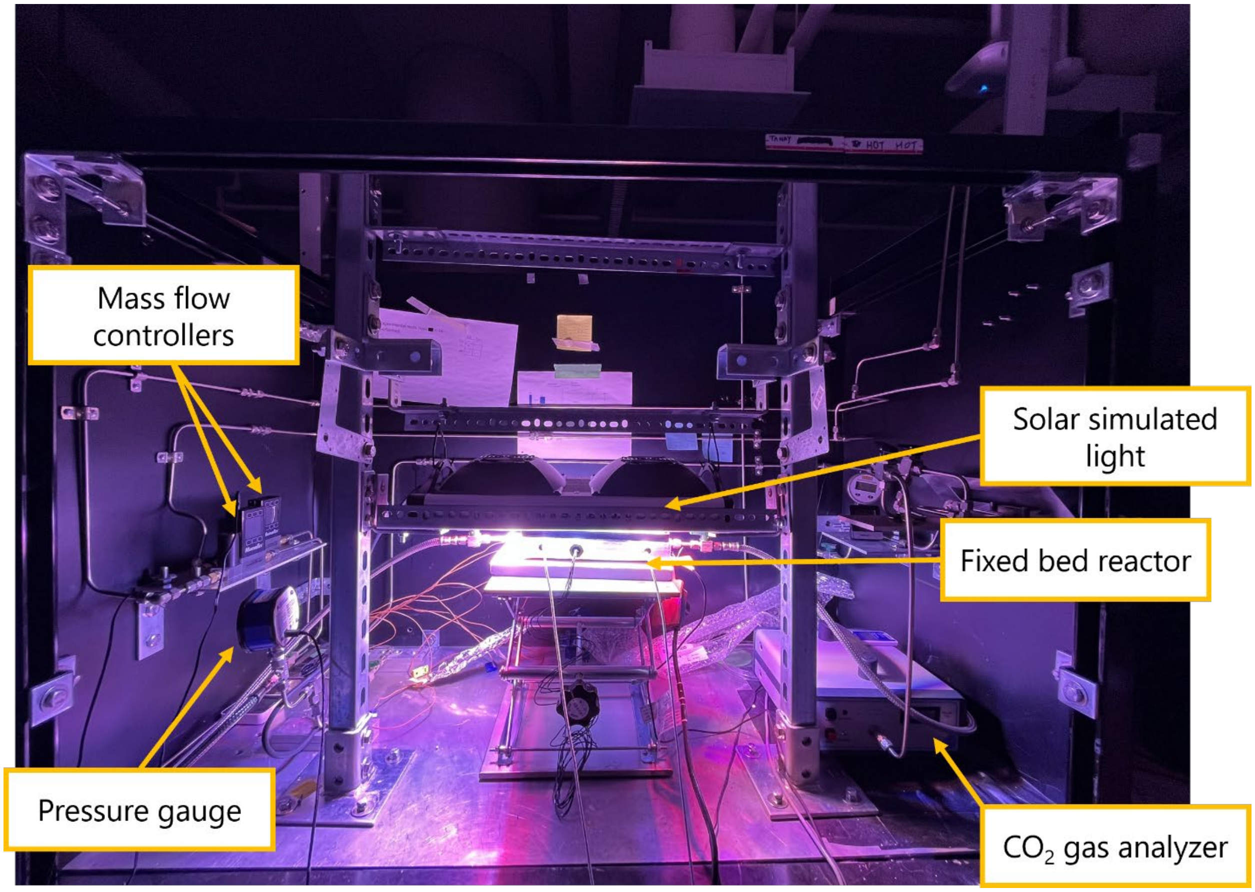


Figure 3.4: Photothermal CO_2 desorption setup with solar simulated light.

The incident light, which had an intensity of 2.3 kW/m^2 was positioned directly above the fixed-bed reactor, which featured a transparent glass lid to allow direct irradiation of the adsorbent surface. As a result, heat was generated locally within the adsorbent bed, raising the temperature to approximately 110 °C and triggering CO_2 desorption and the flow rate during desorption was the same as the adsorption condition. The light intensity of 2.3 kW/m^2, which is twice the intensity of the AM1.5 solar spectra, is easily achievable with the LED light (SolarSystem 550; California Lightworks, USA). The duration of the desorption phase of the experiments was set to 92 mins because the outlet CO_2 concentration returned to a level similar to the inlet concentration.

3.3 Energy Modeling and Regeneration Efficiency

The energy delivered to the system during light-driven regeneration depends on both the power of the light source and the absorption characteristics of the adsorbent material. The energy consumption associated with photothermal regeneration, relative to the functional unit (1 kg CO_2 desorbed), is modeled as:

$$\mathrm{Q_{pt-total}} = \frac{\mathrm{Q_{light}}}{\mathrm{M_{co_2}}} = \frac{\mathrm{I_{light}} * \mathrm{A_{abs}} * \mathrm{t_{des}}}{\mathrm{M_{co_2}}} \tag{3.2}$$

where Q_{light} is the total light energy absorbed and converted into heat (kJ), I_{light} is the intensity of the incident light (W/m²), A_{abs} is the effective surface area of the adsorbent bed that absorbs light (m²), t_{des} is the duration of the light exposure (s) during desorption and M_{CO_2} is the total mass of CO_2 released during desorption stage (kg CO_2).

The photothermal regeneration efficiency is calculated using:

$$n_{pt} = \frac{\Delta \mathrm{H}}{\mathrm{Q_{pt-total}}} \tag{3.3}$$

where n_{pt} is the photothermal regeneration efficiency, $Q_{pt\text{-}total}$ is the photothermal energy requirement (J/kg CO_2), and ΔH is specific heat of desorption (kJ/kgCO_2).

For comparison with the photothermal regeneration method, an electric heating system serves as the heat source for regeneration in what is termed the thermal regeneration method. The energy consumption for this thermal regeneration method is modeled as:

$$\mathrm{Q_{t-total}} = \frac{\mathrm{P_t} * \mathrm{t_{des}}}{\mathrm{M_{CO_2}}} \tag{3.4}$$

where $Q_{t\text{-}total}$ is the thermal energy required (kJ/kg CO_2), P_t is the power supplied to the electric heater (W) and t_{des} is the desorption duration (s).

The thermal regeneration efficiency is then defined as:

$$n_t = \frac{\Delta \mathrm{H}}{\mathrm{Q_{t-total}}} \tag{3.5}$$

where n_t is the thermal regeneration efficiency, calculated as the ratio of the desorption heat requirement to the total thermal energy consumed.

3.4 Summary

This chapter presented a methodological framework for evaluating the life cycle environmental impacts of low-temperature direct air capture systems integrated with solar energy. The study's approach combined established LCA methods with experimental validation to provide a robust assessment of different DACCS configurations. The LCA methodology followed ISO standards using OpenLCA software and the Ecoinvent database, with the ReCiPe Midpoint impact assessment method applied to quantify GWP. The analysis compared three distinct heat supply configurations for adsorbent regeneration: (1) natural gas heat, (2) solar-thermal heat, and (3) a hybrid system combining solar and electric heating. These configurations were evaluated across multiple geographical regions with varying electricity grid mixes and climate conditions, including provinces in Canada, as well as China, Greece, and Switzerland. The functional unit of the assessment was defined as 1 ton of CO_2 captured and stored, facilitating consistent comparisons across different scenarios. The experimental component involved the synthesis and testing of silica-supported polyethyleneimine (SI-PEI) adsorbents. Solar-simulated LED light was used for the photothermal approach, while an electric heater was employed for the comparative thermal regeneration method, allowing for a direct comparison of efficiency and performance between the two regeneration techniques. Energy modeling equations were developed to quantify photothermal and electric heating energy consumption per unit CO_2 desorbed. Regeneration efficiencies were calculated for both methods to enable direct comparison of their environmental and operational performance. These insights are foundational for assessing the viability, scalability, and trade-offs of solar-integrated DAC systems in future carbon removal strategies.

Chapter 4

Results and Discussion

4.1 Life Cycle Analysis

The LCA of the LT-DAC plant reveals significant variations in environmental performance across different geographical locations and system configurations. Analysis of the GHG emissions demonstrates that system layout selection and geographical location substantially influence the overall carbon removal efficiency of DAC operations (Figure 4.1). The solar-only configuration consistently achieves the lowest GHG emissions across all studied regions, with values ranging from 79 kg CO_2-eq/tCO_2 captured and stored in the Canada-Quebec scenario to 366 kg CO_2-eq/tCO_2 in the China scenario. This difference of 287 kg CO_2-eq reflects the influence of regional factors such as solar irradiance, grid electricity carbon intensity, and infrastructure emissions. As shown in Figure 4.1, regions with cleaner grids and stronger solar resources tend to perform significantly better in overall climate impact. These findings align with conclusions from Breyer et al. (2020), who emphasized the critical influence of regional energy mixes on the feasibility of low-emission DAC deployment.

The Canada-Quebec scenario's superior performance in the solar-only configuration can be primarily attributed to its clean electricity grid, with hydropower comprising over 95% of its generation mix. This results in minimal indirect emissions from auxiliary power use, as modeled in the GHG-focused LCI. While construction-phase emissions of hydroelectric infrastructure are accounted for in the Ecoinvent database, the ReCiPe Midpoint (H) method used in this study does not account for broader ecological impacts such as habitat disruption, and long-term land use change—factors which have been highlighted as critical omissions in recent assessments of hydropower sustainability (Gracey & Verones, 2016; Kuriqi et al., 2021). In contrast, the higher emissions observed in the China scenario stem from its coal-dominated electricity grid, which contributes significantly to indirect emissions even in solar-based systems. Intermediate performance of the GHG emissions were observed in Switzerland (80 kg CO_2-eq/tCO_2), Canada-Ontario (100 kg CO_2-eq/tCO_2), and Greece (262 kg CO_2-eq/tCO_2), reflecting their diverse energy mixes and solar resource availability.

The integration of auxiliary heating systems introduces additional complexity to the emissions profile, with values ranging from 90 kg CO_2-eq/tCO_2 in Canada-Quebec to 987 kg CO_2-eq/tCO_2 in China. This configuration shows an average dramatic increase of 253 kg CO_2-eq/tCO_2 compared to solar-only systems across all regions, representing an average 97% increase in emissions.

Notably, the relative performance ranking of regions remains unchanged from the solar-only configuration, suggesting that local grid characteristics continue to play a dominant role in determining overall system performance. Canada-New Brunswick and Canada-Alberta show particularly interesting results in this configuration, with emissions of 364 and 792 kg CO_2-eq/tCO_2 respectively, reflecting their unique combination of solar resources and grid characteristics.

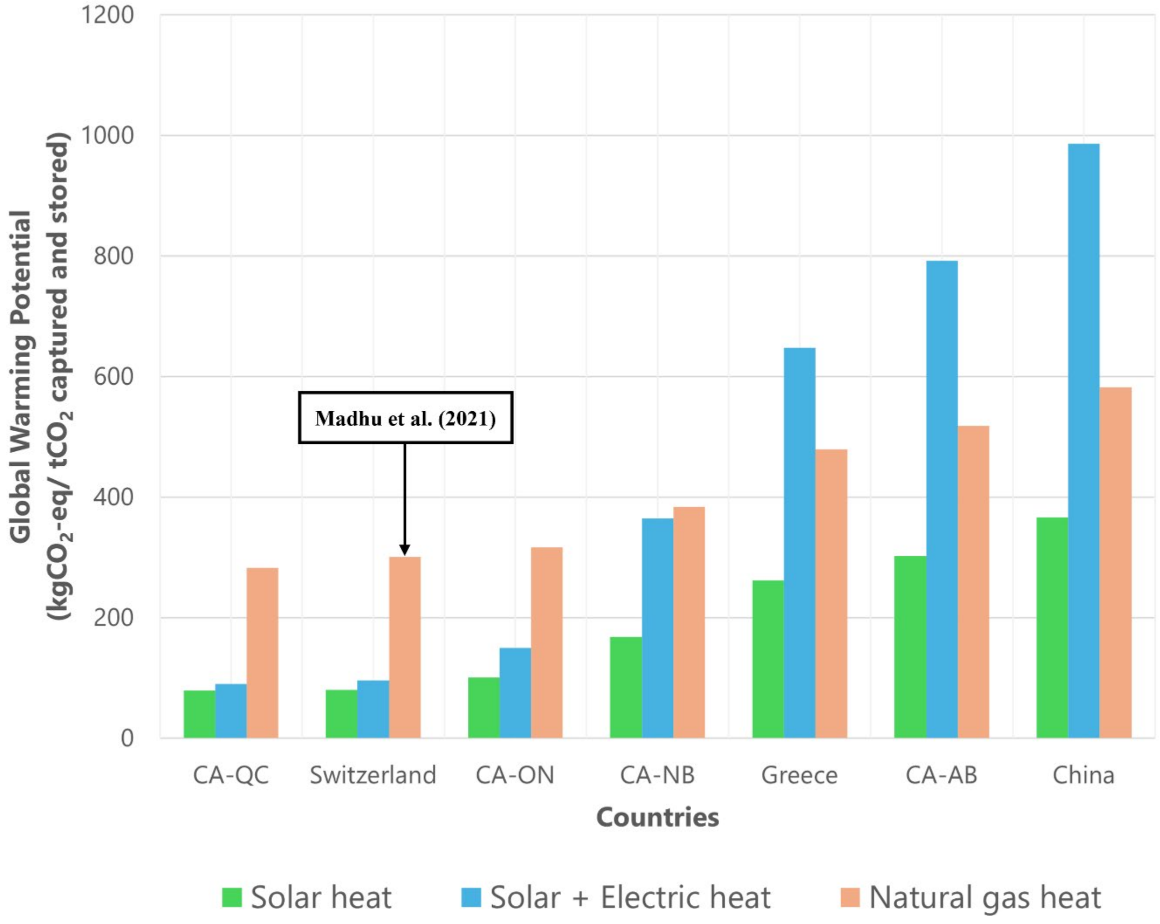


Figure 4.1: Life cycle GHG emissions (kg $CO_{2\text{-eq}}$/ tCO_2) for LT-DAC systems using different energy sources and layouts across selected countries and regions.

To provide a clearer reference point for comparison, the result from Madhu et al. (2021) study which used natural gas as heat source for their LT-DAC reference case in Switzerland is included as a baseline in Figure 4.1, with a GHG emission of 300 kg CO_2-eq/tCO_2. This baseline serves as a useful benchmark to contextualize the emissions associated with different LT-DAC system configurations.

The district/industrial heating configuration utilizing natural gas demonstrates substantially higher emissions across all regions, ranging from 283 kg CO_2-eq/tCO_2 in Canada-Quebec to 582 kg CO_2-eq/tCO_2 in China. When compared to the solar-only configuration, this represents an increase of approximately 200 to 220 kg CO_2-eq per ton of CO_2 captured and stored, depending on the region. This added impact is primarily driven by direct CO_2 emissions from natural gas combustion, which account for 40% to 80% of total life cycle emissions in this configuration. This observation is consistent with Fasihi et al. (2019), who noted that DAC configurations powered by fossil heat sources exhibited much higher GHG footprints even when other system parameters were optimized.

Notably, regional variations in the solar configuration with auxiliary electric heating are more pronounced, with a standard deviation of 337 kg CO_2-eq/tCO_2 — compared to 108 kg CO_2-eq/tCO_2 for the solar-only configuration and 110 kg CO_2-eq/tCO_2 for the natural gas system. This higher variability can be attributed to the hybrid system's dual dependence on two distinct energy sources: intermittent solar availability and grid electricity, whose carbon intensity varies widely by region. In regions with high solar irradiance and clean grids (e.g., Canada–Quebec), the auxiliary system performs similarly to the solar-only configuration. However, in regions with carbon-intensive electricity (e.g., China), the increased reliance on grid electricity during low-sunlight periods substantially raises emissions. Thus, the hybrid system exhibits greater sensitivity to local energy profiles, highlighting the importance of region-specific energy mixes when evaluating DAC deployment strategies.

The sensitivity analysis of regeneration energy requirements in Ontario reveals critical insights into system optimization potential. For every 1 GJ/tCO_2 increase in regeneration energy requirement, emissions increase by approximately 8 kg CO_2-eq/tCO_2 in solar-only systems, 45 kg CO_2-eq/tCO_2 in solar with auxiliary heating, and 168 kg CO_2-eq/tCO_2 in natural gas systems, as shown in Figure 4.2. This linear relationship demonstrates the crucial importance of minimizing regeneration energy demand, particularly in fossil fuel-based systems, where the carbon intensity of the heat source amplifies the climate impact of every additional unit of energy consumed.

The analysis across a regeneration range of 0–12 GJ/tCO_2 shows that emissions in solar-only systems increase moderately (up to 50 kg CO_2-eq/tCO_2), while natural gas systems exhibit the steepest rise, with emissions increasing by as much as 1010 kg CO_2-eq/tCO_2. This sharp escalation is primarily due to the high direct CO_2 emissions associated with natural gas combustion, which scales proportionally with energy demand. In contrast to solar-based systems where incremental energy use draws from a low-carbon source, natural gas systems emit approximately 56 kg CO_2 per GJ of heat produced. Demonstrating that even small increases in regeneration energy translate into substantial increases in life cycle emissions. Consequently, inefficient thermal designs or poorly optimized sorbents in fossil fuel-based configurations can significantly undermine the climate benefits of DAC systems, potentially doubling baseline emissions at high regeneration loads.

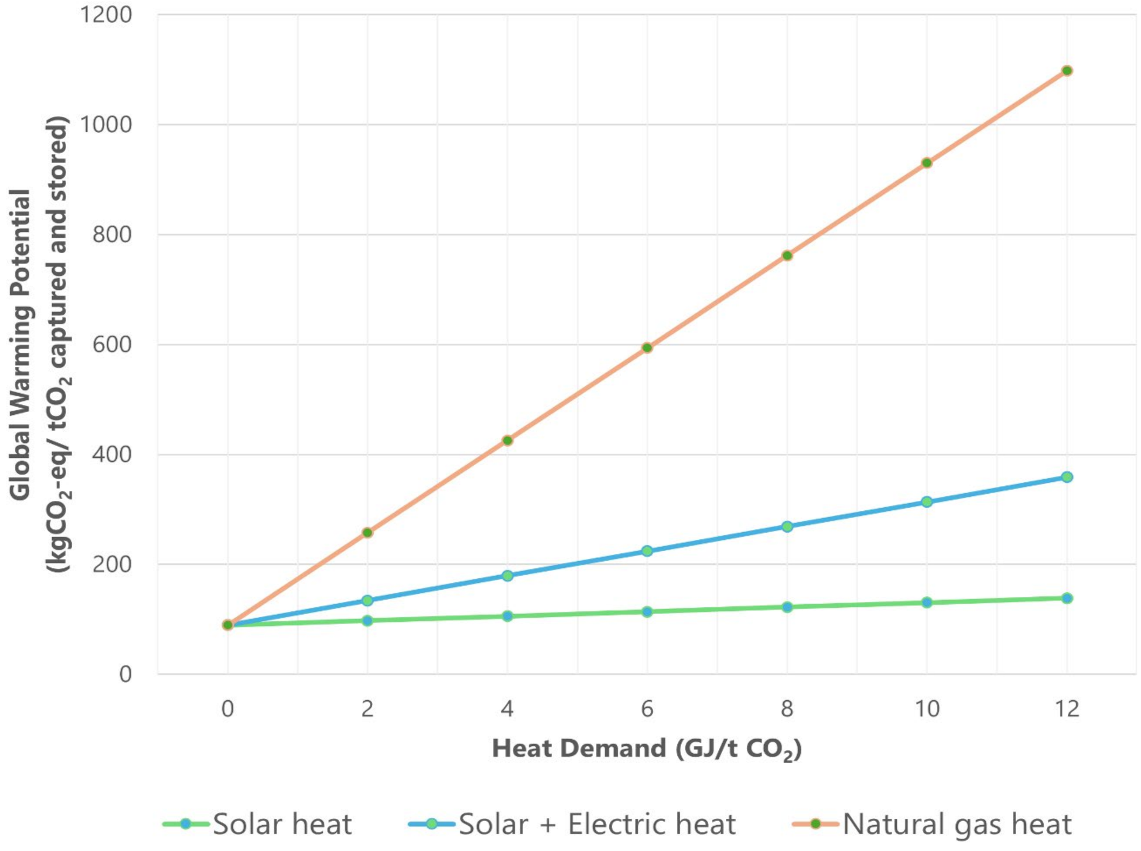


Figure 4.2: Impact of regeneration energy (0–12 GJ/tCO_2) on life cycle GHG emissions (kg CO_2-eq/tCO_2) for LT-DAC systems in Ontario, Canada, across different system layouts.

Process contribution analysis reveals significant differences in the overall impacts of system components, processes, and inputs on emissions per ton of CO_2 captured and stored. As shown in Figure 4.3, in solar-only configurations, the solar collector system contributes 3-40% of total emissions, with notable variations across regions. Switzerland shows the lowest solar system emissions at 7.7 kg CO_2-eq/tCO_2, while Ontario exhibits the highest at 40 kg CO_2-eq/tCO_2, reflecting differences in manufacturing emissions, system installation and construction, the energy used for production and operation of the solar collector, technical specifications, and maintenance. DAC unit infrastructure accounts for 6-27% of emissions across all configurations, ranging from 20 to 23 kg CO_2-eq/tCO_2, with variations primarily driven by regional manufacturing and construction emissions factors.

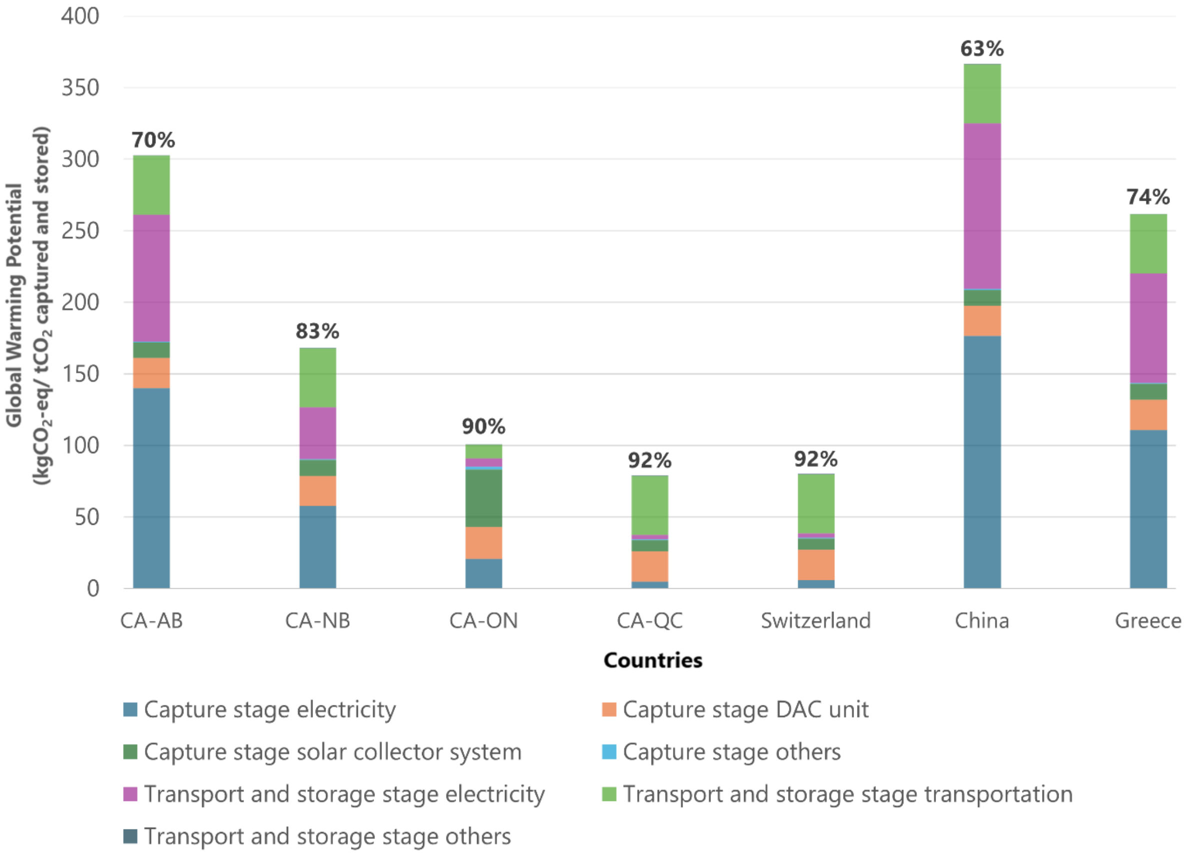


Figure 4.3: Contribution analysis of life cycle GHG emissions (kg CO_2-eq/tCO_2) and efficiencies (%) for LT-DAC systems with solar collector layouts across selected regions.

Life cycle efficiency calculations demonstrate the net carbon removal effectiveness of different configurations. Solar-only systems achieve the highest efficiencies, ranging from 92% in Canada-Quebec and Switzerland to 63% in China. This represents a significant advantage over natural gas systems which, as shown in Figure 4.4, exhibit efficiencies between 72% (Canada-Quebec) and 42% (China). The high life cycle efficiencies observed for solar-powered systems in regions with clean electricity grids (e.g., 90–92% in Canada–Quebec, Ontario, and Switzerland) align closely with the modeling results of Terlouw et al. (2021), who analyzed autonomous DACCS configurations powered by Fresnel solar collectors and photovoltaic electricity, and reported life cycle carbon efficiencies in the range of 84–91%. The natural gas configuration demonstrates the lowest and most variable efficiencies, spanning from 48% in Alberta to 72% in Quebec. These variations, as illustrated in Figure 4.4, reflect both regional energy system characteristics and natural gas supply chain emissions.

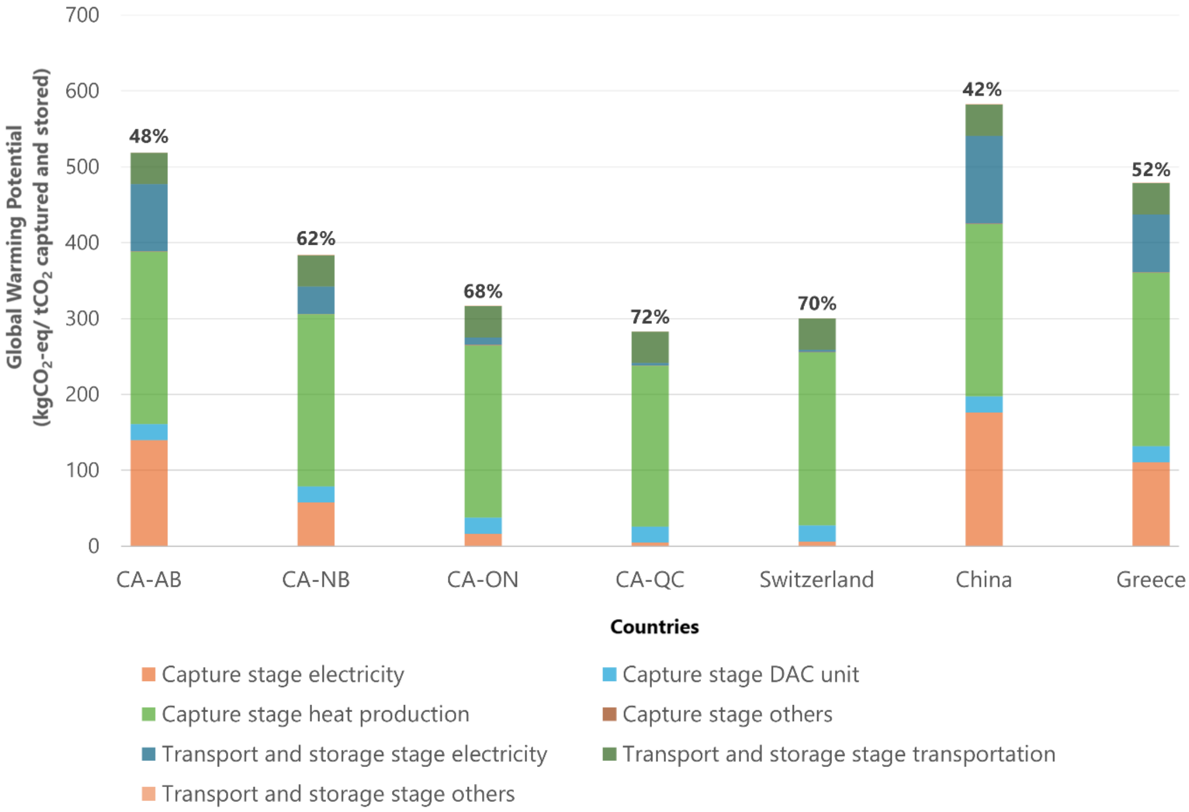


Figure 4.4: Contribution analysis of life cycle GHG emissions (kg CO_2-eq/tCO_2) and efficiencies (%) for LT-DAC systems using district and industrial heating powered by natural gas in various regions.

As illustrated in Figure 4.5, the integration of auxiliary heating systems shows varying efficiencies across regions while maintaining similar component contribution patterns. The efficiency gap between solar and natural gas systems averages 21% across all regions. These efficiency variations directly correlate with grid carbon intensity, with regions having cleaner electricity grids showing larger efficiency advantages for solar-based systems.

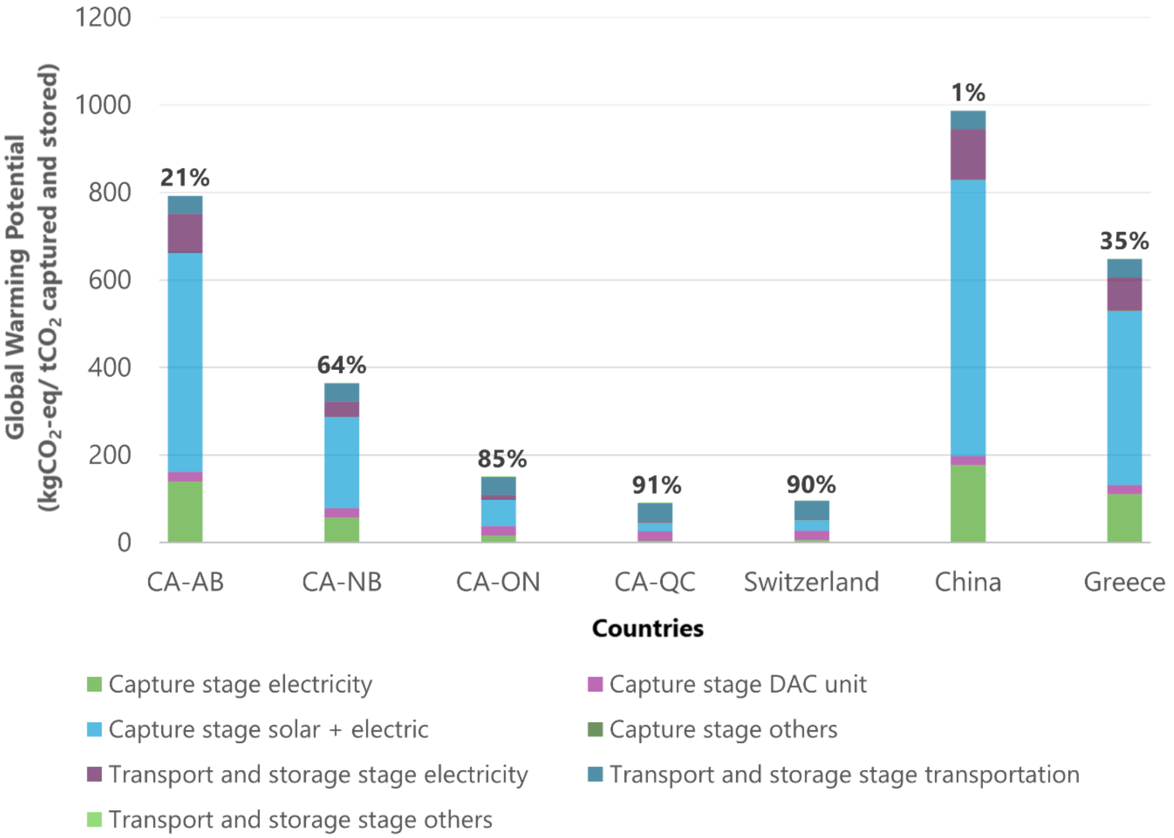


Figure 4.5: Contribution analysis of life cycle GHG emissions (kg CO$_2$-eq/tCO$_2$) and efficiencies (%) for LT-DAC systems using solar collectors with auxiliary heating across various regions.

The regional analysis of the life-cycle efficiency trends reveals the following interesting patterns. Canadian provinces show a consistent efficiency gradient from East to West, with Quebec achieving the highest efficiencies across all configurations, followed by Ontario, New Brunswick, and Alberta. This pattern correlates strongly with grid carbon intensity and solar resource availability. In contrast, European locations (Switzerland and Greece) demonstrate variable efficiency levels despite different climatic conditions, suggesting that grid carbon intensity may be a more dominant factor than solar resource availability in determining overall system efficiency. However, it is also important to note that the life cycle modeling approach used in this study, based on ReCiPe Midpoint (H), emphasizes operational GHG emissions and may underrepresent the embodied or ecological impacts associated with renewable energy infrastructure. As such, these results should be interpreted within the context of a GWP-focused analysis.

The CO_2 transport and storage processes contribute substantially across all configurations, ranging from 15-56% of total emissions (15-157 kg CO_2-eq/tCO_2) for solar based systems and 15-27% of total emissions (44-157 kg CO_2-eq/tCO_2) for natural gas-based systems. Similar component-level variance was noted in the meta-analysis by Terlouw et al. (2021), where CO_2 transport and storage contributions ranged from 11% to 52%, primarily driven by electricity demands for compression, recompression, and injection, as well as CO_2 leakage losses during long-distance pipeline transport. These results emphasize the critical influence of infrastructure layout, transport distances, and regional energy mixes on the total life cycle impact of DACCS.

Notably, regions with more developed CO_2 transport infrastructure, such as Ontario, Quebec, Switzerland, and New Brunswick, show lower transport-related emissions (average 45 kg CO_2-eq/tCO_2) compared to regions requiring new infrastructure development, such as Alberta, China, and Greece (average 135 kg CO_2-eq/tCO_2). This highlights the importance of existing infrastructure in determining overall system performance and highlights a key limitation: the absence of CO_2 pipeline networks or nearby geological storage may significantly constrain the geographic feasibility of DAC deployment, particularly in remote or infrastructure-poor regions.

A detailed examination of electricity consumption patterns reveals significant regional variations in operational emissions. In solar-only configurations, electricity-related emissions range from 8 kg CO_2-eq/tCO_2 in Canada-Quebec to 290 kg CO_2-eq/tCO_2 in China, representing an approximate 36-fold difference. This spread is also the same in configurations with auxiliary heating, emphasizing the compounding effect of grid carbon intensity on system performance. The analysis shows that regions with clean electricity grids can achieve up to 97% lower operational emissions compared to regions with carbon-intensive grids, holding all other factors constant. This finding aligns with the results of Qiu et al. (2022), who projected that as electricity sectors undergo decarbonization, the environmental impacts associated with DACCS operation could decrease by more than 80% by 2050 relative to 2020 levels.

Global DAC deployment is already gaining momentum, with projects spanning Europe, North America, Japan, the Middle East, and even emerging in Africa, reflecting the location flexibility of DAC technology. As of 2025, 27 DAC plants have been commissioned globally, capturing nearly 0.01 Mt CO_2 per year (IEA, 2025). However, to remove approximately 25 Gt CO_2 from the atmosphere between 2025 and 2030, an estimated 5,000 DAC plants, each capturing 1 Mt CO_2 per year would be needed. A single 1 MtCO_2/year facility would require approximately 0.2 km² of land, equivalent to 28 soccer fields (Ozkan et al., 2022). For comparison, a Climeworks DAC unit with a 4 kt CO_2/year capacity occupies around 800 m^2, the size of four typical urban homes (Ozkan et al., 2022).

In the Canadian context, the government aims to reduce national GHG emissions by 40–45% below 2005 levels (572.85 Mt CO_2) by 2030, an approximate reduction of 240 Mt CO_2 (Environment and Climate Change, 2022). This trajectory places DAC in a potentially important role within Canada's climate portfolio. For instance, if scaled linearly from the LCA findings in this study, solar-powered DAC systems such as the one modeled for Quebec and Alberta can achieve an average net reduction of 800 kg CO_2 per tonne captured (i.e., 80% life cycle carbon efficiency). Meeting Canada's 2030 target (Environment and Climate Change, 2022) with such systems would theoretically require approximately 60 DAC plants operating at 1 Mt CO_2/year capacity, starting in 2025. Quebec emerges as an ideal location due to its clean electricity grid and favorable solar potential, while Alberta, with its carbon-intensive grid, reflects a worst-case scenario for DAC deployment efficiency.

4.2 Experimental Analysis

This section presents the experimental characterization of the SI-PEI adsorbent, including its optical properties and the comparative analysis of thermal and photothermal regeneration methods for CO_2 desorption.

4.2.1 UV-Vis

The UV-Vis reflectance spectra of unmodified silica gel and polyethyleneimine-functionalized silica (SI-PEI, 30 wt%) are presented in Figure 4.6. The spectra span the near-infrared (NIR), visible (Vis), and ultraviolet (UV) regions (1400 nm to 200 nm), offering insight into the optical behavior of the materials.

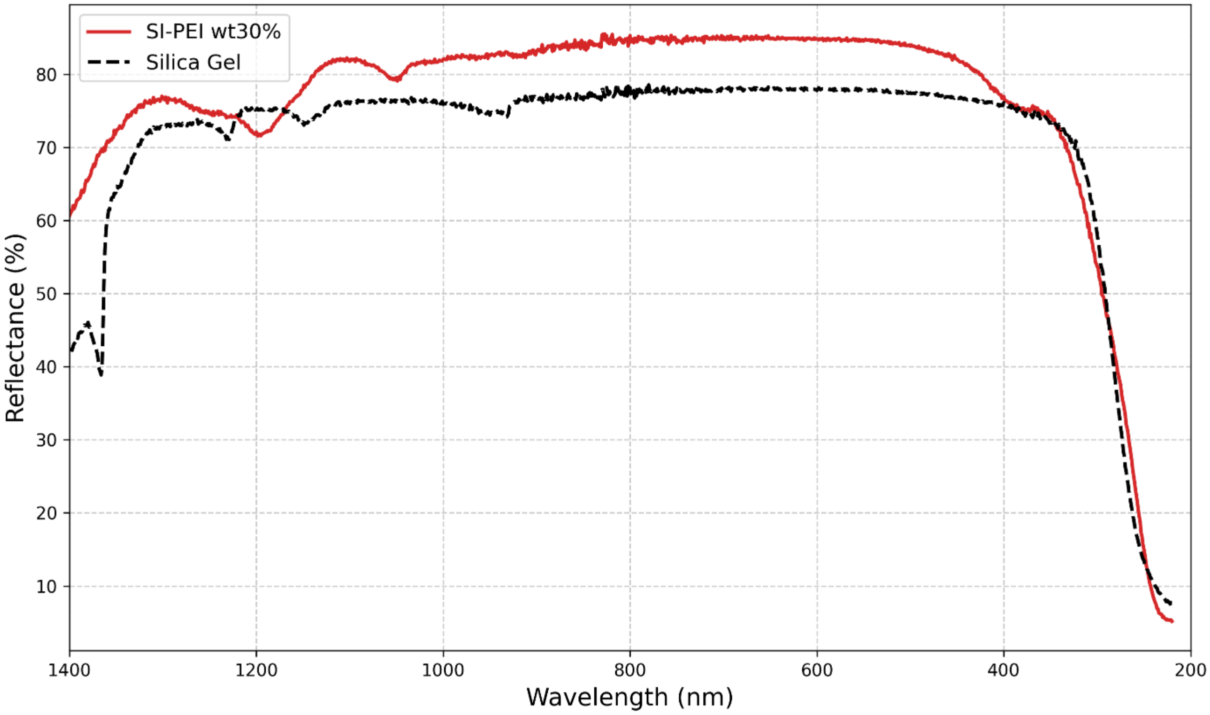


Figure 4.6: UV-Vis reflectance spectra of unmodified silica gel (black dashed) and PEI-functionalized silica (SI-PEI, 30 wt% PEI loading; red solid). Both samples show high reflectance in the visible and NIR regions, while the SI-PEI sample exhibits slightly enhanced absorption below 400 nm.

In the NIR and visible regions (>400 nm), both samples exhibit high reflectance values ranging from 45% to 85%, with the SI-PEI sample showing marginally higher reflectance overall. This indicates that the PEI functionalization does not significantly alter the baseline scattering or color properties of the material, which is consistent with its macroporous silica matrix.

Below 400 nm, a steep decline in reflectance is observed for both samples, indicating strong absorption in the UV region. The SI-PEI sample exhibits a subtle shoulder that is not present in unmodified silica, which may indicate the presence of adsorbed PEI contributing to enhanced UV absorption in this region. While this increased absorption may be attributed to the presence of nitrogen-containing amine groups in PEI, it is also possible that intrinsic features of silica such as defect states or quantum confinement effects, contribute to the observed UV activity. Without additional spectra from isolated PEI or different PEI loadings, this interpretation remains tentative.

Interestingly, Saini et al. (2024) reported that PEI-based polymeric nanoparticles exhibited UV absorption peaks attributed to both π→π* transitions of aromatic sp^2 bonds and n→π* transitions from C=N groups, supporting the idea that amine-rich materials such as PEI can contribute to UV absorption. Although the SI-PEI structure in this study differs, the increased UV absorption below 400 nm is consistent with such findings.

To contextualize the spectral features, Table 4.1 summarizes typical electronic transitions responsible for UV absorption in organic materials. Transitions such as π→π* and n→π* are common in systems containing C=C bonds and amine groups, respectively, and may account for the UV absorption observed in the SI-PEI sample.

Table 4.1: Typical electronic transitions contributing to UV absorption in nitrogen-containing organic materials.

Transition	Involves	Typical Groups	Relative Strength	UV-Vis Impact
π→π*	Pi → Pi*	C=C, aromatic	Strong	Strong absorption
n→π*	Lone pair → Pi*	-NH, $-NH_2$, -OH	Weaker	Moderate absorption

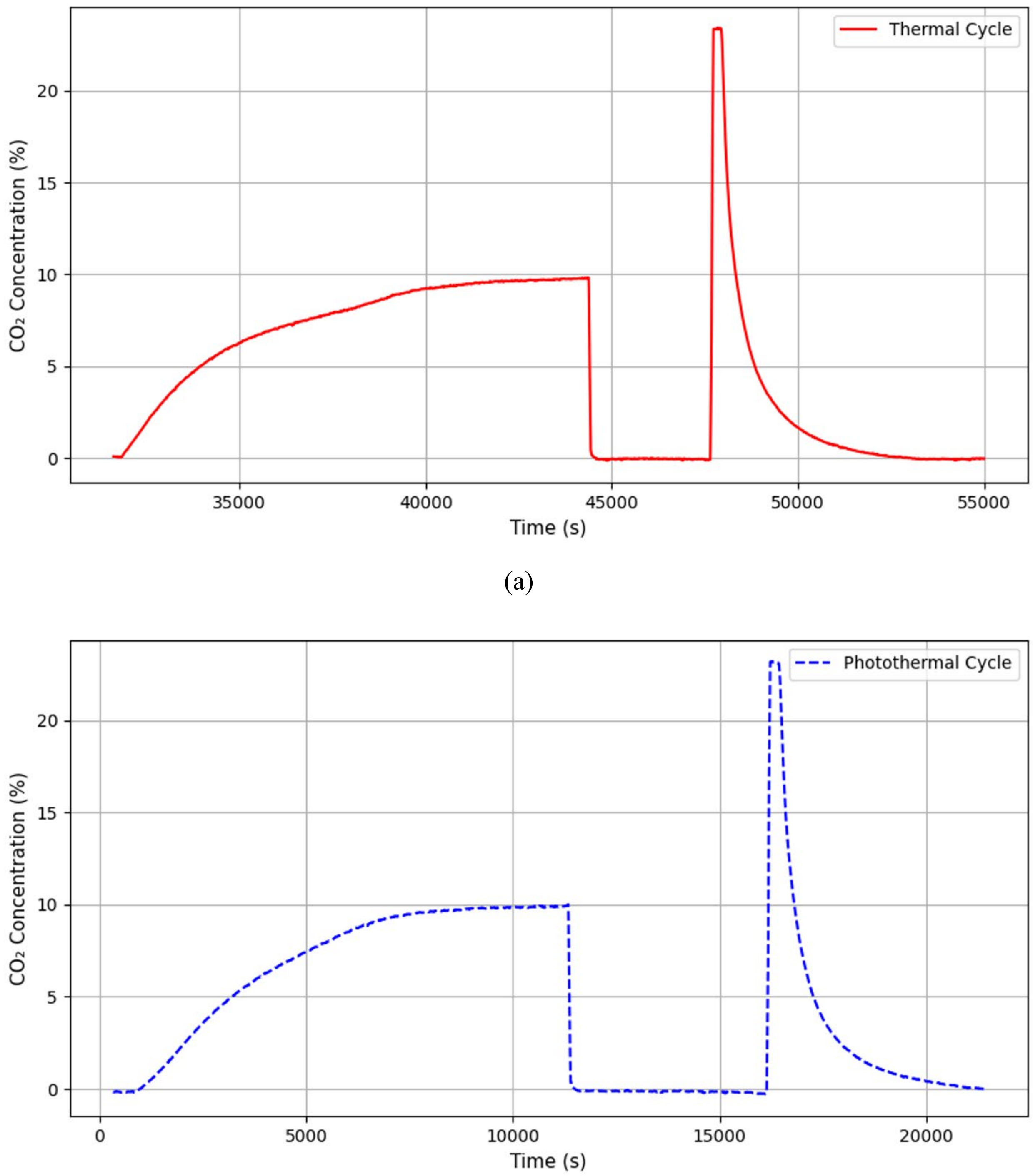
These transitions are representative of electronic excitations in nitrogen-containing organic groups and are included here to contextualize the observed UV absorption. However, the specific contributions in the SI-PEI sample remain uncertain.

Although these transitions provide a plausible explanation for the increased UV absorption in the SI-PEI sample, the specific origin remains uncertain. Further study with control samples would be required to confirm the contribution of PEI.

Overall, the UV-Vis analysis suggests that functionalization with PEI may enhance UV absorptivity while preserving the high visible-range reflectivity of silica. These characteristics could be beneficial for photothermal applications or UV-sensitive process designs, though more targeted optical studies are needed to isolate the effect of the PEI component.

4.2.2 Adsorption and desorption performance

Figure 4.7 illustrates the breakthrough curves for both adsorption and desorption processes, with adsorption conducted at 25°C and desorption at 110°C. The photothermal regeneration method utilized solar simulated light as described in section 3.2.3, while thermal regeneration process employed an electric heating system as the energy source.



(a)

(b)

Figure 4.7: CO_2 adsorption–desorption cycle profiles for (a) thermal and (b) photothermal regeneration.

For better visual comparison, the adsorption and desorption breakthrough curves were plotted separately in Figures 4.8 and 4.9, respectively. This separation allows for direct performance evaluation between the two regeneration techniques under identical temperature conditions, since they are almost identical. The integration plots presented in Figures 4.10 and 4.13 were used to quantify the mass of CO_2 adsorbed and desorbed, which formed the basis for calculating the energy requirements of each system.

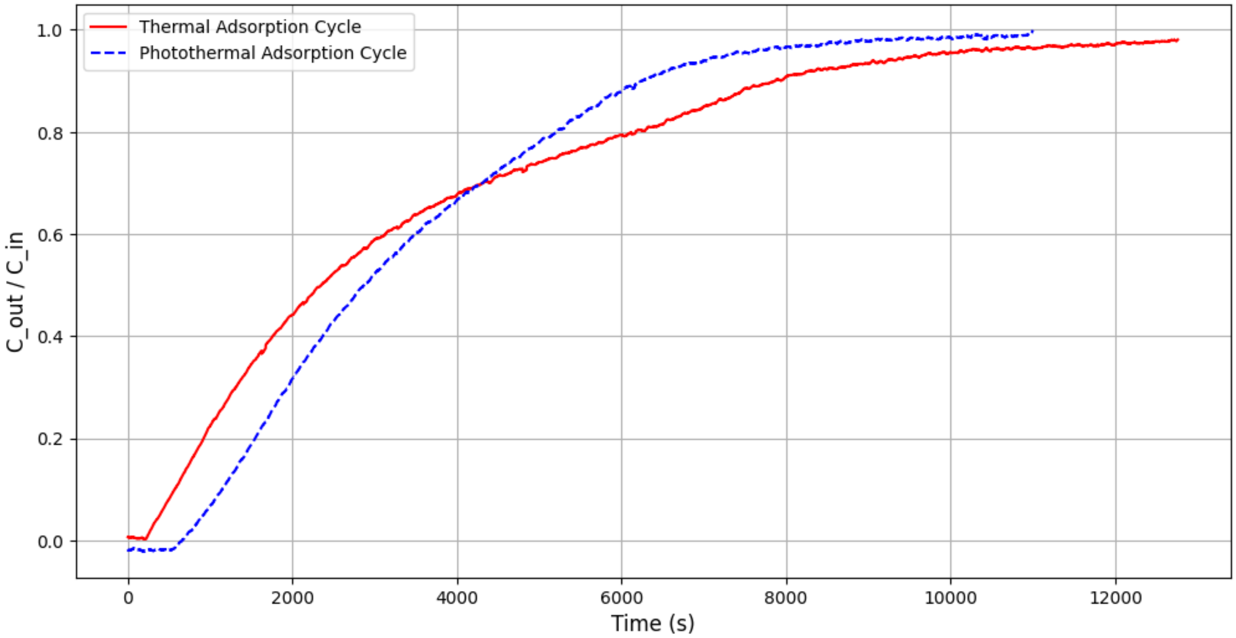


Figure 4.8: Thermal and photothermal CO_2 adsorption breakthrough curves. These breakthrough curves illustrate the CO_2 adsorption performance of SI-PEI, showing the outlet CO_2 concentration as a function of time.

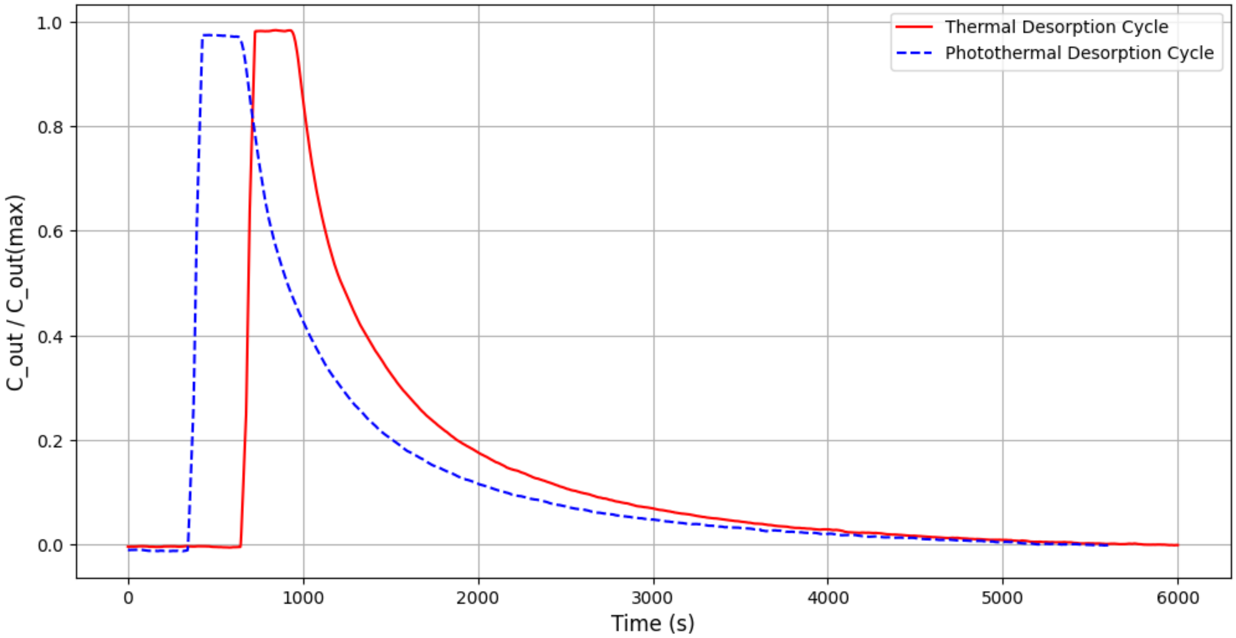
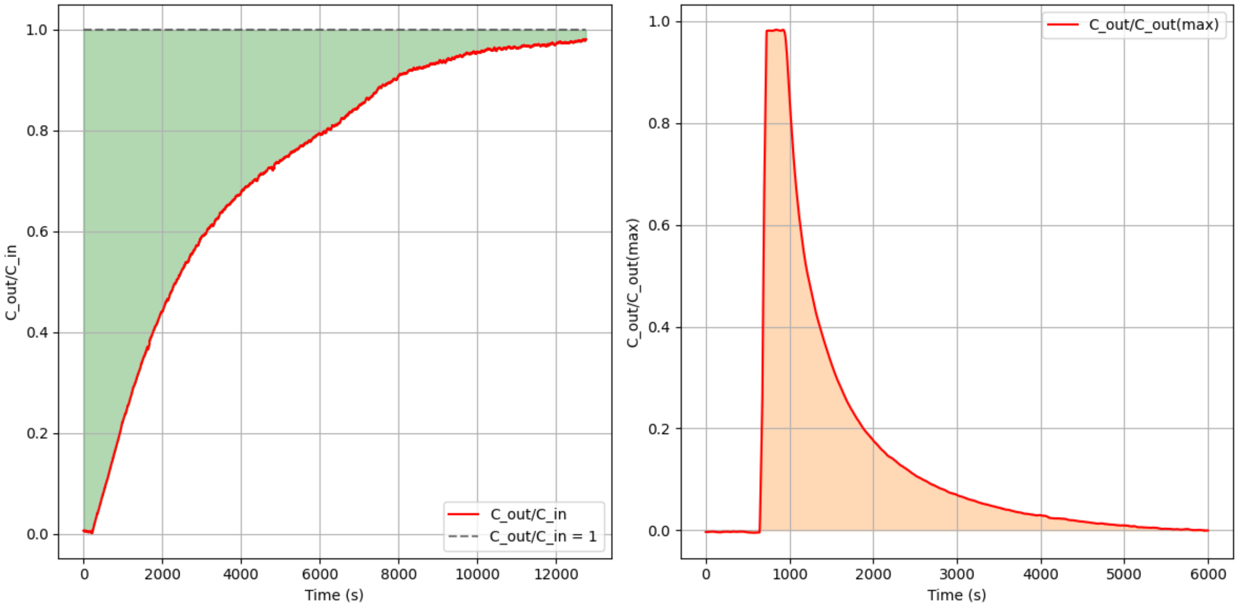


Figure 4.9: Thermal and photothermal CO_2 desorption breakthrough curves. This figure illustrates the desorption profiles for both regeneration methods, highlighting the rate and extent of CO_2 released over time.

4.2.3 Thermal energy requirement

The total thermal energy requirement was determined using Equation 3.4, incorporating key experimental parameters from the electric heater-based desorption process. In this experiment, 40 g of SI-PEI adsorbent was packed into a fixed-bed reactor, and the regeneration was carried out by supplying continuous heat using an electric heater rated at 254 W. The desorption time for the thermal method was 6000 seconds, and integration of the plot in Figure 4.10b revealed a working capacity of 1.28% for the adsorbent based on the mass of CO_2 desorbed.



(a) (b)

Figure 4.10: Thermal cycle CO_2 breakthrough curves showing (a) adsorption and (b) desorption profiles.

Using these experimental values, the thermal regeneration energy requirement was calculated as shown in Figure 4.11.

$$\text{Power to the electric heater} = 254\ \text{W}$$

$$\text{Mass of CO}_2\ \text{desorbed} = 0.0128\,\frac{\text{kgCO}_2}{\text{kg-sorbent}} * 40\ \text{g sorbent} = 0.512\ \text{gCO}_2$$

$$Q_{t-total} = \frac{254\ \text{W} * 6000\ \text{s}}{0.000512\ \text{kg CO}_2}$$

$$Q_{t-total} = 3.0\,\frac{\text{GJ}}{\text{kg CO}_2}$$

Figure 4.11: Calculation note for thermal energy requirement.

The thermal regeneration efficiency (n_t) was determined using the specific heat of desorption (2045 kJ/kg CO_2) from Zhang et al. (2014) as shown in Figure 4.12.

$$n_t = \frac{2045}{3000 * 10^3} \frac{\frac{\text{kJ}}{\text{kg CO}_2}}{\frac{\text{kJ}}{\text{kg CO}_2}} * 100\%$$

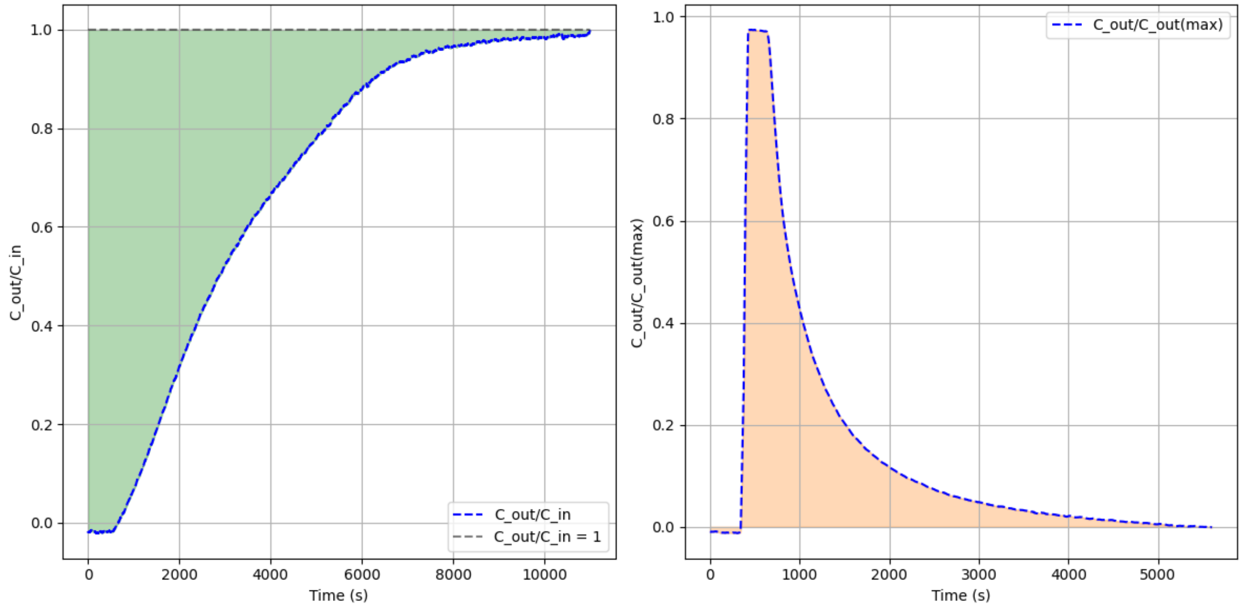
$$n_t = 0.07\%$$

Figure 4.12: Calculation note for thermal regeneration efficiency.

This low regeneration efficiency reflects the mismatch between energy supplied and the energy strictly needed for CO_2 desorption, with significant losses likely due to heat dissipation and limited thermal control in the lab-scale setup.

4.2.4 Photothermal energy requirement

For the photothermal regeneration method, the experimental setup utilized light at an intensity of 2300 W/m² incident onto the adsorbent. The area of the adsorbent bed when projected onto the horizontal plane (the plane parallel with the surface of the LED light source) is 275 cm². The desorption phase lasted 5500 seconds and based on the integration of the curve in Figure 4.13b, the mass of CO_2 desorbed was 0.0123 kg.



(a) (b)

Figure 4.13: Photothermal cycle CO_2 breakthrough curves showing (a) adsorption and (b) desorption profiles.

The photothermal energy requirement was calculated using Equation 3.2 as shown in Figure 4.14, by dividing the total light energy input by the mass of CO_2 desorbed, resulting in 707 MJ/kg CO_2.

$$\text{Surface area of adsorbent bed} = 25\ \text{cm x } 11\ \text{cm} = 275\ \text{cm}^2 = 0.0275\ \text{m}^2$$

$$\text{Light intensity} = 2300\ \text{W/m}^2$$

$$\text{Mass of CO}_2\text{ desorbed} = 0.0123\ \frac{\text{kgCO}_2}{\text{kg}-\text{sorbent}} * 40\ \text{g of sorbent} = 0.492\ \text{gCO}_2$$

$$Q_{\text{pt}-\text{total}} = \frac{Q_{\text{light}}}{M_{CO_2}} = \frac{2300\,\frac{W}{m^2} * 0.0275\ m^2 * 5500\ s}{0.000492\ \text{kg CO}_2}$$

$$Q_{\text{pt}-\text{total}} = 707\,\frac{\text{MJ}}{\text{kg CO}_2}$$

Figure 4.14: Calculation note for photothermal energy requirement.

The photothermal regeneration efficiency (n_{pt}) was then calculated using Equation 3.3 and the same heat of desorption (ΔH = 2045 kJ/kg CO_2) as shown in Figure 4.15.

$$n_{pt} = \frac{2045}{707 * 10^3}\ \frac{\frac{\text{kJ}}{\text{kg CO}_2}}{\frac{\text{kJ}}{\text{kg CO}_2}} * 100\%$$

$$n_{pt} = 0.29\%$$

Figure 4.15: Calculation note for photothermal regeneration efficiency.

Although higher than the efficiency observed for the electric thermal regeneration method (0.07%), the photothermal efficiency remains relatively low. This indicates potential energy losses through reflection, non-absorbed radiation, or non-uniform heat distribution, emphasizing the need for further system optimization. Table 4.2 shows the key experimental values and calculated regeneration efficiencies for thermal and photothermal desorption methods using SI-PEI adsorbents. It is important to note that these efficiency values represent preliminary estimates based on experimental measurements with inherent uncertainties. Key sources of uncertainty include CO_2 mass quantification, light intensity, power input, and surface area measurements. The relative uncertainties for both thermal and photothermal regeneration efficiencies are expected to be below

10%. Accordingly, the reported values are rounded to two significant figures (0.07% and 0.29%) to reflect their precision and support a fair comparison.

Table 4.2: Comparison of thermal and photothermal regeneration parameters for CO_2 desorption

Parameter	Thermal Regeneration	Photothermal Regeneration
Energy input	3.0 GJ/kg CO_2	707 MJ/kg CO_2
Efficiency	0.07%	0.29%
CO_2 desorbed	0.512 g	0.492 g
Power source	Electric heater	LED light (2300 W/m²)
Desorption time	6000 s	5500 s

4.2.5 Discussion of energy requirements and regeneration efficiency

The comparison of the two experimental desorption methods as shown in Table 4.2 highlights important differences in regeneration performance and energy efficiency. The thermal regeneration method using an electric heater required a total energy input of 3.0 GJ/kg CO2, which significantly exceeds the practical energy targets for LT-DAC systems, generally reported between 2.3–7 GJ/tCO2 in literature. The resulting thermal regeneration efficiency was only 0.07%, reflecting considerable system-level losses. These losses are likely due to the lab-scale setup's lack of insulation, uncontrolled heat dissipation to the reactor body and surroundings, and inefficient delivery of energy directly to the adsorbent surface.

In comparison, the photothermal regeneration system demonstrated a lower energy requirement of 0.71 GJ/kg CO2 and an improved efficiency of 0.29%, which is over four times greater than the thermal method. While still far from practical efficiency targets, this result indicates that localized, surface-targeted heating through light irradiation can reduce some of the inefficiencies associated with conventional bulk heating methods.

While the difference in performance is promising, further research is necessary before definitively establishing the superiority of either approach. The current laboratory setup has not been optimized for thermal or photothermal efficiency, heat transfer, light utilization, or sorbent loading configurations. Additionally, both systems were tested under controlled but non-optimized conditions, making direct extrapolation to large-scale applications premature.

Nevertheless, the photothermal process has been proven feasible. It is important to emphasize that the experimental setup was designed to demonstrate the concept of CO_2 regeneration via light irradiation, not to maximize efficiency. A few studies have suggested the potential of releasing CO_2 via light-driven processes, with the claim that photothermal desorption could significantly reduce the heat energy required in DAC regeneration. However, these works did not provide quantified values or systematic analysis. To the best of my knowledge, this is the first study to experimentally quantify energy input and efficiency for photothermal regeneration using simulated solar irradiation.

This preliminary result, despite its modest performance, highlights the significant improvement potential of photothermal DAC systems. For instance, improving regeneration efficiency from 0.3% to 2% would reduce energy requirements by over 80%, directly enhancing life-cycle carbon efficiency and reducing associated GHG emissions, particularly when powered by low-carbon energy sources. Future studies should focus on optimizing reactor design, enhancing optical absorption of the adsorbent, and improving heat confinement strategies to fully realize the benefits of this approach. As global deployment needs approach thousands of DAC units by 2030 (IEA, 2025), small-scale innovations such as photothermal regeneration become increasingly critical for enabling scalable, low-energy carbon removal solutions.

Another critical area for future work is the development of integrated energy systems that enhance the reliability of solar-driven DAC operations. Exploring hybrid renewable energy configurations, such as combining solar thermal energy with battery storage or thermal storage systems like molten salt repositories, could mitigate the intermittency challenges associated with solar energy. Energy system modeling using tools such as EnergyPLAN or HOMER would help identify optimal configurations for different geographical contexts, while pilot-scale demonstrations would validate these integrated approaches under real-world conditions and identify practical implementation challenges.

To fully assess the scalability of DAC, future research should also investigate the implications of large-scale DAC deployment on other environmental impact categories such as land use, water consumption, and resource depletion. While DAC has a relatively small land footprint compared to alternatives like afforestation or BECCS, the material and energy inputs required for large-scale implementation must be carefully assessed to avoid unintended environmental trade-offs. Life cycle sustainability assessment methodologies that extend beyond environmental impacts to include social and economic dimensions would provide a more comprehensive understanding of DAC's sustainability profile, while circular economy principles applied to system design could maximize resource efficiency throughout the technology lifecycle.

4.3 Summary

This chapter provided a detailed examination of both the life cycle environmental impacts and experimental regeneration performance of LT-DAC systems. The life cycle analysis revealed that solar-only configurations consistently deliver the lowest GHG emissions and highest carbon efficiencies across all evaluated regions, particularly in locations with clean electricity grids such as Quebec, Ontario, and Switzerland. In contrast, systems utilizing natural gas or auxiliary electric heating demonstrated significantly higher emissions—up to 987 kg CO_2-eq/tCO_2 in carbon-intensive regions—highlighting the critical influence of grid carbon intensity and energy source on DAC sustainability.

Sensitivity analyses confirmed that regeneration energy demand is a key driver of system-wide emissions, with fossil-based systems showing steep emission increases at higher energy loads. Contribution analyses further emphasized that infrastructure, CO_2 transport, and storage can account for up to 56% of total life cycle emissions, underscoring the importance of considering full system boundaries and regional infrastructure readiness in DAC deployment strategies.

On the experimental side, the comparison between thermal and photothermal regeneration methods revealed that while both approaches currently operate at low efficiencies (0.07% and 0.29% respectively), photothermal regeneration offers a promising pathway to reduce energy inputs through localized heat delivery. Though the experimental setup was not optimized, this study is among the first to provide quantified values for photothermal desorption energy and efficiency using simulated solar irradiation. These findings validate the proof-of-concept and demonstrate the feasibility of light-driven CO_2 release for DAC applications.

In summary, the results support a strategic focus on optimizing regeneration efficiency and leveraging low-carbon, renewable energy sources for heating. Future DAC designs should prioritize solar-thermal integration, material-level improvements, and region-specific deployment to achieve meaningful climate benefits. The findings also inform policy considerations by demonstrating that emissions can vary by a factor of up to 4 depending on configuration and location, reinforcing the importance of aligning technological and geographical strategies for effective carbon removal.

Chapter 5

Conclusions and Future work

5.1 Conclusions

This research has provided an in-depth analysis of the environmental performance and efficiency of DAC technology, particularly with the integration of solar thermal energy. Through a comprehensive LCA, this study has evaluated the GHG emissions, energy consumption, and overall sustainability of DAC under different configurations and geographical scenarios. The findings indicate that DAC has the potential to contribute significantly to climate change mitigation efforts, particularly when powered by renewable energy sources such as solar thermal energy.

A key conclusion drawn from this study is that the choice of energy source for sorbent regeneration plays a critical role in determining the environmental impact of DAC systems. The analysis revealed that conventional DAC systems relying on fossil fuels for heat generation contribute substantially to life cycle GHG emissions, with emissions reaching up to 987 kg CO_2-eq per ton of CO_2 captured in regions with carbon-intensive electricity grids, such as China. However, when DAC systems are powered by solar thermal energy, emissions are significantly lower, ranging from 79 kg CO_2-eq/t CO_2 in Canada-Quebec to 366 kg CO_2-eq/t CO_2 in China, representing a 363% variation between the best and worst-performing regions. These results highlight the importance of siting DAC facilities in regions with abundant renewable energy resources, making them a more sustainable option for long-term carbon removal strategies.

This research identified that DAC systems using natural gas for heating exhibit significantly higher emissions, with values ranging from 283 kg CO_2-eq/t CO_2 in Canada-Quebec to 582 kg CO_2-eq/t CO_2 in China. These emissions result in an efficiency reduction of up to 50% compared to solar-based systems, emphasizing the need to transition away from fossil-fuel-powered DAC configurations. These findings align with and extend previous research by Madhu et al. (2021) and Terlouw et al. (2021), who reported similar trends but with less geographical specificity and without the direct experimental validation provided in this study.

The experimental results comparing thermal and photothermal regeneration further highlighted the potential benefits of alternative regeneration pathways. While both methods were tested in an unoptimized lab-scale setup, the photothermal approach demonstrated a lower energy requirement (707 MJ/kg CO_2 vs. 3.0 GJ/kg CO_2 for thermal) and higher regeneration efficiency (0.29% vs. 0.07%). Although these values are still well below commercial benchmarks, they affirm the feasibility of using light as a regeneration source and suggest substantial room for improvement through system optimization. For example, increasing photothermal regeneration efficiency to just 2% could reduce energy input by over 80%, offering major life cycle benefits, particularly when paired with clean electricity.

A sensitivity analysis of regeneration energy requirements demonstrated that for every 1 GJ/t CO_2 increase in energy demand, emissions increased by approximately 8 kg CO_2-eq/t CO_2 in solar-only systems, 45 kg CO_2-eq/t CO_2 in solar with auxiliary heating, and 168 kg CO_2-eq/t CO_2 in natural gas systems. These findings highlight the crucial importance of minimizing energy consumption through improved sorbent materials and energy-efficient system designs. It also highlights the importance of system design, site selection, and energy integration in optimizing DAC's environmental performance. Regions with high solar radiation and clean electricity grids are particularly well-suited for DAC deployment, as they can achieve greater net-negative emissions with lower energy penalties.

In addition to energy source and regeneration method, regional infrastructure also emerged as a key driver of system performance. CO_2 transport and storage processes contributed between 15–56% of total emissions, with lower impacts observed in regions like Quebec and Switzerland where pipeline infrastructure and storage capacity are more readily available. This highlights a critical constraint on DAC scalability: the absence of transport and storage infrastructure may limit feasibility in certain regions despite favorable solar or grid conditions.

Despite the promising potential of DAC, several significant challenges remain that must be addressed before widespread implementation. These include high energy intensity, limited regeneration efficiency, and high capital and operational costs. Although solar-powered configurations offer notable environmental benefits, their effectiveness is constrained by day-night cycles and seasonal fluctuations in solar irradiance, necessitating the development of efficient energy storage solutions or hybrid approaches. Furthermore, scaling DAC from laboratory setups to industrial applications demands thorough planning around land use, infrastructure reliability, and seamless integration with CO_2 transport and storage networks.

For DAC technology to achieve its potential as a climate mitigation tool, specific policy interventions will be necessary. These include carbon pricing mechanisms that accurately value the environmental benefits of direct air capture, research and development subsidies targeted at reducing capital costs and improving energy efficiency, regulatory frameworks that incentivize the deployment of DAC in optimal geographical locations, and integration of DAC into broader climate policy portfolios, including carbon markets and offsetting schemes.

In summary, this research affirms the environmental promise of solar-integrated DAC systems and introduces the first quantified experimental validation of photothermal regeneration using simulated solar light. Although current efficiencies are modest, the demonstrated feasibility provides a foundation for future technological refinement. With continued technological innovation and appropriate policy frameworks, DAC has the potential to become an increasingly important tool in the global effort to achieve net-zero emissions and eventually net-negative emissions in the coming decades.

5.2 Future Work

While this research has provided valuable insights into the environmental performance of DAC, there are several areas that require further investigation to improve the feasibility and effectiveness of this technology.

Future research should focus on optimizing sorbent materials to enhance capture efficiency and reduce regeneration energy requirements. Computational methods like molecular dynamics simulations and machine learning-assisted screening could accelerate the discovery of novel materials. One of the key limitations in this study was the lack of LCI data for proprietary or novel sorbents such as SI-AEATPMS. To address this, closer collaboration between chemists, materials scientists, and LCA practitioners to improve the transparency and availability of data related to synthesis routes, energy consumption, precursor inputs, emissions, degradation mechanisms, and end-of-life scenarios for advanced adsorbents. Providing such data, even in standardized or modular forms, would enhance the robustness of sustainability assessments and accelerate material screening for climate-relevant applications.

Complementing material innovation, future work should also focus on process engineering advancements, such as modular reactor designs, process intensification, and the integration of hybrid energy systems. Pilot-scale testing of optimized photothermal reactors is needed to validate lab-scale findings and refine operational parameters for real-world conditions. Additionally, investigating pathways to enhance heat confinement, optical absorption, and energy recovery will help improve system efficiency. In addition to solar and electric heating configurations, emerging renewable energy storage technologies should be explored to address solar intermittency in DAC operations. Options such as gravity-based storage, molten salt thermal reservoirs, compressed air systems, or hydrogen-based energy carriers could offer low-carbon, dispatchable heat or power sources for continuous adsorbent regeneration. Integrating these solutions and evaluating them through system modeling could improve the resilience and scalability of DAC under variable weather and grid conditions.

Beyond CO_2 metrics, large-scale DAC deployment must be evaluated through a comprehensive sustainability lens. Future studies should assess potential trade-offs in land use, water consumption, and resource depletion. While DAC has a relatively small land footprint compared to alternatives like BECCS or afforestation, the material and energy demands of widespread deployment warrant further scrutiny. Life cycle sustainability assessment methodologies that extend beyond environmental impacts to include economic and social dimensions will be critical. Circular economy principles, applied to sorbent design and plant infrastructure, could further enhance resource efficiency throughout the system lifecycle.

To ensure the long-term financial viability of DAC, future LCA studies should be complemented with techno-economic analyses that explore cost-reduction opportunities through economies of scale, process optimization, and supply chain improvements. Standardized frameworks for comparing different CDR technologies would facilitate more objective assessment of their relative merits, while multi-criteria decision analysis could provide a more holistic evaluation of DAC's societal value. Assessing co-benefits such as job creation, air quality improvements, and potential integration with industrial carbon capture and storage hubs could strengthen the case for policy support and investment.

Widespread DAC deployment will also depend on supportive policy engagement and stakeholder collaboration. Developing clear regulatory frameworks, carbon pricing mechanisms, and financial incentives can drive investment and accelerate adoption. Research on policy design should explore innovative approaches such as carbon removal credits, public procurement commitments, and technology-specific support mechanisms to address the unique challenges of scaling DAC technology. Particular attention should be paid to harmonizing measurement, reporting, and verification (MRV) standards for carbon accounting across jurisdictions. . Interdisciplinary collaborations between engineers, economists, policymakers, and social scientists will be essential for translating technical progress into equitable and scalable climate solutions. Engaging stakeholders through participatory research will also ensure that DAC development aligns with broader societal priorities and environmental justice goals.

In conclusion, while DAC presents a promising solution for atmospheric carbon removal, realizing its full potential will require integrated research spanning material science, system engineering, policy, and economics. By addressing the multifaceted challenges outlined above through interdisciplinary collaboration and innovation, DAC can become a practical and scalable component of global climate change mitigation strategies.

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