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Ollscoil na hÉireann, Corcaigh
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**Direct Air Capture in Ireland: Research into Technical and
Environmental Insights for its Application**

Thesis presented by
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For the degree of
Doctor of Philosophy

**Process & Chemical Engineering, School of Engineering and
Architecture**
&
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University College Cork

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Declaration

I declare that this thesis is my own work and that has not been submitted in another degree, either at University College Cork or another institution. All external references and sources are clearly acknowledged and identified within the contents. I have read and understood the regulations of University College Cork concerning plagiarism and intellectual property.

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Abstract

Direct Air Capture (DAC) technology offers a promising solution for reducing atmospheric CO₂ concentrations and mitigating climate change. This thesis provides a comprehensive analysis of DAC, focusing on its potential application in Ireland, where renewable energy resources and geological storage capacities are ideal. The research explores the development of DAC technologies and assesses their viability through a site-specific Life Cycle Assessment (LCA). The LCA results show that Ireland is uniquely positioned to benefit from DAC, thanks to its future abundant wind energy and the availability of depleted gas fields for CO₂ storage.

Additionally, this thesis investigates material improvements for DAC systems, particularly through experimental testing of solid sorbents in a lab scale Pressure Swing Adsorption (PSA) apparatus. Materials such as Lewatit and amine-based adsorbents were found to exhibit promising performance, though challenges related to energy efficiency and humidity sensitivity remain.

The findings suggest that while DAC technology holds great potential, significant advancements in material efficiency, energy consumption, and policy support are required to scale DAC effectively. The research contributes to the growing field of DAC by offering insights into the technical and environmental challenges of deploying this technology in Ireland, as well as proposing solutions to enhance its future viability.

Thesis Outputs

Journal articles

- Direct Air Capture in the Republic of Ireland. Is it necessary?

<https://www.sciencedirect.com/science/article/pii/S2352484722016389>

- The impact of Direct Air Capture during the last two decades: A bibliometric analysis of the scientific research, part I

<https://www.sciencedirect.com/science/article/pii/S2772826922000086>

- The impact of direct air capture during the last two decades: A bibliometric analysis of the scientific research Part II

<https://www.sciencedirect.com/science/article/pii/S277282692300010X>

- Life cycle assessment of a direct air capture and storage plant in Ireland

<https://www.nature.com/articles/s41598-023-44709-z>

- Materials for Direct Air Capture: A comparative study of Zeolite 13-X, Lewatit and amine functionalised SBA-15 (Under review)

Conferences & Presentations

- **Environ2023, Letterkenny**

Poster presentation: *Carbon Capture and Direct Air Capture in Ireland: A pathway to a clean future*

Oral presentation: *Carbon Capture and Direct Air Capture in Ireland: A pathway to a clean future*

Oral presentation: *Biomass with Carbon Capture and Storage in Ireland: A techno-economic review*

- **Environ2024, Waterford**

Oral presentation: *Life Cycle Assessment of a Direct Air Capture plant in Ireland*

- **2024 AIChE Annual Meeting, San Diego**

Poster presentation: *Research in Novel Solid Sorbents for Direct Air Capture and Carbon Capture*

<https://aiche.confex.com/aiche/2024/meetingapp.cgi/Paper/689535>

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1 Introduction

1.1 Background

1.1.1 Global climate context and CO₂ emissions

In April 2016, during the Paris Agreement, the world committed to limit the global temperature rise to 1.5°C above pre-industrial levels. However, in 2023 and 2024, following the effects of El Niño, global temperatures surpassed this critical threshold for the first time (K. Li et al., 2023). The Intergovernmental Panel on Climate Change (IPCC) has emphasised the importance of rapid and sustained reductions in Greenhouse Gas (GHG) emissions across all the sectors through the end of this decade (IPCC, 2023). While this breach does not immediately signify the failure of the agreement, scientists now warn that limiting warming to 1.5°C may no longer be possible (Milman, 2024). The consequences of a warming planet are already evident, with increasing occurrences of flash floods, droughts, and food insecurity (Gibbons, 2024).

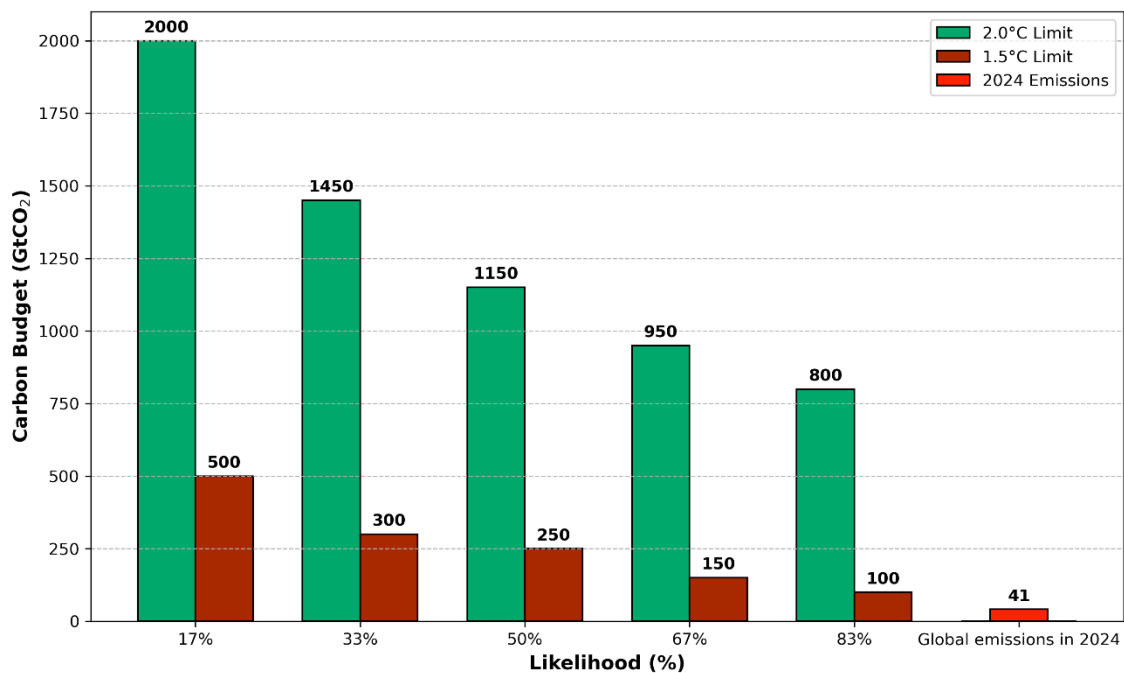


Figure 1.1. Carbon budget likelihood to limit global warming to 1.5 °C (dark red) & 2.0 °C (green) and CO_{2eq} emissions emitted in 2024 (red). Source: (Forster et al., 2023)

At the 16th session of the Conference of Parties (COP16) held in Cancun in 2010, an agreement was adopted to limit global warming to 2 °C. This statement do not suggest abandoning the ambitious targets of the Paris agreement, but rather emphasizes that each

tenth of degree avoided matters, and the term “net zero” should not become meaningless. The Global Carbon Budget reported that in 2024, global CO₂ emissions are expected to rise to 41.6 GtCO₂ (Forster et al., 2023). Currently, around 60% of the anthropogenic emissions are absorbed by oceans and land ecosystems (Forster et al., 2023). These natural processes are crucial to regulating Earth's climate, yet there are alarming signs that some of these ecosystems are under severe stress and may be nearing collapse (Ke et al., 2024).

Although societies have made progress toward renewable energy adoption, and CO₂ emissions have plateaued in the last decade (Hausfather & Friedlingstein, 2024), these efforts began too late and remain insufficient to fully address climate change. Tackling this crisis requires a multidisciplinary approach that synthesises ideas from science, engineering, social sciences, and humanities, underpinned by the three pillars of sustainability (Middleton & Springer, 2011). Certain industries, particularly those that produce essential goods and services, face significant challenges in decarbonising (Casaban & Tsalaporta, 2022) and are expected to continue emitting GHGs in the coming years. The historical accumulative CO₂ emissions are around 2390 ± 240 GtCO₂ (Hausfather & Friedlingstein, 2024). Even if emissions were to cease tomorrow, it would take millennia for the planet to cool back to pre-industrial levels (Zickfeld et al., 2013). This is where Carbon Dioxide Removal (CDR) technologies, such as Direct Air Capture (DAC), become critical.

1.1.2 Direct Air Capture, a key mitigation technology

If the planet surpasses the 1.5°C threshold, the IPCC has stated the necessity of deploying CDRs solutions, specifically technological ones (IPCC, 2023). DAC is designed to directly remove excess of CO₂ from the atmosphere and store it safely, while traditional Carbon Capture focuses on capturing emissions from point sources from sectors that are struggling to decarbonise (Casaban & Tsalaporta, 2022). Net Zero scenarios project the need for an average of 30 DAC plants, each capturing 1 Mt/year to be scaled up between 2020 to 2050 (IEA, 2022b). Capturing CO₂ from the air is more expensive than from point sources. The CO₂ in the atmosphere is much more dilute than in as for example, a flue gas from a power station (IEA, 2022b). This contributes to DAC's higher energy needs and costs relative to Carbon Capture.

Currently, there are two main technological approaches for DAC: solid sorbents and liquid solvents. Solid DAC is based on adsorbents that operate at medium temperatures (Wiegner et al., 2022). In contrast, liquid DAC relies on aqueous basic solutions, such as potassium hydroxide, which captures CO₂ and then releases it by a series of high-temperature processes (between 300 °C and 900 °C) (Brethomé et al., 2018). Both approaches have their advantages and disadvantages. For example, a DAC plant based on solid sorbents do not requires water and its regeneration temperatures range between 80 °C to 100 °C, depending on the material utilised. However, the capture capacity of this approach is lower to an aqueous DAC plant (Brethomé et al., 2018). Despite this, solid sorbent DAC plants are more flexible in terms of being powered by low-carbon energy sources, making them an attractive solution (Terlouw et al., 2024). Technological improvements in materials and energy efficiency are being made, and they will reduce the overall cost of DAC, alongside policy interventions and financing (Holmes et al., 2024).

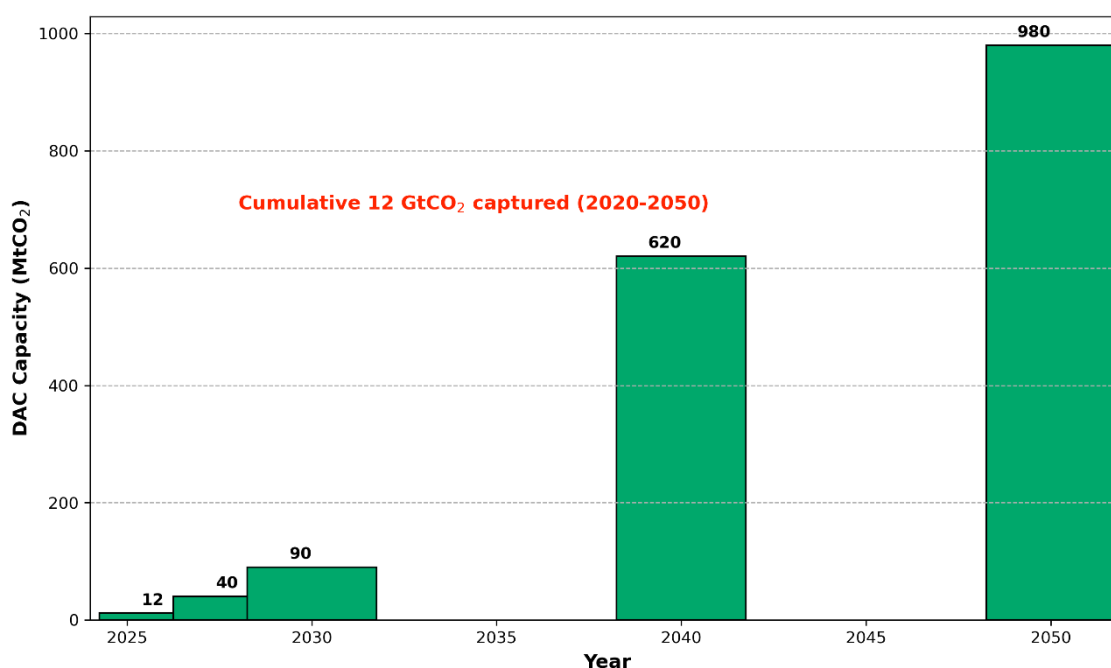


Figure 1.2. Projected evolution of global DAC capacity in 2030, 2040 and 2050. The bar chart illustrates estimated annual CO₂ removal capacity (in megatons of CO₂) based on technological scaling forecast. Source: (IEA, 2022b)

DAC plants are currently operating at small scale, but plans for growth are already in motion (IEA, 2022b). At the present, there are around 27 DAC facilities plants, being the largest one opened in 2024 in Iceland, capable of capturing 36,000 tons of CO₂ per

year (Climeworks, 2022b). Although the technology to reach 1GtCO₂/year by 2050 exists (IPCC, 2023), DAC remains too costly to be a viable large-scale investment at present. Policy interventions, financing, and public support will be crucial in driving these costs down (Webb et al., 2023). Several companies and countries are advocating for these solutions, with the United States currently leading the charge (Casaban et al., 2022).

1.1.3 Global progress and gaps in Direct Air Capture deployment

While DAC is not a new concept, it has lagged behind other technologies in development (Casaban et al., 2022). . Early research on CDR technologies began in the 1990s (K. Lackner et al., 1999), and since then, significant advances have been made in key approaches (Casaban & Tsalaporta, 2023b). Currently, there are approximately 130 DAC projects globally, but only 15 of these facilities are in advanced stages of development (IEA, 2022b). Some of the largest projects are under development in the United States, from which one is expected to be operating in mid-2025 with a capture capacity of 0.5 MtCO₂/year (1PointFive, 2024b). If all planned projects move forward and operate at full capacity, DAC deployment could reach around 3 MtCO₂ per year by 2030 - over 500 times the current capture rate, but still under 5% of the 80 MtCO₂ required to align with Net-Zero targets (Figure 1.2).

The United States has established several policies and programs to support DAC, including the 45Q tax credit (Carbon Capture Coalition, 2023). The 2022 Inflation Reduction Act expands and extends the 45Q tax credit with the basic amount of \$180/tCO₂ captured via DAC and permanently stored (U.S. Department of Energy, 2023a). The 2021 Infrastructure Investment and Jobs Act includes \$3.5 billion in funding for the development of 4 large-scale DAC hubs, each with the capacity to capture, sequester or utilise at least 1 MtCO₂/y, along with related transport and storage infrastructure (U.S. Department of Energy, 2022b, 2024a). In August 2023, the United States allocated \$1.2 billion to two large-scale DAC hubs (U.S. Department of Energy, 2023b). Companies like Climeworks, Heirloom and Batelle are leading Project Cypress in Louisiana, which aims to capture more than 1 MtCO₂/y and permanently store it (U.S. Department of Energy, 2024b). Similarly, the Canadian company Carbon Engineering is working on the South Texas DAC Hub, which aims to capture and sequester 30 MtCO₂/y, with a storage capacity of up to 3GtCO₂ (1PointFive, 2024a).

In addition, the United States Department of Energy launched a \$35 million initiative to advance technologies that reduce emissions from hard-to-abate sectors such as aviation and shipping, by removing CO₂ directly from the atmosphere (U.S. Department of Energy, 2023c). In September 2023, Canada released its carbon management strategy, which includes a focus on DAC (Natural Resources Canada, 2023). In terms of funding, the 60% of the investment will be for DAC from 2022 to 2040. However, remain vulnerable to political changes, as evidenced by potential shifts depending on election outcomes (Hiar, 2024). Companies must ensure that DAC is not misused as a means to justify continued emissions (Samala et al., 2023).

The European Commission has been supporting the development of CO₂ value chains through Carbon Capture projects and innovation programmes, with funding up to €3.5 billion. These projects focus on industrial decarbonisation, such as reducing emissions from the cement industry, biomethane production, or storage of CO₂ into depleted oil and gas fields (European Commission, 2024). Recently, the European Union has also enacted policies and projects to support DAC (Erbach, 2021). DAC projects are eligible under the “Horizon Europe” and Innovation Fund,” which together allocate around €100 billion for research and innovation across all sectors (IEA, 2022b).

In the United Kingdom, in October 2021 the government introduced a Net Zero Strategy which aims to achieve Net Zero Emissions by 2050 (Net Zero Strategy: Build Back Greener, 2021). It identifies the need for around 5 MtCO₂ of “engineered” removals, including DAC, by 2030, increasing to 80 MtCO₂ by 2050 (U.K. Government, 2021). Mission Zero, a UK-based company uses an electrochemical adsorption approach with the aim to reduce the energy consumption by 3-5 times compared with to other established DAC technologies, which involve heating a solid material to extremely high temperatures (Mission Zero, 2024). Moreover, the UK government is also providing up to £21.7 billion for CCS projects, including DAC ventures (Twidale, 2024).

Though DAC has faced criticism for potentially enabling continued fossil fuel dependence, it remains vital in addressing the imbalance of atmospheric CO₂ and limiting global warming (IPCC, 2023). Only a few countries have significantly funded and researched DAC technologies, with most investments occurring in the last decade. While the technology is still in its early stages, recent developments and substantial investments have accelerated its growth. Ireland is emerging as a player in the DAC industry, with companies like Neg8 and Air-In-Motion, coupled with the country's unique geological

advantages, positioning Ireland as a promising hub for this technology (Air in Motion, 2024; NEG8, 2024).

1.1.4 Ireland's potential in the Direct Air Capture industry

Ireland is making progress toward a sustainable future. In the past year, the country has removed peat from its energy mix (SEAI, 2023). Ireland has also set a new record for renewable electricity generation, primarily from wind energy (Wind Energy Ireland, 2023). However, the country's energy supply remains heavily dependent on fossil fuels, with 83% of its energy coming from fossil sources (SEAI, 2023). The overall share of renewable energy is still below 15%, whereas the target for 2030 is to achieve a 43% share (SEAI, 2023). Similarly, the gap in solar energy remains significant. By 2030, the country aims to have 8 GW of installed solar-PV capacity, but currently, only 1.2 GW is installed (Irish Solar Energy Association, 2024). This gap is expected to close by the end of the decade as the country plans to significantly boost solar and offshore wind generation (SEAI, 2024). However, sectors such as transport are expected to remain reliant on fossil.

In 2024, Ireland reduced energy-related emissions by 5% (SEAI, 2023). However, total GHG emissions have remained relatively constant, rising from 55 Mt_{CO2} in 1990 to 60 Mt_{CO2} 2021 (Environmental Protection Agency, 2024). CO₂ and methane (CH₄) emissions have been increasing, driven primarily by the transport and agriculture sectors. Energy industries accounted for 16.6% of total GHG emissions, and emissions from this sector have been steadily rising since 1990 (Environmental Protection Agency, 2024). This highlights the complexity of reducing GHG emissions and demonstrates that switching to renewable sources alone is insufficient. Nonetheless, Ireland's continuous efforts to transition to renewable energy are essential and critical not only for reducing its carbon footprint but also for supporting the future DAC industry, which should be powered by excess renewable energy to fill gaps in energy demand (Casaban & Tsalaporta, 2022).

Powering a DAC industry requires several factors, the most critical of which are energy sources and suitable locations. Ireland is uniquely positioned to contribute to DAC efforts. The British Isles are home to geological formations with a theoretical capacity to

store up to 90,000 MtCO₂, though further surveys are needed to assess the real potential (SEAI, 2022). The LCA conducted on DAC plants in Ireland highlights the country's unique potential for CO₂ storage, particularly in depleted gas fields such as Kinsale and Corrib. These geological formations offer significant storage capacity, estimated to be sufficient for large-scale DAC deployment. However, policies and geological studies are necessary to ensure that CO₂ is safely trapped and remains stored permanently (Finnigan et al., 2024). Reservoir simulations can help predict how CO₂ will behave once injected, including its migration patterns and potential pressure build-up (Jaiswal, 2024). In addition, laboratory tests should be performed to assess how the CO₂ interacts with the geological properties of the basin and ensure the safety of the storage process (Romanov et al., 2024).

The LCA results further emphasize that renewable energy - especially from wind power- will be crucial in reducing the energy intensity of DAC operations. Using Ireland's wind energy resources to power DAC plants could make the technology more economically viable and sustainable. However, for DAC to succeed in Ireland, several additional factors must be considered, including public acceptance, the policy framework, and the choice of capture technology (Casaban & Tsalaporta, 2023a). Ireland's humid climate presents a challenge, as humidity can affect the material performance of DAC plants based on solid sorbents such as zeolites.

1.1.5 Technological Approaches in Direct Air Capture

Adsorption is a separation process in which, depending on the nature of the solid sorbent, molecules from gases can be attracted and separated upon contact. Several techniques have been explored for DAC using this approach: Temperature Swing Adsorption (TSA), Vacuum Temperature Swing Adsorption (VTSA), Pressure Swing Adsorption (PSA), Vacuum Swing Adsorption (VSA), Humidity Swing Adsorption (HSA), and Electric Swing Adsorption (ESA).

TSA involves passing air through the material at lower temperatures, where the CO₂ binds to the adsorbent (Lee et al., 2023). Once the material is saturated, the system is heated, causing the material to release the attached CO₂. This process has several advantages because many materials have high adsorption capacity at ambient

temperatures, and the purity of the CO₂ is higher, as it does not require a purge gas and is released solely by heating (Zhang et al., 2022). However, TSA requires an additional energy input to heat the material, but this can be mitigated with a source of waste heat, solar power, or heat pumps (Casaban & Tsalaporta, 2023a; Ji et al., 2023). VTSA combines vacuum with heating to regenerate the material (Deschamps et al., 2022). While the combination of heat and vacuum enhance CO₂ capture capacity, it also increases capital costs due to the energy demands of the vacuum pump (Deutz & Bardow, 2021; Ji et al., 2023).

PSA operates by binding the CO₂ to the material at high pressures (Riboldi & Bolland, 2017). When the material becomes saturated, the CO₂ concentration at the outlet begins to rise, eventually matching the inlet concentration, indicating full saturation. PSA is considered energy efficient because it does not require thermal processes. VSA, a subset of PSA, evacuates the column by drastically reducing the gas pressure once the material is saturated with CO₂ (Zhang & Xu, 2023). However, VPSA has an energy limitation due to the use of a vacuum pump, which is energy-intensive (W. Liu et al., 2022). Some materials also require heat to desorb the attached CO₂.

HSA operates by adsorbing CO₂ under dry conditions and desorbing it by exposing the adsorbent to humid air (Xie et al., 2024). This mechanism works particularly well with adsorbents like amines, which change their capture capacity in relation to humidity levels (Wang et al., 2023). Moreover, HSA can operate with low energy inputs, making it well-suited for climates like Ireland. However, desorption can take up to 12 hours for small samples, and the efficiency can decrease significantly with changes in humidity, making it highly dependent on specific materials (Bouaboula et al., 2024).

ESA captures CO₂ via oxidation/reduction redox cycles. A solid quinone is incorporated into conductive carbon nanotubes as the working electrode, with ferrocene as the opposite electrode, and both are separated by an ionic liquid electrolyte (Bouaboula et al., 2024). CO₂ capture occurs when the quinone is negatively charged, and desorption happens when the voltage source is reversed, oxidizing the quinone electrode. The main advantage of this approach is its lower energy requirement and the ability to deliver CO₂ at 100% purity (Voskian & Hatton, 2019). However, large-scale implementation faces significant barriers due to the degradation of redox-active carriers, which are extremely sensitive to oxygen (Seo & Hatton, 2023). Other limitations of redox-active systems include the use of toxic and expensive electrolytes (Y. Liu et al., 2020).

1.2 Thesis aim & theoretical framework

The overarching aim of this thesis is to provide a comprehensive understanding of DAC technologies (from systems and materials perspective) by combining bibliometric analyses, environmental assessment, and experimental material testing. In particular, it seeks to evaluate the performance of solid sorbent materials for CO₂ capture in a lab scale PSA apparatus, while also situating this work within the broader research landscape and assessing the environmental impacts through a LCA.

- Chapter 2 provides a foundation overview of DAC and Carbon Capture technologies, exploring their relevance within the Irish climate context. It examines the specific challenges related to decarbonise the country from the hard-to-abate sectors, particularly agriculture, and discusses the potential role of DAC in achieving national climate goals. The chapter also introduces emerging opportunities for CO₂ utilisation, including its conversion to e-fuels.
- Chapters 3 and 4 aim to map the evolution and structure of scientific research in DAC. Chapter 3 presents a bibliometric analysis based on Scopus data, with particular focus on solid sorbents, identifying key publication trends, leading countries, and prolific authors. Furthermore, chapter 4 narrows the scope further by focusing on “CO₂ capture” and “DAC” research, offering a wider perspective on how this field has evolved over time. These analyses help position experimental and environmental work in the latter chapters within the global research context, highlighting knowledge gaps in materials testing.
- Chapter 5 compiles available information on this emerging industry, presenting a Life Cycle Assessment (LCA) study that evaluates the environmental impacts of implementing a DAC plant in Ireland under idealised conditions (specifically, using excess of renewable energy and available waste heat). The analysis includes transportation and storage of CO₂ in depleted gas fields (such as Kinsale), helping to assess the feasibility and sustainability of a DAC plant tailored to Ireland’s geography and energy profile.
- Chapter 6 explores the experimental evaluation of CO₂ adsorption materials using a Pressure Swing Adsorption (PSA) setup. Numerous solid sorbents, from the known Zeolite 13-X, to the novel Lewatit as well as amine-functionalised silica supports, are tested under “pushed” DAC conditions. The objective is to study the

characterisation of these materials and compare their adsorption capacities and stability across cycles, contributing empirical data to support material selection for future DAC systems.

In summary, the global commitment to limiting temperature rise to 1.5°C has been challenged by the slow pace of renewable energy adoption and rising emissions, with recent temperature records underscoring the urgency of addressing climate change. While renewable energy is crucial, it alone will not be enough to reverse the damage done to the atmosphere. CDRs technologies like DAC have become essential tools in the fight to restore atmospheric balance, despite their infancy and the challenges they face in terms of scale and efficiency.

Ireland's unique geological characteristics provide a promising opportunity to contribute to the development and deployment of DAC technology. However, significant research is still required to optimise CO₂ sorbents, adapt DAC systems to local conditions, and ensure safe and permanent storage of captured CO₂. The success of these technologies depends not only on scientific advancement but also on supportive policies and coordinated international efforts.

Together, these studies aim to provide actionable insights into the role of DAC in global climate strategies, contributing to the broader goal of reducing atmospheric CO₂ levels and mitigating the impacts of climate change.

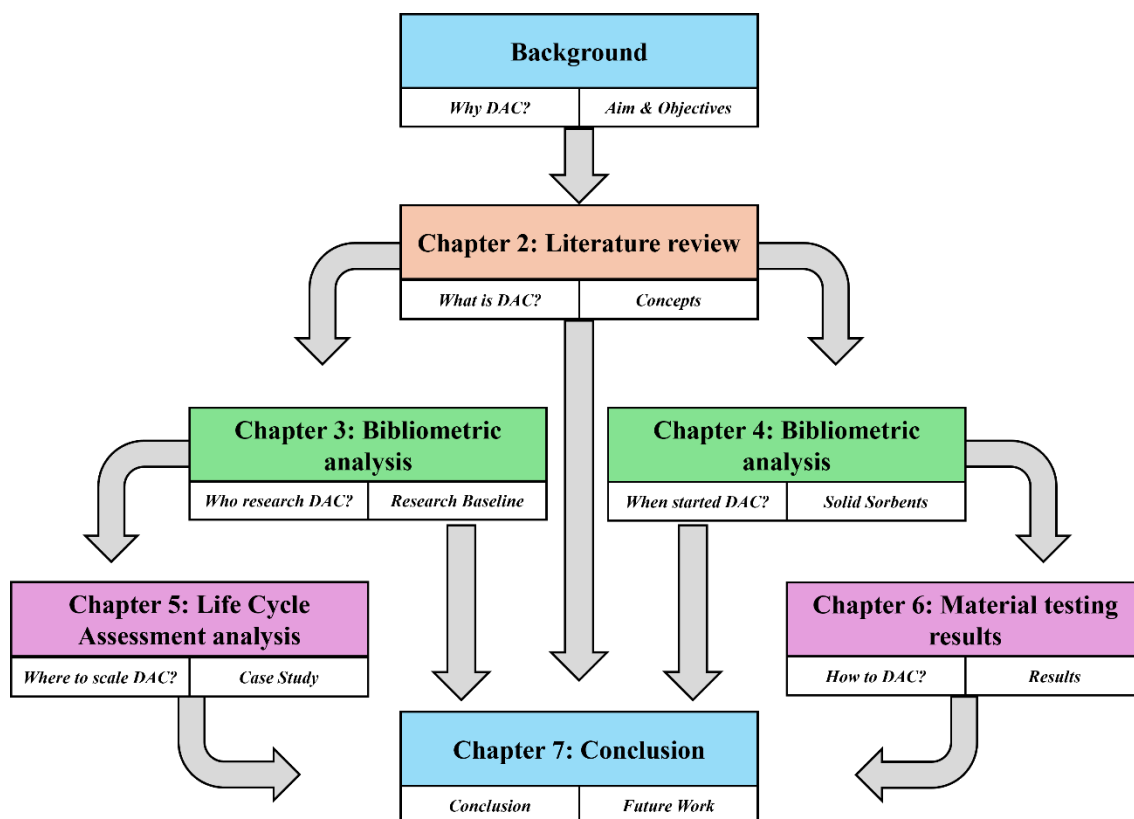


Figure 1.3. Flow diagram illustrating the structure and key research questions of this thesis. Each chapter addressed a fundamental question surrounding DAC, to progressively build a comprehensive understanding of the technology, its context in Ireland, as well as its technical and environmental evaluation.

1.3 Role & Collaborations

This thesis represents my own original research. I am the primary author of all the publications that comprise the chapters of this thesis. All aspects of the research have been reviewed by Dr. Elena Tsalaporta, my research supervisor. Each publication has been carefully reviewed by the co-authors, with particular emphasis on the final chapter.

Chapter 5 concludes the research presented in this thesis and is especially significant for its overall dissertation. Dr. Elena Tsalaporta contributed to the design of the DAC module. Additionally, Dr. John Hanrahan and Dr. Joe McGrath suggested key materials for testing, such as Silica 60 and Lewatit, as well as providing guidance on new approaches for amine-modified SBA-15. They also assisted in analysing the results and provided valuable feedback on the figures and structure of the chapter.

Chapter 2: Direct Air Capture of CO₂ in the Republic of Ireland. Is it necessary?



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Review article

Direct air capture of CO₂ in the Republic of Ireland. Is it necessary?

Daniel Casaban  , Elena Tsalaporta 

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2 Direct Air Capture of CO₂ in the Republic of Ireland. Is it necessary?

Abstract

Global temperature will surpass the threshold of 1.5 °C in the next decade. The implementation and the development of modern technologies, such as Direct Air Capture (DAC) will play a key role in mitigating climate change. The article exhibits the most updated information to emphasise the importance of expanding its commercialisation and social acceptance in the Republic of Ireland. This literature review provides background on the energy demand and the emissions related to the consumption across the country. The study highlights the methods to remove permanently the CO₂ from the atmosphere and its uses for sectors that find difficulties to decarbonise. The Republic of Ireland set an ambitious plan to finish its dependence on fossil fuels. Yet, its emissions will remain constant regardless of the measures taken. Thus, there is a strong reliance on fossil fuels from specific sectors that will keep polluting in the near future. Certain countries and business are funding projects to remove the CO₂ from the atmosphere. Meanwhile, the Republic of Ireland is putting aside these alternatives. The time scheduled to switch towards a free carbon economy is tight. Hence, could the Republic of Ireland meet its climate goals without DAC? Overall, we conclude that there are numerous gaps to fill only with renewables and ignoring alternatives such as DAC can be counterproductive. Therefore, the information provided aims to assess the current state of the technology, as well as show the benefits of investing in DAC. Future studies should focus on the viability of implementing DAC in the Republic of Ireland.

Keywords: *Direct Air Capture, Climate Change, E-fuels, Carbon Dioxide Removals; Republic of Ireland, Circular Economy*

Nomenclature

Abbreviations

AA	Acid acetic
BECCS	Bioenergy with Carbon Capture Storage
CDR	Carbon Dioxide Removals
DAC	Direct Air Capture
DMC	Dimethyl carbonate
DME	Dimethyl ether
e-fuels	Electrofuels
EU	European Union
FE	Formaldehyde
Flh	Full Load Hour
GHG	Greenhouse Gas
GtCO ₂	Billion tonnes of carbon dioxide
ICE	Internal Combustion Engine
IEA	International Energy Agency
IPCC	Intergovernmental Panel on Climate Change
MOF	Metal Organic Framework
MtCO _{2eq}	Million tonnes of carbon dioxide equivalent
NETs	Negative Emission Technologies
OECD	Organisation for Economic Co-operation and Development
ROI	Republic of Ireland
tCO ₂	Ton of CO ₂
UK	United Kingdom
USA	United States of America

Symbols

$\Delta H^{\circ}_{298\text{ K}}$	Enthalpy change at 298 K
GJ	Gigajoule
kWh	Kilowatt hour
MWh	Megawatt hour
TWh	Terawatt hour

2.1 Introduction

The excessive concentration of carbon dioxide (CO₂) in the atmosphere is reaching alarming levels (NASA Climate, 2020). International agencies such as the IPCC and IEA stated the necessity to implement Carbon Dioxide Removals (CDR) such as Direct Air Capture (DAC) at an industrial scale (IEA, 2021; Masson-Delmotte V. et al., 2021). Although the world is making progress to walk towards a low carbon dependency, the pace of the change is uncertain because of numerous aspects: geopolitical, extraction of materials, access to innovative technologies, and meet energy demands among others (Masson-Delmotte V. et al., 2021). In the meantime, how is the increase in atmospheric CO₂ concentration likely to change? If we cease emitting now and we switch to renewable energy, can the natural carbon sinks alone restore the balance?

Despite being necessary to stop pressure on these ecosystems, the short answer is no; it will take time because the half-life of the CO₂ molecule in the atmosphere ranges between 100 and 200 years (Archer et al., 2009). Thus, during the last two centuries, the concentration of CO₂ in the atmosphere has been dramatically increasing from 280 ppm to 418 ppm up till now (NASA Climate, 2020). According to the last reports from the IPCC, the current path is leading to an increase between 2.1 to 3.5 °C and, since today, the estimated concentration of CO₂ accumulated in the atmosphere is 2390 ± 240 GtCO₂ (Masson-Delmotte V. et al., 2021). So, how can we handle this problem?

There is not a unique solution. Switching to renewable energy sources in combination with a global effort and implementing Negative Emission Technologies (NETs) will be the turning points to reverse this unwanted situation (IPCC, 2018). The non-implementation of CDRs could end in an overshoot of 0.8 °C (Realmonte et al., 2019). This situation will not only aggravate irreversibly the global negative effects, but it is currently inflating the costs of the power systems (Fuhrman et al., 2021; Galán-Martín et al., 2021). Yet, the European Union (EU) and the government of the United States (USA) are "giving green light" to fund and develop DAC projects (Gates et al., 2022; Initiative & Earthshots, 2022). The Republic of Ireland (ROI) is already developing plans and fully relying on renewable energies; however, it does not have a mid-term program to reduce its emissions with a bridge technology (Government of Ireland, 2021).

The international assumption of the ROI -the "green emerald"-, is because of the local landscapes. Still, is the country as "green" as its image? The answer is no: the ROI was on the 11th out of 15 European countries with significant improvements in the social, education, peace, and justice areas. Regardless of the "green image" of the island, the ROI was at the lowest position in the environmental dimension -aside from Spain and Luxembourg- (Clark, C.M.A., Kavanagh, C., Lenihan, 2020). The ROI is one of the largest pollutants per capita, and in 2019, its emissions reached 14.2 metric tons per person (European Commission, 2025). Even with such rates, the ROI is developing efforts to cut carbon emissions by 2030: wind power will supply most of the energy (IWEA, 2021). Will this be enough to meet climate goals?

Unfortunately, there are sectors that have two key issues to reduce their emissions: there is a lack of time to adapt them to other energy options and are heavily reliant on fossil fuel (BP, 2020). As a result of its dependant nature, in 2050 between 40% to 60% of the final consumption in the transport sector will rely on oil, biofuels and gas (BP World Energy, 2019; IEA, 2020). Therefore, to support hard-to-decarbonise sectors such as the transport, it is necessary to consider alternative approaches. CDRs could provide it by not only producing negative emissions in favour of reducing CO₂ concentration, but it will also support these sectors that could experience difficulties in decarbonising (Osman et al., 2021). Thus, creating synthetic fuels will help sectors such as the aviation or the chemical industry (Marchese et al., 2021). Using e-fuels could store the excess of energy from renewable sources, such as the wind (Sherwin, 2021). In addition, implementing e-fuels could ease the energy demand of these sectors while simultaneously become carbon neutral (Becattini et al., 2021).

Although the country is increasing its wind capacity (IEA, 2020), this tendency does not exist in the CDR area (Government of Ireland, 2021). The strong focus on the construction of wind farms is overshadowing other solutions that could help these clean energy sources (Howley et al., 2020). The ROI could take advantage of the excess of future wind energy generated and create e-fuels or store the CO₂ (Bos et al., 2020). As mentioned before, not considering these alternatives in mind will lead to new global emissions - 40 GtCO₂ per year-, from which some of them will remain in the atmosphere for at least 100 or 200 years (Archer et al., 2009). Hence, there is an urgency to deploy negative emission such as DAC in favour of reducing the global concentration of CO₂ and to meet the energy demands (Hanna et al., 2021).

Through the above statements, there are noticeable conveniences with implementing DAC. Today, non-renewable energies -such as coal, oil, and natural gas- account for 84% of the global energy consumption (BP, 2020). In the case of the ROI, the transport, residential and heat sectors highly depend on fossil fuels (SEAI, 2020a). Delaying further action in the DAC path will not only have implications for the national energy systems, but it will also miss the environmental benefits of its implementation (Galán-Martín et al., 2021).

This study will bring insights into the local energy reliance on fossil fuels and its consequences. It will also provide knowledge about the companies taking the lead and the benefits of implementing a DAC industry. Other researchers and local authorities studied the problematic situation related to CO₂ emissions. However, there are no studies that included CDRs as part of the solution. Hence, the purpose and motivation of this article is to address the importance of DAC in the ROI. The country is deploying diverse plans to mitigate climate change, yet CDRs did not receive the proper attention. Therefore, the study will help to understand how DAC could fit to reduce this dependence on fossil fuels and its environmental benefits.

2.2 Energy security in the ROI

To understand the increment of GHGs emissions, first it is important to underline the origin of these anthropogenic emissions. The average energy consumption per citizen in the ROI is 70 MWh (Brophy et al., 2021). The ROI had the lowest value in energy efficiency between the OECD state members (H. Sun et al., 2019). In 2018, the energy demand in the ROI was 143.05 TWh, from which oil accounted for 57%, electricity 19%, natural gas 16%, and the remaining 3.8% to renewables. Household, industry, services, agriculture & fisheries, and transport were the final demand areas (SEAI, 2020b). In relation to the transport sector, private cars dominated the final energy use (SEAI, 2020a).

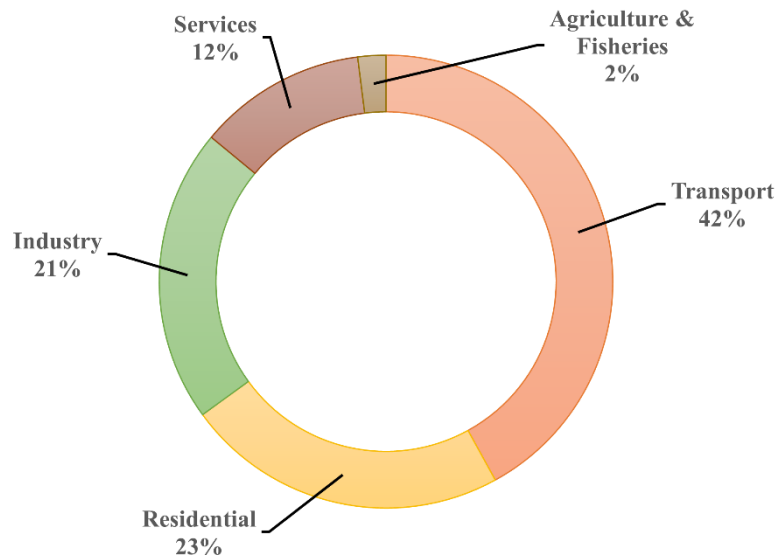


Figure 2.1 The ROI's energy demand consumed by sectors in 2019. Transport sector (in orange) dominated the demand of energy in 2019, whereas the residential (in yellow) and the industry (in green) accounted the other half.

In 2019, Ireland's natural gas supply amounted to about 52.10 TWh, from which indigenous output provided roughly 25 TWh, meeting 47% of total supply. In the last decade, the ROI's gas consumption from the power sector was 28.62 TWh. The local gas fields from Corrib field in Co. Mayo and Kinsale field in Co. Cork helped to reduce the use of most carbon-intense fossil fuels. Kinsale's exploit started in the late 70s, but its production has been in downturn since 1995 (Kinsale Energy, 2020). In the same period, the construction of two interconnectors in 1993 and in 2002 connected the ROI to the UK (Gaslink, 2011). In 2015, the production from Kinsale only accounted for 3% of the national demand (Kinsale Energy, 2020). Subsequently, the commercialization of the Corrib field in the same year allowed the ROI to displace enormous quantities of importing gas (Gas Networks Ireland, 2017). However, the Corrib field production decreased after it peaked in 2018 (Thatcher et al., 2021). The projections estimated that after 2022, the Corrib field will only meet 27% of the ROI demands, and the remaining percentage will come from the UK (Gas Networks Ireland, 2021). The Brexit situation and gas dependence on the UK could have a negative effect on the ROI (Abdul-Salam, 2019) and as a result of this, the gas imports from the UK will increase throughout the next decades (Government of Ireland, 2019). However, this last statement might not be true if the government approves new licenses to explore and exploit new local gas fields.

In contrast with gas, the ROI is a minor consumer of oil contrary to other members of the EU: in 2021, the oil imports' share of the country was inferior to 5% (European Commission, 2022b). Yet, the ROI has a high dependency in oil as a primary energy in the transport sector (Dineen et al., 2021). The ROI imports all its demand as crude oil (36%) and refined products (64%) (SEAI, 2020b). The country's refinery in Whitegate Co. Cork can process 75,000 barrels per day, which is the equivalent to 40% of ROI's inland oil demand (SEAI, 2020b). Although there was a reduction of the energy used in this sector due to the pandemic, the consumption rebounded to pre-covid levels in the 2021 (Dineen et al., 2021). Governmental bodies expect a rapid transition towards electric vehicles. Despite the uncertainty of the path to transition, this action could decrease the dependency in oil. However, the problem will transfer to the electrical grid, which is already under pressure.

As opposed to the oil consumption, the pandemic did not affect the electricity consumption (Dineen et al., 2021). Although there was energy reduction related to the transport sector, the energy use from the residential sector skyrocketed (Dineen et al., 2021). In consequence, the gross electricity use increased 81% since 1998 (Sen et al., 2021). In 2020, the electricity demand was 7.36% higher than the 2008 peak (Dineen et al., 2021). Services accounted for 45% of the demand, followed by the residential and industry sectors with a share of 31% and 21% respectively (Dineen et al., 2021). Before the pandemic, the electricity demand increased in all sectors except in the industry (SEAI, 2020b). This electricity consumption forecast expects a steady growth in the current decade (SEAI, 2020b). Hence, the immense electricity demand and the increment of the gas prices over the coming years are leading to energy supply problems (Riegel, 2021).

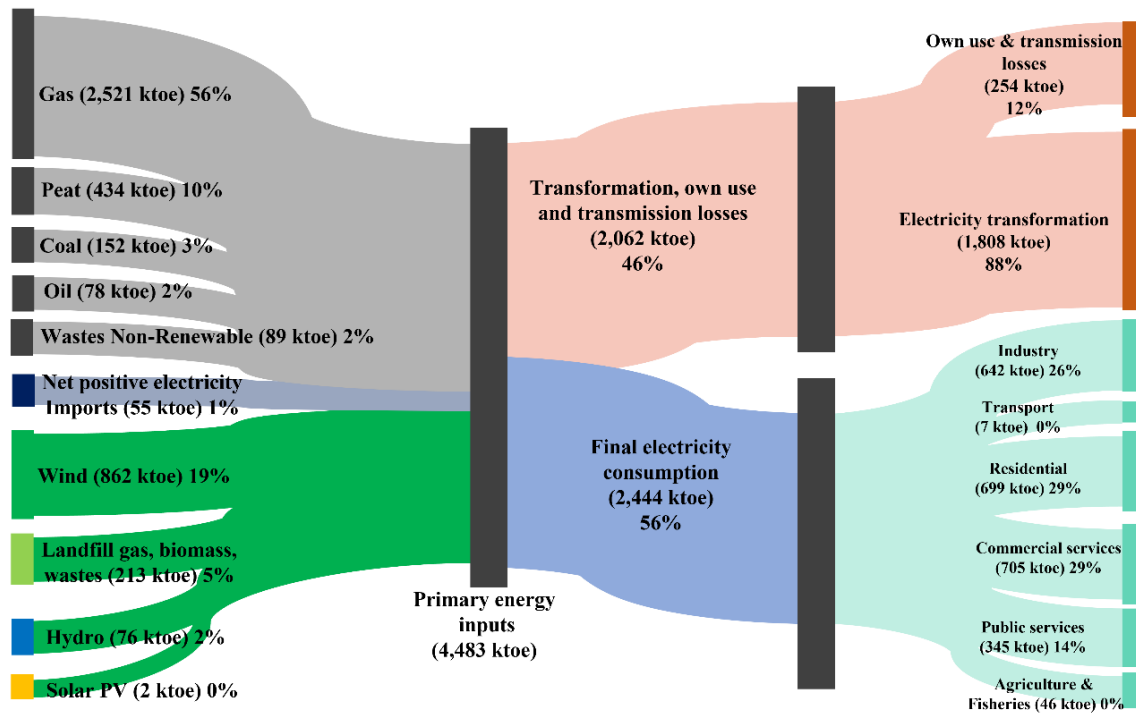


Figure 2.2. The ROI's flux of energy in electricity generation during 2019. Most of the electricity generated came from non-renewable sources, where gas accounted 56% of the energy inputs. Wind generation represented 20% of the primary energy but is incrementing its capacity each year. Data source: SEAI (SEAI, 2020b)

2.3 The historic carbon emissions of the ROI

In pre-pandemic years, the energy related emissions in the ROI accounted for 59% of the total GHG's, in which transport was the most pollutant sector -Figure 2.3- (Sustainable Energy Authority of Ireland, 2018). As previously mentioned, there is a high dependency between the transport sector and oil (Dineen et al., 2021). Before 2020, oil accounted for more than half of the total GHG's emissions (Sustainable Energy Authority of Ireland, 2018). Because of the lockdown in 2020, the GHGs emissions fell due to the travel restrictions (Dineen et al., 2021).

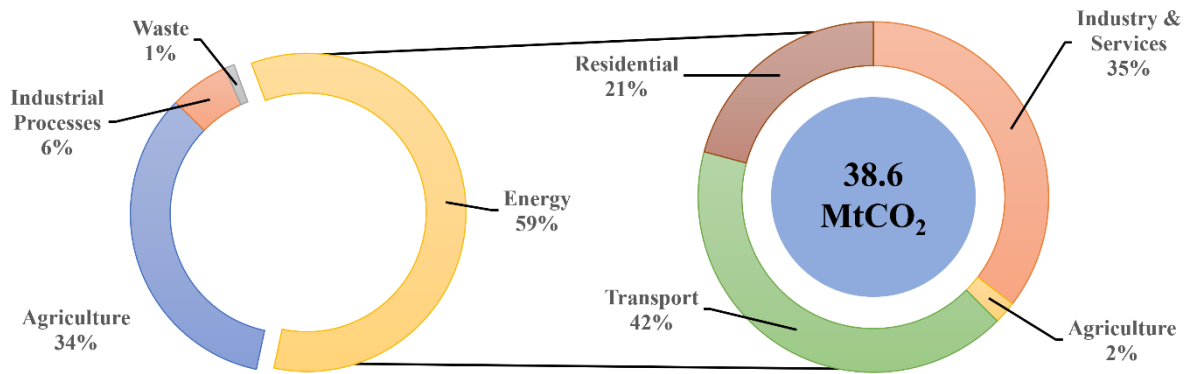


Figure 2.3. In 2018 in the ROI, the agriculture sector accounted for 34% of the total emissions, yet the energy sector encompasses 59% of the total share. Within the energy sector, transport accounted for roughly 42%, highlighting the challenge of decarbonising this sector.

The ROI is not an isolated country, yet since 1990 its local GHGs emissions per year ranged between 50 MtCO_{2eq} and 70 MtCO_{2eq} (Greenhouse & Emissions, 2018). The local natural carbon sinks can absorb 44.73 MtCO_{2eq} (Norton et al., 2018). At the end of 2019, the historical accumulation of GHGs in the country was about 510.56 MtCO_{2eq} (Greenhouse & Emissions, 2018). Figure 2.4 exhibits the accumulated historic emissions from the different sectors in the ROI (Greenhouse & Emissions, 2018), and the emissions predicted for the coming years (Environmental Protection Agency (Ireland), 2020). If the uptake capacity of the carbon sinks will not change (Norton et al., 2018), the accumulated emissions of the ROI emitted will range from 850 to 600 MtCO₂ by 2040 (Environmental Protection Agency (Ireland), 2020).

However, the continuous discharge of pollutants is not only a local issue and the total GHGs emissions rebounded to pre-pandemic levels (Jackson et al., 2022). Global natural sinks can capture 5 GtCO₂ per year (Houghton, 2003; Keenan & Williams, 2018; Le Quéré, 2010; Le Quéré et al., 2018; Manning & Keeling, 2006), yet recent studies suggest that nature is capable to uptake the double amount. Annually, the amount of carbon captured by forest ecosystems is 7.6 GtCO₂ (Harris et al., 2021). In contrast, oceans can remove 3.5 GtCO₂ per year (Watson et al., 2020).

Other study mentioned that between 1994 and 2007 the ocean carbon sinks increased its capacity to absorb CO₂ (Gruber et al., 2019). However, human activity emitted 407 GtCO_{2eq} throughout this period. The 45.45% of these emissions -185 GtCO₂- are in the atmosphere. The biosphere could capture 114.7 GtCO₂ (28.18%), and the oceans

assimilated 107 Gt_{CO₂} (26.36%). This indicates that the biosphere and oceans captured 8.82 Gt_{CO₂eq} and 8.23 Gt_{CO₂eq} per year, respectively.

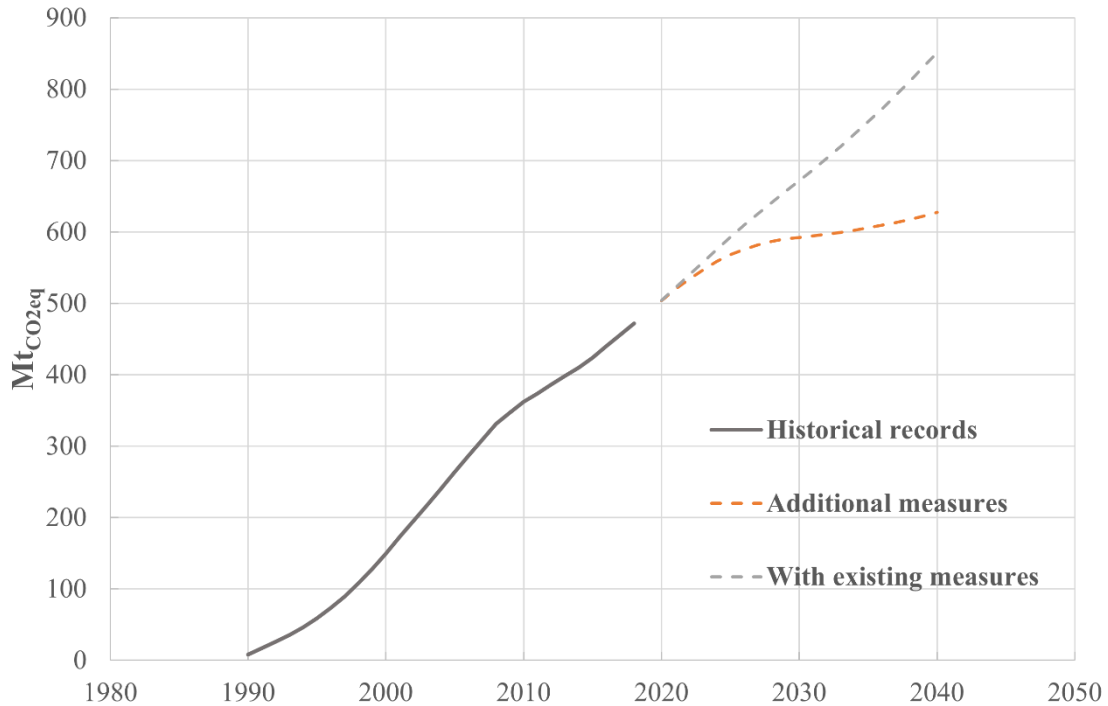


Figure 2.4. Historical GHGs accumulation and future projections without including a DAC plant according to the EPA. Source: ROI's Final Greenhouse Gas emissions 1990-2018 (63) & Greenhouse gas emissions projections (Environmental Protection Agency (Ireland), 2020)

Although the increment in the ocean's capacity to uptake CO₂ increased, there are downsides: one of the main issues is the acidification and the saturation in the upper layers of the ocean (Findlay & Turley, 2021). Still, the continuous emissions will affect the capacity for absorbing more CO₂ (UNESCO, 2021). Even if the global community ceases discharging pollutants into the atmosphere, the temperature of the planet would remain constant because of the cumulative emissions (Randers & Goluke, 2020). Nevertheless, it is possible to reverse the acidification of the oceans and to break the continuous increment of the temperature with the use of CDRs and carbon capture solutions. Although CDRs cannot overturn other negative consequences, such as the Sea level rise, this technology is highly necessary to achieve net zero CO₂ (Masson-Delmotte V. et al., 2021).

2.4 The state-of-the art in DAC

In the ROI there are already commercial companies exploring the field of NETs, which is a big step in contrast with other nations. However, it is quite contradicting that the local government ignored the necessity to develop this technology (Government of Ireland, 2021) when international agencies such as the IPCC and IEA remarked its importance (IEA, 2021; Masson-Delmotte V. et al., 2021).

Trinity Green Energies is a local company located in Dublin. In a personal communication, a team member shared their goals, expectations and future scope. The company goal is to reduce the CO₂ content in the atmosphere to sustainable levels between 2040 and 2050. They aimed to uptake 10 MtCO₂ per year by using non-toxic materials with an exceptional affinity to CO₂ and at a low operational cost (Neg8, 2021). Another of their top priorities is to control further CO₂ emissions in balance with natural carbon sinks. To achieve it, the company expects to apply their technology in cooperation with social, political, and industrial stakeholders, as well as the broader public. Although they expect to capture 230 tonnes by 2030, the company in collaboration with global partners, has laid out an ambitious roadmap to uptake 1 GtCO₂ annually between 2040 and 2050 (Neg8, 2021). The future scope of the company is in tune with the present study. In 2040, it will be necessary to uptake 8 GtCO₂ per year. Trinity Green Energies states that ongoing policies and methods such as transport electrification or renewable power will not solve the problem. Only CDRs can remove permanently the CO₂ from the atmosphere and bury it in subterranean strata or utilise as chemical feedstock.

Another local company in the DAC market is 'Carbon Collect' (Carbon Collect, 2022). Their aim is to accelerate the implementation of CDRs in the current decade. To achieve it, the company expects to plummet the cost to \$100/tCO₂. The company uses what they called "mechanical trees." These machines differ vastly from other DAC processes. The apparatus consists in a 10-meter-tall mechanical structures filled with sorbents materials. The material adsorbs the CO₂ carried by the wind at ambient conditions. Thus, the structure does not require energy to capture the carbon. It is only necessary a small amount of energy to desorb the CO₂ in each cycle. The scale up of this technology exhibits a significant advantage in contrast to other DAC systems. Yet, the land use is the major disadvantage of this technical benefit: according to the company for a 4 MtCO₂ per year DAC farm, it is necessary 120,000 units. At the global level, there are three companies leading now: Carbon Engineering, Climeworks and Global Thermostat.

Carbon Engineering is the first big company in DAC and its creation dates to 2009 (Carbon Engineering, 2019). The goal of the company is to prove the feasibility of a megaton-scale DAC plant. Thus, Carbon Engineering expects to capture 1 Mt_{CO2} from the atmosphere per year. This company uses KOH as a solvent to uptake the CO₂ from the atmosphere, combined with a caustic calcium regeneration process (Keith et al., 2018). In 2015, they built the first pilot-scale plant in Canada. They stated that to capture one tone of CO₂; it is necessary 366 kWh of electricity with 5.66 GJ of Natural Gas (Carbon Engineering, 2019; Keith et al., 2018).

Climeworks is a Swiss company which runs DAC modules with solid sorbents to capture the CO₂ (Keith et al., 2018). As well as most of the companies, their aim is to reduce the global concentration of CO₂ in the air. In 2017, they opened the first commercial enterprise in Hinwil with the aim of capturing 900 tonnes of CO₂ per year (Evans, Simon, 2017). The electricity demand in Hinwil is almost 300 kWh per tonne of CO₂, combined with a heat demand of 2000 kWh (Beuttler et al., 2019; Evans, Simon, 2017; Keith et al., 2018). In October 2020, they stored the CO₂ captured underground in Iceland (Beuttler et al., 2019). Recently, Climeworks opened its first DAC scale plant called ORCA (Climeworks, 2022a). In 2025, they set an aim to capture 225Mt_{CO2}, which is the 1% of the global emissions and partly the equivalent to the historical emissions of the ROI (Beuttler et al., 2019). In its ambitious plan to achieve a megaton capacity by 2030, the company is constructing its 18th project; 'Mammoth' (Climeworks, 2022c). When fully operational, this new installation expects to capture 36,000 tons of CO₂ per year.

In 2010, a company founded in the USA called Global Thermostat started working with CDRs. The company has two pilot plants with a capacity to uptake 40000 tonnes of CO₂ per year (Zero Carbon Systems, 2021). As Climeworks, Global Thermostat uses amine-based sorbents to enhance the CO₂ capture (Zero Carbon Systems, 2021). The aim of the company is to develop a low-cost solution in the DAC field. Global Thermostat reported an electricity and heat cost lower compared to Climeworks, which range between 150-260 and 1170-1410 kWh per tonne, respectively (Ping, E., Sakwa-Novak, M., Eisenberger, 2018).

Despite these three companies hold a decade of research, there are multiple new companies appearing at this moment that will compete with them. The majority use novel and interesting techniques to enhance the CO₂ capture. All of them share the same goal

and formed a coalition (Direct Air Capture Coalition, 2021). Table 2.1 summarizes the methods used by these companies as well as their goals and future projects.

'Carbon Capture' is a company from the USA that uses the same procedure as Climeworks or Global Thermostat (Carbon Capture, 2022). The company is studying the use of zeolites and other known materials in molecular sieves to capture the CO₂. Recently, they signed a partnership with a solar company called Heliogen to speed and scale up DAC. In the same way, 'Sustaera' is using an alkali metal to capture the carbon through an adsorption process (Sustaera, 2022). Sustaera's ambition is to remove 0.5Gt_{CO2} by 2040. In the short term, the company expects to commercialise modules with a capacity of 1t_{CO2}/day. Powered by renewable energy, the company aims to scale a pilot plant by 2023 with an adsorption capacity of 10 t_{CO2}/day.

'Verdorex' uses a novel method called electro-swing adsorption (Nancy W. Stauffer, 2020). The company is at an early stage and therefore they limit information about their goals and expectations. However, they aim to develop a leading technology for DAC and low concentration point sources. The innovative technique uses quinones as solid sorbent that connect to a ferrocene. Then, by applying voltage, the electrons move from the ferrocene to the quinone through a wire connection. During this step, the negative charge of the quinone can adsorb the CO₂. Once the solid reaches the saturation step, the voltage's direction goes backwards and the electrons flow from the quinone to the ferrocene. Because of the positive nature of the quinone, the material loses all the CO₂ affinity. Then, the use of a purge gas helps to desorb the CO₂.

Other companies prefer to use solvent methods because of its facility to scale a DAC plant. 'Noya' uses a chemical blend of water to enhance the absorption of CO₂ in cooling towers (Noya, 2022). The start-up aims to revolutionize the concept of DAC and accelerate the transition to a sustainable future. To achieve this goal, Noya plans to turn 2 million more of cooling towers at industrial sites across USA into DAC modules. This novel method will enhance the implementation of DAC and, therefore, reduce its costs.

Furthermore, 'Mission Zero' is developing a new solvent compound that requires four times less energy than potassium hydroxide (Mission Zero, 2022). If they achieve this milestone, the company will reduce the energy costs, allowing other competitors to benefit from their research (Mission Zero, 2022). To prove it, Mission Zero expects to open a scale pilot plant in the current year. Yet, in 2023, they aim to commercialise a plant and to uptake 1kt_{CO2} per year.

'Carbon Infinity' study the use of promising materials such as Metal Organic Frameworks (MOFs) while are developing apparatus to perform the adsorption of CO₂ from the atmosphere (Carbon Infinity, 2022). Even if solid sorbent systems have disadvantages to scale-up in contrast with absorption ones, various companies bet on adsorption solutions due to low operational costs. Thus, solid sorbent DAC industry is almost new, and will expand its influence over the next 10 years. However, there are alternative methods that do not require the use of solid sorbents or solvents for DAC.

'Heirloom Carbon' uses a calciner to remove the CO₂ from the atmosphere (Heirloom Carbon, 2022). Their goal is more ambitious than the rest of the companies. In 2035, they expect to capture 1GtCO₂. Thus, from 2022, they plan to capture 1 ton of CO₂ per day. The technique comprises the exposition of stacks of MgO or CaO to ambient air through a passive contactor. These stacks capture the CO₂ by a carbonation process that takes two weeks. Then, the MgCO₃ or CaCO₃ formed goes to a calciner where the products of the process are CO₂ and MgO or CaO. The last step consists in the recycling of MgO or CaO for another use, while the CO₂ is permanently stored or used as raw material for other products.

All the companies aim to tackle climate change by capturing the excess of CO₂ from the atmosphere. As stated above, the DAC industry is in its infancy and the goals and expectations will depend on its social acceptance. However, removing the excess of CO₂ is as important as switching to renewable energy sources (IEA, 2021; Masson-Delmotte V. et al., 2021). If governments do not give the proper attention to this issue and accelerate its implementation, the problem will remain. The ROI is fortunate to have two companies, yet the government did not consider CDRs as a priority in its National Development Plan (Government of Ireland, 2021).

2.5 Storage and utilisation of CO₂

The primary goal and environmental benefit of DAC is to remove and store permanently the CO₂ from the atmosphere. There are two principal forms to bury the CO₂ depending on the geographical location: geological or oceanic storage (Metz & O. Davidson, H. C. de Coninck, M. Loos, 2005).

In both forms, the injection of CO₂ in bedrocks that currently keeps or previously held fluids such as natural gas or oil (Mishra et al., 2021). Once in the bedrock, the

injected CO₂ switches to liquid phase due to the ambient pressure and temperature (Jain, 2021). In this state, the CO₂ density range between 50-80% of the water density, similar to various hydrocarbons (Mishra et al., 2021). The density ensues driving forces to push the CO₂ and moving it upwards (M. Chen et al., 2021). Hence, the importance of selecting a well-sealed rock is vital to keep the CO₂ store in the depleted gas field (Jain, 2021). After incorporating the CO₂ into the bedrock, the CO₂ fills the pores and displacing other gases (Dai et al., 2017). The geochemical trap occurs once the CO₂ dissolves in water and reacts with the fluids from the bedrock (Hamza et al., 2021). Then, the increment in the density of the CO₂-water blend sinks the CO₂ at the bottom of the bedrock rather than moving upwards (Hamza et al., 2021). Later, the dissolved CO₂ and minerals create ionic species, transforming the captured CO₂ into solid carbonates and traps it for millions of years (Metz & O. Davidson, H. C. de Coninck, M. Loos, 2005). But as with every process operation, there are risks that are important to highlight.

Table 2.1 Overview of key DAC companies, summarising their capture methods, technological approaches, current scale, stated goals, future expectations and geographical locations.

Company	Technology	Method	Scale	Goals	Expectations	Future scope	Country	Reference
Carbon Capture	Adsorption	Use of zeolites and other solid sorbent materials in molecular sieves	Pilot	To reverse climate change at low-cost using renewable energy	-	Partnership with Heliogen to speed and scale up a DAC plant	USA	(Carbon Capture, 2022)
Carbon Collect	Passive adsorption	The wind carries the CO ₂ that enters in contact with the sorbent tiles from the mechanical tree	Pilot	To speed up the global climate effort and contribute to reverse CO ₂ emissions over the next decades	To reduce the cost to \$100/t _{CO2} captured at the end of this decade	Carbon Collect aims to capture 3.8 Mt _{CO2} /yr by scaling the technology in Arizona	ROI	(Carbon Collect, 2022)
Carbon Engineering	Absorption	CO ₂ separated directly from the atmosphere by using a potassium hydroxide solution	Pilot	To demonstrate the feasibility of a megaton-scale DAC plant	To capture 1Mt _{CO2} /yr with their first-large scale plant	Construction of a scale plant with a capacity of 1Mt _{CO2} /yr	Canada	(Carbon Engineering, 2019; Keith et al., 2018)
Carbon Infinity	Adsorption	Carbon infinity studies the use of MOFs to reduce the cost-effective of DAC	-	To remove the CO ₂ from the atmosphere and provide a green CO ₂ feedstock for the sectors with difficulties to decarbonise	To commercialise DAC modules by the end of 2023	The company is engineering an apparatus to facilitate the implementation of DAC in the market	UK	(Carbon Infinity, 2022)
Climeworks	Adsorption	CO ₂ separated directly from the atmosphere by using solid sorbents	Large	To develop a climate-positive world by removing the CO ₂ from the atmosphere	To build a multi-megaton capacity by 2030 and gigaton capacity by 2050	Climeworks has set an ambitious target of removing 225 Mt _{CO2} /yr in the near future	Swiss	(Beuttler et al., 2019; Evans, Simon, 2017; Keith et al., 2018)
Global thermostat	Adsorption	CO ₂ separated directly from the atmosphere by using solid sorbents	Pilot	To develop a lower cost solution to tackle climate change through DAC	-	Global thermostat aims to implement scale plants with a capacity of 4000 Mt _{CO2} /yr	USA	(Zero Carbon Systems, 2021)
Heirloom Carbon	Calciner	Loop cycles of carbonation and decarbonation of MgO or CaO to separate the CO ₂	Bench	To pull billions of tons of CO ₂ from the atmosphere to enhance the energy transition	To remove 1Gt _{CO2} from the air by 2035 with the most efficient DAC system	Heirloom plans to capture a ton of CO ₂ per day in 2022	USA	(Heirloom Carbon, 2022)
Mission Zero	Electrochemical separation	Use of a water-soluble solvent that require 4 times less energy than current hydroxide base solvents	Bench	To develop a low-cost DAC system and enhance its industrial scale	2022: An industrial scale pilot to remove 365 t _{CO2} /yr	In 2022, the company aims to scale an industrial pilot plant to remove 365 t _{CO2} /yr. In 2023, the company will improve its capacity to 1kt _{CO2} /yr	UK	(Mission Zero, 2022)
Noya	Absorption	Chemical blend added to the water content in a cooling tower	Pilot	To accelerate the transition of the industry towards a sustainable future	To scale their technology in 2 million of cooling towers to pull around 8.5Gt _{CO2} /yr	The company aims to minimize the cost per tonne by maximizing the CO ₂ production	USA	(Noya, 2022)
Sustaera	Adsorption	Use of an alkali metal based solid to enhance the capture	Pilot	To remove 0.5Gt _{CO2} by 2040	To commercialise DAC systems starting at 10t _{CO2} /day	In 2023, they plan to establish partnerships to scale a 10 t _{CO2} /day plant	USA	(Sustaera, 2022)
Trinity Green Energies	Adsorption	Use of porous materials with great affinity to CO ₂	Bench	To become the leading inventor of carbon capture systems powered by renewable sources	To deploy 25,000 CO ₂ removal systems in the ROI and capture 10Mt _{CO2} /yr in 10 years	Between 2040 and 2050, the company plans to collaborate in the capture of 1 Gt _{CO2} /yr with global partners	ROI	(Neg8, 2021)
Verdorex	Electro-swing adsorption	Use of quinones connected to a ferrocene	Bench	To remove the CO ₂ from industrial sources and air	To reduce a 70% the cost from the present DAC systems	The company expects demonstrate the feasibility of their method by scaling-up a pilot plant in the next years	USA	(Nancy W. Stauffer, 2020)

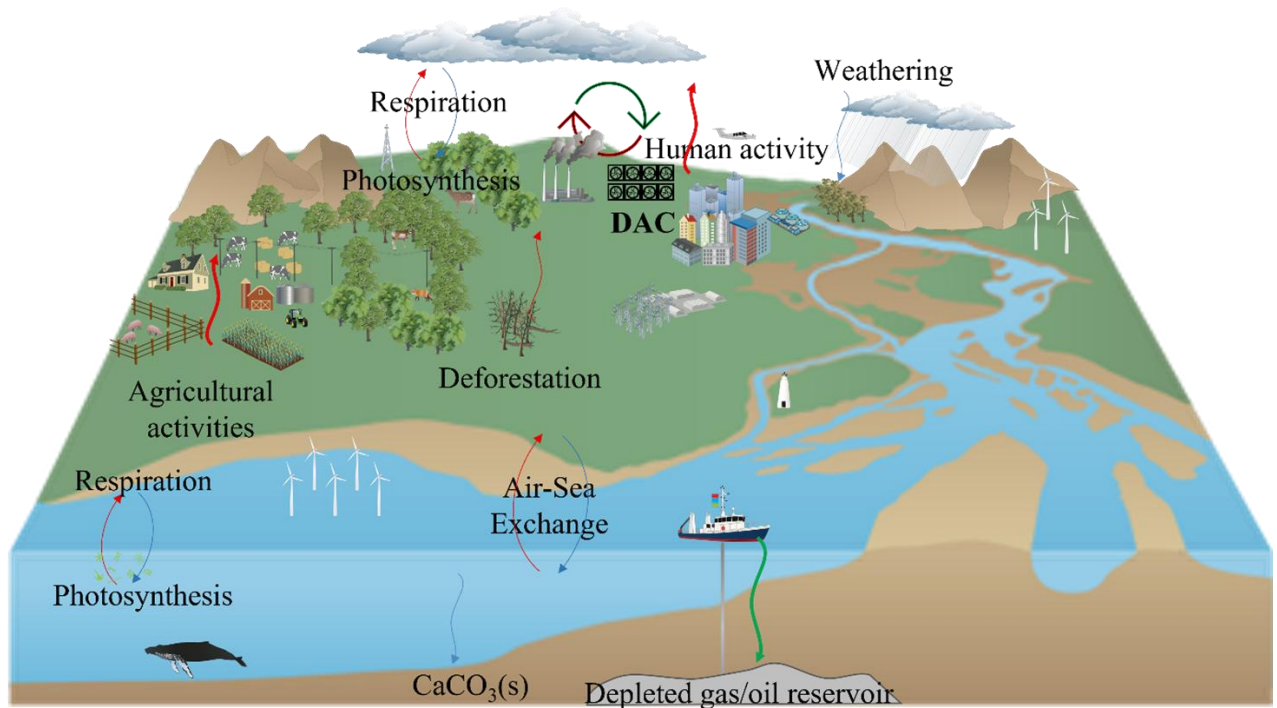


Figure 2.5. Simplified carbon cycle. Red arrows represent the emissions of CO₂ into the atmosphere, whereas blue arrows illustrate the CO₂ uptake by the natural sinks. The green arrow shows an anthropogenic CO₂ uptake and storage.

Researchers studied the risk associated with atmospheric CO₂ sink in the past. If there is a geological risk in the reservoir structure, it could lead to leakages of CO₂ (Tiwari et al., 2021). The first step to store CO₂ is to reduce the possibility of damage to the reservoir structure during the drilling operations (Kassem et al., 2021). Diminishing the pressure in the reservoir and at the bottom hole of the injector will reduce the potential risk of CO₂ leak (S. Li & Zhiwei, 2021). Hence, there are pressure thresholds that are important not to surpass to avoid leakages (Kassem et al., 2021).

Another problem already studied is the formation of a "CO₂ shadow." A liquid solvent carbon study stipulated a capture area about the size of all the ROI, with a capture velocity of 1 cm/s in 5 separate places: Nevada, the Gobi Desert, Equatorial Pacific Ocean, Antarctica and Northern and Southern Pacific Ocean (Johnston et al., 2003). They demonstrated an uptake of 3 GtCO₂ per year (Johnston et al., 2003). The results showed that the environmental risks are like seasonal and diurnal CO₂ fluctuations (Johnston et al., 2003). Aside from the environmental benefits, there are economic gains in developing a DAC industry.

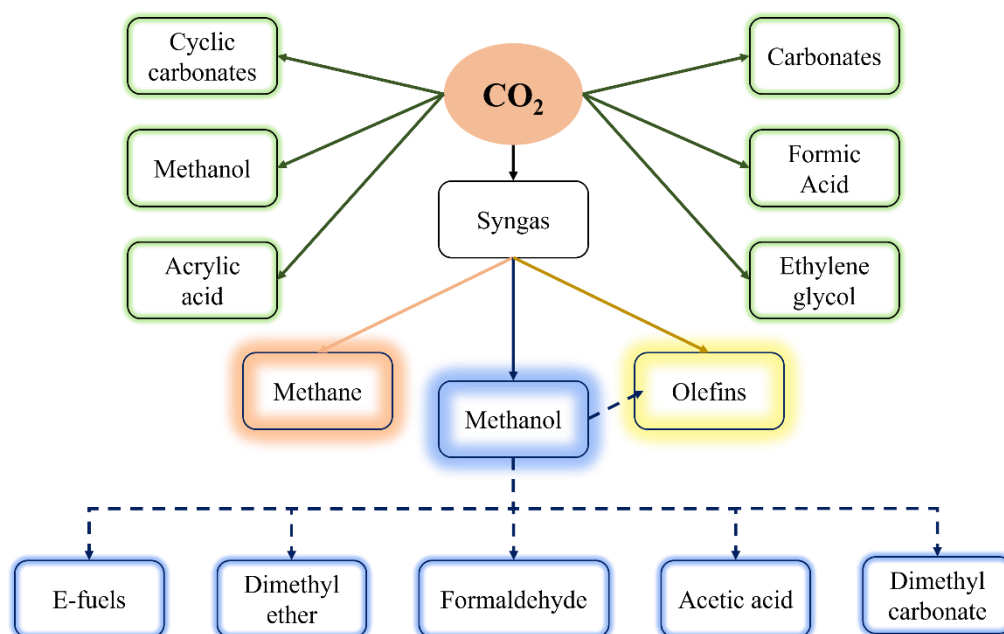


Figure 2.6. Overview of a potential range of products derived from CO₂ as raw material, including fuels, chemicals, and materials. These pathways illustrate the versatility of CO₂ as a feedstock for valued-added products and the integration opportunities for DAC and carbon capture technologies.

Suggested by authors in the scientific literature, Figure 2.6 shows some novelty pathways to transform the CO₂ captured into diverse products. Powered by renewable energy in combination with the use of CO₂ as raw material, the production of methane could feed certain industries (Breyer et al., 2015). In 2050, synthetic natural gas should be 75% of the European gas demand (Blanco et al., 2018). To substitute the use of fossil gas, it is necessary to investigate new techniques and materials. One of the focal points in this process it is the research in catalyst. Nickel catalysts are showing high rates of CO₂ conversion (Mebratu et al., 2021). Therefore, nickel is an excellent material to enhance the methanation process (M. Zhu et al., 2021). During the reaction, the methanation depends more on the content of H₂ than the amount of CO₂ (Champon et al., 2019). To reduce the environmental impact from the electrolysis of H₂O, the energy supplied should come from renewable sources (Guilera et al., 2021). The environmental gains of H₂ are remarkable in contrast to CH₄. Still, the H₂ has multiple disadvantages related to the storage step or the low energy density that the CH₄ molecule does not have (Egeland-Eriksen et al., 2021; J. Zhang et al., 2019). Thus, the use of CO₂ to produce syngas as basic material can further make more products than methane. Although this technique can replace fossil gas in the long term, in the near future, the impact of producing synthetic natural gas is still higher than fossil gas (Guilera et al., 2021). This is due to the use of

fossil CO₂ from combustion sources. Therefore, DAC systems could avoid this disadvantage.

Table 2.2 Principal reactions during the methanation process

Reaction	Stoichiometry	Enthalpy
Sabatier reaction	$CO_2 + 4H_2 \leftrightarrow CH_4 + 2H_2O$	$\Delta H^\circ_{298\text{ K}} = -165\text{ kJ/mol}$
	$CO + 3H_2 \leftrightarrow CH_4 + H_2O$	$\Delta H^\circ_{298\text{ K}} = -206\text{ kJ/mol}$

Methanol is the most important material in the chemical industry, with a wide range of goods produced (Olah et al., 2011). Fossil fuels are not required for the production of 'green methanol'. The hydrogenation of CO₂ can be an alternative way to produce this product -Table 2.3-(Bos et al., 2020). This method is similar to capturing CO₂ by using solid sorbents. Previous studies suggested that a scale plant production of 'green methanol' could abate 1.6 tonnes of CO₂ per tonne of methanol produced (Van-Dal & Bouallou, 2013). Currently, many by-products made of methanol keep using fossil fuels in their manufacturing processes. Moreover, it is difficult to move from conventional catalyst produced from earth-abundant materials. Using these catalysts in the production of 'green methanol' requires enormous amounts of water at the outlet of the reactor (Bowker, 2019). At the present, researchers are investigating this problem to enhance a sustainable production of methanol (An et al., 2017).

Table 2.3 Principal reactions during the methanol synthesis

Reaction	Stoichiometry	Enthalpy
CO ₂ hydrogenation	$CO_2 + 3H_2 \leftrightarrow CH_3OH + H_2O$	$\Delta H^\circ_{298\text{ K}} = -50\text{ kJ/mol}$
Reverse water-shift reaction	$CO_2 + H_2 \leftrightarrow CO + H_2O$	$\Delta H^\circ_{298\text{ K}} = 39\text{ kJ/mol}$

Most of the olefins come from the thermal cracking reactions of the naphtha. Block materials and plastics use the most known olefins -ethylene and the propylene-. However, there is an alternative to the conventional production. The combination of bio-fixation and catalytic cracking of biomass can produce these goods and stop the use of fossil fuels (Zhong et al., 2021). Recent studies are making efforts to enhance the reaction efficiency and to prolong the lifetime of the catalyst which will increase the product

selectivity (Z. Shi et al., 2021). Nonetheless, the production of coke during the process is problematic (Mores et al., 2011). Yet, the conversion of coke into active carbonaceous intermediates could solve this problem (Zhou et al., 2021). There have been important advances in this specific field, but it is necessary to keep investigating novel techniques to increase the production of olefins. As well as the olefins, the dimethyl ether (DME) production relies on the use of fossil fuels.

At present, most of the DME manufactured comes from natural gas. There are two primary uses of the DME: as propellant in the chemical industry and as fuel in compression ignites diesel engines (Azizi et al., 2014). Alternatively, it is possible to produce DME from methanol -Table 2.4- via dehydration in gas phase over acid catalyst such as zeolites (Peinado et al., 2020). However, during the conversion of DME, there was a decrease in the catalyst activity due to the formation of coke deposits. As well as in the methanol synthesis, scientists are researching new methods to develop new catalysts to enhance the conversion (Godini et al., 2022). Other industries also depend on the methanol production to deliver its goods.

Table 2.4 Principal reaction during the methanol dehydration

Reaction	Stoichiometry	Enthalpy
Methanol dehydration	$2CH_3OH \leftrightarrow CH_3OCH_3 + H_2O$	$\Delta H^\circ_{298\text{ K}} = -37 \text{ kJ/mol}$

In the food industry, the acid acetic (AA) is indispensable as food additive or preservative agent. There is an enormous demand for this product to make goods from other business: pharmaceuticals, plastics, photographic chemicals, pesticides, or rubber among a wide range (Deshmukh & Manyar, 2021). The most common technique used to produce AA is via methanol carbonylation, which uses CO from fossil fuel sources. The good news it exists a novel and green technique called methanol hydrocarboxylation - Table 2.5. This process requires the use of CO₂, which could come from DAC (Qian et al., 2016). As well as the olefins and the DME, the study of new catalyst is important to boost the production of AA (Ahmad et al., 2021).

Table 2.5 Main reaction during the methanol hydrocarboxylation

Reaction	Stoichiometry	Enthalpy
Methanol hydrocarboxylation	$CH_3OH + CO_2 + H_2 \leftrightarrow CH_3COOH + H_2O$	$\Delta H^\circ_{298\text{ K}} = -138\text{ kJ/mol}$

The production of formaldehyde (FE) is essential for manufacturing paper, plywood, or formaldehyde resins (Abdollahi & Hosseini, 2014). Currently, the production of FE relies on a combination of non-oxidative and oxidative dehydrogenation reactions of methanol -Table 2.6- (Maiwald et al., 2003). These reactions require the use of a heated copper oxide catalyst. The progress in the recent years helped to improve the catalytic dehydrogenation of methanol (Malik et al., 2021). Some of them could reach an optimistic 90% of selectivity towards FE (Thrane et al., 2021).

Table 2.6 Principal reactions during the methanol dehydrogenation

Reaction	Stoichiometry	Enthalpy
Non-Ox. Methanol dehydrogenation	$CH_3OH \leftrightarrow CH_2OH + H_2$	$\Delta H^\circ_{298\text{ K}} = 84\text{ kJ/mol}$
Ox. Methanol dehydrogenation	$CH_3OH + \frac{1}{2}O_2 \leftrightarrow CH_2OH + H_2O$	$\Delta H^\circ_{298\text{ K}} = 160\text{ kJ/mol}$

Another hopeful process is the production of Dimethyl carbonate (DMC). This compound has specific uses, e.g., as raw material for organic synthesis and reactions such as carbonylation (Naqvi, 2017). DMC is an environmentally friendly solvent used to make adhesives and cleaning agents. In the past, the phosgene handled the production of DMC (Tundo & Selva, 2002). But a novel approach proposes to synthesize methanol with CO₂ to produce DMC (M. Zhang et al., 2021). The problem of this innovative approach relies on the stability of CO₂ and the low conversion rates (Garcia-Herrero et al., 2016). Although the process is promising, it requires further investigation to reduce the disadvantages (Faria et al., 2021; Garcia-Herrero et al., 2016; Ohno et al., 2021; M. Zhang et al., 2021). There is a wide range of products in the chemical industry that depend on these compounds, yet other sectors are contingent on fossil fuels. A good obvious example of the last statement is the transport industry. Thus, it is vital to explore free-carbon

methods to produce and expand alternative fuels such as methane, methanol, ethanol and DME (Seyam et al., 2021).

2.6 Discussions

The benefits of our lifestyle have as a result a continuous emission of GHGs; and the ROI has become one of the most pollutant European countries per capita (European Commission, 2025). Thus, the insufficient detail and unclear guidelines for how each sector should reduce their emissions is a serious problem (George Lee, 2022). Some authors in the scientific community studied the assessment of scaling carbon capture and storage and NET technologies as a solution to solve part of the climate change challenges (Haszeldine et al., 2018). Furthermore, other studies pointed out the costs and energy analysis of these CDR solutions (Fasihi et al., 2019; Sinha & Realff, 2019). The present study offers a novel approach linking the society's necessities and CO₂ emissions from the ROI to DAC technology. This literature review is a wake-up call for local authorities to enhance further research in the CDR field. With the information reviewed from different sources, the study found a strong link between CO₂ emissions and energy consumption.

The ROI relies on the use of fossil gas to generate electricity. This is not new, and the country has set targets over the coming years to switch towards a free carbon economy. Some of the welcomed actions aim to implement new offshore wind farms in order to satisfy the future energy demand. Other policies are more ambitious and seek to substitute all the Internal Combustion Engines (ICE) cars fleet for electric vehicles (IWEA, 2021). However, there is an uncertainty about the pace to fulfil these objectives. What is the main reason? Many important sectors that supply goods and services are complex to decarbonise. The continuous use of fossil gas in the ROI will cover a significant part of its energy demands in the next decades (O'Halloran, 2021a). The size of this demand is uncertain and will depend on the pace of the shift in the power and heat sectors. Thus, the electricity generation and heat sector will continue to require the use of gas in 2030. Moreover, the country relies in the gas fields from the North Sea (Sesini et al., 2022). The interconnector between the ROI and the UK handles the gas supply to the island. Hence, the study emphasizes the critical dependency of the gas imports that come from UK which could risk the future energy security (Riegel, 2021). In addition, other researchers stated

that a gas disruption from Russia to the continent will not affect the ROI (Yusta & Beyza, 2021). However, the price of the gas imports and the GHGs emissions will increase because of the use of solid fuels (Abdul-Salam, 2019; Deane et al., 2017).

In the beginning, we underlined the dependency of oil in the transport sector and how the ROI aims to electrify it. In 2030, the EU expects to hit 3 million of public charging points. Yet, there is an uncertainty in the affordability of the electric vehicles: absence of an overall roadmap for charging points, network capacities, batteries, economic stimulus, raw materials, and incorporated policies on vehicles (European Court of Auditors, 2021; Robinson & Erickson, 2016). Moreover, there are many controversial aspects related with lithium batteries: the expected high demand, the location of the mines, the immense quantities of water required to produce it, local necessities, pollution of the extraction zone, poverty, and exploit of local resources (Nature, 2021). Thus, with all these associated problems, ICE cars will remain on the roads after 2030 and incrementing the CO₂ concentration in the atmosphere.

At present, developing countries are emitting over 60% of the global emissions, which reflects the difficulties of these nations to follow the established pace (Henriques & Borowiecki, 2017; Höhne et al., 2011). Yet, emissions per person from developed countries are higher in contrast to developing ones (European Commission, 2025). Hence, the wellbeing of the ROI will also depend on the global response (Nature, 2021). If the global community does not act as one to tackle climate change, the GHGs emissions will keep rising each year (Tracker, 2021). Indeed, the last reports did not include these emissions, where current targets and policies declared that the temperature would increase between 1.8 and 2.4 °C (Vice-president & Timmermans, 2021). For this scenario, the forecast expects an energy reduction between 3% to 9% related to the energy content in the wind speed at 120 meters (Nolan, 2015). This decline is analogous to the current energy drop from UK's wind farms (Morison, 2021). To avoid blackouts because of the low wind speeds, the ROI's government ordered last year to open the power plants of Whitegate in Cork, and Huntstown in Dublin (Barry O'Halloran, 2022b; O'Halloran, 2021b). So, will ROI meet its climate goals and satisfy its energy demands? We do not oppose to the measures stated above, but this study notices many gaps and local authorities set aside alternatives related to CDRs (Government of Ireland, 2021).

Table 2.7 Overview of novel methods and techniques to synthesise CO₂ into valuable products for specific industries.

Product	Applications	Novel method	Raw materials	References
Acid acetic	• Pharmaceutical industry • Food industry • Chemical industry • Plastic industry	Methanol hydrocarboxylation	Methanol, CO ₂ and H ₂	(Ahmad et al., 2021; Deshmukh & Manyar, 2021; Qian et al., 2016)
Dimethyl carbonate	• Battery electrolyte • Cleaning agents • Green solvent	Direct synthesis of Methanol	Methanol and CO ₂	(Faria et al., 2021; Garcia-Herrero et al., 2016; Naqvi, 2017; Ohno et al., 2021; Tundo & Selva, 2002; M. Zhang et al., 2021)
Dimethyl ether	• As fuel in diesel engines • Aerosol propellant • Solvent	Methanol dehydration	Methanol	(Azizi et al., 2014; Godini et al., 2022; Peinado et al., 2020; Seyam et al., 2021)
E-fuels	• Aviation transport • Road transport • Shipping	Fischer-Tropsch process	CO ₂ and H ₂	(Balasubramanian et al., 2018; Marchese et al., 2021; Rosanna Viscardi et al., 2021; Sabatino et al., 2021)
Formaldehyde	• Paper industry • Household products • Glues and adhesives	Catalytic dehydrogenation of methanol	Methanol and O ₂	(Maiwald et al., 2003; Malik et al., 2021; Thrane et al., 2021)
Methane	• Electricity production • Chemical feedstock • Heating in buildings	Methanation	CO ₂ , CO and H ₂	(Egeland-Eriksen et al., 2021; Mebrahtu et al., 2021; J. Zhang et al., 2019; M. Zhu et al., 2021)
Olefins	• Building block materials • Plastic industry • Detergents	Bio-fixation and catalytic cracking biomass	CO ₂ , CO and H ₂	(Z. Shi et al., 2021; Zhong et al., 2021; Zhou et al., 2021)

Even if the world achieves all its current targets related to renewable energy deployment, the IPCC and IEA highlighted CDR and carbon capture technologies as a crucial part in the mitigation plans (IEA, 2021; Masson-Delmotte V. et al., 2021). Additionally, limiting global warming to 2 °C will require a minimum global uptake of 6 GtCO₂ per year (Haszeldine et al., 2018). But even some academics question its viability (Sudipta & Kuo-Wei, 2020). However, various nations and companies demonstrated its viability despite the negative support from certain sectors. The study enumerated a few number of companies from which some of them have more than 10 years of experience. Although the elevated cost to remove one ton of CO₂ remains a problem at the present, each year the research in this area reduces the gap to make it more viable. The companies summarised in this study are receiving support from their respective countries yet, this is not the case of the ROI. Norway started working on a project called Longship in which they will inject CO₂ in depleted oil and gas fields (Helgesen et al., 2021). This process requires shallow and depleted hydrocarbon reservoirs or saline formations, and the ROI has two: the Kinsale and Corrib gas fields. However, the use of these depleted reservoirs will require further study because of earthquake risk (Zoback & Gorelick, 2012). As with known technologies in its initial phases, its capacity is inferior. Yet, the price per tonne of CO₂ captured will plummet after the firsts scale plants are running. Because of this, the price will drop drastically in the mid-century with an average cost of €50 per tonne (Fasihi et al., 2019).

So far, we showed the environmental gains but not the economic benefits if the ROI invests in DAC. The transport sector uses oil products, which is one of the point sources of CO₂ emissions (O'Halloran, 2021a). If it is not possible to substitute the ICE cars at the pace predicted, it will be necessary to explore alternatives that will act as a bridge in the meantime (Balasubramanian et al., 2018). Fischer-Tropsch processes could make a wide range of hydrocarbons that can further be refined into e-fuels (Marchese et al., 2021). It could produce diesel, petrol, kerosene, or other derivatives for the transport sector (Rosanna Viscardi et al., 2021). To perform the process, it is needed a continuous feedstock of syngas and CO₂ (Grim et al., 2019). The production of e-fuels generated by renewable sources in regions with high Flh of wind -as is the case of the ROI-, would decrease the associated economic costs and GHGs emissions (Fasihi et al., 2016; Soler, 2019). E-fuels could play a vital role in establishing an artificial carbon cycle however, the high cost to produce them at the moment remains an obstacle. Yet, these alternative

fuels will help to fill the gaps in the energy transition. An important aspect of the e-fuels is that they do not need the construction of additional infrastructure or technology to supply them. In addition, modern technologies, such as data centres, require significant amounts of energy. Furthermore, to achieve a NET Zero Carbon Gas Network in the ROI, it is essential to supply 50% of natural gas abated with CDRs, 37% of bio-methane and 13% of Hydrogen (GNI; Ervia, 2019). Implementing synthetic natural gas could store the excess of energy from renewable sources in lower production hours. Thus, producing a synthetic natural gas cycle at a local scale would abate the gas imports and reduce the energy dependency.

If the ROI wants to cover future energy needs and to tackle climate change, it is necessary to include NETs from CDRs in its future National Development Plans (Beuttler et al., 2019; Gambhir & Tavoni, 2019; Haszeldine et al., 2018; Realmonte et al., 2019; Rogelj et al., 2015). However, implementing DAC in the ROI would become more energy intense because of the presence of moisture. Yet, it is still essential to develop and implementing the technology in the country. DAC has multiple advantages in contrast to other NETs, such as Bioenergy with Carbon Capture Storage (BECCS). One limitation of BECCS is the land use, which shares it with food production and ecosystem protection (Creutzig, 2017). Powered by renewable energies, DAC could act as a bridge in the energy transition. Now that governments and private companies are funding the research and scale up of DAC, the ROI should not lag and start investigating it (Gates et al., 2022; Initiative & Earthshots, 2022).

As explained in this study, nature itself has an immense capacity to absorb CO₂, yet the time it will take to capture all the accumulated emissions will extend to the next century. To make it more complex, there is no global consensus and many of the actions proposed will take time, which will extend the GHGs emissions (Taconet et al., 2020; Tracker, 2021). Thus, to tackle climate change and support emerging nations to not commit the same mistakes, it is essential to develop DAC and fill the gaps that renewable energy cannot. A global decline in the energy demand and allowing new technologies in the market would help to mitigate climate change and meet the society demands.

2.7 Conclusion

Despite ROI's green landscapes, the country's energy consumption and GHGs emissions are far to be "green." In the last years, the country improved its standard of living due to the expansion of several industries and the economic growth. This situation had positive benefits, allowing more people to live in the country; yet there is always a downside. The increment of the energy demand in the last years could risk the energy security of the ROI.

All the communication channels are resonating the message of preserving and caring for the environment. However, the shortage risk of electricity in the coming years could affect different businesses and the power sector. It could be a beneficial decision to transition towards renewable energy. The geographical advantage of the ROI could allow to meet most of its energy demand by wind. Yet, the storage problem related to renewable energy remains an obstacle. This means that if there is no wind -and it has already happened recently-, fossil gas will take its place in the lower production hours. The calm context we had at the end of 2021 led to a low energy production from all the wind farms in the North Sea and, therefore, in a continuous consumption of fossil gas. These new emissions will remain in the atmosphere for at least 125 years until the natural carbon cycle can absorb it. This calm situation will leave the ROI's grid in a stressful cycle. Also, the increment of the gas prices could lead to undesirable consequences such as electricity shortages.

Despite the unsettling previous statements, the ROI could position itself as a role model for other European countries. The geological conditions of the ROI have the great capacity to reduce the concentration of CO₂ in the atmosphere. In the meantime, the production of e-fuels could take advantage of the excess of renewable energy. The ambitious targets related to the wind offshore farms in the ROI are not enough alone, but it could help to deploy a scale DAC plant and fill the gaps.

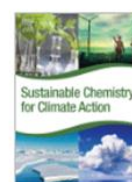
It is crucial to mention that with the non-implementation of DAC, the ROI would only lag the rest of Europe. With its undeniable terrain capacity, the country has the potential to depend on itself. If other countries such as Norway or the United States are doing it, why not the ROI as well?

Chapter 3: The impact of Direct Air Capture during the last two decades: A bibliometric analysis of the scientific research, part I



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The impact of Direct Air Capture during the last two decades: A bibliometric analysis of the scientific research, part I

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3 The impact of Direct Air Capture during the last two decades: A bibliometric analysis of the scientific research, part I

Abstract

While concerns have been raised about the ability of technologies like DAC to meet projected capture demands, such criticisms remain subject to debate. In the past, the vast investment in renewable technology is allowing today's rapid deployment. Why is this not currently happening in the CO₂ capture area? This bibliometric analysis which focused on the use of solid sorbents in the CO₂ capture field between 2001 to 2021, aims to answer these questions. The study reviewed three capture methods: post-combustion, pre-combustion and DAC, with particular emphasis on the latter. To understand the evolution of DAC, this novel approach highlights which authors and countries have been investigating the use of solid sorbents. The outcomes of this research showed that, during the first decade, there was a minor interest in funding and investigating solid sorbents for DAC solutions. It was only at the end of the second period when the use of these materials in the topic emerged to the surface. Acting as an example, the United States, China and the United Kingdom spent more financial help to investigate the use of sorbents. However, all of today's CO₂ capture plants are working for enhanced oil recovery. In the Republic of Ireland, there are a few articles exploring the use of these materials to uptake CO₂. It is possible that certain articles were not considered by the software. Upcoming analysis will answer this question and include all the existing materials in the wide spectrum of solid sorbents at the CO₂ capture field.

Keywords: *Direct Air Capture, Carbon dioxide removal, Bibliometric analysis, Carbon capture, solid sorbents.*

Nomenclature

Abbreviations

CCS	Carbon Capture and Storage
CDRs	Carbon Dioxide Removals
COP26	Conference of the Parties
DAC	Direct Air Capture
e-fuels	Electrofuels
GHG	Greenhouse Gas
GtCO ₂	Billion tonnes of carbon dioxide
IEA	International Energy Agency
IPCC	Intergovernmental Panel on Climate Change
MEA	Monoethanolamine
MOFs	Metal Organic Frameworks
MtCO _{2eq}	Million tonnes of carbon dioxide equivalent
PEI	Polyethyleneimine
RH	Relative Humidity
ROI	Republic of Ireland
SBA-15	Santa Barbara Amorphous-15
TEPA	Tetraethylenepentamine
TVSA	Temperature Vacuum Swing Adsorption

Symbols

GW	Gigawatt
kWh	Kilowatt hour

3.1 Introduction

The increase of the global temperature is an unavoidable truth. Even if the global community is able to fulfil all climate pledges in the coming years, the planet will be on course of increasing its temperature between 1.8 and 2.4 °C (European Commission, 2021). Before the 26th Conference of the Parties (COP26), the global temperature was in process of an increment between 2.1 and 3.5 °C (IPCC, 2021). In order to reduce greenhouse emissions, moving towards renewable energies is a key factor. Yet can it be that simple? The answer to such question is even more complicated: many sectors still depend on fossil fuels, and such obstacle could create more barriers to these industries (Casaban & Tsalaporta, 2022).

To make it more intricate, several countries were and are not counting their emissions properly because of the use of inaccurate data (Mooney et al., 2021). The estimated emissions not considered range between 8.5 GtCO₂ to 13.3 GtCO₂ and this could affect the measures taken by policymakers (Mooney et al., 2021). The Intergovernmental Panel on Climate Change (IPCC) and the International Energy Agency (IEA) stated the necessity to remove the excess of carbon dioxide (CO₂) from the atmosphere by Carbon Dioxide Removals (CDR) (IEA, 2021; Masson-Delmotte V. et al., 2021). Even more recently, after the COP26, Europe and the United States declared the urgency to invest in Carbon Capture and Storage (CCS) and CDR technologies such as Direct Air Capture (DAC) (Gates et al., 2022; U.S. Department of Energy, 2022a).

The history of DAC was never a straightforward path, and has its origin linked to the CCS sector. Both technologies are necessary to tackle climate change (P. R. Shukla et al., 2022). Yet, DAC and CCS are two different things: DAC aims to remove the CO₂ from the atmosphere while CCS capture it from point sources (P. R. Shukla et al., 2022). Moreover, DAC is a promising technology that will create a new industry by mitigating climate change (Gates et al., 2022). Before exploring more the past of DAC, it is pertinent to highlight the gap between the renewable sector and DAC. CCS emerged in the 1920s as a solution to separate the CO₂ from CH₄ in gas fields -Figure 3.1. It was not until the early 70s when the first plant to separate CO₂ for enhanced oil recovery opened (Global CCS Institute, 2020). After the energy crisis of the 70s caused by the “oil crack”, many countries funded and deployed renewable solutions.

In 1970, Dr Elliot Berman and experts in the energy sector reduced the price of the solar cells (Shu, 2019). Few years later, the NASA and the United States Department of Energy developed a research program which concluded with the construction of many wind turbines with Mega-watt capacity (Asmus, 2001). During 1974, the United States developed a 10-year research funding focused on materials such as Polycrystalline Silicon Solar Cells (U.S. Department of Energy, 1986). The approximate total cost was \$227.8 million of current year (U.S. Department of Energy, 1986). The National Science Foundation and the NASA funded the research, development and support of an experimental wind turbine generator in 1975 with a capacity ranging between 10 kW to 3 MW (Thomas et al., 1975). For a 5-year-period, the cost was \$26.3 million (Thomas et al., 1975). In 2015, the global investment in renewable technology was \$270 billion, where China was the major contributor followed by the United States (Ritchie Hannah et al., 2021). In an overall view, the national funding in renewable energy of the United States from 1978 to 2018 was \$27.65 billion of dollars (Clark, 2018). Thus, the estimated funding per year was around 0.7 billion, but it was in the early years when a major part of the capital expenditure was completed.

The interest for both technologies should be the same, yet the economic funding makes the difference: while renewable energies received the finance, DAC solutions stayed behind (IEA Greenhouse gases, 2012). CCS and DAC solutions have been forgotten in the past, where between 1993 and 2019, the cumulative public funding in the United States for carbon sequestration was \$3.7 billion for a 26-year period (Hezir et al., 2019). In addition, the investment in DAC was only \$11 million; for an 11-year period (Hezir et al., 2019). Although neglected from being financed in the last years, the European commission will be investing €820 million to research and develop CDRs such as DAC between 2022 and 2026 (Gates et al., 2022). Recently, the United States has passed the proposed Inflation Reduction Act, which will enhance DAC and CCS projects (Evergreen Action, 2022; Jenkins et al., 2022). This bigger boost will allow \$1.8 billion for a new clean industry, of which \$1 billion will be invested in carbon removal research, development, and demonstration (Luke Johnson, 2022).

Thus, the cost per tonne of CO₂ captured is expected to decrease in the current decade (Climeworks, 2022c). One of the most attractive solutions is the use of solid sorbents to remove CO₂; yet there is still a path to walk to plummet the costs (Fasihi et al., 2016). The study of the characteristics of the materials to retain CO₂, the number of

cycles and its pore size, are three of the key factors to scale-up solid sorbents in DAC technology. Among the materials studied for CO₂ capture, metal-organic frameworks (MOFs) and amine-based sorbents stand out as promising candidates for DAC applications.

MOFs are crystalline elements with exceptionally large surface areas, ranging from 1000 to 10000 m²/g and adjustable pore sizes between 3 to 20 Å (Anastasiou et al., 2018). One of its essential properties is the high selectivity to aim certain gases among others in a blend. It is possible to control the adsorption process of CO₂ by modifying the pore size and shape of the organic ligands from its composition (Anastasiou et al., 2018). Cyanides, nitriles, halides, and anionic organic molecules constitute the MOFs' organic side (O'Keeffe & Yaghi, 2012). The structure of the nodes are metal ions, such as Al³⁺, Cu²⁺ or Zn²⁺ (O'Keeffe & Yaghi, 2012). Despite its unique structural characteristics which caught the interest of the scientific community, researchers investigated how to neutralise the weaknesses of this material by improving its chemical, thermal, and mechanical stability. Chemical stability answers the resistance to various chemicals, such as moisture or acids, while thermal and mechanical stability are related with the capacity of MOFs to keep its structural integrity after the exposition to heat or pressure treatment (Ding et al., 2019).

Similarly, amines have their peculiarities which differentiates them from MOFs. First, it has a high capacity to adsorb CO₂, which does not require higher temperatures to regenerate the solid (Brilman & Veneman, 2013). Its mechanical and thermal stability allows the material to work in presence of higher levels of humidity (Brilman & Veneman, 2013). But the most important economic factor is its low manufacturing cost (Brilman & Veneman, 2013). These properties make the amines an excellent material to blend with certain sorbents to cover their disadvantages and enhance the CO₂ uptake. The most common amines used today are the Polyethyleneimine (PEI) and the Tetraethylenepentamine (TEPA) (Chaikittisilp et al., 2011; Holewinski et al., 2015; Subagyono et al., 2011; Q. Wang et al., 2017).

In the last decade, the research in new materials and methods to reduce the cost of CCS and CDRs has increased. However, as mentioned above, there are obstacles to overcome in order to help its commercialisation. Thus, the aim of this article is to perform a bibliometric analysis to know which countries were investigating the use of solid

sorbents in the DAC area. This study also has the intention of boosting the public interest of DAC in the Republic of Ireland (ROI).

3.2 Methodology

Web of Science and Scopus are the two main databases for bibliometric analyses. Both have many similar features, yet Scopus is able to cover more up-to-date research articles. Hence, the decision to use Scopus as a database in this work was due to the novelty of the technology. The range of the search includes any type of CO₂ capture related with the use of liquid solvents or solid sorbents, in particular, MOFs and amines.

The nine keywords established for this analysis helped to determine which articles were investigating solid sorbents for CO₂ capture purposes. The keywords are: (TITLE-ABS-KEY (carbon AND capture) OR TITLE-ABS-KEY (co2 AND capture) AND TITLE-ABS-KEY (direct AND air AND capture) OR TITLE-ABS-KEY (dac) OR TITLE-ABS-KEY (liquid AND solvent) OR TITLE-ABS-KEY (solid AND sorbent) AND TITLE-ABS-KEY (metal AND organic AND frameworks) OR TITLE-ABS-KEY (mofs) OR TITLE-ABS-KEY (amine)). This command gave 952 results between 2001 and 2022. VOSviewer was the software used to analyse the data obtained from Scopus. The use of Microsoft Excel to inspect the data helped to improve the accuracy of the study.

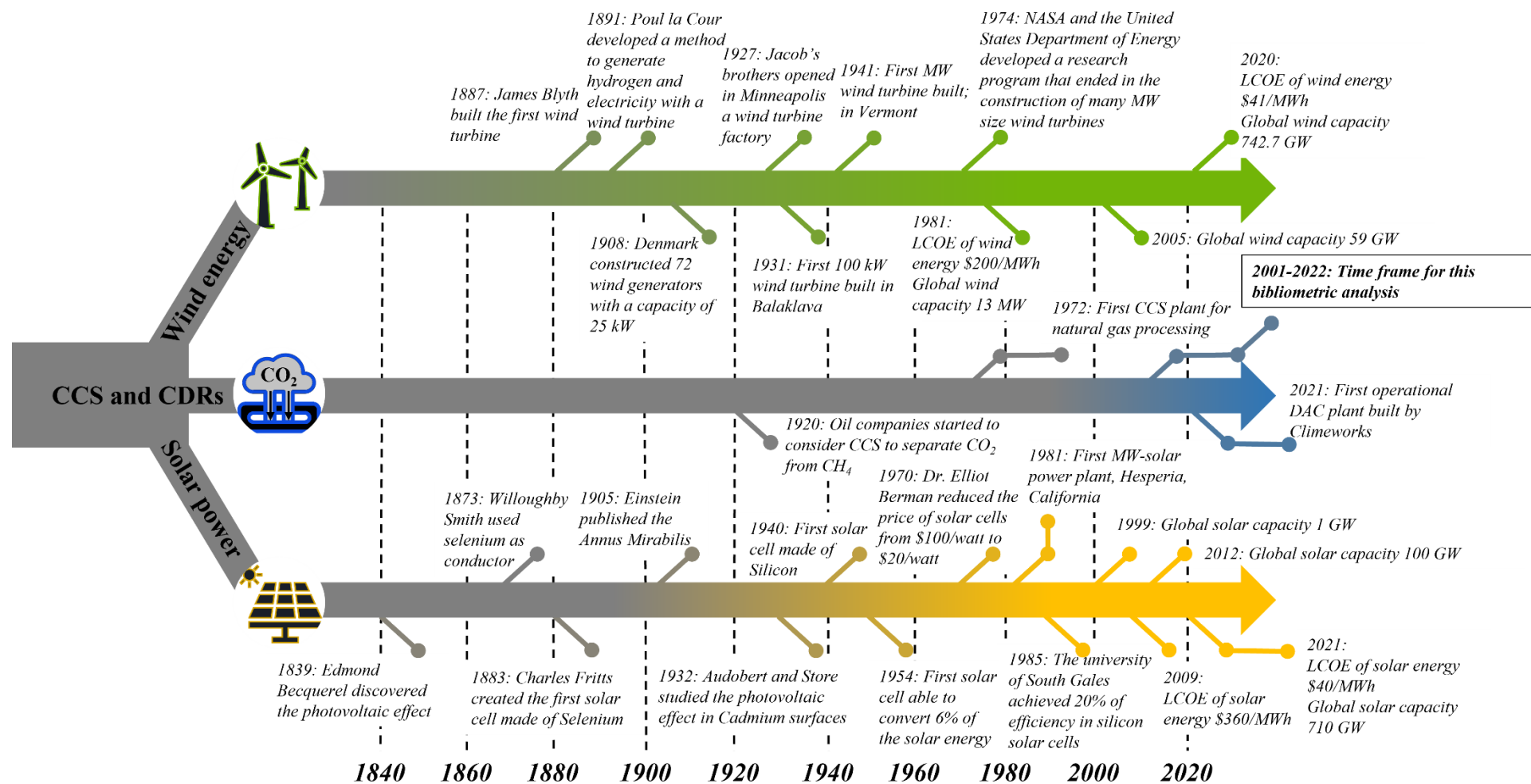


Figure 3.1. Timeline graph of the renewable and CCS/CDRs historical development. The top and bottom arrows emphasise the research and advances in wind and solar energy. In contrast, the middle arrow highlights the most important achievements related to CCS and CDRs. Every arrow illustrates the evolution of each technology from research to commercialisation and expansion. The graph shows the lack of public research and interest in CCS/CDRs during the last century. Only in the last two decades is when the research in the CCS/CDRs area started.

3.3 Results and discussion

The outcomes of the bibliometric analysis on solid sorbents from 2001 to 2022 are organised in different categories of interest: country, authorship and purpose of the research. Under the country-based category, this study will highlight which countries and institutions were investigating the use of solid sorbents materials to capture CO₂. Later, it will be examined what authors had most impact in the field. The last category explores the purpose of the research in the solid sorbents. This section studies the keywords and abstract to understand the purpose of the research.

As shown in Figure 3.2, before 2007, there were a few numbers of publications linked with CO₂ capture and solid sorbents. This could be due to the low repercussion and importance given in the past. On the other hand, the number of citations decreased, which resembles a normal distribution. The evolution trend is quite common: after a successful article published in a certain year, the upcoming studies will reference it, increasing the number of publications on the topic (Sebastian & Ramiro H. Gálvez, 2017). The outcomes showed that the number of citations reflect two peaks in the years 2012 and 2016.

Prior to 2005, the IPCC released a special report for policymakers, scientists, and engineers about the potential of CCS to capture and storage the CO₂ in geological sinks (Metz & O. Davidson, H. C. de Coninck, M. Loos, 2005). Two years later, the IPCC stated in its synthesis report the necessity of developing technologies to capture the carbon from the main emission points (Reay et al., 2007). Following these statements, the number of publications related with post combustion capture incremented after a period. A highly relevant and complete publication mentioned the use of activated carbons or zeolites to uptake the CO₂ from flue gas streams and power plants (Samanta et al., 2012).

Later in 2011, the company Climeworks started developing an adsorbent to enhance the uptake of CO₂ from ambient air (Gebald et al., 2011). During that year, the IPCC scheduled an international meeting on Lima (Perú) where they discussed about DAC (IPCC, 2012). After three years, the IPCC urged reducing the concentration of CO₂ from the atmosphere by using natural sinks or engineering processes (IPCC, 2014). These key messages during that period may encouraged the beginning of the research in CO₂ capture. This inevitably ended in various scientific articles focused on solid sorbents. Two

of the most cited articles were written by Sanz-Pérez et al. and Lalit A. Darunte et al. in 2016 where in both of them Christopher W. Jones is co-author (Darunte et al., 2016; Sanz-Pérez et al., 2016). In the same year, the Paris Agreement established not to surpass the 2 °C and to limit global warming at 1.5 °C since the industrial revolution (European Academies Science Advisory Council, 2018). The research on solid sorbents has risen steadily year after year; reducing the gap and helping to scale post combustion and DAC processes.

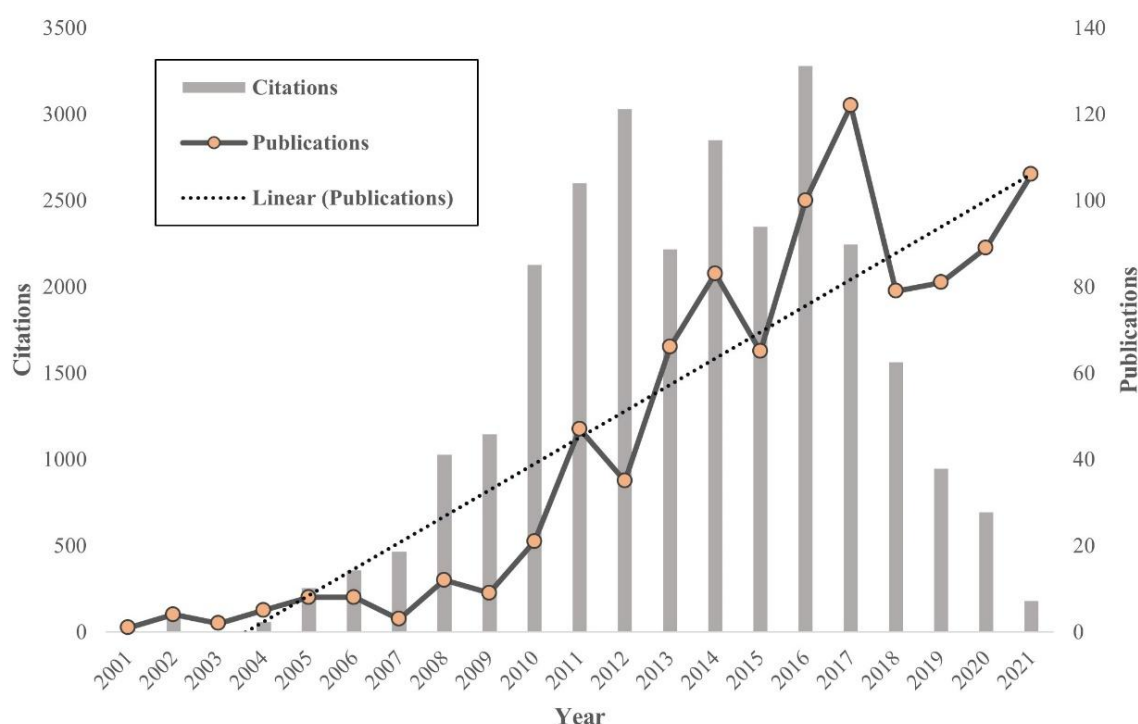


Figure 3.2. Evolution of the interest in the capture and CO₂ removal topic in the scientific field over the past two decades. In the figure it can be observed the increase in the number of publications and how after 2009, the number of citations increased, until 2021 following a normal distribution. This is expected, as citations of more recent articles typically take time to accumulate and reach comparable levels of attention.

3.3.1 Countries investigating the use of solid sorbents in the CO₂ capture area

Despite the lack of investment in DAC, the indexes showed certain countries are contributing more than others to enhance the use of solid sorbents in the CO₂ capture area. An overall of 48 countries in the world were and are currently investigating the use of solid sorbents. Only 20 of them have at least 10 publications -Figure 3.3-. The country that holds a higher number of publications is the United States with 260, followed by China and the United Kingdom with 184, and 53 publications, respectively. At the same

time, the most cited countries were United States (9893 citations), China (3768 citations), Canada (2917 citations), Spain (2122 citations), United Kingdom (1434 citations).

The United States and China are currently the top global emitters, yet both countries are incrementing their efforts to cut its emissions. As a result of that, both countries are leading the research in solid sorbents -Figure 3.4a-; in contrast, there is a significant group of 18 countries with less than 5 publications. These two top countries have a high number of plants working in Enhanced Oil Recovery and geological storage of CO₂; together with the facilities from the other countries, they can capture 40 MtCO₂ each year (Global CCS Institute, 2020). United States, China and United Kingdom, hoard numerous CO₂ capture projects in the current decade. Most of them are related to the use of solid sorbents in post combustion processes (Global CCS Institute, 2020).

The vast majority of the publications from Asian continent comes from one country, China. Although this industrialised nation had significant discrepancies during the COP26, it has set as aim to become carbon neutral by 2060 (Jianqiang Liu, 2021). Thus, this can be seen in the results of the current study, where the research in solid sorbents is a fundamental part to achieve such ambitious target. In addition, China started the development of demonstration projects to capture 150,000 MtCO₂ by CCS in the Shaanxi province (Global CCS Institute, 2020).

In the European continent, the countries that spent resources investigating solid sorbents are the United Kingdom, Norway, and France -Figure 3.4b. The United Kingdom cancelled many projects in the past due to many private companies refusing to work with the budget offered (Rathi, 2017). In October 2021, the United Kingdom selected two CCS projects for state funding of £1 billion. The aim of these projects is to store between 20-30 MtCO₂ per year by 2030 (Kulovic, 2021). Norway is currently working on the “Longship project” which aims to capture 1.5MtCO₂ per year (Helgesen et al., 2021). Programs like the previously mentioned ones are vital to reduce beforehand the costs of CCS; however, these projects do not place special emphasis in DAC. The budget to abate the CO₂ from the United States was inferior in contrast with other technologies. Hence, the public-economic contribution from European countries is expected to be inferior (Hezir et al., 2019). The ROI only had two publications related with solid sorbents. Yet, this situation could change in the current decade with the new funding plans from the European Union and the United States (Gates et al., 2022; U.S. Department of Energy, 2022a).

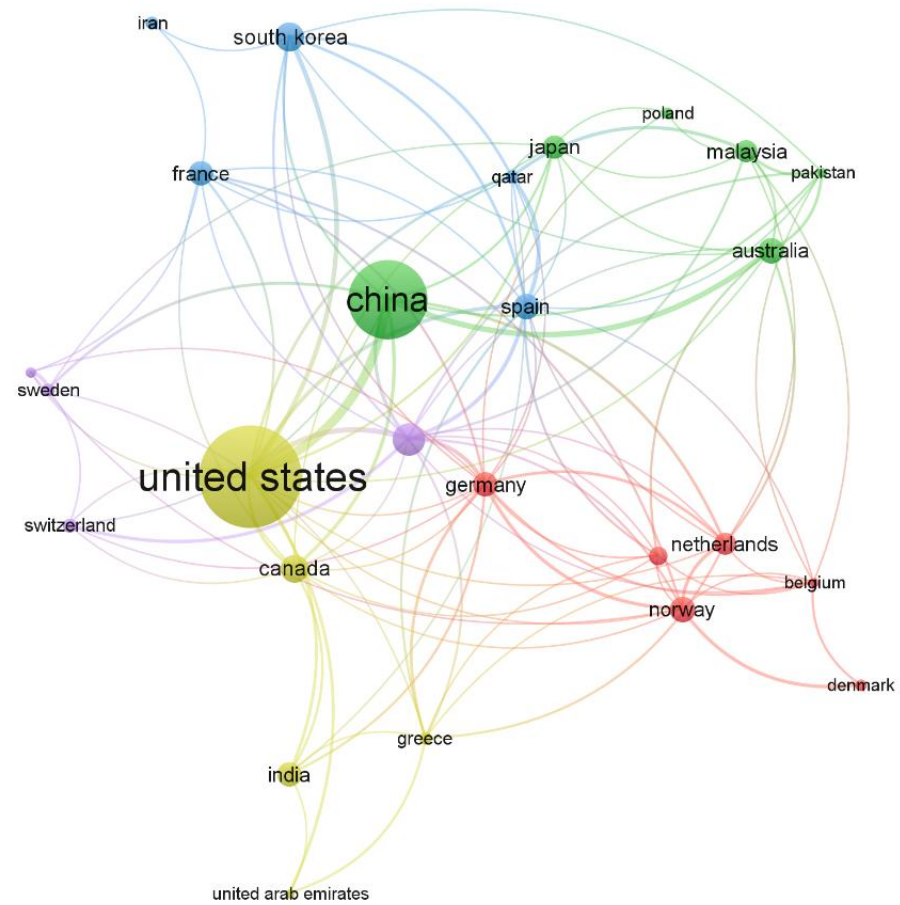
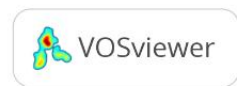


Figure 3.3. Links and strength (dots) approach across different countries with at least 10 publications on the associated search between 2001 and 2021.

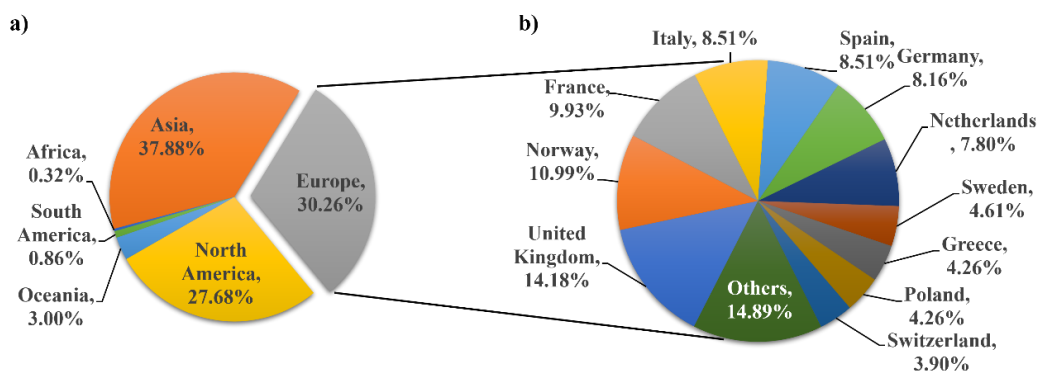


Figure 3.4. Contribution from different countries in the search of novel materials to enhance sorption processes (a), and in the European continent (b).

3.3.2 Authorship participation

In the scientific research, the author's number of publications and citations are a form to show the prestige and expertise of a certain person on the topic. The outcomes of this analysis showed that Christopher W. Jones from Georgia Institute of Technology hold the maximum number of publications and citations. Professor Jones works on carbon sequestration projects by using synthetic materials to enhance the adsorption of CO₂. The impact from authors is determined by the level of contribution in the field (Sandström & Van Besselaar, 2016). Hence, there is a strong relation between the number of articles published with the number of citations -Figure 3.5-.

As stated before, an article that had a significant impact on the topic could contribute to the scientific community as well as providing new knowledge in the field (Sebastian & Ramiro H. Gálvez, 2017). Other authors suggested that many publications and few number of citations could implicate that the researchers are not providing any novel insights (Loebbecke et al., 2007). For example, there could be authors with just one publication; yet if it is highly cited, future works after this publication can be overshadowed by the rest of publications (Sanz-Pérez et al., 2016). Because of the issues from this duality, it was necessary to filter authors with a minimum of 1 publication and at least 100 citations.

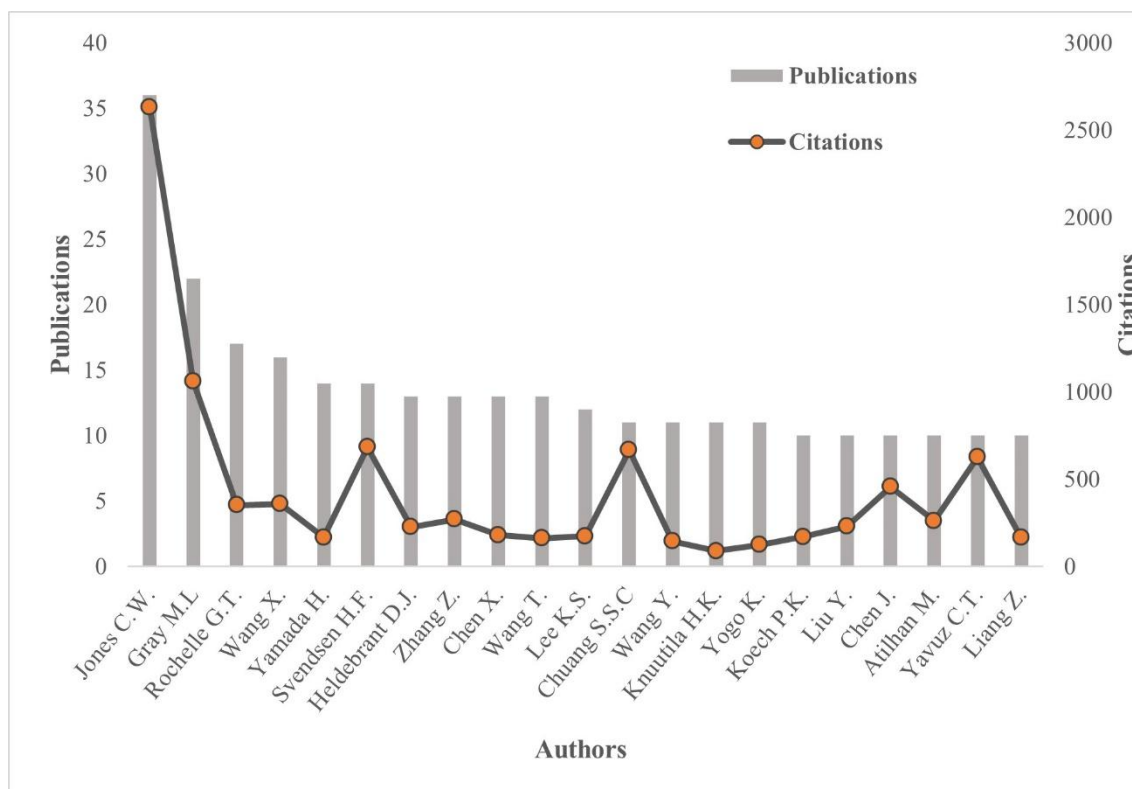


Figure 3.5. Participation of authors with a minimum of 10 publications from 2001 to 2021.

The size of each node from Figure 3.6 represents the number of publications, on the other hand, the width of the lines shows the total co-authored publications. The colours represents the more up-to-date papers, whereas the “Total link strength” shows the cooperation between different authors. Christopher W. Jones was the most associated author, with a value of 82 for a total link strength. The network created by this core encloses other authors, such as Sanz-Pérez or A. Darunte. It is one of the strongest and more modern groups. There is another large cluster that gathers the Asian institutions with authors such as Ying Wu from Nanjing Tech University (China) or Xiaoping Chen from the Southeast University (China). As stated above, researchers from China are investigating solid sorbents to enhance CO₂ capture (Wu et al., 2020). Great part of the other clusters do not have a strong relation with the two major networks. In consequence, these authors are poorly collaborating with researchers from United States and China.

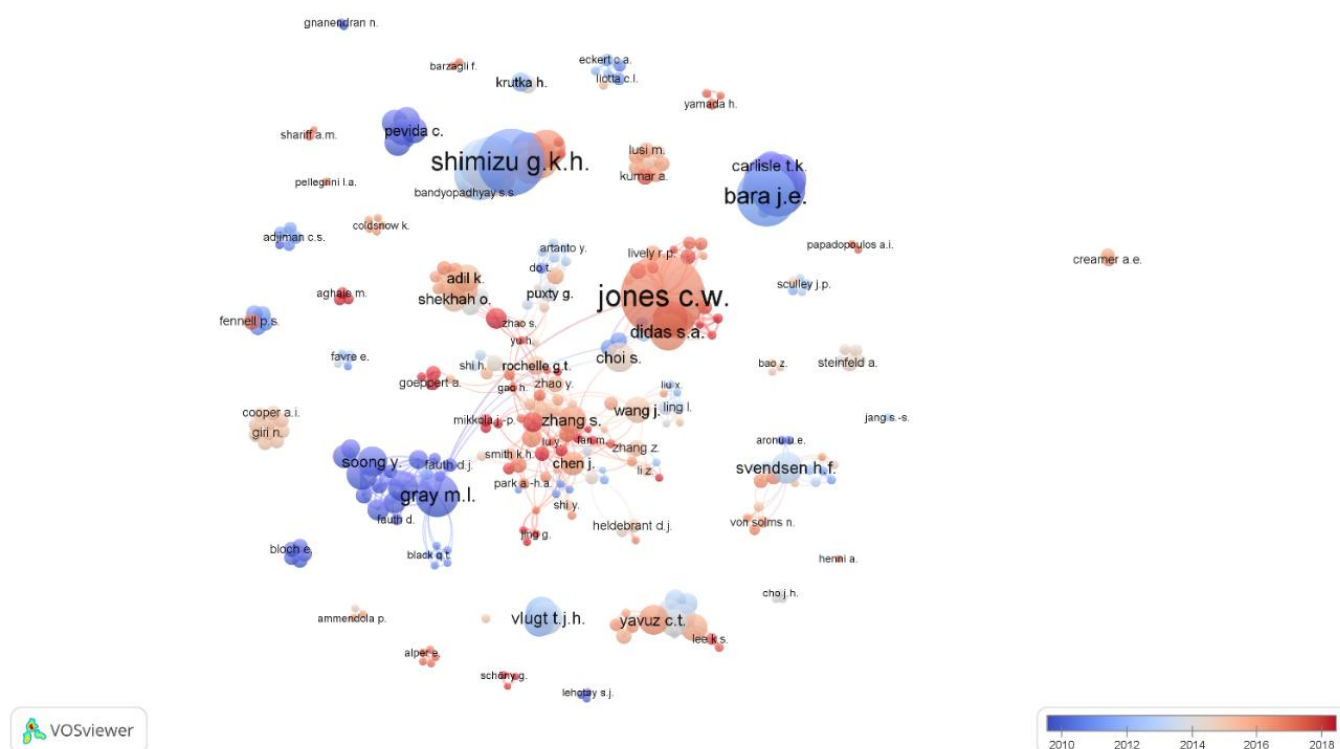


Figure 3.6. Co-authorship net with at least 1 publication and 100 citations from the period 2001 to 2021. The size of the node represents the number of publications, whereas the colour represents the novelty of the articles.

To explore the interrelationship between authors, conducting a co-citation study is essential. The academic link of an article that holds numerous citations will be stronger than the other ones. Thus, the threshold for this co-citation research was set at 20 minimum citations, from which 1549 of 37,643 authors fulfil the limits. The software structured the authors in 4 different clusters: red, green, blue, and yellow. The most cited authors were Christopher W. Jones, Rochelle G.T., Sayari A. and Svendsen H.F. -Figure 3.7-. Yet, other authors as Anthony E.J with fewer citations, had a notable strong link with only 256 citations.

The outcomes of this analysis showed that each author cluster divides the research topic into diverse techniques. Due to the nature of this bibliometric analysis, the largest group was the red cluster. This group contains all the information related to the use of MOFs, amines, or solid sorbents to enhance the adsorption of CO₂. The strength from the links of Christopher W. Jones was the most remarkable (217684). Professor Jones uses different solid sorbent materials to enhance the uptake of CO₂. The interconnection of his

links with other clusters shows that a comprehensive list of authors considers the contribution from Professor Jones.

However, Edward John Anthony was a research scientist focused on natural resources. Currently, he is a professor in energy systems at the University of Cranfield (England), working in calcium looping cycles. By including the words *post combustion* and *direct air capture*, Scopus considered other types of CO₂ sequestration. The investigations in calcium loop from Professor Anthony is not less important than solid sorbents and his link strength does not go unnoticed (106118). Exploring other methods of CCS and DAC can help to reduce the gap of these technologies. His research is taking into account from numerous experts. He is a guest professor at the Southeast University of Nanjing, which explains his links with other Asian authors.

In the blue cluster, Gary Thomas Rochelle is the most important author with a link strength value of. 74170. Professor Rochelle works at the University of Texas in Austin (United States). The aim of his studies is related with the use of amines to capture CO₂ from post combustion power plants by absorption processes. As well as in the case mentioned with Professor Anthony, Scopus considered another method of separation: the use of *amines* is not excluded for other processes. This explains the “poorly” connections from the authors of the blue cluster where many of them are “isolated” in small groups. Hence, many other authors from this group share the same research area in CO₂ capture as Professor Rochelle.

On the other hand, the yellow cluster involves the use of ionic liquids to absorb CO₂ from flue gases. Aside from that, Joan Frances Brennecke from the University of Texas at Austin (United States) had the strongest link with a value of 101355 (Uni et al., 2010).

As afore mentioned, the work from the green, blue and yellow cluster is not secondary. The CO₂ capture method from these groups differs from the red one. If the purpose of this analysis was not limited to the use of solid sorbents in adsorption processes, undoubtedly their link strength would be higher. Including many other keywords could help to explore all the existent methods of CO₂ capture.

3.3.3 Purpose of the research

It is essential to explore the aim of the studies from the researchers. The keywords and the abstract contains this information which helps to comprehend the purpose of their study. In order to filter the large number of keywords, this study set a threshold where a keyword should be included in at least five different articles. From 1704 keywords, 90 of them have a minimum of five co-occurrences -Figure 3.8. Thus, the software generated six main clusters: CO₂ capture, carbon capture, carbon dioxide, adsorption, absorption and ionic liquids.

The CO₂ capture cluster had a total number of 197 occurrences and is connected with a large number of keywords. Carbon capture and carbon dioxide keywords formed two groups with 60 and 59 occurrences, respectively. Although there are different techniques to uptake CO₂, the purpose from most of the articles is identical. Other keywords named same concepts diversely while other are related with methods or materials: adsorption (59 occurrences), absorption (55 occurrences), and ionic liquids (40 occurrences). These six specific clusters encompass the rest of words. The outcomes showed that important keywords such as “amines” (28 occurrences), “Direct Air Capture” (21 occurrences), and “metal-organic frameworks” (5 occurrences), are interconnected with the adsorption cluster. This suggests that there are a few number of articles linking the use of these two solid sorbents to DAC applications.

However, this analysis could drag a significant error. As stated above, authors could have named a same concept with a different word. This implicates that some research articles related to a specific method could end up ignored by the software and subsequently, increasing the error. Due to this, these words can appear in the abstract. Thus, as we were aiming to identify the purpose of the authors in their articles, this study explored the abstract’s words.

Scopus suggested that from 952 articles, only 482 articles contained the words “Post combustion”, “DAC”, “Air capture” or “Pre combustion” -Figure 3.9-. A few programmed commands were necessary to explore these keywords in the abstract of the 952 documents.

The outcomes revealed several articles designed to post combustion capture (380 publications and 12474 citations). Meanwhile, the number of publications associated with DAC or air capture were 90, with at least 3458 citations. As expected, there were a few numbers of publications linked to the use of the different sorbents in pre combustion

processes (12 publications and 220 citations). Enhanced oil recovery is a mature technique, and its research has been done in the past. Yet, from the 482 articles, only 90 are associated to DAC in the last two decades. Upon review, the remaining 470 papers do not explicitly state the specific application context of their material research. It is likely that many focus on fundamental material properties relevant to CO₂ capture; and therefore, these publications only used keywords such as CO₂ capture or carbon capture. Although studying material properties is vital, these papers do not specify a particular deployment context, such as DAC or post-combustion. However, the number of DAC publications in the last years gained the sufficient attention to be at the same level as the post combustion research -Figure 3.10-.

In 2011, Christoph Gebald and Jan Andre Wurzbacher, the founders of the company Climeworks, published an important article (Gebald et al., 2011). In their work, they produced a novel amine to uptake the CO₂ from the atmosphere. The adsorption capacity at 40% of Relative Humidity (RH) was about 1.4 mmol/g and its capacity remained constant after 20 adsorption-desorption cycles. This article encouraged researchers to investigate materials such as amines, however, it was not until 2016 when an article written by Eloy S. Sanz-Pérez et al. “gave the green light” to investigate the use of solid sorbents (Sanz-Pérez et al., 2016). The review article per se was very complete and included the outcomes of varied materials studied by other researchers. As the authors mentioned, it is essential to investigate and improve the performance of these materials to scale DAC. Figure 3.10 shows the impact this article had; after its publication, similar research articles followed the topic, and the citations of DAC in 2016 were above from those associated with post combustion processes.

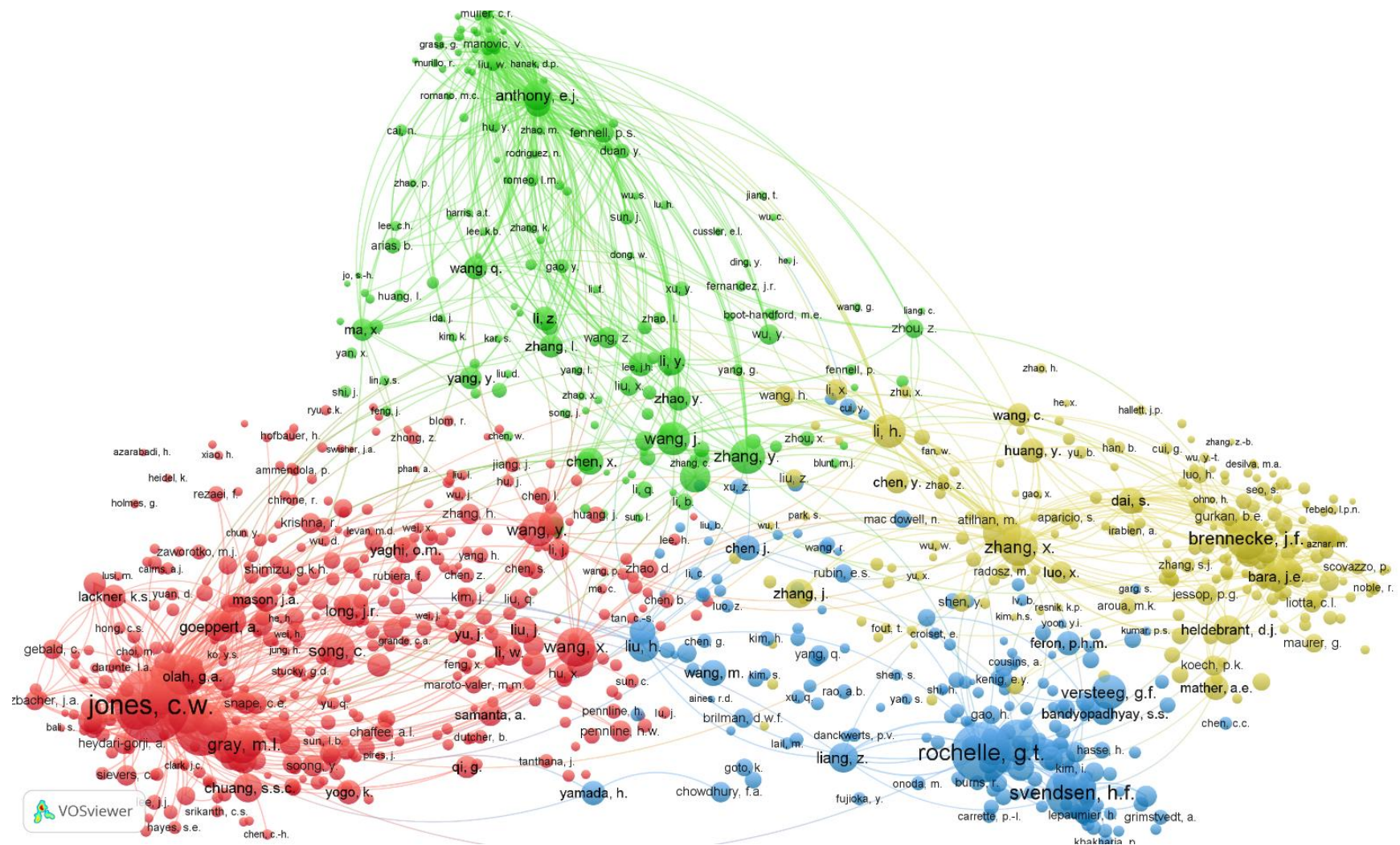


Figure 3.7. Co-citation network of authors with at least 20 publications during the period 2001 to 2021. Four clusters can be observed for the for types of CO₂ capture methods.

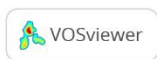
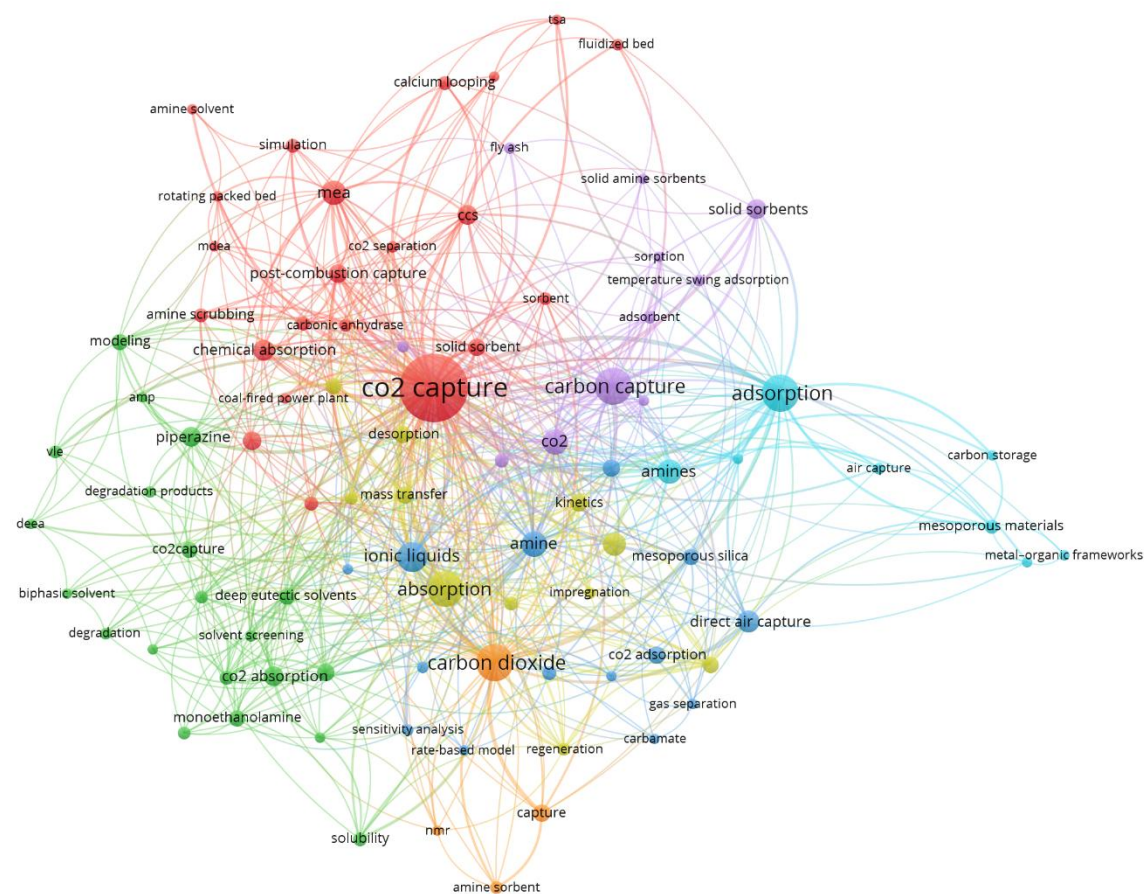


Figure 3.8. Co-occurrence system with at least 5 appearances in the publications related with the search during the period 2001 to 2021. At the central point it can be observed how the word “CO₂ capture” emerged as a central point, whereas adsorption, amines and mesoporous materials (in light blue) are linked to direct air capture

In 2012, the citations from post combustion articles were on the rise. The article written by Samanta Arunkumar et al. could be the cause of this interest (Samanta et al., 2012). Their work was very akin to the one from Eloy S. Sanz-Pérez et al., which became a model for the latter authors. As aforementioned, the investigation in post combustion received the attention from international groups such as the IPCC years before. Meanwhile, at the beginning of 2010, DAC research was in its infancy. One of the most critical features to scale DAC is linked with the research of novel materials to capture the CO₂ at low operational cost. Authors from different countries heard this message and the interest in DAC bloomed at the end of the last decade.

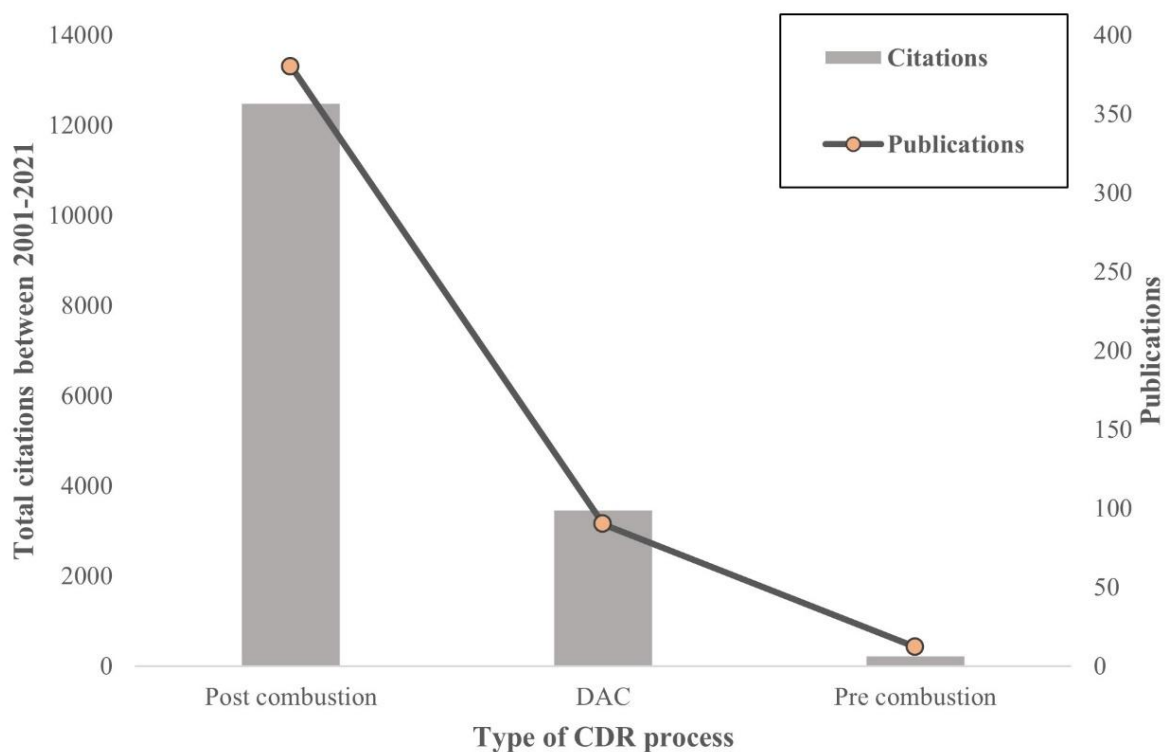


Figure 3.9. Variance between the articles focused on the study of novel materials for Post-combustion, DAC, and Pre-combustion systems during the period 2001 to 2021.

Giancarlo Piscopo and Stefan Loebbecke focused their research on the use of MOFs to capture CO₂ (Piscopo & Loebbecke, 2020). As they stated, MOFs are materials with grand expectations to reduce the atmospheric concentration of carbon. Many aspects in relation with MOFs are necessary to improve: the adsorption capacity, the CO₂ selectivity, the material stability after a certain number of adsorption-desorption cycles, the energy requirements to desorb the CO₂ from the solid and moisture. The latter will be essential to fix it to scale DAC

in the ROI. This problem involves an elevated energy demand to desorb the CO₂ from the solid. Hence, as the authors mentioned in their research, is essential to keep investigating to increase the bond of CO₂ with MOFs in presence of water.

Other researchers as Muhammad Munir Sadiq et al. demonstrated a pilot scale DAC plant by using MOFs (Sadiq et al., 2020). Their novel approach secured the adsorption process against the moisture content in the air. Hence, the energy requirements for desorbing the CO₂ from the material were minimal -1.6 kWh/ kg of CO₂. After 2680 cycles, the CO₂ purity ranged between 70-80%. Their results encouraged them to produce another small device with an uptake capacity of 6 kgCO₂ per day.

Authors Ali Kiani et al., performed a technoeconomic analysis to study the use of DAC with amines to generate renewable H₂ (Kiani et al., 2021). Although the purpose of their study is not linked with solid sorbents, they suggest using waste heat to reduce the energy cost. The authors proposed to integrate a methanation unit in the DAC system. However, the study emphasises the elevated cost associated to the H₂ production.

In Finland, Jere Elfving et al. studied different techniques to improve the regeneration step (Elfving et al., 2021). The research compared numerous methods to enhance the CO₂ capture in a fixed-bed reactor. The best conditions were achieved by using a Temperature Vacuum Swing Adsorption (TVSA) unit at 60 °C. After 23 cycles, the adsorption capacity remained stable. In a recent work, the authors considered the humidity. Jere Elfving and Tuomo Sainio estimated the equilibrium CO₂ capacities with amines in humid and dry air conditions (Elfving & Sainio, 2021). They studied the adsorption capacity and calculated the CO₂/H₂O ratio by changing the content of moisture: 0.1 vol%, 1 vol% and 2 vol% H₂O. After 17 cycles with a 2 vol% H₂O and a CO₂ concentration about 380 ppm, the CO₂ adsorption-desorption capacity decreased 0.73% per cycle.

Zhu et al. tried to reduce the cost of amines by using a functionalised aqueous amine miscible in organic Mg_xAl-CO₃ (X. Zhu et al., 2021). In contrast to SBA-15 supports, amine-tethered Mg-Al-CO₃-LDH were 30% more efficient. At 30 minutes, the adsorption capacity was at 70% and the regeneration temperature was inferior to 80 °C. After 50 adsorption-desorption cycles, the adsorption capacity remained stable -0.912 mmol/g-. Yet, they did not consider the effect of moisture.

In The Netherlands at the University of Twente, Schellevis et al. optimised the operational variables of a TVSA system: gas velocity, desorption temperature and working capacity (Schellevis et al., 2021). The solid sorbent used was a commercial amine functionalised material. Their outcomes showed that the desorption temperature defined the

thermal stability of the solid sorbent used. In their outcomes, the gas velocity does not have a higher impact in the energy consumption and increases the productivity. At higher desorption temperatures, the working capacity rose and therefore reduced the desorption time. As the authors stated, incrementing the temperature does not implicate to intensify the energy cost. Yet, a vacuum pump to desorb the sorbent and the weather conditions could affect the final energy cost.

Numerous authors studying materials in the CO₂ capture field are emerging over the world with a same goal: reduce the costs of solid sorbents and scale DAC. The world aims to capture 100 MtCO₂ by 2030, yet the global funding is six order of magnitude inferior to the expected (Mission Innovation, 2022). While these researchers are exploring this solution, there was a few number of articles from the ROI. Although there are two companies in the country, public funding is essential to encourage research programs and dispel all the existing doubts (Casaban & Tsalaporta, 2022).

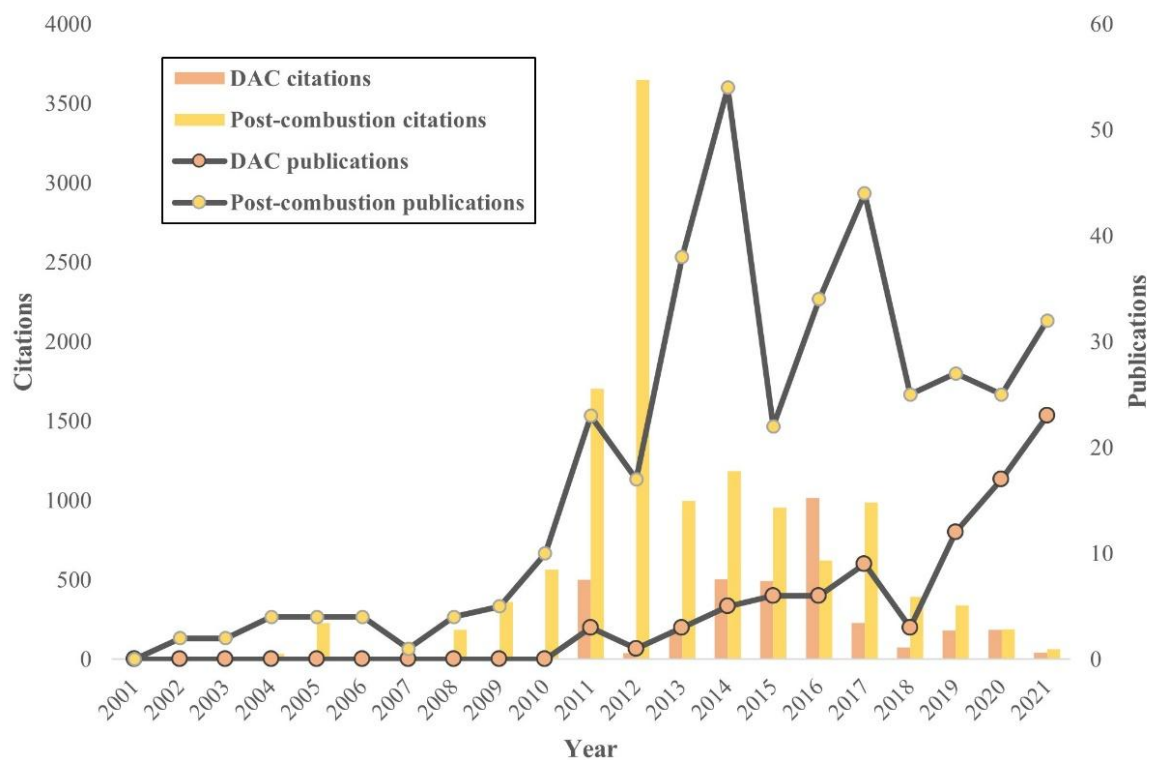


Figure 3.10. Comparison between the evolution of DAC and Post-combustion articles in the research field for the period 2001 to 2021.

3.4 Conclusion

The misconception about CCS and DAC efficiency is not helping its research and deployment. Although the process to separate CO₂ is already known, the IPCC stated that without CCS and CDRs, tackling climate change would cost double. While the CO₂ concentration in the atmosphere is rising each year, this study corroborates the poor public perception on these solutions in the last two decades. Hence, there is more than enough evidence to increase its funding.

The research related to DAC is the one most affected, yet the results demonstrated that the concern for the topic augmented at the end of the last decade. Nations are exploring and investigating in solid sorbent solutions, but there are two of them that are taking the lead: China and the United States. This study visualised the strong link between both countries and their influence among the rest. Thus, the collaboration of authors as Christopher W. Jones with other researchers from Europe is accelerating the process. However, the outcomes did only show two articles from the ROI.

Recently, the European Union gave its approval to research and scale-up DAC and hydrogen in the next few years. Though there are no confirmations for this next statement, it is possible that the ROI will be out of the budget due to the poor engagement in the topic. These circumstances will lead the ROI to the end of the line in the “fight” against climate change. Research in materials and methods may not benefit the country in the short term, but it will help to accomplish future goals of this novel technology: exploring new materials and methods and discard those which are not working. Yet, it is crucial the collaboration among nations and researchers.

Numerous articles named by authors are focused on using different solid sorbents at lab scale to enhance “CO₂ capture.” However, to boost DAC industry, future studies should engage in feasible materials that could work at atmospheric conditions. Several countries -similar to the ROI- have higher percentage values of RH. The presence of water is an issue that will increment the energy cost to uptake the CO₂. Hence, there are several conditions for a sorbent to be economically viable: the CO₂ adsorption capacity, the life cycle of the material, its abundance in nature, and other technical factors such as the energy consumption or the adsorption process. Thus, if there is a material with an excellent adsorption capacity, yet it can

only perform a few numbers of cycles or if the material is not sustainable; this sorbent would not be the ideal to scale DAC.

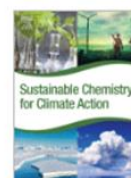
This novel analysis explored the use of solid sorbents in the CO₂ capture field. However, there are various and different solid sorbents not included in this study, as for example activated carbons. On some occasions, authors named related articles in a broader manner without putting special emphasis on the respective carbon capture technologies, but focusing more on specific materials. Maybe some authors are communicating their research outputs by alternative channels or platforms. These arguments could have limited the present research and maybe the software overshadowed studies from researchers of the ROI. This study is not exhaustive; this is the first of a series of bibliometric analysis, and future work will focus on other known materials and the existing technologies, in order to further map the scientific work in this area. As for example, the work from Edward John Anthony (Blamey et al., 2010), (Bui et al., 2018), who is working in a relative topic of calcium loops, or the work from MacElroy (Tsalaporta & MacElroy, 2020), (Keshavarz et al., 2021), (Niall et al., 2014). Yet, the message is clear: there is trend in the scientific field towards CO₂ capture, DAC and carbon capture.

Chapter 4: The impact of Direct Air Capture during the last two decades: A bibliometric analysis of the scientific research, part II



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The impact of direct air capture during the last two decades: A bibliometric analysis of the scientific research Part II

Daniel Casaban , Elena Tsalaporta  

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4 The impact of Direct Air Capture during the last two decades: A bibliometric analysis of the scientific research, part II

Abstract

This bibliometric analysis is a following part that aims to expand the information from our previous study. Direct Air Capture (DAC) technologies are not progressing as fast as expected and time is limited. Research and shared knowledge can accelerate the implementation of DAC and Carbon Capture Utilisation and Storage (CCUS) solutions. Numerous authors publish their findings on different platforms and journals. Thus, this bibliometric analysis is using Web of Science as data base. During this study, two cases of study were performed: one merges concepts such as “carbon capture,” CO₂ capture and DAC, and a second one only focused on removing the CO₂ from the atmosphere; being the latter at the principal topic of this analysis. CCUS and DAC + S share the same approach to capture CO₂, yet the conditions are quite different. Many articles related to DAC were overshadowed by words such as “CO₂ capture” and “carbon capture.” However, the results showed that in 1999, Professor Lackner was the first scientist who suggested the implementation of DAC technologies. After him, a great number of authors followed up and researched on the topic. Based on the outcomes of this study, United States, Europe and China are leading the research. The United States holds the major number of publications and has strong links with the rest of the countries, which implicates that authors from other nations collaborate with the North American country. Yet, it is necessary to increase its social acceptance to accelerate the deployment of these technologies.

Keywords: Direct Air Capture, Carbon dioxide removal, Bibliometric analysis, Carbon capture, CO₂ capture.

4.1 Introduction

The concentration of carbon dioxide (CO₂) in the atmosphere has dangerously returned to pre-pandemic levels, and it is dramatically increasing each year (NOAA, 2022). Despite the 2015 agreement of COP21, where governments agreed to limit the temperature rise to 1.5 °C, (United Nations, 2015), no substantial action has been taken towards this direction; and now, we are in the risk of surpassing this 1.5 °C threshold (McGuire, 2022). In order to control global warming to the limits established in the Paris agreement, the emissions should be reduced by a 45 percent each year and there is an emission gap of 23 GtCO₂ annually (United Nations, 2022). Therefore, the grid infrastructure must be modernised by using renewable sources; yet this is not enough for certain sectors that depend on fossil fuels.

Although renewable energy is growing exponentially, the world's primary energy consumption remains stable (Ritchie Hannah et al., 2023). The outsourcing of European industry can be a problem if materials such as aluminium or rare earth metals are required for the energy transition (Liestbet et al., 2022). These minerals are necessary for wind turbines, solar panels, electric vehicles and maybe for the upcoming Carbon Dioxide Removal (CDR) and Carbon Capture industry. Some researchers believe that lithium-ion batteries have peaked, which may constitute a problem for certain emerging technologies (Patrick George, 2022). This unstable situation has led to the redistribution of the CO₂ emissions to other countries (Baumert et al., 2019). Thus, will the CO₂ concentration in the atmosphere keep increasing? As pessimistic as it may sound, recent studies affirm that limit global warming to the 1.5 °C threshold is impossible (Diffenbaugh & Barnes, 2023). Global CO₂ concentration has only dropped during recession periods (NOAA, 2022). In order to avoid these undesirable circumstances, many changes around diverse sectors must be done in the near future: accelerate and implement renewable sources of energy, smart consumption, transport alternatives and decarbonisation of the industry, among others (IPCC, 2022). Until we tick all these boxes, the CO₂ concentration will keep increasing due to hard-to-abate emissions and, according to the last report from the IPCC, every fraction of degree should be addressed with CDRs (IPCC, 2023).

Direct Air Capture (DAC) is a novel CDR that can remove and store permanently the CO₂ from the atmosphere or transform it into valuable products (Casaban & Tsalaporta, 2022). DAC concept emerged in 1999, where Professor Klaus Lackner suggested in a conference to reduce the CO₂ concentration of in the air (Lackner et al., 1999). And, in 2021, the first

operational scale plant was built in Iceland (Climeworks, 2021). Right now, there are only a few DAC plants operating and it is necessary to deploy and investigate more to reduce the costs of this technology (IEA, 2022a). DAC funding has been downgraded in the last two decades (Casaban et al., 2022). Avoiding emissions are indispensable but CDRs are required to limit global warming (World Economic Forum, 2021). Therefore, DAC is a key technology in the CDR portfolio, but this technology only received the 0.04% of the world investment on the energy transition (BloombergNEF, 2022). Hence, CDRs can remove 2 Gt_{CO2} from the atmosphere from which only a 0.1% is coming from novel methods such as DAC (Smith et al., 2023). Industrial deployment and research are essential to understand the objectives and challenges ahead and, therefore, reduce the costs (McQueen et al., 2021).

Until today, numerous advances have been done since DAC was recommended. During this window of time, novel solvents and solid sorbents have been the main line of investigation (Gregory de Temmerman; & François de Rochette, 2023). At the moment, DAC plants based on solvents have higher capacity yet, this method is more energy intense and requires the use of large amounts of water (Custelcean, 2022). On the other hand, solid sorbent technology operates at ambient pressure with maximum temperatures of 120 °C and its plants are easy to scale (Wiegner et al., 2022). Moreover, there are other and new technological techniques that will require time to mature as for example, electro swing adsorption or a DAC approach based in membranes (Beaumont, 2022; Castro-Muñoz et al., 2022).

Therefore, this study aims to cover the gaps and extend beyond the research previously done (Casaban et al., 2022). In contrast with our previous work which only explored the use of solid sorbents, this bibliometric analysis is focused only on DAC, and its differences with carbon capture. Although carbon capture is also important and aims to remove the CO₂ from point sources or to re-utilise it; DAC efforts have to be directed to remove the concentration from the atmosphere and store it safely underground (Congressional Research Service, 2021).

4.2 Methodology

In contrast with the previous study -where Scopus was used as database-, Web of Science was selected as database for the current analysis. The reason of this change is that numerous researchers publish their works on multiple platforms, and this was identified as one limitation from our first study. Hence, the use of Web of Science as a database in this work

helped to obtain more documents and extend the range of the analysis. Two different searches were done: one focused on any type of CO₂ capture and another one with a special emphasis on DAC. R studio was used to analyse the data obtained from Web of Science. From the data collected, it was analysed the authors keywords, title words and trends in order to know the interest in the topic. It was also studied which authors contributed more to the scientific research as well as which countries are spending more resources on these topics. These analyses helped to understand the evolution of carbon capture and DAC in the last 20 years.

As aforementioned, the first search was focused on any type of CO₂ capture method. Any document containing in their titles, author's keywords and keywords plus the following code was selected for our first analysis: ((TI = ("Direct Air Capture" OR "Carbon capture" OR "CO₂ capture" OR "atmospheric carbon capture" OR "atmospheric CO₂ capture" OR "CO₂ Capture from Air" OR "CO₂ Capture from the Ambient Air" NOT ("CO₂" OR "capture" OR "air" OR "direct" OR "carbon"))))

OR (KP = ("Direct Air Capture" OR "Carbon capture" OR "CO₂ capture" OR "atmospheric carbon capture" OR "atmospheric CO₂ capture" OR "CO₂ Capture from Air" OR "CO₂ Capture from the Ambient Air" NOT ("CO₂" OR "capture" OR "air" OR "direct" OR "carbon")))

OR (AK = ("Direct Air Capture" OR "Carbon capture" OR "CO₂ capture" OR "atmospheric carbon capture" OR "atmospheric CO₂ capture" OR "CO₂ Capture from Air" OR "CO₂ Capture from the Ambient Air" OR "Carbon Dioxide Extraction from Air" NOT ("CO₂" OR "capture" OR "air" OR "direct" OR "carbon")))). This command gave 8315 results between 1999 and 2022.

As expected, the vast majority of the articles are investigating capture CO₂ from point sources. Because of this, it was necessary to filter the data in order to know exactly how many articles were and are exploring the CO₂ capture in the DAC field. Therefore, the following search improved the refinement: TS = ("Direct Air Capture" OR "atmospheric carbon capture" OR "atmospheric CO₂ capture" OR "CO₂ Capture from Air" OR "CO₂ Capture from the Ambient Air" OR "Carbon Dioxide Extraction from Air"). A total number of 599 documents were obtained between 2006 and 2022.

4.3 Results and discussion

In this study, three categories of interest were proposed to highlight the most relevant information in the DAC and carbon capture area: labelling words, most important authors in the field, and country contribution.

4.3.1 Labelling keywords and titles

Authors tend to name in different forms the same concept and this is one of the most characteristic limitations of these bibliometric studies. This leads to unclear outcomes where the contribution from others can be missed (Haustein & Larivière, 2015). Therefore is important to establish a common niche of words in order to help and identify the studies from researchers. “Direct Air Capture” could be the central word from which many others branch out. “Carbon Capture” is the dominant expression to refer to all the CO₂ capture processes. As explained earlier, CO₂ capture ranges from Carbon Capture and Storage (CCS) or Carbon Capture Utilisation and Storage (CCUS) at point sources, to CDRs, DAC + S or atmospheric capture. Despite their similarities (European Commission, 2022a), there are many parameters that differentiates the processes: relative humidity, CO₂ concentration, pressure, etc. From the bulk of the 8315 documents, “carbon capture” and “carbon capture and storage” were mainly used whereas there were only 150 occurrences of the word “direct air capture” (Figure 4.1a). Therefore, and by including the word “dac,” the search aimed to highlight DAC studies showed 6 more occurrences (Figure 4.1b).

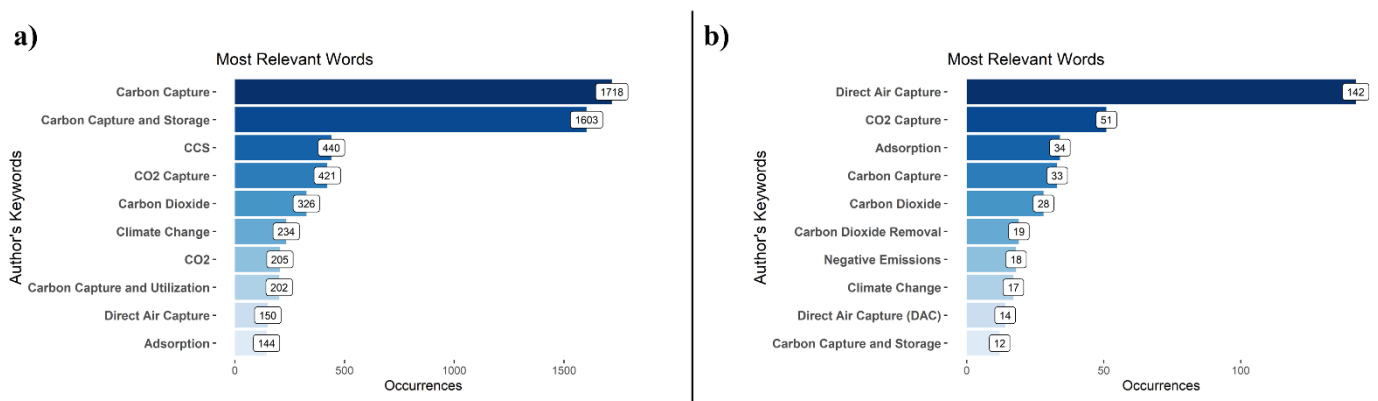


Figure 4.1. Most relevant keywords by the authors on carbon capture (a) and direct air capture (b).

If author's keywords does not show the aim of the study, the titles of the documents can determine the purpose of the research. "Carbon capture" and "carbon dioxide capture" words were the most commonly used in the results of the first search, whereas "direct air capture" only appeared 169 times (Figure 4.2a). From the second search, the title's words obtained ranged from "carbon dioxide capture" to "air capture technologies" (Figure 4.2b). Yet, it was observed that most documents are adopting the word "direct air capture" in the most recent documents. Analysing the titles' words have an additional and important limitation than the authors' keywords does not have. In the case of the first search, the titles were analysed by a sequence of two adjacent words, while in the search focused on DAC were studied by groups of three consecutive words. This distinction was necessary because many articles identified in the initial search used previously mentioned terms interchangeably. Thus, it is important to emphasise the difference between "carbon dioxide capture" and more specific approaches such as "carbon dioxide removal" or "direct air capture." As observed in Figure 4.2b, some of title's words include the sequence "air capture processes." Adding this expression to the "direct air capture" expression gives the exact number of occurrences as in the large search.

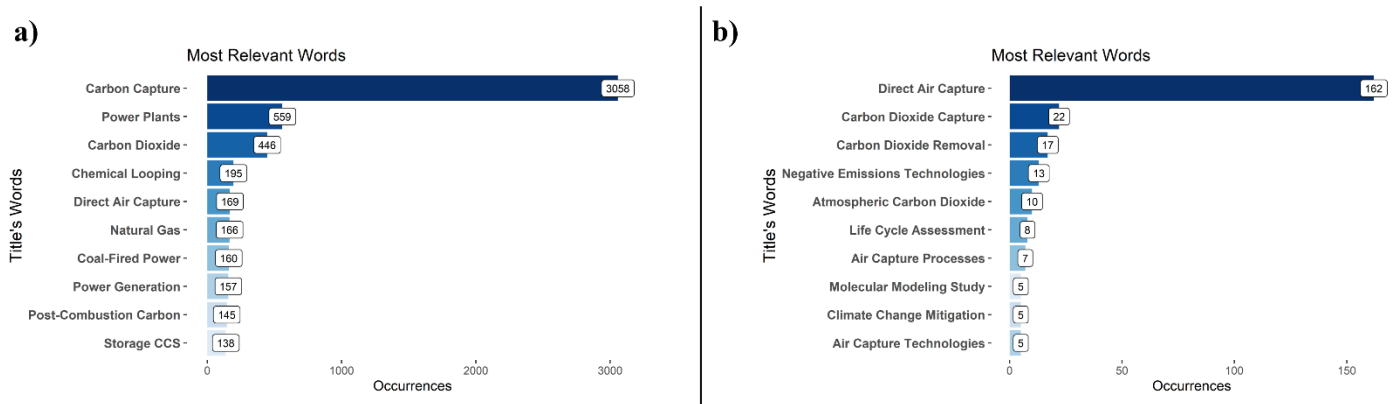


Figure 4.2. Most relevant words included in the publications' titles by the authors on the topic of carbon capture a) and direct air capture b)

Although the three previous analyses showed the minor impact of DAC in the CO₂ capture area, the trend in the last years shows the exact opposite (Figure 4.3). Even though the idea of cleaning the atmosphere emerged in 1999, it was at the beginning of 2015 when the interest in the research topic boosted, and the role of DAC, was considered indispensable to meet the future climate goals (Realmonte et al., 2019). Important economic studies were done to clarify the doubts of these pathways (Fasihi et al., 2019). Yet, the critics points out the high cost of the materials and the energy, as well as doubting of its efficiency (Sudipta & Kuo-Wei,

2020). Although DAC can share the same background as carbon capture, this technology has emerged in the last few years (Erans et al., 2022). During that moment, researchers were studying methods to enhance the process (Gebald et al., 2011).

The articles of DAC grew in 2011, where researchers such as Christopher W. Jones and MacElroy started investigating the use of amines, silicas, metal organic frameworks among other solid sorbents (Choi et al., 2011; Tsalaporta et al., 2017). As many of them, international authors were and are currently investigating materials and processes that could work at atmospheric conditions.

4.3.2 Most relevant authors

In this section, the study aims to highlight the most relevant authors between the carbon capture and DAC field. This analysis does not intend to discredit the work from the researchers. As explained in the previous section, it is extremely difficult to include the work of all the authors in the DAC sphere. It is possible that many good articles have not been included in the analysis due to quoting reasons. Thus, in contrast with the previous work, this bibliometric did not consider the number of citations and publications as a threshold. However, due to the large number of authors, it was necessary to reduce and filter the number of publications.

There are two main metrics to measure the impact of the authors: h-index and g-index. The h-index assesses the productivity and citation impact of a specific publication. This implicates that h-index links the number and the impact of each paper (Hirsch, 2005). Per contra, the g-index allows highly cited articles from an author that can strengthen the rest of their publications. For example, if one of the ten publications has 100 citations and the rest have none, the author g-index will be equal to 100; whereas for an author h-index equal to 100, it is necessary that each of the 10 publications have at least 10 citations (Egghe, 2006).

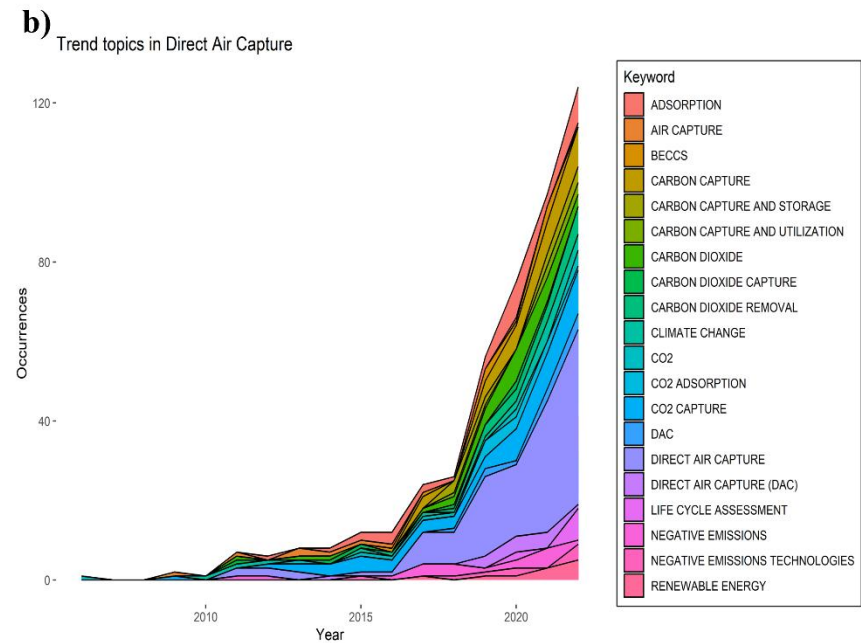
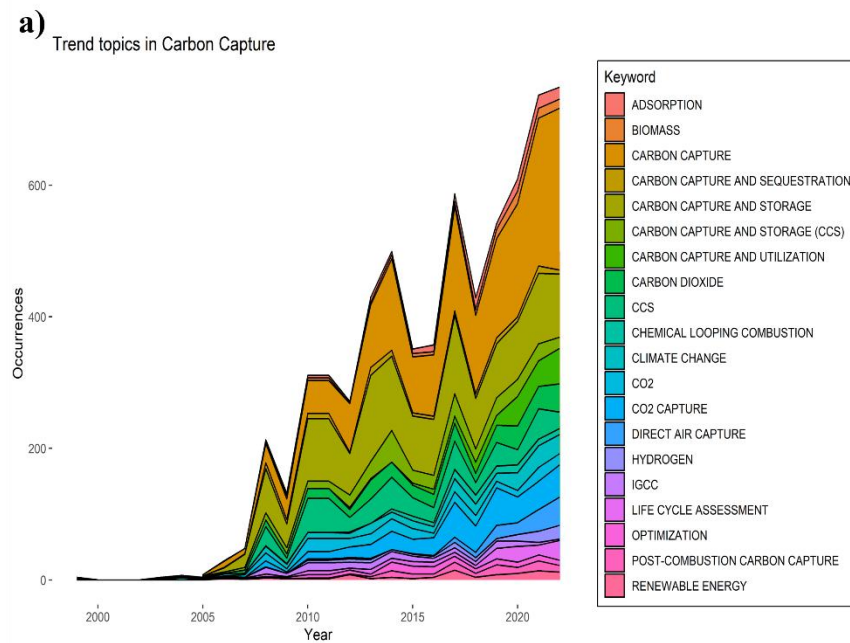


Figure 4.3. Research trends since 2009 in the carbon capture and DAC search with a word minimum frequency value of 5. In both figures it can be observed that the research topic of DAC and CO₂ capture have been increasing over the past two decades. In addition, it is noticed how authors name the same process distinct.

Figure 4.4a and Figure 4.5a present the h-index and the g-index for the top 15 most-cited authors identified from the initial search results. As expected, many authors from this part of the analysis are studying how to capture CO₂ from point sources. Professor Cormos is a chemical engineer studying the integration of CCS technology in the power sector (Cormos, 2012). Vasilije Manovic is a professor of Carbon Systems engineering at Cranfield University. His work includes CO₂ capture by calcium looping cycles and CO₂ utilization (Z. Zhang et al., 2020). Professor Manovic collaborated with Professor Edward John. Both are studying how to reduce the emissions from power plants (Manovic & Anthony, 2008). Professor Mac Dowell have multiple collaborations with other authors and as well as Professor Cormos, whose work is focused on decarbonizing the power and industrial sector (Dowell et al., 2017). Yet, recent works suggest the use of DAC to produce synthetic fuels for sectors with a high dependency on fossil fuels (Daggash et al., 2018).

Although there are authors in the CCS sector that are also investigating the DAC area, naming the process of removing the CO₂ from the atmosphere as “carbon capture” or “CO₂ capture” can outweigh specific publications. Professor Jennifer Wilcox name can be observed in both figures. She is a distinguished professor of chemical engineering and energy policy. Her work is not only focused on exploring the role of CCS, but she also studies NETs such as DAC (House et al., 2011; Robert Socolow et al., 2011). Professor Wilcox points out the necessity of reducing the CO₂ emissions and not fully relying on large scale CDRs. This article was written in 2018, three years prior to the launch of the first operational DAC plant. However, as she mentioned, it is important to reduce the cost to scale CDRs technologies and if in future they become commercially available, these technologies will have the potential to revert the global temperature (Fuss et al., 2018). In order to address this issue, Professor Christian Breyer collaborated with Mahdi Fasihi in a techno-economic study (Fasihi et al., 2019).

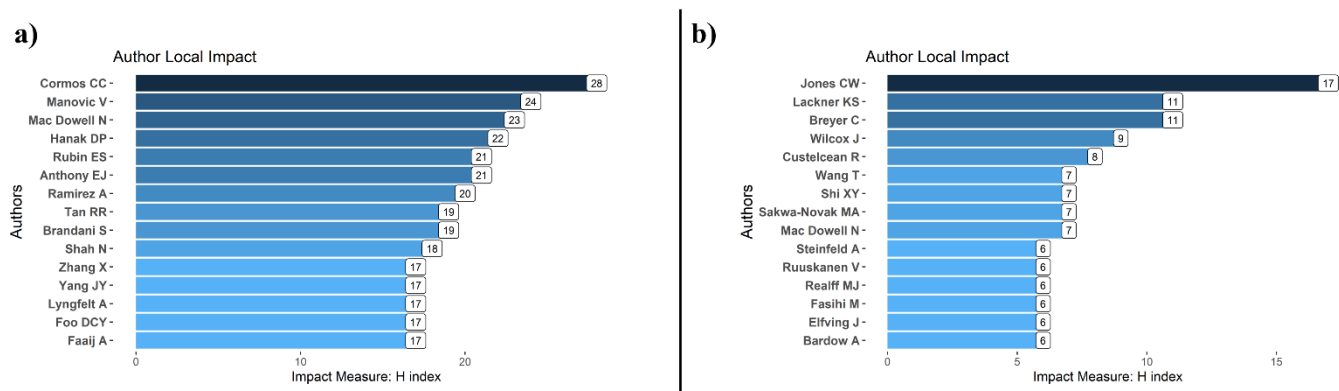


Figure 4.4. H-index of the authors in the carbon capture a) and direct air capture b) areas.

Yet, if someone is worth of stressing, is Professor Klaus Lackner. He was the first who suggested cleaning the atmosphere (Lackner et al., 1999). All the scientific contribution from Professor Lackner have been concentrated on removing the CO₂ from the atmosphere and its subsequent mineral sequestration (Lackner et al., 1995; Lackner, 2009). He studied methods to enhance the performance of the process and reduce the associated costs (T. Wang et al., 2011). As well as Professor Mac Dowell, he foresaw the dependency of certain sectors from fossil fuels (Graves et al., 2011). Because of the importance of finding sustainable materials, Professor Lackner collaborated with Dr Xiaoyang Shi in the study of sorbents for DAC (X. Shi et al., 2020). Most of the work from Dr Shi is directed around removing the CO₂ from the atmosphere (X. Shi et al., 2017; Song et al., 2018).

In our previous work, we remarked the contribution from Professor Jones and this can be seen in Figure 4.4b and Figure 4.5b where Christopher W. Jones is the author with the highest impact (Casaban et al., 2022). Among other authors, such as Gebald, Wurzbacher, and Professor Steinfeld (Gebald et al., 2011), he studied the use of amines and solid sorbents to capture the CO₂ from ambient air (Choi et al., 2011). His work is not only aimed at the DAC field (Sanz-Pérez et al., 2016), but he also studies the use of these materials to enhance the capture of CO₂ from point sources (Choi et al., 2009; Hicks et al., 2008). His research interests ranges between two materials: Metal-Organic Frameworks and functionalised amines (Bae et al., 2010; Qi et al., 2011).

In contrast with Professor Jones, senior researcher Radu Custelcean investigate the use of aqueous alkaline solutions for DAC purposes (Custelcean, 2022). His interests encompasses the crystallisation and CDRs areas (Custelcean, 2021). One of his recent works studied the use of a promising sorbent for DAC purposes via crystallisation (Stamberga et al.,

2022). To perform it, he explores the use of amino acids to capture CO₂ and, later, that it can be regenerated by guanidine compounds (Kasturi et al., 2021).

Researcher Tao Wang collaborated with Professor Klaus in the study of an amine sorbent to absorb CO₂ (T. Wang et al., 2011). Thus, his research interests are focused on the use of amines in absorption processes (T. Wang et al., 2016). As mentioned before, these processes are more energy intense than the adsorption ones, yet there are companies and other authors that are exploring this technique (Yu et al., 2019).

Besides Climeworks, Global Thermostat is a company that also aims to reverse climate change by reducing the concentration of CO₂ from the air. Miles Sakwa-Novak is the lead scientist at the R&D department. He studied materials such as mesoporous carbons to enhance the CO₂ adsorption (Kuan Huang et al., 2015). Along with Professor Jones, he explored the use of polyethylenimine in solid sorbents (Pang et al., 2017). In one of his works, he emphasises the importance of the research in novel materials: the co-adsorption of the water molecule during the process is a vital issue that is necessary to address in order to scale DAC technologies (Didas et al., 2014). Conjointly with Christopher Jones, they hold numerous patents of materials for CO₂ adsorption (Jones et al., 2022).

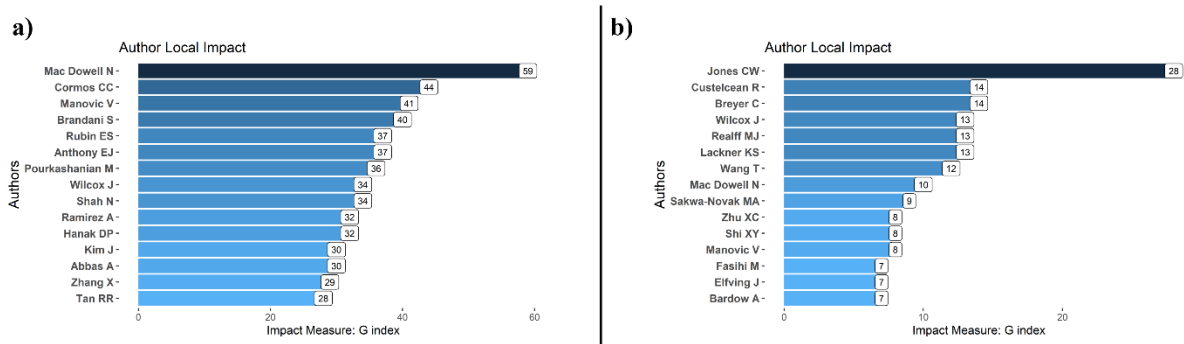


Figure 4.5. G-index of the authors in the carbon capture a) and direct air capture b) areas.

Andre Bardow is a Professor in Energy and Process Systems Engineering. As well as Professors Anthony John, Mac Dowell and Tao Wang, he took part in the paper: “Carbon capture and storage (CCS): the way forward” (Bui et al., 2018). He examines the use of “carbon capture” as mitigation tool against climate change (Kätelhön et al., 2019). Again, the word “carbon capture” can be ambiguous and enclose post-combustion and DAC processes. Yet, many of his articles are focused on life cycle assessments of “carbon dioxide capture” and “DAC” (Deutz & Bardow, 2021; Von Der Assen et al., 2013). Furthermore, powered with wind

energy, he is exploring the production of synthetic fuels for the sectors that are finding difficulties to decarbonise (Deutz et al., 2018).

As stated before, DAC and CCS can share the same methodology, but they are different. In Figure 4.6, it can be observed again that most of the authors are focusing their studies on removing CO₂ from point sources; Professor Lackner, previously identified as a pioneer in the field, appears exclusively in the red cluster. Although decarbonising the industry will play a crucial role in the mitigation of climate change, research and development of new materials, techniques and process for scaling DAC, are essential to meet the climate targets. Thus, there were authors that have inherited and became aware from the suggestion from Professor Klaus (Figure 4.7).

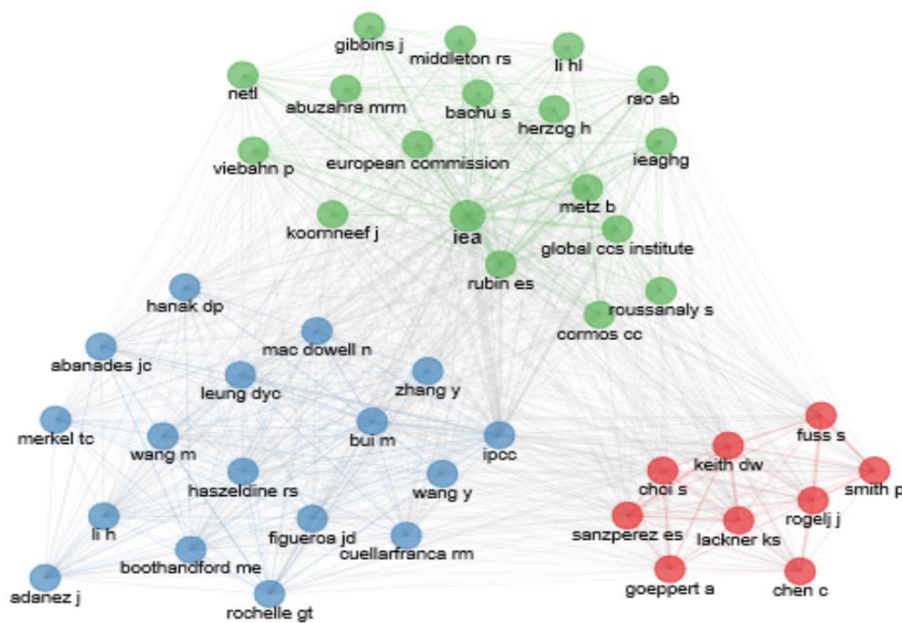


Figure 4.6. Collaboration network between the authors in the carbon capture area.

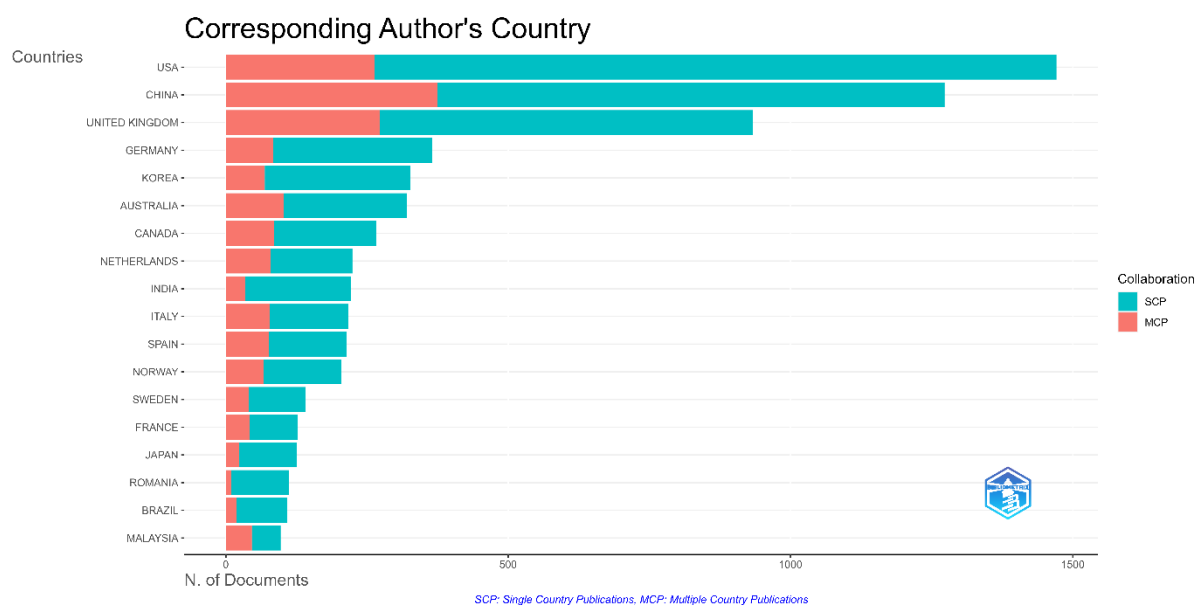


Figure 4.8. Country collaboration publication by author's country in the carbon capture area.

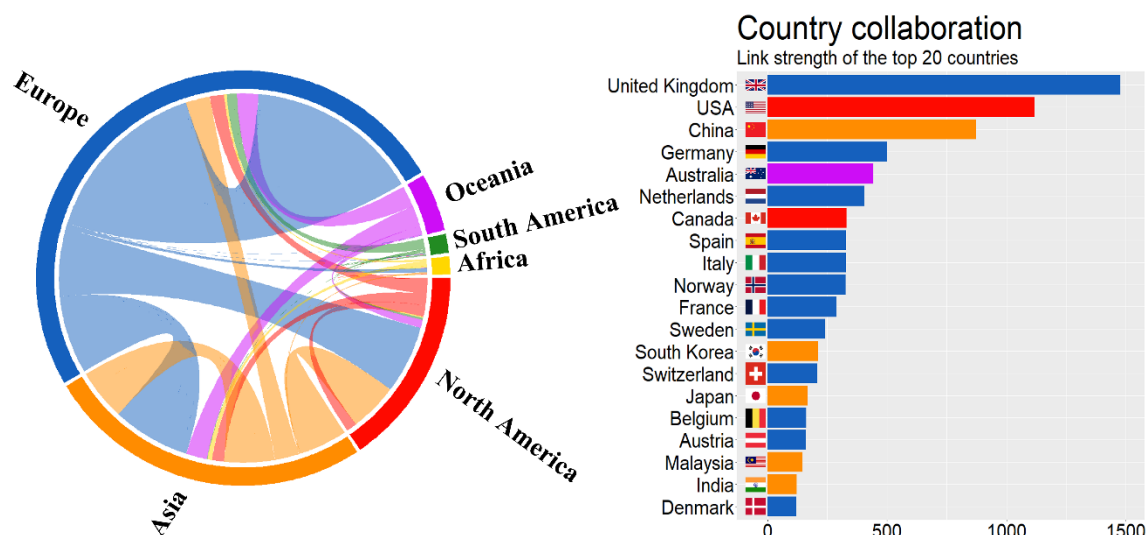


Figure 4.9. Chord diagram of the collaboration between continents and its link strength from the carbon capture area.

While exploring the DAC documents in the second search, the results showed that the three leading countries were the United States (221 documents), the United Kingdom (94), and Germany(60 documents) Figure 4.10. Along with the first search, Europe was the continent with the biggest collaboration strength. Authors from Asia interacted with authors from Europe and authors from Europe, interacted with authors from North America Figure 4.11.

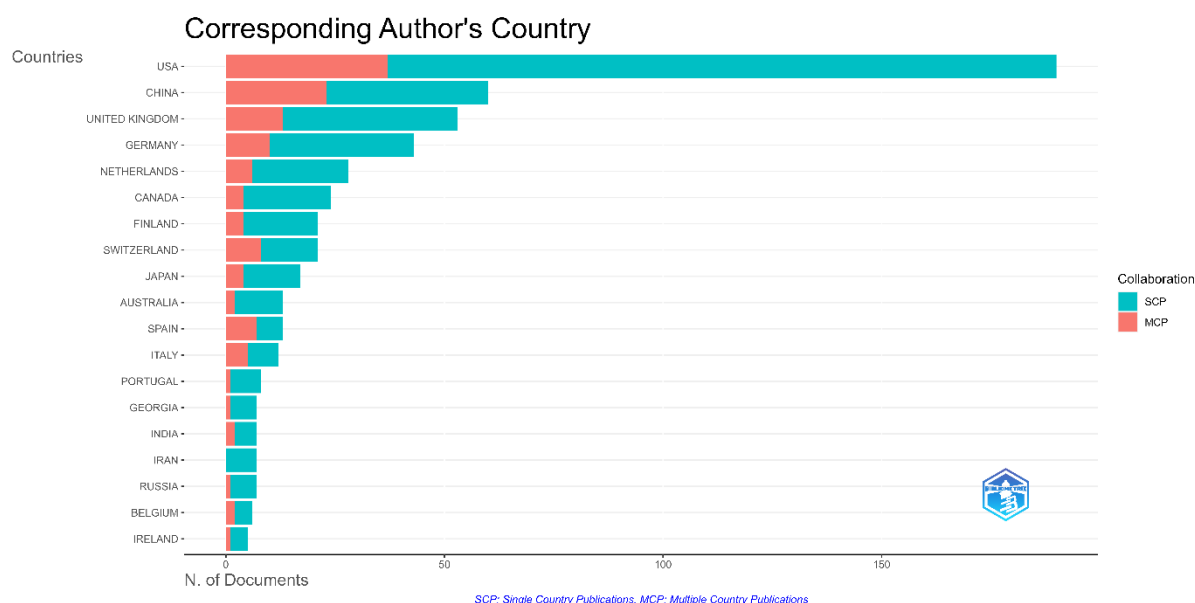


Figure 4.10. Country collaboration publication by author's country in the direct air capture area.

In Figure 4.13, shows the cooperation between the nations. To improve readability of the network map, links with a strength of two or lower were omitted; yet each dot in the map highlights that the country have authors that collaborated in the research. South America countries interacted with countries such as Portugal, Spain, United Kingdom and United States. Nations from Africa tend to co-produce with China and other European countries. The major country from Oceania, Australia, has taken part in publications with countries from Asia, Europe and North America. In the European continent, the core of the DAC research is located at the United Kingdom, Germany, Switzerland and Italy.

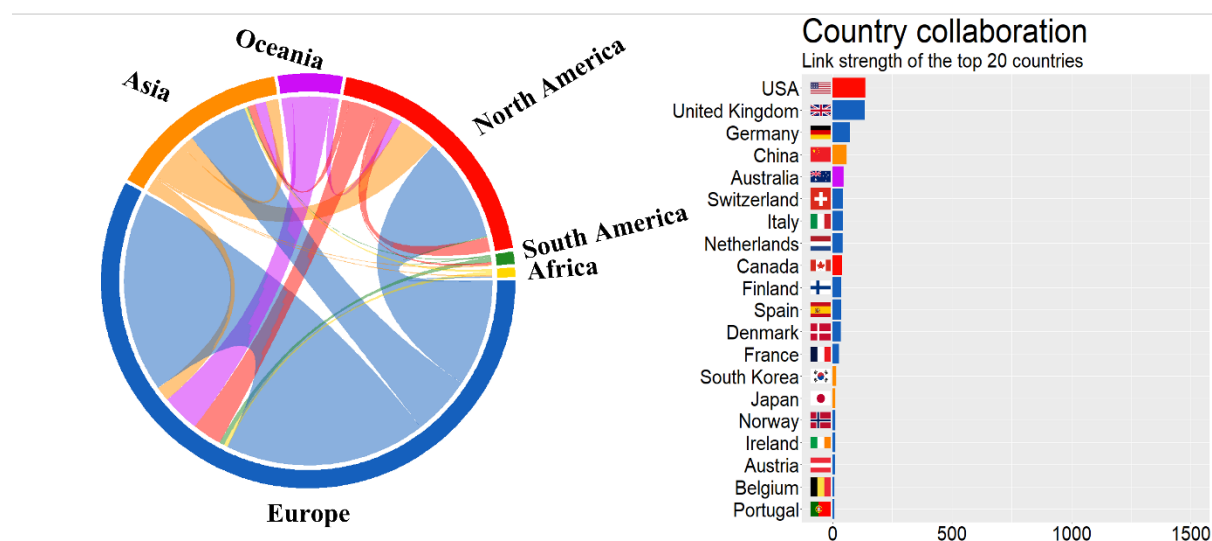


Figure 4.11. Chord diagram of the collaboration between continents and its link strength from the direct air capture area.

Professor Haszeldine from United Kingdom is exploring the implementation CO₂ capture in the industry and for DAC purposes (Alcalde et al., 2018; Brownsort et al., 2016). As well as Miles Sakwa-Novak, Professor Garcia is investigating the effect of the co-adsorption of water using amines in temperature vacuum swing adsorption processes (Young et al., 2021). Together with Kári Helgason from CarbFix (CarbFix, 2022), these researchers explained the benefits of implementing a DAC industry in the country. Therefore, CCS and NETs such as DAC will improve the three fundamental pillars of sustainability of Scotland (Rebecca Bell & Nicholas Ashmore, 2022). The United Kingdom has included multiple DAC projects in their national development plans (Department of Business Energy & Industrial Strategy, 2022). Some of them are exploring the synergies of DAC powered by nuclear energy. Others, as Mission Zero technologies, are currently designing a pilot plant. Bluebox Energy aims to deploy a DAC technology within the communities and farms or greenhouses to produce biochar. All these projects are still in developing phase, yet they are expected to scale in the coming years.

Total CO₂ emissions in the European Union have been reduced by 1 billion of tonnes during the last ten years (Figure 4.12). It is important to separate emissions reduction and Carbon Dioxide Removal (Climeworks, 2023). By sharing the same goal, both approaches are expected to fulfil different objectives in the climate crisis. Thus, DAC + S and CCUS are predicted to play relevant roles. Yet, countries as for example, Germany, are one of the most fossil fuel dependent due to its industry and therefore, its emissions are between the highest. Social acceptance is a key element in the DAC research and Germany's budget in these technologies were exceptionally low in the past (Viebahn et al., 2019). However, this was noticed, and the government explored and boosted this solution (Block, 2022).

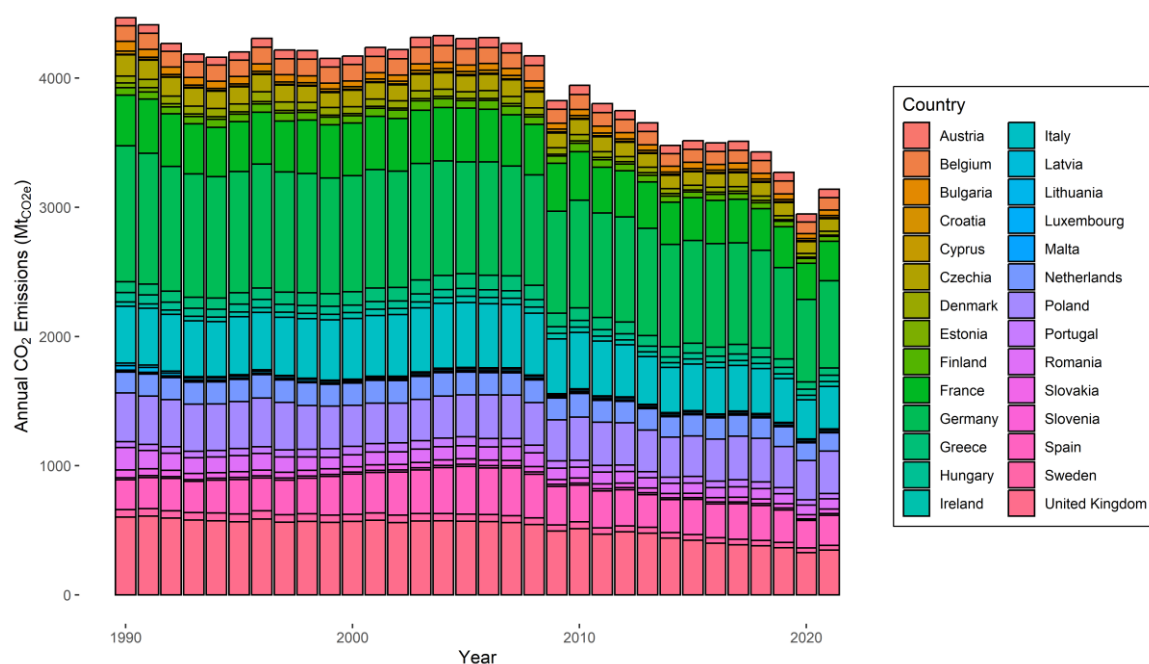


Figure 4.12. EU-28 CO₂ emissions from 1990 to 2021 (Land uses are not included). Source: (Ritchie Hannah & Roser Max, 2022)

Switzerland was the country where the company Climeworks researched DAC technologies. ORCA was the first scaled DAC plant in the world (Climeworks, 2021), and currently, they are building its second plant, Mammoth (Climeworks, 2022c). This could be the main reason of why numerous countries collaborated with Switzerland. Together with the United States, they investigated materials and techniques to reduce the associated cost of the process. In addition, the leading audit and advisory company in Switzerland is committed to not only support initiatives to switch towards renewable energies, but it also aims to reduce the concentration of CO₂ from the atmosphere (Climeworks, 2022e).

At Troia, in Italy, the STORE&GO project has three demonstration facilities (Store&Go, 2018). The aim of this plant is to produce liquified renewable gas. They combine a micro methanation reactor with a liquefaction unit. The CO₂ for this process is obtained via DAC. The plant takes advantage of the high PV production capacities available in the region. They produce and transform the excess of renewable energy in other form for the sectors that cannot easily switch to renewable energies. This is an obvious example of how Ireland can take advantage of the wind production.

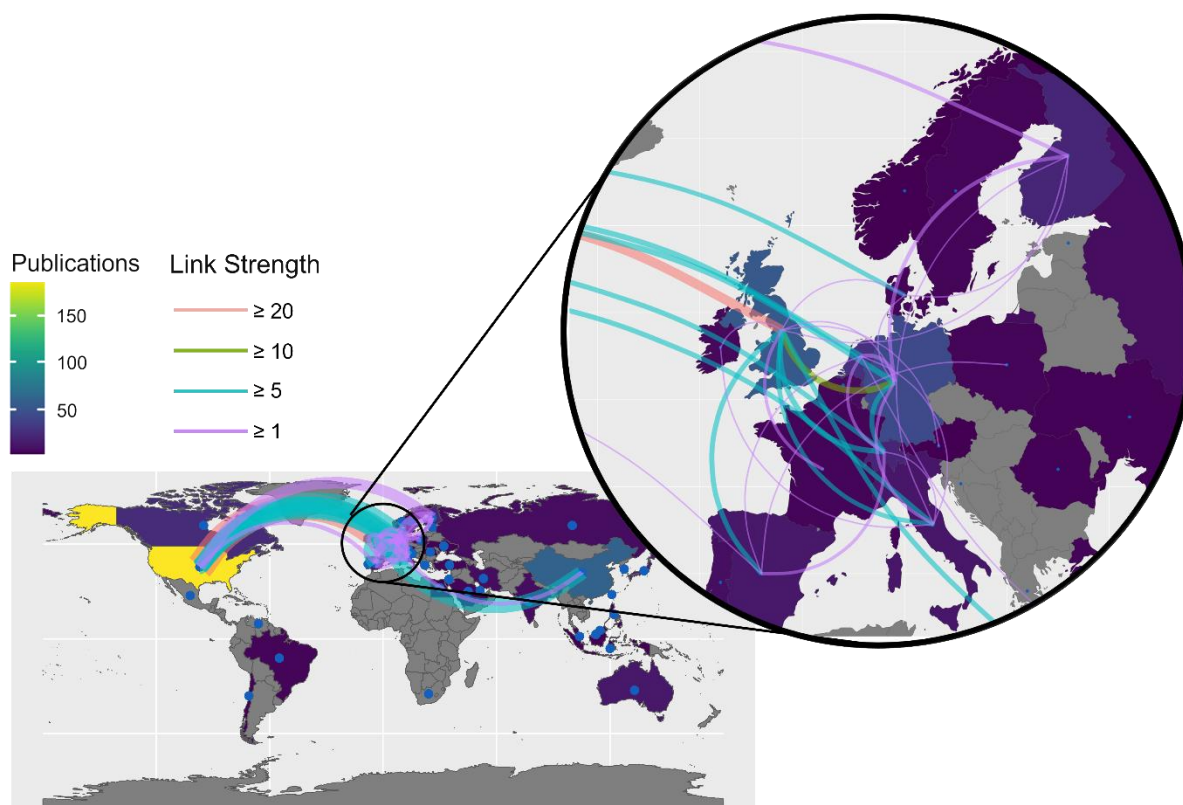


Figure 4.13. Link strength between countries with publications related to DAC. Each dot represent that authors from the country has collaborated in DAC publications with other nations. The link spectrum colour represent the strength between the countries; from modest (violet) to strong (red).

In Ireland, only 8 publications related to this field were identified, despite the presence of a DAC company working in this area, Neg8 (Casaban & Tsalaporta, 2022). The first paper is a collaboration with Professor Breyer (Breyer et al., 2022). In this work, the authors explain the synergies between renewable energy and CDRs, such as DAC. The potential of producing synthetic fuels for the sectors that cannot decarbonise is essential for a net-zero future. Science Foundation Ireland supported the work of Dr Realmonte and enhanced the links between Italy and Ireland. In her first work, researcher Realmonte explains how DAC can help the countries' climate targets (Realmonte et al., 2019). To achieve it, she points what is necessary to scale a DAC plant: research and innovation in solid sorbents (Chatterjee & Huang, 2020). Yet, they also mention that the manufacturing of these materials cannot be compared to the current commercialisation of sorbents. They are confident that research towards this field will reduce the costs and DAC should be considered a solution in the near and long term. Thus, investigating new materials is on the research interests from Dr Madden (Madden et al., 2017). He has been studying physisorbent materials such as Zeolite 13-X, SBA-15 or Mg-MOF-74, among others (Kumar et al., 2015). However, the work from Professor MacElroy was not included in the search. He also studied materials to reduce the concentration of CO₂ from the

atmosphere (Tsalaporta et al., 2017). As well as him, there could be authors that are difficult to find their work, and this is one limitation of these analyses and the reason of setting a common word.

4.4 Conclusions

Our previous work was limited by the use of Scopus as database. Another restriction was the focus on solid sorbents for adsorption processes; therefore, there were only 90 articles related to DAC from a total of 952. During this bibliometric study, we tried to address every known paper by highlighting these limitations. Thus, the total number of articles increased to 8315, of which 600 focused on capturing CO₂ from the air. It could be observed that the number of countries investigating these areas was expanded. Some articles may refer to “direct air capture” simply as “carbon capture” or “CO₂ capture,” using the terms interchangeably. As a result, relevant work focused on DAC could be overlooked in this study. Both methods are necessary to mitigate climate change. Yet it is essential to separate the concepts in order to find the desirable information. This was the main reason of filtering the 8315 documents with a second search. Further studies with CO₂ air removal purposes should include the word “Direct Air Capture.” This will help other authors to find their studies in wide spectrum of “carbon capture.” However, the trend is clear and in the last few years, the research towards DAC has increased, and numerous studies from authors of different countries are contributing to reduce this gap to scale this technology.

All their work is extraordinarily complex to summarise in this article, and their research is not underrated. However, it was important to highlight researchers such as Professor Klaus, one of the first people to stress the importance of reducing the atmospheric CO₂. Investigating new methods and materials in this decade is crucial for developing this technology in the near future. Climate change is not the problem of one nation. As well as in our first study, the United States is the country with the major number of publications on the topic. As stated on multiple occasions through these series of studies: research and investment will help to reduce the breach of this technology. The government of Ireland should not turn a blind eye and include this solution in its National Development Plan.

Chapter 5: Life Cycle Assessment of a Direct Air Capture and Storage plant in Ireland

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Life cycle assessment of a direct air capture and storage plant in Ireland

[Daniel Casaban](#)  & [Elena Tsalaporta](#) 

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5 Life Cycle Assessment of a Direct Air Capture and Storage plant in Ireland

Abstract

Despite the efforts to transition to a low carbon economy, greenhouse gas emissions are surging to critical levels. Carbon Dioxide Removals (CDR) methods, such as Direct Air Capture (DAC), have been gaining substantial public attention in the last few years. DAC is essential in curbing CO₂ concentrations and achieving climate targets. It is said that DAC can be deployed anywhere, but thorough a life cycle assessment (LCA) is imperative to prove its viability. Therefore, this paper aims to explore the feasibility of constructing a 1 Mt_{CO2} plant in Ireland, using Kinsale and Corrib gas fields as storage points. The results showed that the country is an ideal candidate for scaling up this emerging industry. The efficiency is primarily influenced by the construction of the pipeline section, given a reliable source of heat and electricity. The study highlights the significant impact of distances to the storage points on feasibility, favouring counties near of the gas fields. In conclusion, Ireland has the potential to establish its own DAC industry.

Keywords: *Direct Air Capture, Climate change, Carbon Capture and Storage, Carbon Dioxide Removal, Ireland.*

5.1 Introduction

Numerous countries have formulated strategies to tackle climate change; however, the world continues in track towards surpassing 2°C threshold above pre-industrial levels (Diffenbaugh & Barnes, 2023). Global Greenhouse Gas (GHGs) emissions rebounded after 2020, reaching a staggering 52.8 GtCO_{2e} (Anne et al., 2022), which is affecting our society (Marina et al., 2022). Low carbon emitting technologies (LCET) are anticipated to serve as a bridge towards a net-zero economy (World Economic Forum, 2022). In line with this, the Intergovernmental Panel on Climate Change (IPCC) has recommended the adoption of Carbon Dioxide Removals (CDRs), such as Direct Air Capture (DAC), to remove 6 GtCO₂ annually by 2050 (R. Shukla et al., 2022). These CDR methods are considered essential in addressing the ongoing climate crisis.

Over the last two decades, scientists have devoted significant research efforts on investigating adsorption and absorption techniques to extract CO₂ from the atmosphere (Casaban & Tsalaporta, 2023b). In adsorption processes, ambient air is passed through a solid material with a high affinity for retaining CO₂, while absorption involves the use of an aqueous solution (Casaban et al., 2022). Notably, one of the key advantages of absorption over adsorption is the capacity to operate under continuously (Keith et al., 2018). Conversely, adsorption excels in its ability to perform the process with reduced water and energy needs (Casaban et al., 2022). Both methods have been demonstrated feasible, leading to the construction of the first industrial plants by certain companies, with ongoing developments in this field (Climeworks, 2021; Matt McGrath, 2021).

In order to execute the process and mitigate the most severe effects of climate change, a substantial portion of the captured CO₂ must be stored (IEA, 2007). Depleted oil and gas fields emerge as one of the most effective solutions for CO₂ storage (Hannis et al., 2017). Lessons and insights gleaned from prior experiences demonstrated the feasibility of this approach (IEAGHG, 2023). Additionally, deep saline aquifers have been investigated as geological formations suitable for CO₂ storage (Celia et al., 2015). These reservoirs are estimated to possess larger storage capacities compared to oil and gas fields (IPCC, 2005). However, the storage capacity of these geological formations hinges on the specific characteristics of the aquifer and the complexity of the storage operation (Bachu, 2015).

Structural storage involves the underground pumping of CO₂; because of its lighter density compared to water, the gas ascends through the porous rock until it reaches the top of the geological formation, where it becomes trapped and securely sealed (Iglauer, 2018). In the case of saline aquifers, CO₂ storage is achieved by a process known as saline trapping. In this case, rocks act as a sponge, where the injected CO₂ remains trapped within the pore spaces (Joodaki et al., 2020). To enhance its stability, CO₂ can be dissolved into saline water to increase its density and causing it to sink to the lowermost regions of the rock formation, thereby permanently entrapping the CO₂ (Irfan et al., 2018). Moreover, CO₂ dissolved in saline water exhibits reduced acidity and can react with minerals in adjacent rocks, facilitating the formation of new minerals (Gadikota, 2021). Currently, numerous projects and experiments are being investigated in Iceland, where they are storing CO₂ in basalt rocks (Matter et al., 2011). Furthermore, some studies suggested converting CO₂ into cement through mineral carbonation using Ca⁺ and Mg⁺ rich materials (Winnefeld et al., 2022).

Thus, the DAC industry is poised to grow its capacity in the current decade. As previously mentioned, the primary objective of this emerging industry is the permanent storage of CO₂. Nevertheless, certain sectors are expected to keep emitting GHGs in the coming years. In response, the captured CO₂ can be re-utilised to offset emissions from agriculture or be synthesised into alternative fuels for aviation or chemical applications (Casaban & Tsalaporta, 2022). To comprehensively evaluate the efficiency of this process and its environmental implications, a Life Cycle Assessment (LCA) is essential for gaining insights and know the effectiveness of the overall process (Daggash et al., 2018; van Leeuwen & Mulder, 2018).

Climeworks is expected to open a new DAC plant in Iceland, taking advantage of the surplus of geothermal energy (Climeworks, 2022c). Their LCA study highlighted carbon dioxide removal efficiencies of 90%, affirming the efficacy of their process (Deutz & Bardow, 2021). The adaptability to build a DAC plant in any location depends on the accessibility to a rich renewable energy source (IEA, 2022a). Moreover, it is imperative to recognise other factors during its construction and operation that can affect the performance of the CO₂ capture. As Terlouw et al. stressed, the transportation and storage of CO₂ can have detrimental impacts to the environment (Terlouw, Bauer, et al., 2021). The necessary infrastructure for gas transport, the drilling and injection operations, and the possible CO₂ leakage, can collectively elevate the energy demands, thereby contributing to a higher carbon footprint (Volkart et al., 2013).

The Republic of Ireland has the EU's fastest-growing economy, yet its national development plans did not take into account DAC solutions (Casaban & Tsalaporta, 2022). Ireland overarching objectives is to reduce its carbon emissions by transitioning from fossil gas to renewable energy sources within its electricity grid (SEAI, 2011). Additionally, the government aspires to reduce transport emissions by introducing one million electric vehicles by 2030, while heat pumps are expected to replace oil and gas in the residential sector. These transformative initiatives have the potential significantly reduce the country's reliance on fossil fuels. Nonetheless, it is noteworthy that no LCA study has been conducted to highlight the importance of DAC process in future scenario projections.

From this perspective, the objective of this LCA study is to evaluate the environmental performance of a DAC facility in Ireland by analysing potential implementation sites. The goal is to identify which locations offer the most favourable conditions in terms of emissions and energy requirements. In addition, the country has two potential storage sites the examination of which will provide insights to understand these previously mentioned factors associated with the capture and storage processes (Casaban & Tsalaporta, 2022). A previous LCA from Climeworks underscores the necessity of a low carbon source to power the plant (Climeworks, 2022e). While Ireland lacks geothermal energy resources, it boasts consistently robust wind speeds, offering the potential to generate enough energy to exceed the domestic demand (SEAI, 2011).

5.2 Methodology

During this study, SimaPro was the software utilised for the comprehensive assessment of materials, energy requirements, and environmental impacts throughout the construction and operation of a DAC plant in Ireland. It is important to remark that some of the system boundaries were influenced by the data confidentiality inherent to this emerging technology. The DAC process technique for this study is based on the Climeworks plant, specifically, the Temperature-Vacuum Swing Adsorption process. In the absence of information on air humidity in the referenced studies, the feed air was assumed to be dry. Therefore, this reliance constitutes a notable limitation within the context of this work. To achieve a more detailed analysis, it is crucial to examine all the engineering processes behind of a real DAC plant.

As well as to many industrial processes, the operation of a DAC plant will be influenced by the energy source employed. In order to evaluate the climate change impacts, we utilised the EF 3.0 Method, which incorporates the Global Warming Potential to 100 years (GWP100) as an indicative metric for quantifying the GHGs in terms of CO₂ equivalent (European Commission, 2013). Thus, the primary goal of this study is to quantify the kg of CO₂ emitted per ton of CO₂ removed, encompassing all phases of the process: construction, operation, and storage. In order to assess the carbon efficiency, we established three thresholds at 25%, 50% and 75%. Additionally, we followed the methodology and guidance provided by Terlouw to expose our study case (Terlouw, Treyer, et al., 2021). In this context, and due to data confidentiality, the authors made assumptions to approximate the impacts from the construction of the DAC plant: CO₂ collectors, process unit, steel tanks and a maintenance hall, for control the process.

5.2.1 Construction of a DAC plant

The Collector boxes are composed of materials such as plastic, steel, insulation, and fan, which is remarkably similar to the Life Cycle Inventory (LCI) of a typical passenger vehicle (Van Der Giesen et al., 2017). In account of this similarity, we adopted their approximation as a basis for our assessment, and we selected a 1200 kg compact size petrol/natural gas car as a proxy for the collectors. As stated by Climeworks, 80 collector boxes have the capacity to capture 4 ktCO₂ per year in the plant located at Iceland (Climeworks, 2021). Each collector measures 2 meters in width, height and length, resulting in a total volume of 640 m³, with a corresponding density of 110 kg/m³ (Climeworks, 2022e). These assumptions provided a base for the evaluation of the collector boxes during this study.

In addition to the collector boxes, a passenger vehicle was used as a representative approximation for the process unit. A DAC unit is comprised by two containers, each of which accommodates 9 collector boxes. These containers have a length of 12 meters and a width and height of 2 meters. Collectively, the total volume of the process unit is estimated to be 50 m³. In the case of the DAC plant from Climeworks called ORCA, it needs 9 process units, amounting a total volume of 450 m³ in order to capture 4 ktCO₂ per year (Climeworks, 2021). Additionally, the infrastructure requires the inclusion of one steel tank per 18 collector boxes. These steel tanks have a diameter, height, and thickness of 2 meters, 6 meters and 0.02 respectively.

The construction of the DAC plant requires the transformation of grassland into an industrial area, overlaying it with concrete for the foundations (Viebahn et al., 2019). Climeworks reports a requirement of 90 m² for the DAC plant located at Hinwil (Climeworks, 2022a). Terlouw et al., linearly scale the land use for both, the ORCA plant and a prospective 100 kt plant (Terlouw, Treyer, et al., 2021). Their findings indicated that, for the 4 kt_{CO2} and 100 kt_{CO2} plants, concrete layers of 400 m² and 5000 m², respectively, each with a thickness of 1 meter are imperative. Moreover, to ensure the structural integrity, the concrete should be reinforced with 120 kg steel/ m³ (Deutz & Bardow, 2021).

While the exact dimensions of the maintenance hall remain unclear (Viebahn et al., 2019), Terlouw et al. estimated that this structure should occupy 75% of the total surface area. To validate the accuracy of our results -Figure 5.1, a comparative analysis was conducted with the data from Climeworks and Terlouw. In their research, they reported figures of 14.4 and 13.5 kg of CO₂ emitted per ton capture for the 4kt_{CO2} plant, and 5.8 and 6.7 kg of CO₂ emitted per ton capture for the 100 kt_{CO2} plant. The results obtained in this study aligned with those reported in the literature, indicating emissions of 14.5 kg of CO₂ per ton captured for a 4kt_{CO2} plant and 6.3 kg of CO₂ per ton captured for a 100kt_{CO2} plant. Based on these results, this study scaled the LCI (Table 5.1) to reflect the requirements of a megaton-scale DAC plant. Notably, future installations are anticipated to require less quantities of materials and land use, driven by advancement in the field such as improved techniques or the development of novel sorbents (Sanz-Pérez et al., 2016).

Table 5.1. LCI data for the construction of DAC plants with capacities of 4,000 tCO₂/year and 1MtCO₂/year. The inventory includes material and energy inputs such as concrete, steel, chemicals used as well as land transformation and land occupation (m² per year), which accounts for the duration and area of industrial land use.

Product	Processes	Materials/Assemblies	4kt	1Mt
Collectors	Passenger car, petrol/natural gas	-	70,400 kg	880,000 kg
Process Unit	Passenger car, petrol/natural gas	-	49,500 kg	611,000 kg
Foundations/Hall	Occupation, industrial area	-	48,000 m ² a	116,000 m ² a
	Transformation, from grassland, natural	-	400 m ²	10,500 m ²
	Transformation, to industrial area	-	400 m ²	10,500 m ²
	-	Steel, low-alloyed	48,000 kg	748,490 kg
	-	Concrete, normal	400 m ³	306,770 m ³
	Control building	-	120 m ²	3,000 m ²
Tanks	-	Steel, low-alloyed	34540kg	381,000 kg

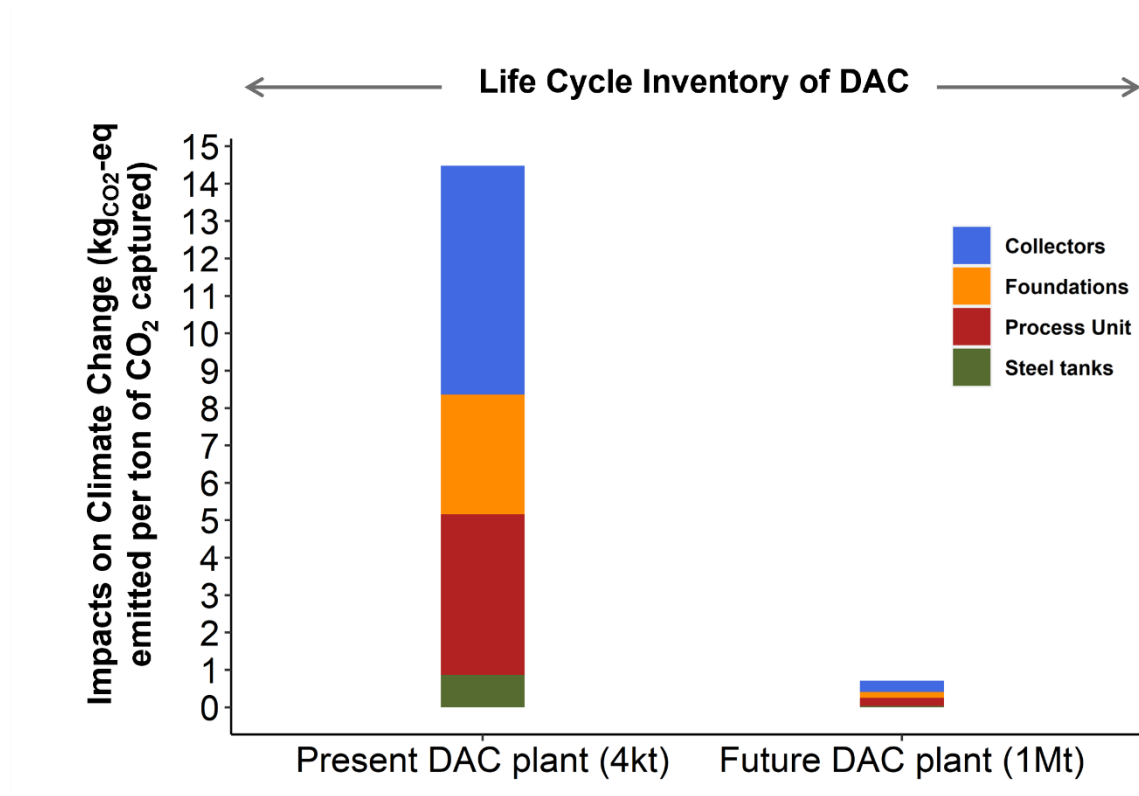


Figure 5.1. Impacts on Climate Change from the Life Cycle Inventory for a DAC plant with of 4kt and megaton capacity.

5.2.2 Operation of a DAC plant

According to Climeworks, their plant currently requires of 500 kWh of electricity and 1500 kWh of heat to operate (Climeworks, 2022a). However, recent studies suggested that ongoing improvements in the process will reduce the energy needs to 444 kWh of electricity and 1333 kWh of heat per ton of CO₂ captured -Figure 5.2- (Hanna et al., 2021). In order to meet the heat demands, our assumption is that the necessary energy for the process can be sourced from a chemical industry or from a waste incineration plant, such as the one located in Dublin (Elsam, 2006). In 2020, approximately 3.2 million tonnes of municipal waste were generated, with 41% of it being transformed into energy (EPA, 2023). The availability of a source of waste heat has the potential to significantly reduce emissions and environmental impacts associated with the process (Fasihi et al., 2016). For the electricity needs, we hypothesised that the renewable energy predominantly originates from wind sources, while non-renewable is sourced from gas plants within the island. This scenario is premised on the expectation that the country's sole coal plant of the country will cease its operations in the near future (Barry O'Halloran, 2022a). However, should Moneypoint coal plant continue to operate beyond 2030, it would result in an increment of the CO₂ emissions from the electricity

requirements, consequently amplifying the associated environmental impacts. Thus, the scenarios from our study are based on the future development plans of the country (Government of Ireland, 2021).

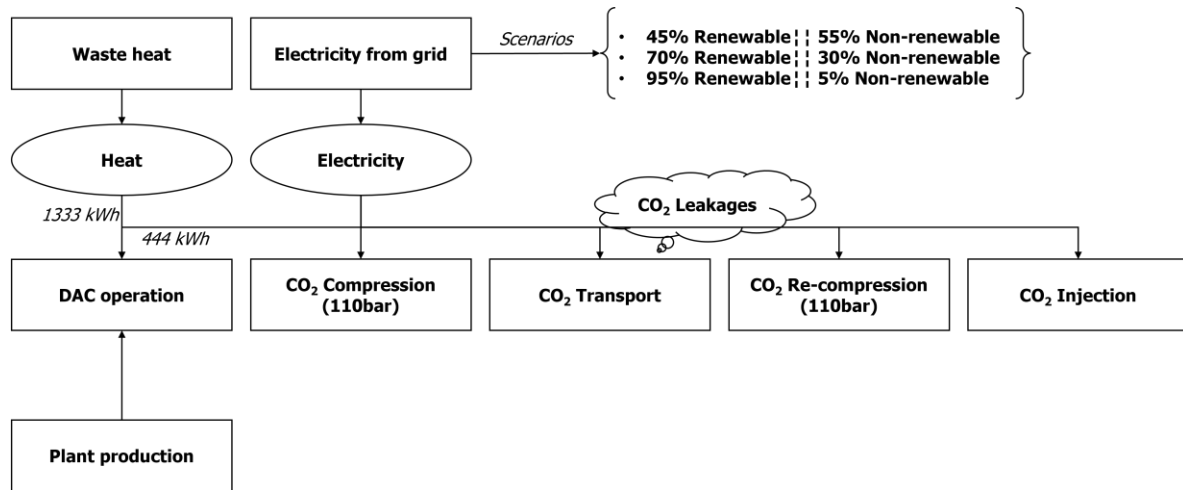


Figure 5.2. Diagram of a DAC+S according to every scenario

- Scenario 1: 55% of the energy comes from fossil origin and 45% from renewable.
- Scenario 2: 30% of the energy comes from fossil origin and 70% from renewable.
- Scenario 3: 5% of the energy comes from fossil origin and 95% from renewable.

5.2.3 CO₂ storage

According to Hendriks et al. (Chris & Wina, 2004), there is a method to estimate the energy needs for the injection step. To transport the CO₂, the gas should be compressed from 1 bar to 110 bar in a four-step centrifugal compression (Musardo et al., 2013). If the capacity of the plant is of about 1 MtCO₂ per year with a Flh value of 8160, the mass flow of the process is 34 kgCO₂/s or 122.4 tCO₂/h. All of these parameters are considered in Equation 5.1, where Ce is a constant with a value of 87.85 kJ/kg.

$$E = C_{el} * \ln \left(\frac{P_{outlet}}{P_{inlet}} \right) * F \quad \text{Equation 5.1}$$

The results indicated that a total of 115 kWh of energy is needed for the combined compression stages. Moreover, an additional input of 12 kWh is required to inject the CO₂ to a depth of at least 2000 meters underground. Consequently, the cumulative energy input from the transport and storage process amounts approximately 130 kWh per ton of CO₂ removed. This energy will be provided depending on the specific energy mix associated to the established scenarios.

In alignment with Terlouw approach (Terlouw, Treyer, et al., 2021), we considered the potential fugitive emissions of CO₂ during its transportation within of the pipelines, categorised as a medium baseline scenario. Moreover, Holloway et al. developed a method to estimate the loss of CO₂ during its transmission (Holloway et al., 2006). According to their research, the CO₂ emissions rate is 1.66 times that of the CH₄-emission rate. Thus, this rate is multiplied by the annual distribution emission factor of CH₄ to calculate the estimate losses of CO₂ for the different DAC plants across the country (Table 5.2) (Carras Pamela M Franklin et al., 2006). It is noteworthy that the maximum depth of Kinsale gas field is 1000 meters, while Corrib can reach depths of up to 3000 meters (Table 5.3). However, storage depths around 2000 meters have negligible effects on the LCA (Institut & Volkart, 2013). Therefore, the potential CO₂ leakage from the injection wells is deemed insignificant in the case of Kinsale (Kelemen et al., 2019).

Table 5.2. Leakage emissions from transporting CO₂ to Kinsale and Corrib via pipeline.

DAC plant	Distance to Kinsale gas field (km)	Distance to Corrib gas field (km)	Leakage to Kinsale gas field (kgCO ₂)	Leakage to Corrib gas field (kgCO ₂)
Castlebar	316	128	45	18.23
Donegal	393	195	55.96	27.77
Dublin	260	323	37.02	46
Kinsale	74	336	10.54	47.85
Tipperary	150	281	21.36	40.01
Wexford	171	377	24.35	53.69

While there are numerous potential CO₂ storage locations in close proximity to Ireland, with a capacity to store 93,115 MtCO₂ -Figure 5.3-, it is important to remark that only a relative small portion of these sites can be considered practical or effective at the moment (1,505 MtCO₂) (Lewis et al., 2009; SEAI, 2008). Therefore, due to the existing pipeline infrastructure, there is enough data to support the selection of Kinsale's gas field as favourable location for CO₂ storage (Gas networks Ireland, 2020).

Moreover, the depletion of the Corrib gas field in the coming years presents a dual challenge and opportunity for Ireland. It poses a potential issue for national energy security, but simultaneously offers an opportunity to transition towards renewable energy sources of energy as well as bolstering a local DAC industry (Casaban & Tsalaporta, 2022; Department of Environment, 2022). While the prospect of storing the abated CO₂ in other locations remains viable, it is imperative to address both political and technical challenges before its implementation. For instance, the UK government has plans to utilise the Irish Sea Basin to store CO₂ (Kirk, 2005). The basin particularly located at Lough Neagh, is one of the most important wetland habitats in Europe and provides multiple ecosystem services (Louise Cullen, 2023). It is important to emphasise that the data pertaining to Lough Neagh only considers the effective capacity of a small area in the basin, and uncertainties exist regarding the thickness of the seal layer, which is essential for an effective CO₂ storage (SEAI, 2008). Therefore, given its significant ecological importance, any consideration of Lough Neagh Basin for CO₂ storage will require an exhaustive and meticulous study to assess potential impacts on this natural ecosystem.

The concern of seismic activity during the CO₂ storage has been a recurring topic of discussion and has been widely explored in the past (Jonathan O'Callaghan, 2018). In 2026, Scotland is going to build the first DAC plant with a megaton capacity (Matt McGrath, 2021). This particularly plant relies in absorption processes, but it is worth noting that solid sorbent DAC plants are rapidly progressing and may soon achieve similar capacities (Climeworks, 2022d; Ella Nilsen, 2023). In light of this, we have assumed that the plant of this current study has a capacity to remove 1 MtCO₂ per year. Furthermore, this study suggested six potential locations for a DAC plant deployment across Ireland: Kinsale, Tipperary, Wexford, Dublin, Castlebar, Donegal.

Table 5.3. Characteristics from some of the practical and effective storage basin locations in Ireland.

Site name	Location	Storage type	Status	Storage capacity (Mt _{CO2})	Reservoir type	Reservoir depth (m)
Kinsale	Celtic Sea Basin	Gas field	Depleted	350	Triassic sandstone	800
Spanish Point	Porcupine Basin	Gas field	Depleted	120	Jurassic Voligian Sandstones	-
Corrib	Northwest Basin	Gas field	Operational	6950	Lower Cretaceous sandstone	2000
East Irish Sea Basin	Irish Sea Basin	Aquifer	-	630	Triassic sandstone	900
East Irish Sea	Irish Sea Basin	Oil & Gas field	Depleted	1050	Triassic sandstone	900
Kish Bank	Central Irish Sea Basin	Aquifer	-	267	Sherwood Sandstone	1750
Lough Neagh	Northern Ireland	Aquifer	-	1940	Sherwood Sandstone	1300

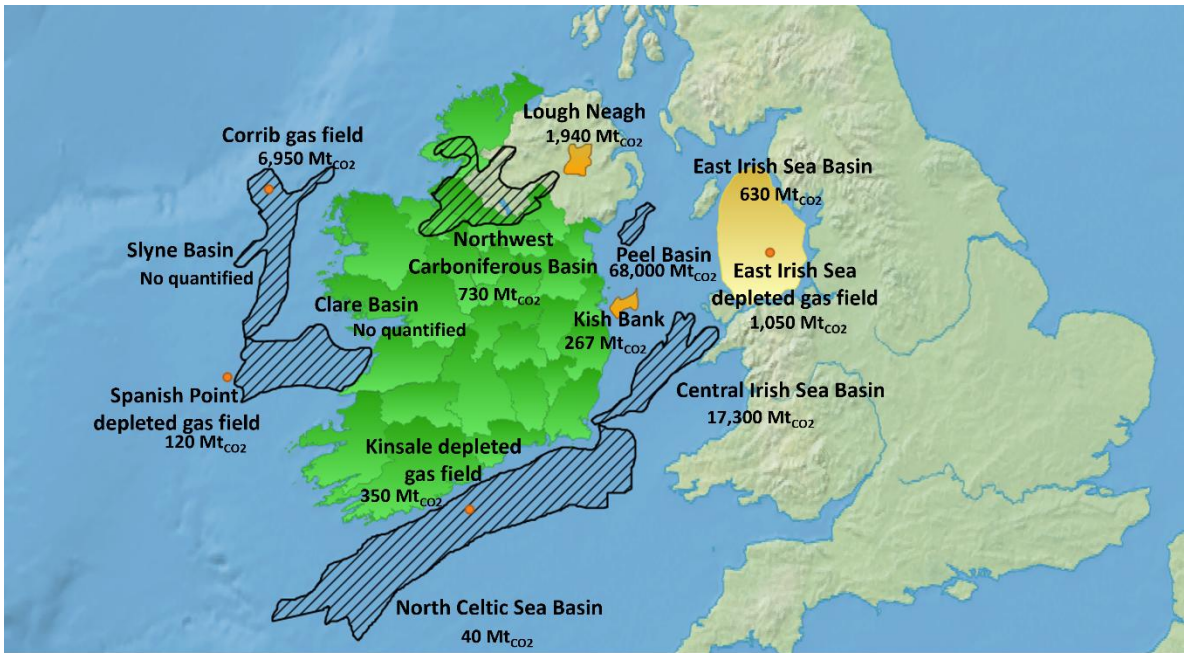


Figure 5.3. Most important storage points near of Ireland. The points represent the oil & gas fields. The hashed basins represent the capacity in theory, and the coloured ones represent the effective CO₂ storage capacity.

5.3 Results and discussion

The displayed results serve to demonstrate the feasibility of establishing a DAC plant in the country. It is noteworthy that solid sorbent processes only require modest levels of energy consumption. However, this study has some limitations due to the lack of detailed data from pilot plants, such as real energy consumption for background processes. One relevant example

is the impact of moisture in the air. The presence of water in high humidity climates such as Ireland, can increase the system's energy demand because of an additional dehumidification step to ensure an effective CO₂ purification. Moreover, certain solid sorbents exhibit a significant decrease in its adsorption capacity when exposed to high levels of humidity, affecting the overall CO₂ capture performance. Currently, Climeworks' DAC facility using solid sorbents has demonstrated a nameplate gross annual capture capacity of 36 ktCO₂ (Climeworks, 2022c). This represents a milestone from the 4 ktCO₂ per year capacity of its previous plant (Climeworks, 2021). This increase over just two years highlights the rapid technological process, anticipating that within the next decade, these plants could achieve the megaton-scale capacity (Matt McGrath, 2021). Even so, Climeworks has reported net removal efficiencies ranging from 75% to 99%, meaning that for a 4ktCO₂ plant like Orca, the actual net removal could be around 3ktCO₂ per year (Climeworks, 2024). Currently, the only DAC plants constructed are strategically located areas endowed with a high presence of renewable energy resources, and the storage points are situated close to the capture plant. Ireland, in alignment with sustainable goals, is moving towards low carbon energy sources energy, and in the near future, the country expect to meet 70% of its electricity demand by renewable sources. Recent developments, such as the successful first major auction to implement offshore wind power, mark a significant milestone of a promising future within the power sector (Shortt, R. 2023). Nevertheless, it is imperative to remark that renewable energy have certain limitations and addressing emissions from sectors that are finding difficulties need CDR and carbon capture methods.

Table 5.4. Summary of environmental impacts related to DAC operation related to the best and worst locations for the different storage points. This includes multiple impact categories beyond GWP100, enabling a broader assessment of environmental trade-offs.

		Unit	Kinsale storage		Corrib storage	
			55%FF/45%RE	5%FF/95%RE	55%FF/45%RE	5%FF/95%RE
Environmental impact			Scenario 1/Donegal	Scenario 3/Kinsale	Scenario 1/Kinsale	Scenario 3/Castlebar
	Climate change	kgCO _{2eq}	3.43E+02	1.09E+02	3.19E+02	1.57E+02
Ozone layer	Ozone depletion	kgCFC11 eq	4.51E-05	2.41E-05	4.29E-05	2.86E-05
	Photochemical ozone formation	kgNMVOCeq	1.43E+00	4.79E-01	1.27E+00	8.07E-01
	Particulate matter	disease inc	2.82E-05	9.82E-06	2.48E-05	1.66E-05
Human toxicity	Non-cancer	CTU _h	5.02E-06	2.13E-06	4.47E-06	3.25E-06
	Ionising radiation	kBqU-235eq	3.79E+01	2.23E+01	3.66E+01	2.49E+01
	Cancer	CTU _h	8.23E-07	2.74E-07	7.19E-07	4.85E-07
	Acidification	mol _{H+eq}	1.43E+00	6.08E-01	1.29E+00	9.00E-01
Eutrophication	Freshwater	kg _{Peq}	7.18E-02	3.45E-02	6.47E-02	4.87E-02
	Marine	kg _{Neq}	4.48E-01	1.52E-01	3.98E-01	2.53E-01
	Terrestrial	mol _{Neq}	4.76E+00	1.61E+00	4.23E+00	2.68E+00
Ecotoxicity	Freshwater	CTU _e	4.90E+03	2.26E+03	4.40E+03	3.26E+03
Resource use	Land use	Pt	1.44E+03	1.17E+03	1.39E+03	1.27E+03
	Water use	m ³	8.56E+01	3.04E+01	7.82E+01	4.51E+01
	Fossils	MJ	8.41E+03	5.09E+03	8.15E+03	5.58E+03
	Minerals & metals	kg _{Sbeq}	2.24E-03	1.32E-03	2.03E-03	1.74E-03

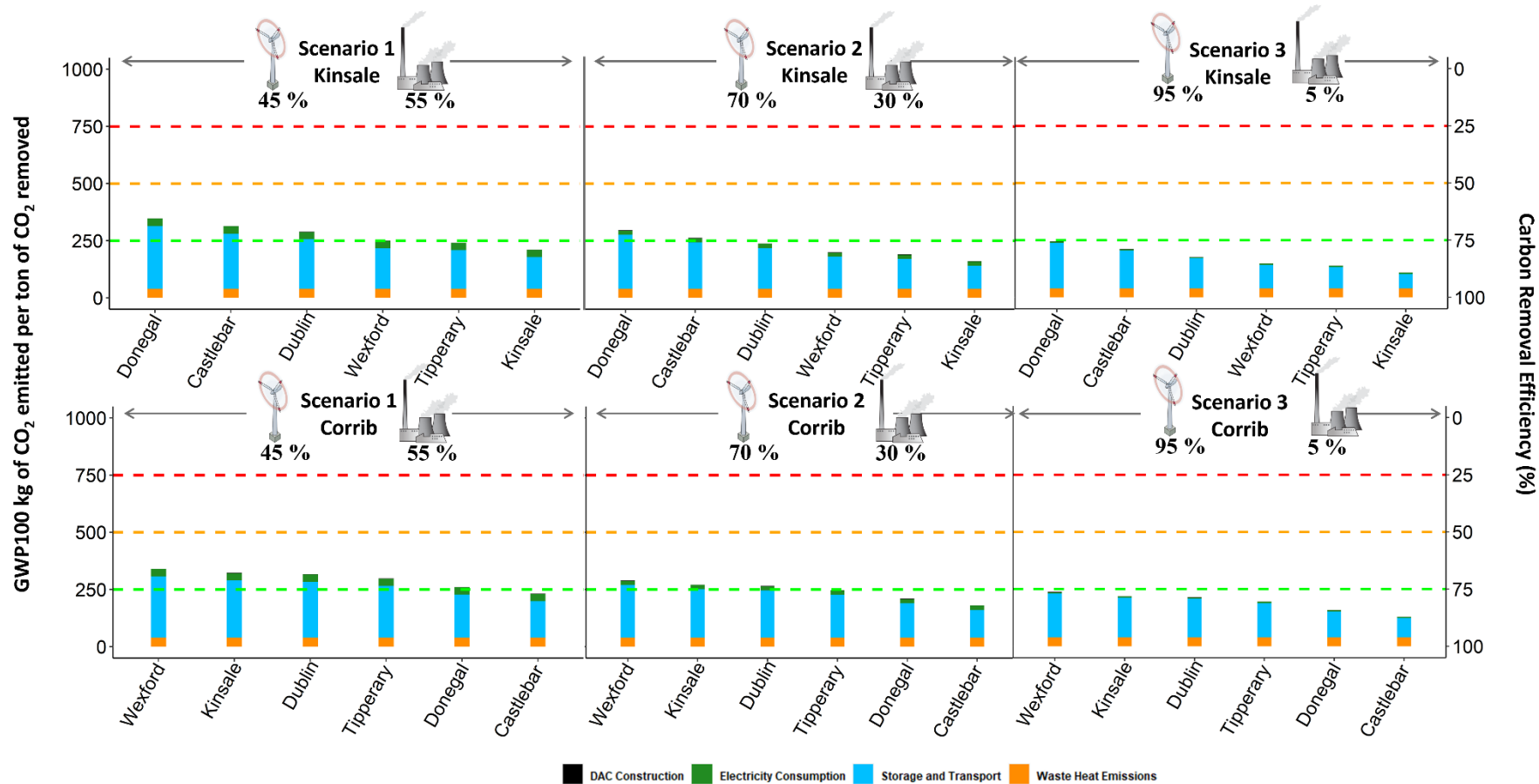


Figure 5.4. Life cycle emissions per ton of CO₂ removed from a DAC plant of one megaton capacity constructed different locations according to each scenario and storage location. The share at the second y-axis represents the efficiency of the process. The colours of the bar display the contribution of each process. In the storage and transport is included the infrastructure requirements and construction impacts.

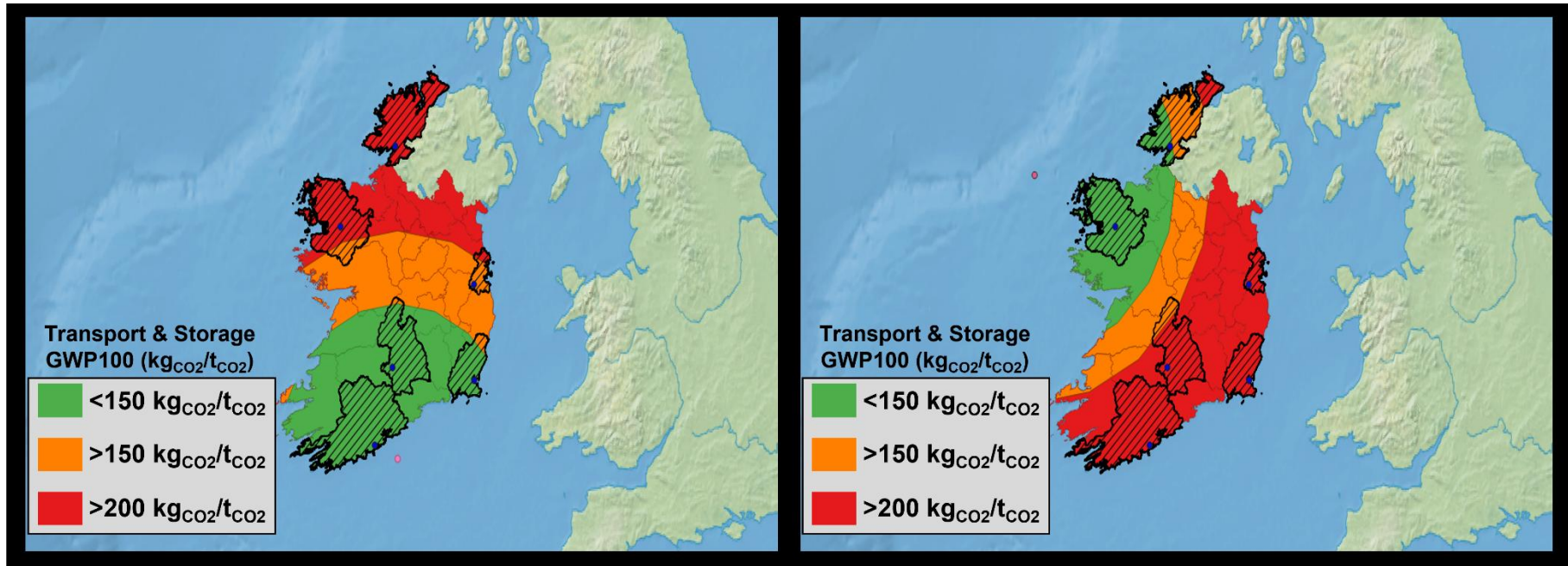


Figure 5.5. GHGs emissions in kg of CO_{2eq} per ton of CO₂ from the plant to the storage place. Kinsale depleted gas field is visible in the first map, located at the south, while the currently active gas field of Corrib appears in the second map, positioned in the northwest. Both storage sites are marked with an orange dot. CO₂ leakage from Table 5.2 during transport is included in the analysis. Counties shaded in red indicate areas where emissions from transportation and storage could reach 200 kg of CO₂ per ton of CO₂ captured.

In our study, conducted across various scenarios within both study cases, it is important to remark that emissions did not surpass the half ton of CO₂ emitted per ton captured as illustrated in Figure 5.4. The construction phase of the plant has a minimum impact during its operational lifetime. Throughout the analysis, it was possible to note a correlation between the source of electricity and efficiency of the plant. Long-term projections, particularly those near Corrib and Kinsale gas fields, indicate a promising trajectory towards enhanced efficiency. As the wind power sector matures in the country, supplying the bulk of the energy demands, it is possible to reach efficiency levels 87% or 89%. Nevertheless, it was revealed that the consequences on the environment from the assembly of the pipeline section had to be taken into account for the future planning of the plant. The energy requisites for both, the operation of the plant and CO₂ transport, were presupposed according to the future development plans (Government of Ireland, 2021). However, it is pertinent to underscore that the emissions from electricity needs can vary depending on the source of electricity. A concurrent study remarks the significance of this important factor to another study and how the carbon efficiency can drop to 10% if the electricity of the grid comes from fossil sources (Cooper et al., 2022). Within their analysis, it was evident that a mere 18.9% energy from renewable sources can affect the whole process. This assertion finds similarities with the work from Madhu et al., who contend that a low-carbon energy source (70 g CO₂eq kWh⁻¹), can elevate the carbon efficiency of the DAC+S system to nearly 86% (Madhu et al., 2021).

The integration of heat derived from waste plants was an important factor in reducing the carbon footprint of the process. Nevertheless, it is imperative to note that these emissions were included in our analysis to comprehensively assess the impacts on the overall process. In locations where waste heat is not available, the emissions generated during DAC operation can reduce the overall capture efficiency of the process (Terlouw, Treyer, et al., 2021). In that case, the consideration of a heat pump becomes pertinent (Deutz & Bardow, 2021). A coefficient of performance of 2.51 is sufficient to meet the operational requirements of the plant, falling within the typical range of heat pumps in the market (David et al., 2017). Thus, a throughout study is vital to identify neighbouring industries capable of provide a rich source of waste heat to support the operational demands of the plant.

The emissions associated with the transportation of CO₂ are linked to the construction of the pipeline section, with emissions increasing proportionally to the distance from the capture point to the storage place. Additionally, the energy requirements for the compression and delivery of the CO₂ also increase with greater distances, necessitating an additional

compression step for distances exceeding 200 km. In the context of both study cases in scenario 1, it becomes evident that the storage and transport processes can reduce the carbon removal efficiency up to 25%. This effect can be visualised in Figure 5.4, underscoring the importance of the emissions during the construction of the piping system. Consequently, to account for the emissions caused during the construction of the transportation infrastructure, we segmented it into three diverse areas based on its carbon footprint (Figure 5.5). It is important to remark that this study possesses a limitation and assumes a direct, linear route for CO₂ transport from the plant to the storage point. Nonetheless, leveraging the current infrastructure can offer opportunities to reduce emissions and render the transport of CO₂ viable for other regions. Other studies have suggested this solution, with the costs associated with CO₂ transportation representing a small fraction of the overall DAC emissions (Gregory de Temmerman; & François de Rochette, 2023). While GWP100 was used to address this study, Table 5.4 showed an additional wider range of environmental indicators. The results show that most categories have relatively low impacts in the lifetime. Even categories with higher intensity like water or fossil energy use, the values remain within a reasonable range. These findings show that DAC systems, when scaled and optimised, can have a low impact on the environment.

The elevated temperatures and pressures at the depths of the reservoirs induce changes in the characteristics of the CO₂, facilitating the storage a larger volume. At a depth of 800 meters, CO₂ reaches the critical point where both its temperature and pressure excess 31 °C and 73 atm, respectively. At this state, the volume of gas contracts to approximately 0.32 times that of its surface state. Given the ideal conditions for building a DAC plant in the country, a megaton scale plant is capable of storing 48.54 million of m³ over its operational lifetime. In light of these considerations, 17 plants of 1 MtCO₂ capacity will be sufficient to fill the Kinsale depleted gas field. If we consider the natural carbon sinks, the accumulated emissions from 1990 to 2023 from Ireland are almost 500 MtCO_{2e} (Casaban & Tsalaporta, 2022). Regardless of the progress in the field or the prospected emergence of future gigaton plants by 2050, this approach shows the potential of DAC to contribute to the mitigation of half of the accumulated emissions. As previously mentioned, DAC remains in its nascent stages of development, yet there are many similarities with the renewable energy sector in the 1990s (Smith et al., 2023). In order to achieve the desired levels of carbon dioxide removal, it becomes imperative explore materials and methods applicable under atmospheric conditions (Casaban & Tsalaporta, 2023). It is important to highlight that the presence of moisture in the ambient air can pose challenges,

reducing the process efficiency and increasing the electricity consumption for dehumidification purposes.

Nevertheless, the findings of this study establish Ireland as an ideal candidate for the deployment of a DAC plant in the near future. Taking advantage of the wind and a reliable source of heat from waste, promises to substantially reduce the energy costs associated to the DAC plant operation, specifically, solid sorbent-based technologies (Chowdhury et al., 2023). However, this represents the optimal scenario, and not every region within the country may meet the requisite conditions. Our analysis revealed that the most important factor lies in the transport and storage process. Counties close to Kinsale and Corrib gas fields have an advantage in contrast with the others, as they do not need the same infrastructure requirements to transport the CO₂. This presents a potential catalyst for the local economy. However, there are viable alternatives for the midland regions of the country, offering opportunities for equitable deployment of DAC facilities.

Deploying a DAC in plant in midlands counties may present challenges when it comes to storing CO₂ in Kinsale or Corrib, yet the Northwest Carbonaceous can store an effective quantity of 730 Mt_{CO2}. A more viable approach would involve the construction of DAC plants in Limerick or Dublin. Despite these counties are relatively distant from the storage points examined of this study, they have other nearest locations where store the CO₂. In the vicinity of Dublin lies the Kish Bank basin -Table 5.3-, with an effective capacity for storing up to 270 Mt_{CO2}. Thus, the CO₂ can be delivered to regional nearby reservoirs, such as the Central Irish Sea Basin or on the other hand, to the North Sea through shipping, which is the biggest storage point in Europe (European Commission, 2019). This strategic shift could enhance the viability of different locations for a DAC plant due to the construction of a pipeline infrastructure will not be a requisite (EPA, 2008) and therefore, reduce the associated emissions.

5.4 Conclusions

Because of its robust economic growth and strategic geographical positioning, Ireland has significant potential in the emerging DAC industry. Notably, the agricultural sector plays a crucial role in the country's economy. In the near future, it is imperative for this sector to reduce its emissions by a substantial 25%. Therefore, there will persist "hard-to-abate" emissions that requires effective mitigation measures. A reliable DAC industry can be instrumental in closing

the carbon cycle by addressing these residual emissions. If the process is powered by renewable energy sources, part of the CO₂ can be converted into e-fuels for the aviation and shipping industry, as for example the production of e-methanol and e-kerosene. These approaches hold the potential to significantly reduce the emissions associated with storage and transport, although they may incur increased energy costs, specifically associated to the use of an electrolyse. Future studies will scrutinise this aspect in order to determine its viability.

Therefore, Ireland can find itself in an advantageous position to become pioneers in the fields of renewable energy and CDR. While renewable energy is expected to expand in the coming decade, CDR and carbon capture lag behind. It is crucial to highlight that no single technology can solve the climate crisis. Renewables face its own challenges, such as energy transmission and storage. Simultaneously, there is a crucial need to curtail the costs associated with the capture of CO₂ from certain CDRs and carbon capture techniques. It is important to stress that CDRs, including DAC, should complement emissions reductions efforts and serve to neutralise any residual emissions from sectors with difficulties to decarbonise. If these technologies are integrated with other climate policies, in combination with renewable energy, can collectively fulfil their respective roles.

Encouragingly, last years have witnessed a shift in perspective. Renewable energy is booming, and CDR and carbon capture technologies are garnering increased recognition and support, with new initiatives emerging. Numerous DAC plants and planned projects are on the horizon this decade, with the expectation of providing valuable insights into this critical technology. Therefore, the availability of data proved to be a significant limiting factor during our analysis, making our LCA reliant on prior studies.

In an effort to minimise the carbon footprint of the operation, we considered that the megaton DAC plant would be powered by waste heat and renewable energy. Our study focused primarily on the utilisation of the depleted gas fields of Kinsale and the future depleted gas field at Corrib. According to every scenario Castlebar and Kinsale emerged as highly favourable locations for the construction of a DAC plant, owing to their proximity to the storage location. Most of the emissions from the process are attributed to the transport and storage of the CO₂. Nonetheless, DAC plants can be established in various locations throughout the country. By leveraging the existing gas transportation network, counties such as Waterford have the opportunity to establish a DAC plant. Additionally, close to Dublin and in the Irish Sea, particularly the Kish Bank Basin and East Irish Basin, offers the substantial storage capacity of 1050 Mt_{CO_{2e}}. Counties such as Donegal can explore the potential of the onshore Northwest Carboniferous Basin, which is estimated to store 730 Mt_{CO_{2e}}. To facilitate these

initiatives, comprehensive exploration operations and environmental risk assessments must be conducted at the known and other un-quantified basins, such as the Clare Basin located in Limerick.

Chapter 6: Materials for Direct Air Capture: A comparative study of Zeolite 13-X, Lewatit and amine functionalised SBA-15

Note: Results from this chapter are expected to be split in two publications: characterisation and adsorption capacities.

6 Materials for Direct Air Capture: A comparative study of Zeolite 13-X, Lewatit and amine functionalised SBA-15

Abstract

In an effort to optimise the CO₂ capture, this study investigates the adsorption performance of amine-modified materials compared to known adsorbents such as Zeolite 13-X and Lewatit. Amine-functionalised supports are known for their enhanced CO₂ chemisorption through carbamate formation. However, under ambient conditions, the performance of the modified materials was suboptimal due to pore blockage, caused by high amine loading, which suppressed physisorption. Zeolite 13-X, typically characterised by its high microporosity, exhibited a type II isotherm, suggesting an increased mesoporous fraction in the material. In contrast, Lewatit, with its amine-functionalised mesoporous structure, displayed a type III isotherm with superior adsorption capacity. Desorption studies revealed a sharp initial release of CO₂ from macropores, followed by slower desorption from chemically bound sites. On the other hand, amine-functionalised materials displayed a desirable desorption curve. The findings highlight the importance of pore size distribution and micropore accessibility in optimizing adsorption performance for CO₂ capture. Future works should focus on exploring materials with high microporosity, as well as controlling pore characteristics and maximizing the interaction between amine groups and CO₂, especially at concentration of 400 ppm and high levels of humidity.

Keywords: Direct Air Capture, Zeolite 13-X, Lewatit, PEI, TEPA, SBA-15.

6.1 Introduction

Between 2023 and 2024, for the first time and due to El Niño phenomenon, global temperatures were in a ten-month record where temperatures surpassed the 1.5 °C threshold (K. Li et al., 2023). If global temperatures drop after the current meteorological effects from El Niño, this increment will be seen as an abnormal outlier from the trending global increment (Cuff, 2023). Yet, if the temperature remains high or rapidly accelerates, will implicate that we are in uncharted territory (Ripple et al., 2023). Renowned scientists stated that this boundary will be exceeded at the beginning of the next decade (Hansen et al., 2023). In order to transition towards a sustainable future, it will be necessary the implement of renewable sources of energy as well as the implement of supporting policies, however, this is not enough. The Intergovernmental Panel for Climate Change (IPCC) stated very clearly that if we overshoot the 1.5 °C limit, Carbon Dioxide Removals (CDRs), such as Direct Air Capture (DAC) would be necessary to reduce the global concentrations of CO₂ (P.R. Shukla et al., 2022).

Different types of methods have been widely explored in CO₂ capture technologies: absorption, adsorption, electrochemical adsorption, passive adsorption, etc (Casaban & Tsalaporta, 2022). Adsorption has attracted the attention as a promising solution due to its low-cost performance (Sabatino et al., 2021). Despite DAC and Carbon Capture or post-combustion are similar, both are different (Casaban & Tsalaporta, 2023b). Post-combustion capture has been demonstrated viable in the recent years (Global CCS Institute, 2024). Yet, the main challenges of DAC are related to enhance and make economically viable the capture of CO₂ in small concentrations (Erans et al., 2022). One of the main challenges associated with is the synthesis of novel materials with a great affinity for CO₂ (Sanz-Pérez et al., 2016).

Among the materials investigated, mesoporous silicas as SBA-15 have emerged as a promising material (G. Zhang et al., 2018). The honeycomb pore structure and its pore volume attracted the attention of this material for CO₂ capture (Dao et al. 2013). Alone, the capacity of this material in CO₂ adsorption is really low (Sanz-Pérez et al., 2016). However, several authors reported that once the SBA-15 is enhanced with amines, its carbon dioxide performance increases (G. Zhang et al., 2018). Dao et al. 2013 prepared a material with 40 wt% of TEPA and 30 wt% of DEA and their study concluded that the CO₂ adsorption capacity of this material was 3.38 mmol/g (Zhao et al., 2020).

Therefore, amines have been used as a coating material in CO₂ adsorption processes. Between them, the most widely used were polyethylenimine (PEI) and tetraethylenepentamine (TEPA) (Sarmah et al. 2013). Drage et al. 2008 concluded that the adsorbent prepared with PEI had a great regeneration stability, yet its CO₂ capacity was low due to the high viscosity of the amine (Drage et al. 2008). Sanz et al 2016 reported that a SBA-15 material loaded with 50 wt% of PEI can show capacities up to 3.98 mmol/g (Sanz-Pérez et al., 2016). On the other hand, TEPA shows a great CO₂ adsorption capacity but a low regeneration stability (Linneen et al. 2013). Zhao et al studied the mass ratio of PEI and TEPA loaded in SBA-15, aiming to promote the retention of TEPA in the polymer chains of PEI (Zhao et al., 2020). Their results showed CO₂ adsorption capacities up to 4.20 mmol/g with a 10% vol of CO₂. These capacities inspired us to study such materials in pellet form and compare them with other commercial ones: zeolite 13-X, Lewatit.

Thus, this study presents a comparative evaluation of these materials under PSA conditions, with the aim of exploring their potential for DAC systems. In particular, the inclusion of Lewatit, which is a commercial ion-exchange resin, representing a novel contribution – at the time of testing, and its performance had received limited attention in the literature. In addition, the use of Zeolite 13-X as a control benchmark provides a reference for assessing the performance and ensuring that the apparatus setup is properly validated.

6.2 Materials & Methods

The materials used in this study included lewatit, Zeolite 13-X, PEI and TEPA from Sigma-Aldrich. In addition, SBA-15 in powder and pellet form was purchased from Glantreo. To pelletise SBA-15 powder, the binders bentonite and methyl Cellulose were required, both also obtained from Sigma-Aldrich. The CAS numbers for the primary materials are as follows: Zeolite 13-X (CAS: 63231-69-6), SBA-15 (CAS: 7631-86-9), PEI (CAS: 25987-06-8), TEPA (CAS :112-57-2), bentonite (CAS: 1302-78-9) and methyl cellulose (CAS: 9004-67-5). Lewatit does not have a registered CAS number, and its product number was 94136.

6.2.1 Materials and amine functionalisation

A ratio of 3:1:1 of SBA-15, bentonite and methyl-cellulose and 10 gr of DI water was used to pelletise the SBA-15 powder. Then, with the help of an extruder the pellets were cut

with a dimension of 2 mm and 1 mm. After this, the pellets were dried at room temperature. After the pellets were fully dried, they were brought to a four-step calcination process in a furnace (*Table 6.1*).

Table 6.1. Four-step calcination process of SBA-15

	Temperature (°C)	Time (min)
Step 1	300	30
Step 2	400	30
Step 3	500	30
Step 4	600	3600

The resulting material and the SBA-15 in pellet form was then rehydroxylated in a solution of 7:1 of DI water and hydrochloric acid (37%) (HCl) for 2 h at 100 °C.

A specified quantity of PEI was dissolved in certain amount of ethanol, with continuous stirring for one hour. Next, SBA-15 was added to the solution, which was then stirred for at least 10 hours at 40 °C until the ethanol evaporated. The resulting viscous material was washed with ethanol and dried in an oven at 80 °C for 15 hours to remove excess solvent and unbound amines, yielding a suitable sorbent for further testing. This material was named SBA-15-xPEI, where x represents the mass percentage of the amine. Following this, an additional adsorbent was synthesised. The previously obtained adsorbent served as the support for the TEPA loading. This amine was impregnated using the same procedure as described above. The final adsorbent was designated as SBA-15-xPEI-yTEPA, where y indicates the mass percentage of TEPA.

6.2.2 Characterisation techniques

To evaluate the structural and chemical properties of the selected materials, a series of characterisation techniques were performed. These analyses are essential to understand textural parameters, surface chemistry, and pore structure that influence CO₂ adsorption capacity.

The Brunauer-Emmett-Teller (BET) method was applied to N₂ adsorption isotherms at 77.3 K to obtain the specific surface area of the materials. This technique is widely used for evaluating mesoporous and microporous materials. In addition, to estimate the micropore volume and micropore surface area, a t-plot analysis was performed. This is particularly important for materials with high microporosity such as zeolites.

Barret-Joyner-Halenda (BJH) method was utilised to determine mesopore size distribution based on the desorption branch of the isotherm. This analysis can provide the distribution and cumulative pore volume, helping to identify the size and range of the mesopores. On the other hand, Density Functional Theory (DFT) analyses were performed to evaluate the cumulative pore volume and surface area as a function of the pore width. Although, this method is less applied than BET or BJH, it offers higher resolution for pore size evaluation into the micro- to mesopore range.

A Fourier Transform Infrared (FTIR) spectroscopy was performed to confirm the successful attachment of the amines into the materials. The appearance of N-H binds and C-N stretching vibrations is an indicator of the presence of PEI and TEPA on the surface of the mesoporous silicas.

6.2.3 Experimental setup and conditions

The adsorption experiments were performed in a fixed-bed reactor stainless steel with a total volume of 25 cm³. Two CO₂ sensors (model: K30 FR Fast Response 10,000 ppm purchased from “CO₂meter”), were placed at the inlet and the outlet in order to monitor CO₂ concentrations across the runs. Given the variability from ambient CO₂ levels, both sensors were calibrated prior each experiment (Figure 6.1). Blank measurements were carried by running nitrogen to calibrate 0 ppm and compressed air at the working pressure (3 bars).

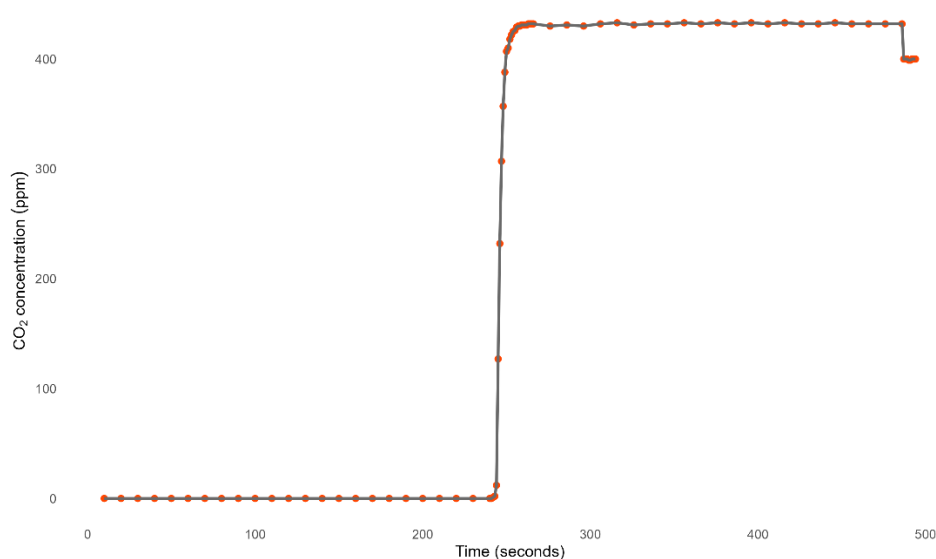


Figure 6.1. Blank measurement of the adsorption column prior experimental runs. The procedure involves flushing nitrogen gas through the column for approximately 4-5 minutes to calibrate the CO₂ sensors to 0 ppm. This procedure is then followed by 5 minutes of flushing compressed air in order to stabilise the readings at ambient CO₂ concentrations. The response time for the sensors to rise from 0 ppm to the outlet concentration is approximately 23 seconds.

Before the adsorption experiments, the adsorbent materials were desorbed in an oven. The desorption temperature and time varied by material: 100 °C for 1 hour for SBA-15/S60, 90 °C and for 1 hour for Lewatit and, 200 °C for 12 hours for Zeolite 13-X. After thermal treatment, the materials were placed in the column which was purged with 2 L/min of N₂ to remove all the traces of CO₂ and moisture.

For the adsorption tests, a flow of 2.5 L/min of compressed air at room temperature (approximately 18-22 °C), was introduced into the column. The air was drawn from outside the building to simulate atmospheric CO₂ concentrations. The breakthrough experiment was considered complete once the CO₂ concentration at the outlet equalled that of the inlet, indicating full saturation of the sorbent bed. Following each adsorption run, the material was regenerated using the same desorption conditions, repeating in each cycle.

While this study follows a DAC framework, the adsorption experiments were conducted under elevated pressure of 3 bars to emulate conditions closer to a PSA process. This choice was made to explore the maximum uptake capacity of the materials, and that the system can hold under favourable conditions. Although not representative of real DAC conditions, the results assess material performance, informing the selection of sorbents for atmospheric-pressure DAC systems.

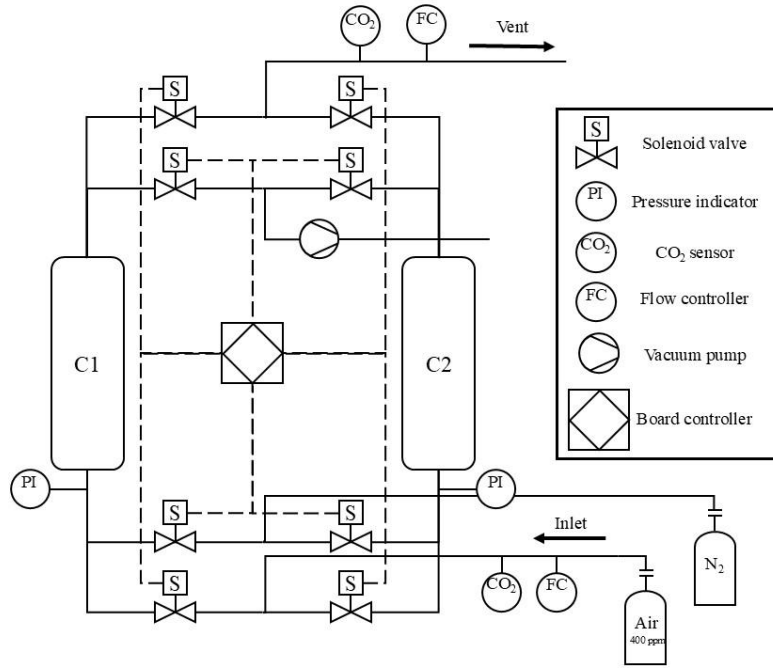


Figure 6.2. PI&D diagram of the PSA-Direct Air Capture setup used for the experimental runs. The system includes two adsorption columns, solenoid valves, pressure indicators, CO₂ sensors, flow controllers, a vacuum pump and a board controller.

The CO₂ adsorption capacity was then calculated by the following formula (Hungerford, 2024):

$$q_t = F \cdot \Delta t \int_0^t \left(\frac{C_0 - C_i}{C_0} \right) dt \quad \text{Equation 6.1}$$

q_t : CO₂ adsorption capacity

C : Concentration of CO₂ at the outlet (ppm)

C_0 : Concentration of CO₂ at the inlet (ppm)

t : Adsorption time (min)

F : Flow rate (L/min)

6.3 Results

6.3.1 Characterisation

Table 6.2 shows the properties of the materials analysed in this study. Analyses were carried by Dr. John McGrath and Dr. John Hanrahan from Glantree. The surface area represents the total area available for adsorption. The total number of pores consists of microporous, mesoporous and microporous, which provides a measure of the overall space available for adsorption. Microporosity affect the material's ability to adsorb CO₂ via physisorption. Pore diameter influences the kinetic, selectivity and efficiency of the adsorption process.

Properties of SBA15-30P-40T and S60-30P-40T were not able to reproduce due to low surface area. Even (Zhao et al., 2020) reported good results with these materials, the coating of both type of amines in both supports overloaded the materials, which resulted in pore blocking by the amines.

The driving force of Z-13X is physisorption, even results showed at Table 6.2 displayed a low micropores area, other studies (Micromeritics, 2020), demonstrated that Z-13X is known to have a high micropore area which make this material an excellent candidate for CO₂ adsorption. In some cases, the micropore volume of this material can reach values of 0.250 cm³/g (Garshasbi et al., 2017a). Therefore, the high adsorption capacity of Z-13X is largely due to these microporous properties. Macroporosity of Zeolite 13-X falls under the range of 20 to 500 Å. However, the BET surface area obtained for Zeolite 13-X (20 m²/g) is significantly lower than the typical values reported in the literature, which range between 600 to 700 m²/g. In this study, the sample was regenerated at 200 °C for 12 hours; yet several studies suggest that complete degassing of Zeolite 13-X may require regeneration temperatures between 250 and 300 °C (Sayilgan et al., 2016; Thompson et al., 2019). This indicates that the material may have been incompletely degassed, which leads to an underestimation of its true surface area and, consequently, limiting this analysis.

Lewatit, as a polystyrene-based ion exchange resin, primarily contains macropores larger than 500 Å, which limits its micropore volume (Low et al., 2025). This material is designed with amine functional groups that excel at capturing CO₂ through chemisorption. While Lewatit's micropore volume is relatively low compared to traditional adsorbents like zeolites or activated carbons, its performance is driven more by its chemical affinity for CO₂ rather than just surface area or microporosity.

Table 6.2 Properties of the studied materials

Material	A_{BET} (m^2/g)	A_{mic} (m^2/g)*	V_t (cm^3/g)**	D_p ($\text{\AA}$)***
SBA15-30P-40T	-	-	-	-
S60-30P-40T	0.0045	0.1374	-	-
Z-13X	20.46	2.03	0.0528	228.46
Lewatit	31.66	-	0.0428	1,021

* Micropore area

** Total pore volume

*** Pore size

To understand the material's properties, adsorption isotherms analyses were performed (*Figure 6.8*). Z-13X and Lewatit both exhibited a narrow hysteresis loop in the isotherms, indicating a uniform pore structure with consistent mesopore size distribution, as reflected in Table 6.2. This uniformity suggests that both materials facilitate efficient adsorption and desorption processes, allowing gases to be adsorbed and released smoothly.

BET plots for Zeolite 13-X and Lewatit (*Figure 6.3*) both showed linear behaviour in the relative pressure range of 0.05-0.3, which indicates a valid application of the BET method. Lewatit exhibited a shallower slope, consistent with its higher surface area reported by other authors ($31.66 \text{ m}^2/\text{g}$) compared to Zeolite 13-X ($20.46 \text{ m}^2/\text{g}$) (Low et al., 2024).

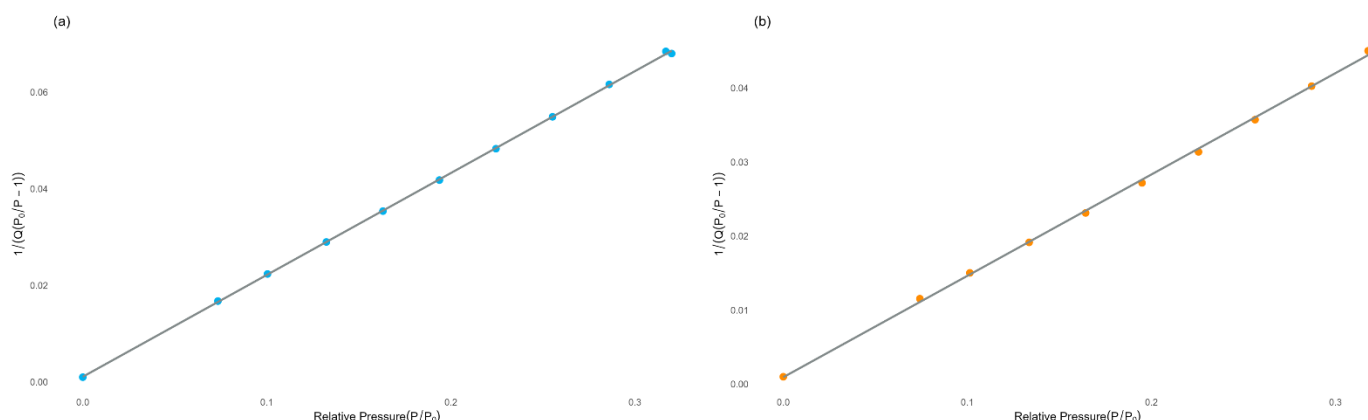


Figure 6.3. BET plots of Zeolite 13-X (a) and Lewatit (b) based on nitrogen adsorption at 77 K. Both materials showed a good linearity in the BET range, confirming a reliable surface area estimation

The BJH pore size distribution revealed a distinct porosity characteristic for both materials. Zeolite 13-X (Figure 6.4a) showed a visible mesoporous distribution with pore volumes concentrated between 20 and 100 Å. In contrast, Lewatit (Figure 6.4b) exhibited a significant pore volume only at very large pore widths (>1000 Å), which indicates a predominant macroporous structure with minimal microporosity.

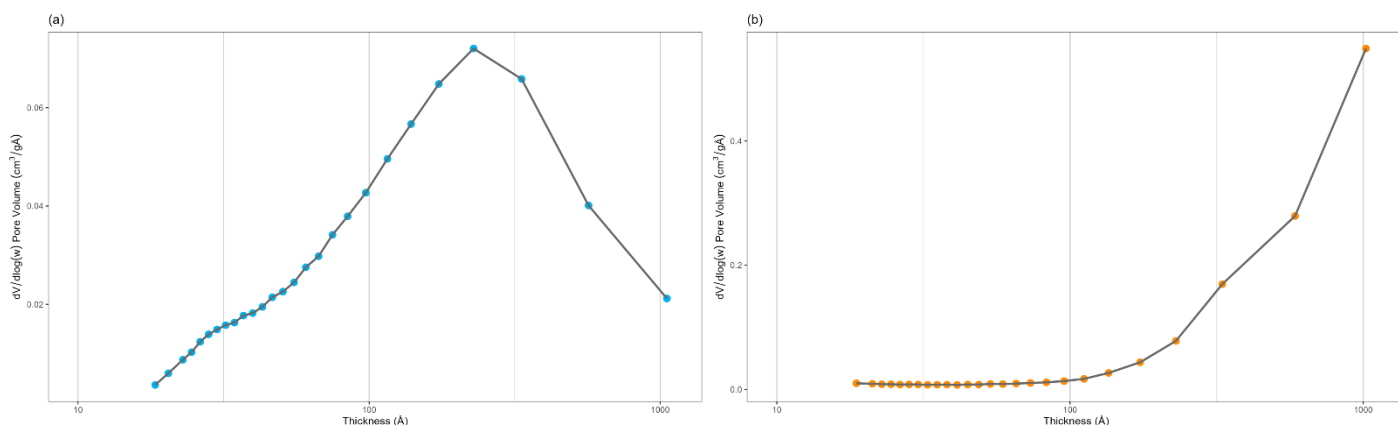


Figure 6.4. BJH pore size distribution for Zeolite 13-X (a) and Lewatit (b) performed with Halsey Faas Correction method. Zeolite 13-X displayed a broad distribution in the mesoporous range, while Lewatit it showed a microporosity dominance with pore volumes concentrated above 1000 Å.

Meanwhile, the DFT of the cumulative pore volume profiles reflect the differences between Zeolite 13-X and Lewatit. Zeolite 13-X (Figure 6.5a) showed a gradual increase related to its pore volume at 40 Å until reaching a pore volume of 0.05 cm³/g. This is an indicative of well-developed mesoporosity. On the other hand, Lewatit (Figure 6.5b) displayed a negligible pore volume accumulation until very large pore widths, with a sharp rise above 1000 Å; confirming that the macroporous are predominant in the material, with a total pore volume of 0.045 cm³/g.

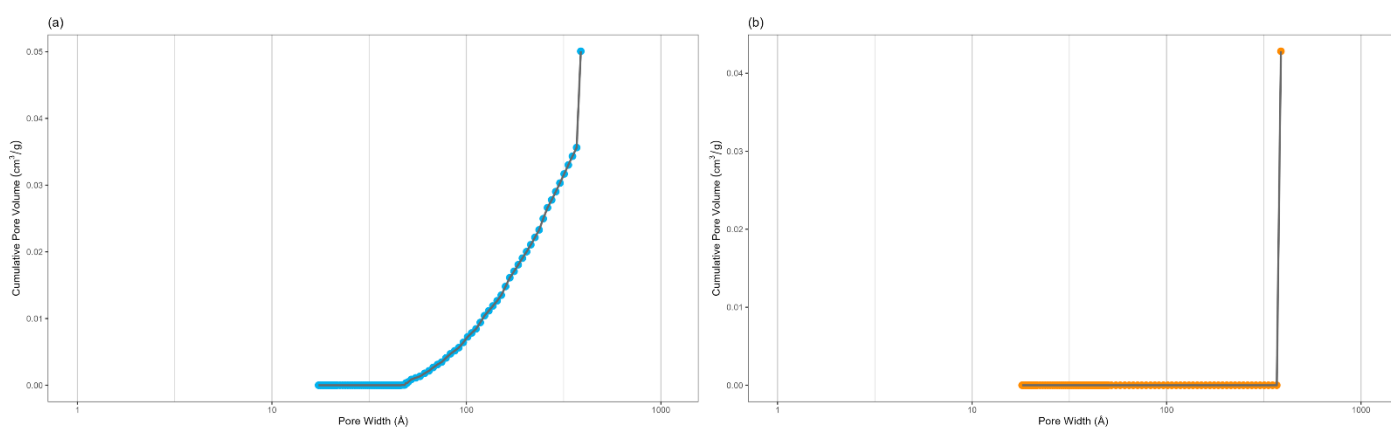


Figure 6.5. Cumulative pore volume as a function of pore width for Zeolite 13-X (a) and Lewatit (b). Zeolite 13-X demonstrated a continuous accumulation within the mesoporous range. Meanwhile, Lewatit exhibited a minimal accumulation until reaching the macroporous region (≥ 1000 Å).

Despite Zeolite 13-X have been widely reported to have a pronounced micropore area (between 353 to 450 m²/g)(Kongnoo et al., 2017), the t-plot analysis (Figure 6.6a) yielded a micropore area of just 2.03 m²/g. As well as with the micropore area, the micropore volume of the analysis displayed values of 0.000928 cm³/g, while this material can report values between 0.25 to 0.3 cm³/g (C. Chen et al., 2014; Garshasbi et al., 2017b). Thus, this significant deviation probably arises from insufficient activation conditions prior the analysis. As aforementioned, the material was regenerated at 200 °C as suggested by many authors (Chan et al., 2012; Jacobs et al., 2023; Y. Sun et al., 2022). Yet, to ensure the complete desorption of pre-absorbed species and full access to micropores, authors recommended push the regeneration temperature up to 300 °C (Fu & Davis, 2023; Naike Chemical, 2024). In contrast, Lewatit (Figure 6.6b) displayed a slightly higher BET surface area, consistent with meso- or macroporosity. Cumulative surface area plots from DFT (Figure 6.7) further suggest minimal contribution from micropores in both materials, reinforcing the likelihood of incomplete pore accessibility of Zeolite 13-X during the analysis.

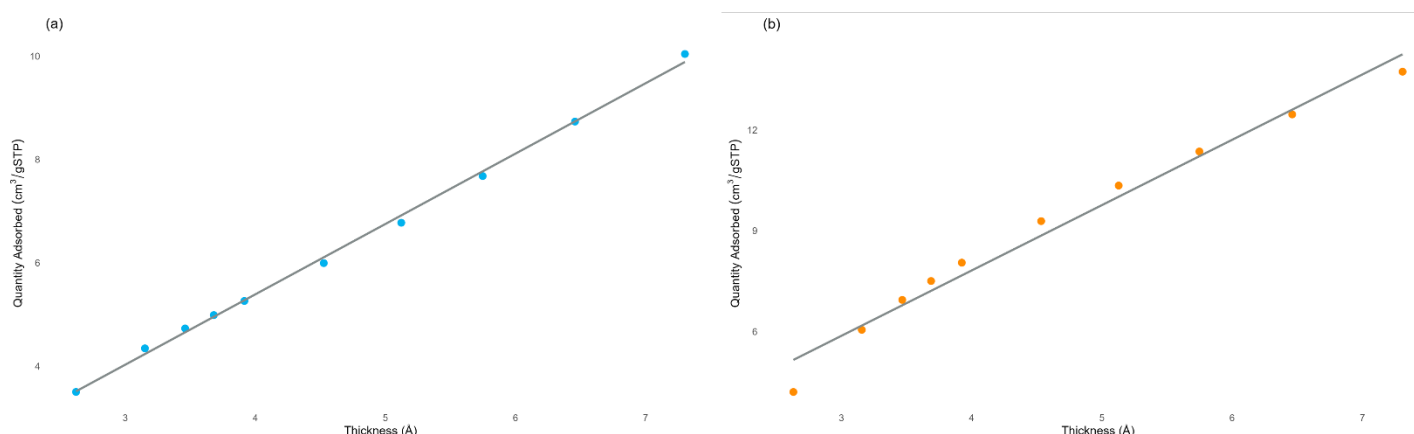


Figure 6.6. T-plot analysis of Zeolite-13X (a) and Lewatit (b). The low intercept from both materials suggests underestimation of the micropore volume. In the case of Zeolite 13-X, potentially linked to an incomplete degassing. Lewatit shows a similar linear profile through the origin, indicating its meso-/macroporous nature.

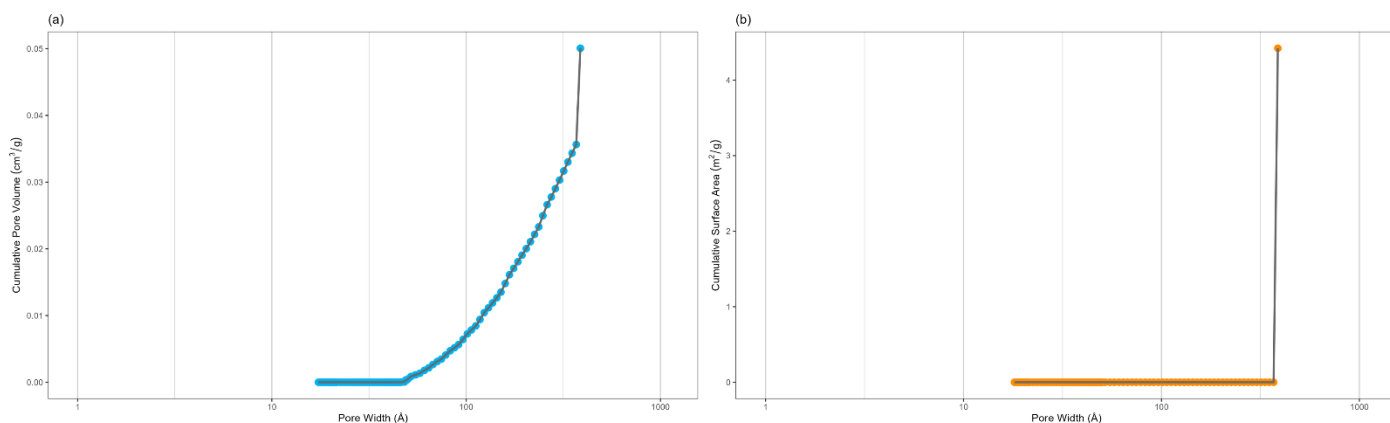


Figure 6.7. Cumulative surface area vs pore width derived from DFT analysis for zeolite 13-X (a) and Lewatit (b). Zeolite 13-X shows surface area contribution mainly from pores $> 40 \text{ \AA}$, with negligible microporosity. Lewatit on the other hand, exhibits a sharp increase above $300 \text{ \AA}$, characteristic of macroporosity.

The final confirmation that Zeolite-13X was not fully regenerated comes from the linear isotherms (Figure 6.8). This material is typically known to exhibit a Type I isotherm (Karka et al., 2019), due to its predominantly microporous structure (pores $< 2 \text{ nm}$), which favours rapid adsorption at low pressures, indicative of monolayer adsorption (Micromeritics, 2022). The observed type II isotherm and low adsorbed volume (only $50 \text{ cm}^3/\text{g}$ STP compared to typical values around $100 \text{ cm}^3/\text{g}$) further supports the hypothesis of incomplete regeneration. Despite its known microporosity, the current results only highlight the external surface area or the presence of mesopores.

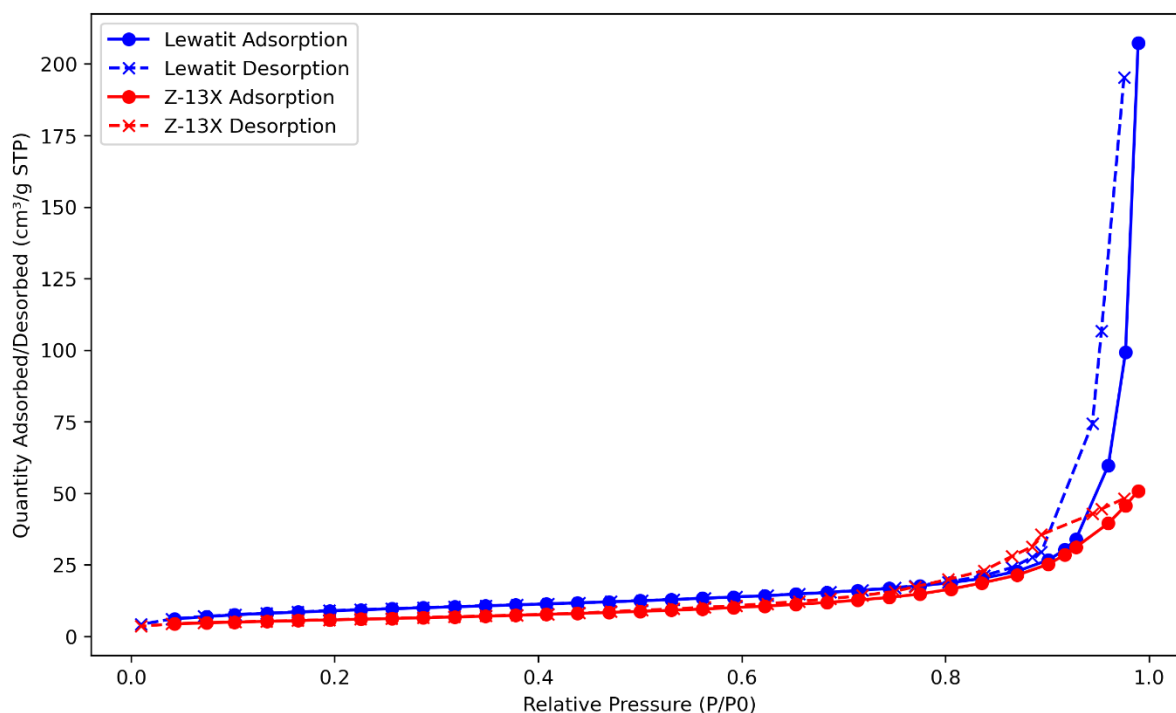


Figure 6.8. N₂ linear isotherms of Zeolite 13-X and Lewatit, measured at 77.3 K. Isotherms for the other tested materials could not be plotted due to invalid data.

In the case of Lewatit, similar quantities of adsorbed nitrogen have been reported by other authors ($\sim 200 \text{ cm}^3/\text{g}$), and the isotherms obtained in this study resemble those found in the literature (Low et al., 2023). The material displayed a Type III isotherm, typical of materials with significant macroporosity. Lewatit contains amine functional groups, which are highly selective for CO₂ capture, but the isotherm measured using nitrogen reflects its mesoporous characteristics. The sharp rise at high relative pressures indicates the onset of multilayer adsorption and capillary condensation within these mesopores, consistent with Type III behaviour. This suggests that Lewatit's mesoporous regions play a crucial role in the adsorption of gases, allowing for the adsorption process to proceed in layers as pressure increases.

Following the characterisation of the materials, the FTIR performed on the S60 and SBA-15 proved the presence of amines groups on the silica surface (Figure 6.9), confirming the success of the functionalization process (Liou et al., 2022). The peaks at 460 cm^{-1} and 1080 cm^{-1} , correspond to the Si-O-Si stretching vibrations, indicating the presence of the silica framework. A band at 950 cm^{-1} , is attributed to the Si-OH bond, which correspond to the silanol groups (Klinthong et al., 2013). The adsorption peaks around 1350 cm^{-1} , are linked to the CH bending vibrations from organic groups, which confirms the presence of hydrocarbon chains

from the amines (Klinthong et al., 2013). The band at 1570 cm^{-1} , correspond to the rocking vibration of the NH bond. The broad band at $>3000\text{ cm}^{-1}$, is associated with the stretching vibrations of the NH bond (Klinthong et al., 2013). As observed in Figure 6.9, two changes were observed after the amine coating. The vibrational peaks at 950 and between 3000 and 3600 cm^{-1} become weakened, indicating that the amine have partially displaced hydroxyl groups. In contrast, the intensity of the bands at $1310 - 1570\text{ cm}^{-1}$ and $2840 - 3000\text{ cm}^{-1}$ are enhanced, indicating an increase in the amount of amine groups in the surface. This result confirms that the amines were successfully loaded onto the support materials.

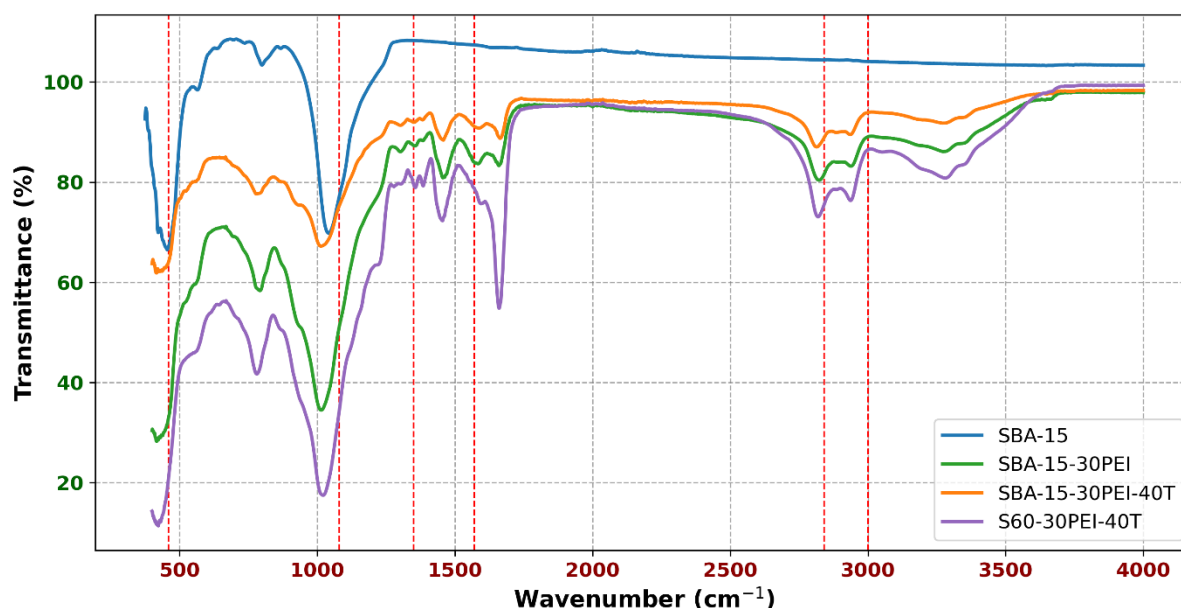


Figure 6.9. FT-IR analyses of functionalised silicas and unmodified SBA-15. The spectrum for unmodified SBA-15 is shown as a reference. Comparisons are made with SBA-15 loaded with 30 wt.% PEI, SBA-15 loaded with 30 wt.% PEI and 40 wt.% TEPA and Silica 60 loaded with 30 wt.% PEI and 40 wt.% TEPA. Characteristics peaks related to amine groups compared with other authors confirm the successful functionalisation of the materials.

6.3.2 CO₂ adsorption experiments

Lewatit VP OC is a commercially available polystyrene polymer that is cross-linked with divinylbenzene and functionalised with amine groups (Anthony P. Tirio & Rudolf, 2015). This material has been recognised as excellent for CO₂ capture processes (Veneman et al., 2015, 2016). These studies have explored the use of Lewatit for carbon capture from point sources. Another study evaluated its performance in CO₂/CH₄ separation (Sonnleitner et al., 2018). Therefore, there is little information about its performance under 400 ppm.

All the experiments (*Figure 6.10*) showed well-defined breakthrough curves, with an initial long period at 0 ppm, followed by a noticeable rise until the concentration reached the inlet value. The material exhibited an average adsorption capacity of 1.64 mmol/g across multiple runs. Notably, the first run yielded a higher capacity of 2.41 mmol/g. This pattern was consistently observed in a repeated run, where the capacity stabilised around ~1.60 mmol/g after the initial run. The higher capacity in the first run may be due to incomplete regeneration or gradual material deterioration, leading to limitations in subsequent cycles. Despite the higher adsorption capacity observed during the initial run, these results are likely outliers. Other authors have reported similar capacities (ranging between 1.5 and 1.6 mmol/g) under comparable operational conditions, such as CO₂ partial pressures of 0.12 kPa and temperatures around to 15 °C, which aligns with the values recorded in the subsequent runs of this study (Low et al., 2023). Therefore, this confirms that the macropores of Lewatit facilitate the transport of CO₂ molecules to the amine sites, where strong chemical bonds are formed, leading to high CO₂ uptake. Despite having a lower micropore volume, Lewatit compensates with a high surface coverage of functional amine groups, making it highly effective for CO₂ capture, particularly at lower concentrations (Vocht et al., 2022).

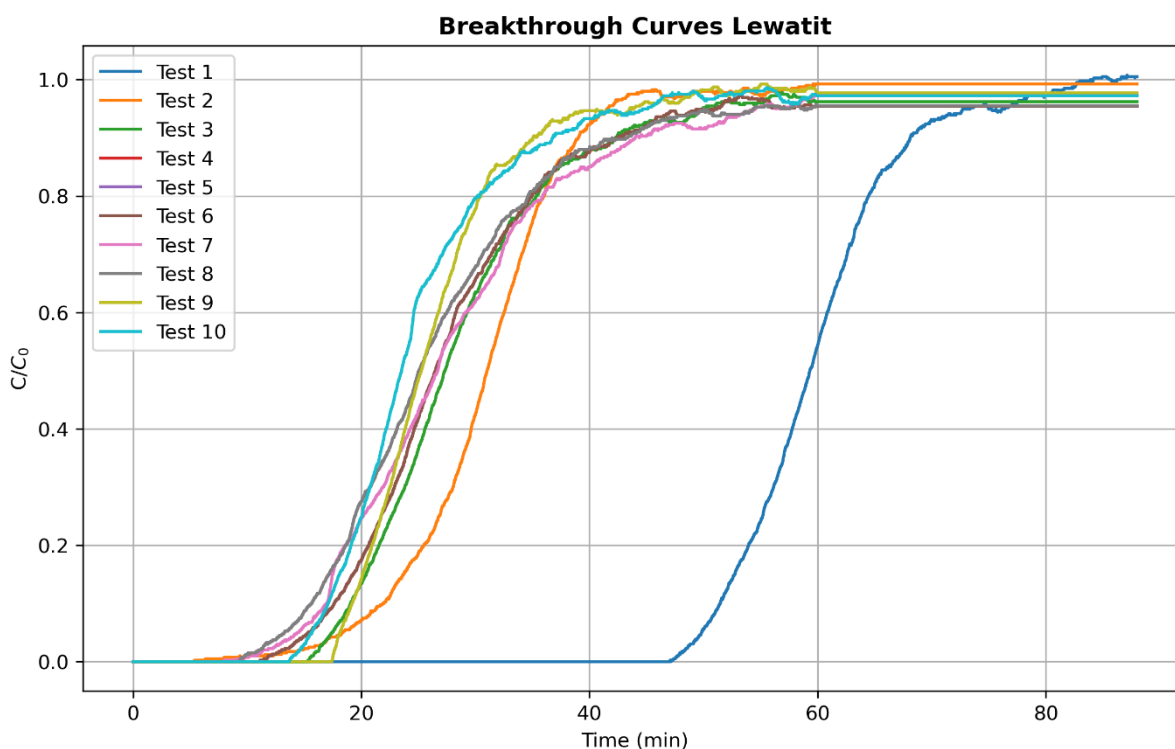


Figure 6.10. Breakthrough curves from 10 consecutive adsorption runs using Lewatit under 0.12 kPa of CO₂ partial pressure at ~14 . °C. The first run exhibits a delayed breakthrough, indicating a higher adsorption capacity compared to the subsequent tests.

Zeolite 13-X has been widely used in industrial scale. This material is known as a good adsorbent of CO₂. Results from *Figure 6.11* confirm this approach, where the material displays a long breakthrough curve with capacities between 0.7 and 1 mmol/g. Similar adsorption capacities for this material have been reported by other researchers and commercial entities, supporting the reliability of the results obtained in this study (Ahmed et al., 2023; Hungerford, 2024). A long breakthrough curve indicates that Zeolite 13-X is an effective CO₂ adsorbent, yet this is an undesired result. These results indicate that Zeolite 13-X requires a longer period to reach the saturation point, which leads to higher associated operating costs (De Oliveira et al., 2023). Moreover, this condition will end in more frequent and intensive regeneration cycles, thereby further increasing the operational costs. Thus, future cycles may be adversely affected if CO₂ is not completely removed from the Zeolite 13-X.

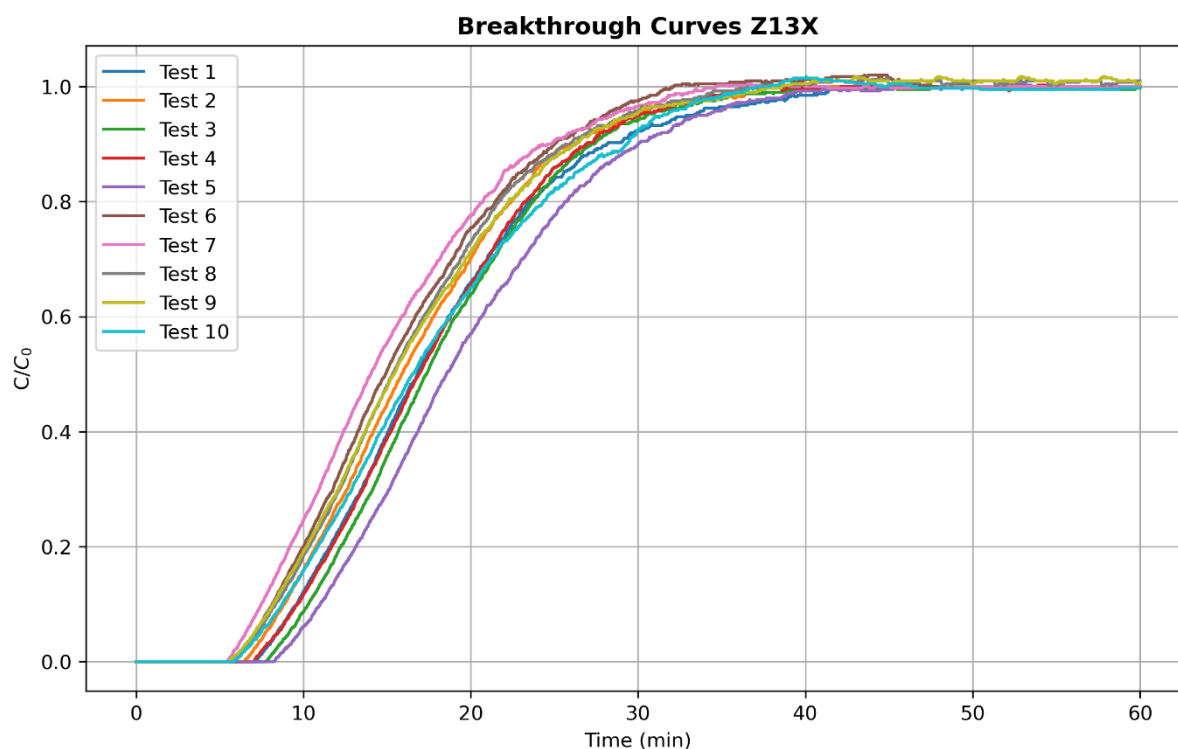


Figure 6.11. Breakthrough results for the 10 runs of Zeolite 13-X. The profiles show consistent behaviour across all runs, indicating stable adsorption performance under repeated cycling.

Breakthrough curves of SBA-15 amine modified are shown in *Figure 6.12*. If SBA-15 is not impregnated with amines, there is a suboptimal performance (Zhao et al., 2020). Therefore, once the material is coated with certain amount of PEI, the adsorption time extends as a result of additional capture sites. According to (Jahandar Lashaki & Sayari, 2018), if the loaded amount of PEI surpasses the 70 wt%, the adsorption capacity decreases due to the excess of amine blocks the pores of the material (Jahandar Lashaki & Sayari, 2018). Diamine SBA-15 imbued with TEPA displays a better performance due to the increase in amino active sites. Part of the loaded TEPA will fill the pores while the other is accumulated in the outer surface of the support. Thus, as reported by (Jahandar Lashaki & Sayari, 2018), the SBA-15-30PEI-40TEPA, displayed a great adsorption capacity. However, the experimental conditions of this study differ, and with an inlet concentration of 400 ppm, the adsorption capacity is approximately 0.5 mmol/g (*Figure 6.14*). Notably, as observed in *Figure 6.12*, the capacity never reaches the inlet concentration. This can be caused to the high loading of amine in the SBA-15, which is blocks the pores. As a result, the material loses some capacity, as physisorption is significantly reduced, leaving chemisorption as the driving force through the process.

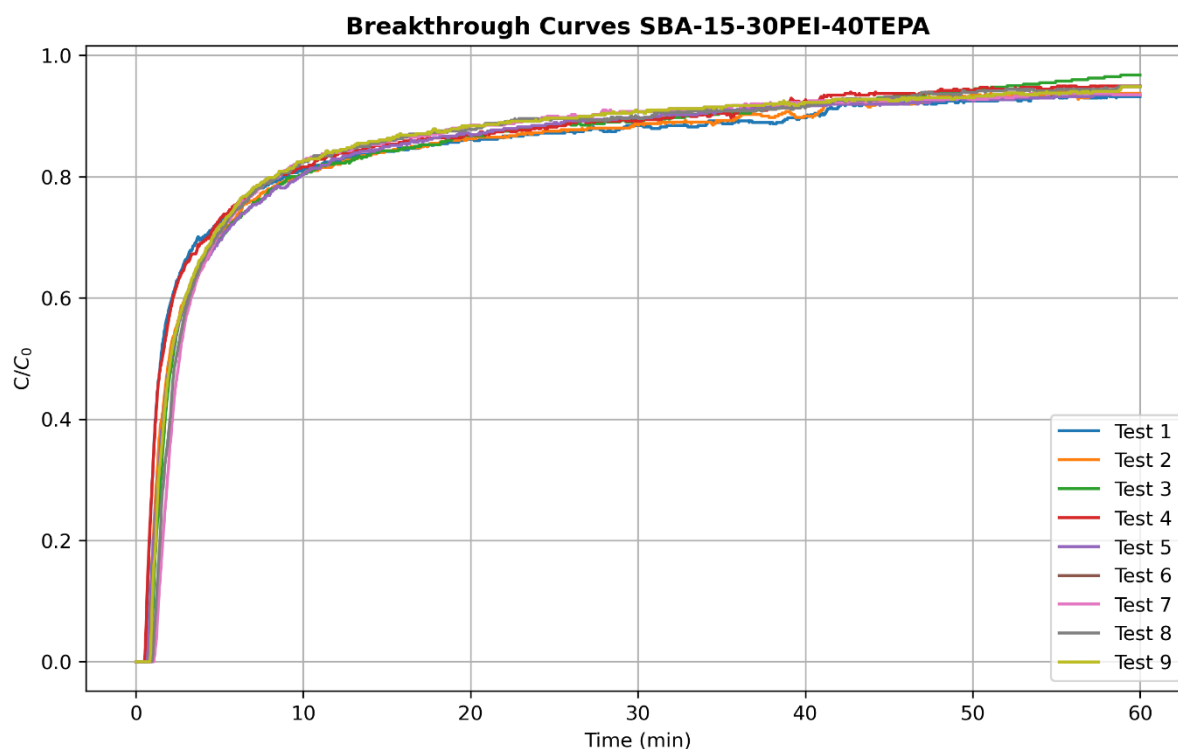


Figure 6.12. Breakthrough results for SBA-15-30PEI-40TEPA over 9 consecutive CO₂ adsorption cycles. The rapid breakthrough and low adsorption capacities reflect the limited performance under low CO₂ partial pressures. The curves displayed reproducible behaviour and good cycle stability.

In an effort to maximize the time at 0 ppm as was observed in both commercial materials and replicate the sharp rise in adsorption seen with the amines, Silica 60 was considered during this study and was modified with amine groups following the same procedure as SBA-15. Silica 60 is a cheaper support which has a pore size of 6 nm and a surface area between 400 and 600 m²/g, making it a promise material for CO₂ adsorption. The results displayed in *Figure 6.13* indicate that, like SBA-15, the high amount of amine loaded onto the support negates physisorption. Although this material demonstrates a higher capacity than SBA-15-30PEI-40TEPA (*Figure 6.14*), it exhibits a long tail in the breakthrough curve, indicating that more time is needed to reach saturation. Silica 60 turned yellow after nine cycles. Typically, after a certain number of cycles with only PEI attached, the material becomes yellowish; however, this did not occur when the support contained both types of amines. Therefore, during the experiments, it was observed that the amine groups were not well attached to the support, resulting in mass loss with each cycle.

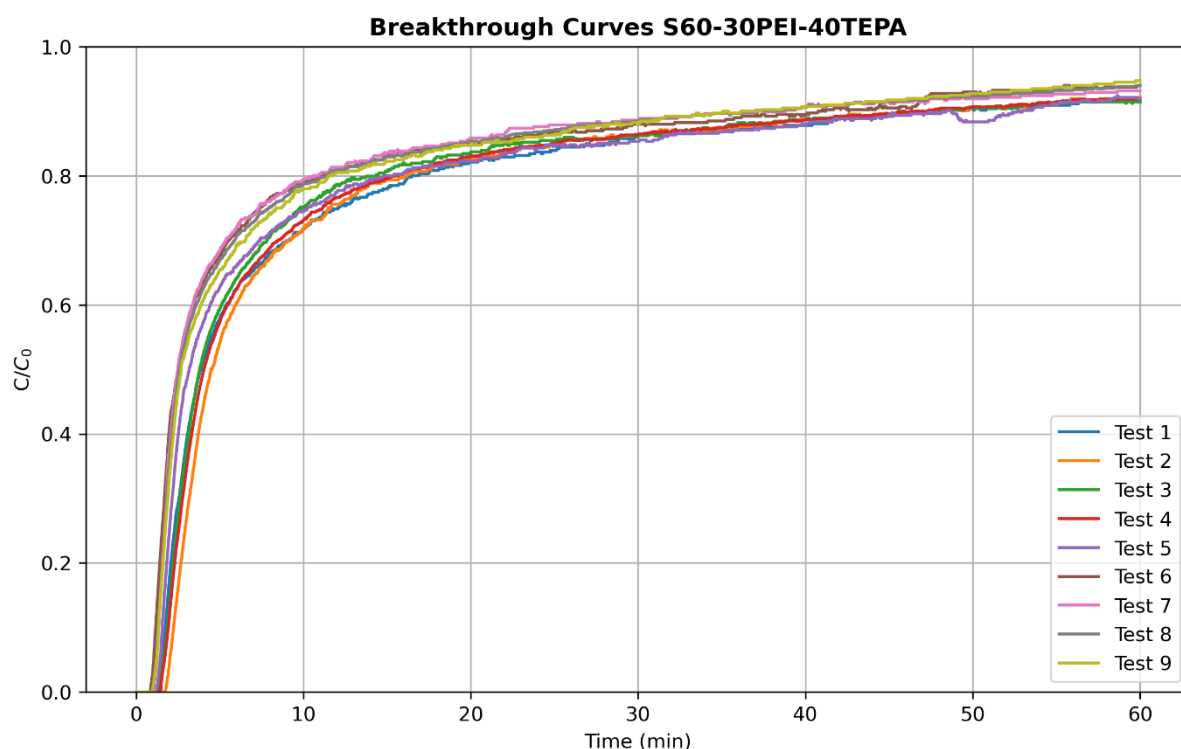


Figure 6.13. Breakthrough results for S60-30PEI-40TEPA across 9 cycles. Slightly higher adsorption capacities were observed compared to SBA-15, suggesting a better amine accessibility or interaction with CO₂.

In the results of this study, Lewatit exhibited a remarkable CO₂ adsorption capacity (Figure 6.14). As explained before, this enhanced capacity is primarily attributed to the presence of amine groups on the surface of the resin, which facilitates a strong interaction with CO₂. Water plays an important role in influencing the CO₂ adsorption mechanism on amine-based materials like Lewatit. In the presence of water vapor, water can act as a free base, promoting the formation of bicarbonate when CO₂ is adsorbed. However, in our case study (dry conditions), when water is absent, carbamate species are formed instead. Under these dry conditions, two amine molecules are required to bind one molecule of CO₂ to form carbamate. Rens Veneman has observed this behaviour (Veneman et al., 2014), who confirmed the formation of carbamate on Lewatit VP OC 1065 during their experiments. The distinction between the bicarbonate formation in humid environments and carbamate formation in dry conditions highlights the sensitivity of CO₂ adsorption mechanisms to moisture and underscores the versatility of amine-functionalised resins for CO₂ capture in varying environmental conditions. This process is significantly stronger than physisorption, as observed in other materials like Zeolite 13-X, where adsorption occurs primarily through

weaker van der Waals forces. A noticeable decline in adsorption capacity was noted for the S60-30PEI-40TEPA after the fifth cycle. During the experiments, partial loss of the material was detected, as it began to contaminate the system. This suggests that the amine was poorly anchored to the silica surface, likely due to weaker interactions between the support and the impregnated TEPA. As a result, amine leaching and material degradation occurred in the subsequent cycles, which compromised the material's long term performance and stability.

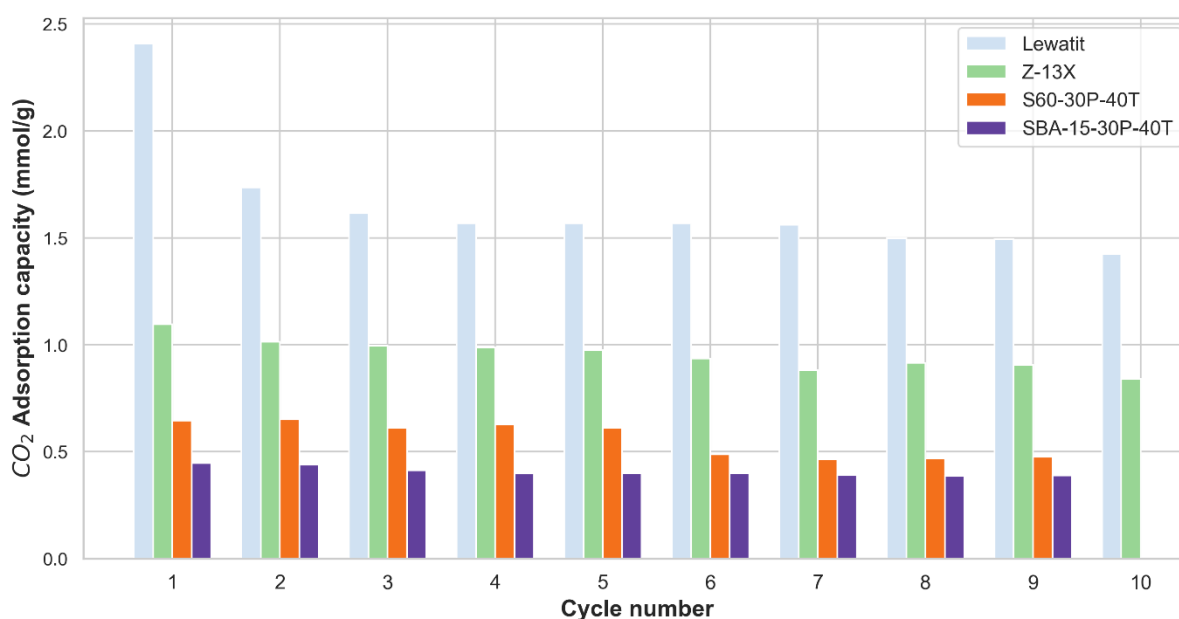


Figure 6.14. Comparison of CO_2 adsorption capacities of the materials tested across each cycle. Bars represent the material performance (mmol/g), highlighting the higher capacities of Lewatit (~ 1.5 mmol/g) and Zeolite 13-X (~ 0.9 mmol/g), compared to lower but steady values observed for S60 (~ 0.6 - 0.45 mmol/g) and SBA-15 (~ 0.4 mmol/g). A sharp drop in adsorption capacity was observed for S60 after cycle #5, likely due to partial material loss and amine leaching.

Desorption behaviour of the amine-modified materials and Lewatit was assessed by thermal regeneration followed by flushing with nitrogen the test column. SBA-15 modified with PEI and TEPA, showed a rapid desorption, with the CO_2 concentration dropping to zero within 5 minutes, indicating a fast release of the weakly bound CO_2 . In contrast, S60-30PEI-40TEPA and Lewatit, released a substantial amount of CO_2 immediately upon nitrogen purging, reaching levels of 10,000 ppm in the case of Lewatit, which surpassed the upper detection limit of the sensor. This sharp spike in desorption can be likely attributed to the rapid release of CO_2 from the more accessible chemisorbed sites in the macropores (Figure 6.15). Macropores provide a faster diffusion pathway for CO_2 , which is less tightly bound compared to CO_2 adsorbed in micropores.

This was followed by a gradual decline in concentration, suggesting the presence of more strongly bound CO₂ or delayed desorption behaviour due to the material structure or amine interactions. The strong desorption signal from Lewatit is consistent with its uptake and denser amine loading, while the slower response of S60 may reflect a combination of poor amine anchoring and incomplete regeneration.

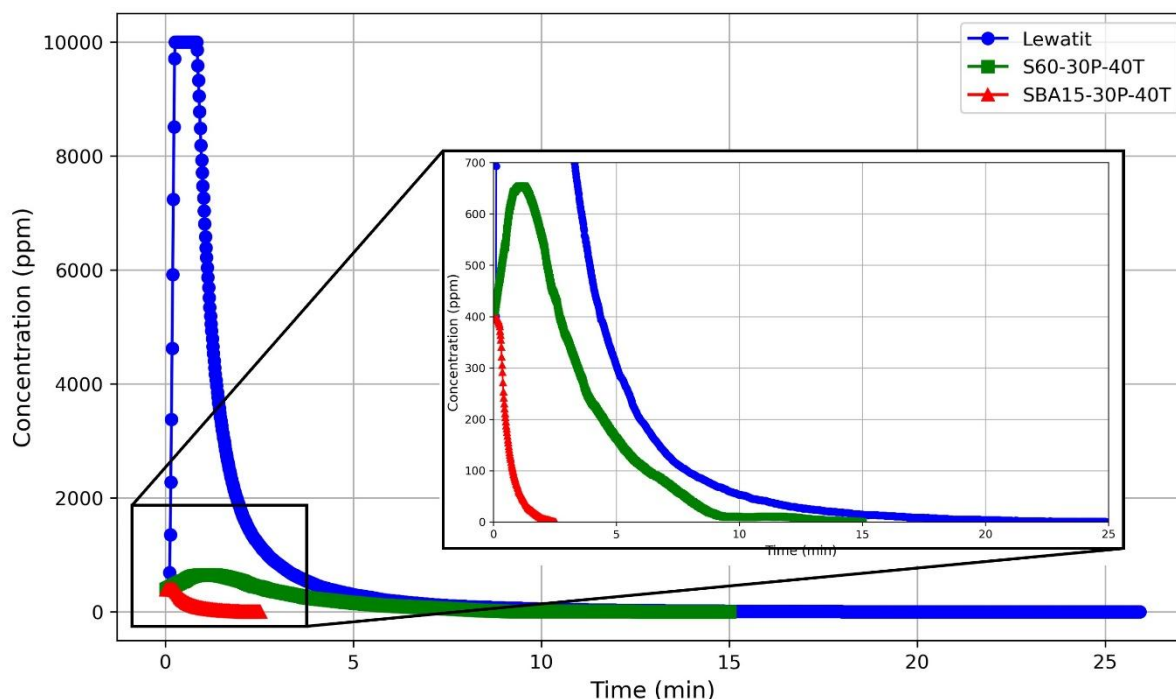


Figure 6.15. Desorption curves of Lewatit, Silica 60 and SBA-15-30PEI-40TEPA after thermal regeneration (1 hour at 90 °C for Lewatit, 100 °C for amine-modified materials) followed by nitrogen flushing. SBA15 exhibited a rapid desorption curve, with CO₂ levels falling below 400 ppm in under 5 minutes. Lewatit and S60 released a significant CO₂ peak at the beginning of the purge (exceeding 10,000 ppm in Lewatit), followed by a slower decline, which indicates a stronger or delayed CO₂ release mechanisms.

6.4 Conclusion

The FT-IR analysis confirmed the successful loading of amine groups on the material supports, as evidenced by the increased intensity of NH bond-related peaks. However, the performance of the amine-modified materials was inferior compared to Zeolite 13-X and Lewatit. While numerous studies demonstrated the potential of amine-modified materials for CO₂ adsorption, the conditions of these studies involve higher concentrations of CO₂. Amine functionalised materials enhance CO₂ adsorption through carbamate formation, via chemisorption. Yet, it was observed that overloading the supports with more than 70% of

amines led to a significant decrease in adsorption capacity. This reduction is attributed to pore blockage in SBA-15, which hinders physisorption and limits the process to chemisorption only.

Associated with its high microporosity, Zeolite 13-X exhibited a type II isotherm in its characterisation, which is atypical for this material since it usually displays a Type I isotherm; suggesting that mesoporosity may be more dominant in the sample, reducing the effectiveness of the microporous structure in adsorbing CO₂. In contrast, Lewatit displayed a type III isotherm characteristic of materials with high mesoporosity and surface-functionalised groups like amines. This mesoporous structure, combined with the presence of amine groups, makes Lewatit the most effective adsorbent in this study. The first adsorption cycle of this material was repeated twice, and similar results were obtained. This could be due to the loss of material after the first experiment. The sharp spike in desorption curves indicates the rapid release of CO₂ from the macropores, followed by a slower desorption from the micropores and the interaction between the amine groups and CO₂. Yet, amine materials displayed a sharp and desirable decrease adsorption curve.

This work explored these materials under a total pressure of 3 bars (0.12 kPa of CO₂ partial pressure) to increase the adsorption capacity of the materials. This “pushed” DAC conditions can increase the operational costs in future DAC plants. Therefore, future works should focus on microporosity and ensure the effective interaction between amine groups and CO₂ under realistic DAC conditions (namely, a total pressure of 1 bar and high humidity). The presence of moisture is known to promote CO₂ adsorption in amine-based materials, making it a key parameter to explore. Materials such as COF-999 are known for its exceptional microporosity, offer promising avenues for improving CO₂ capture efficiency (Yang et al., 2025). Controlling pore size distribution and ensuring accessibility of micropores could significantly enhance the adsorption capacity and regeneration efficiency of the materials.

7 Conclusion

The main objective of this thesis was to provide a comprehensive understanding of DAC technology, assess its potential application in Ireland, and explore improvements through material testing in a lab scale PSA system. This work has shown that while DAC holds strong potential in mitigating climate change, important challenges remain, particularly in scaling the technology, improving its efficiency and ensuring its economic viability.

Chapter 2 provided an in-depth analysis of DAC and Carbon Capture technologies, discussing the possibilities of DAC in removing the CO₂ from the atmosphere and its possible uses in producing e-fuels. This chapter also explored the emergence of DAC companies over the last decade, highlighting how technological advancements and financial support are critical to the technology's success. It showed that DAC's role is increasingly recognised, yet considerable scepticism persists around its economic feasibility and scalability.

Chapters 3 and 4 examined global research on DAC, identifying which countries and institutions are leading in DAC research, with a specific focus on solid sorbents. The analysis revealed that despite growing interest, DAC research and deployment are geographically concentrated, with only a few countries investing heavily. Solid sorbents, though promising, still face issues related to energy efficiency and performance across varying atmospheric conditions. Not all adsorbents perform effectively in every climate, further emphasising the need for region-specific research.

In Chapter 5, a LCA was conducted to evaluate the potential of DAC in Ireland. The results demonstrated that Ireland is uniquely positioned to benefit from DAC due to its future abundant renewable energy resources, particularly wind energy, and its geological storage capacity from basins and depleted gas fields such as Kinsale. However, challenges related to cost, infrastructure, and energy intensity remain. The LCA also highlighted the importance of ensuring that DAC to have a low environmental impact, it must be powered by renewable energy sources. This finding reinforces the importance of aligning DAC development with renewable energy policies and increasing investment in wind energy infrastructure to power future DAC systems. These limitations, along with the available data, provide a solid foundation for further studies on DAC deployment.

The final chapter presented the experimental results of material testing in the PSA apparatus. This study revealed that materials such as Lewatit and amines such as PEI and TEPA

exhibit promising performance for DAC applications. The findings indicate that further research in material science is crucial for enhancing adsorption capacity and reducing energy requirements, ultimately improving the economic viability of DAC. The need for materials that perform well under varying environmental conditions, such as high humidity in regions like Ireland, remains a significant matter for future research.

In summary, this thesis has demonstrated that DAC technology has the potential to play a significant role in mitigating climate change, particularly in countries like Ireland, where renewable energy and geological storage resources are favorable. However, the technology remains in its early stages, and significant advancements are needed in materials, system efficiency, and supportive policy frameworks that will change the public perception and are required to scale DAC effectively.

7.1 Contributions to the field

This thesis contributes to the growing body of knowledge on DAC technology by providing a comprehensive review of its global development, a site-specific LCA study for Ireland, and presenting experimental data on material performance in PSA systems. Historically, DAC has been met with scepticism, much like renewable energy technologies such as wind and solar were in their early stages. This hesitancy delayed their implementation, which has had long-term consequences such as surpassing the 1.5 °C threshold. The same situation should not be repeated with DAC. While some debate persists regarding the prioritisation of certain technologies, it is clear that DAC and renewable energy technologies are complementary, and both are necessary for achieving a sustainable future.

Moreover, this research offers valuable insights into the challenges and opportunities of deploying DAC in Ireland. Although Ireland's renewable energy potential is vast, particularly with plans to expand offshore wind generation, the DAC sector will need to be ready to scale up. The findings presented in this thesis underscore the importance of starting that process now, in parallel with renewable energy developments. For DAC to succeed, substantial planning and investment must begin immediately to avoid further delays in the fight against climate change.

7.2 Limitations and future research

Despite the progress made in this thesis, several limitations must be addressed through future research. First, while the LCA provided a detailed assessment of DAC in Ireland, further refinement of geological storage capacity data is needed. Long-term stability of CO₂ storage in depleted gas fields must be thoroughly evaluated to ensure safe and permanent storage. This includes conducting detailed risk assessments, comprehensive geological surveys, and advanced simulations to predict CO₂ behaviour once injected into storage locations. Lab-based studies on how CO₂ interacts with specific geological formations are also key for ensuring that storage is safe and controlled over time.

Additionally, the experimental work on PSA materials showed promising results, but more extensive testing across a wider range of materials and environmental conditions is necessary to identify the most suitable solutions for DAC systems. Materials like Lewatit performed well in this study, but the limited range of materials tested leaves room for further exploration. For example, while the experiments in this thesis were conducted in dry conditions-ideal for materials such as zeolites, known for their high CO₂ adsorption capacity but amines capacity is boosted in contact with humid air- this does not reflect the humid climate of Ireland. Testing materials under humid conditions will be necessary to identify those that can maintain high performance in real-world applications. Additionally, new materials such as COF-999, which was recently discussed by Klaus Lackner's team and is said to capture CO₂ at rates comparable to trees, could be tested to assess their suitability for deployment in Ireland. However, the performance of these materials under Ireland's humid climate remains to be seen, and further studies are needed.

In addition to material testing, future techno-economic studies should incorporate the latest data on DAC to assess the feasibility of using air-drying techniques with materials such as zeolites, exploring whether the combination could reduce costs while maintaining high CO₂ capture efficiency. Further simulation-based research, like the scenario analysis conducted in Chapter 4, should explore potential pathways for DAC deployment and its impact on Ireland's carbon budget.

Future research should focus on improving material efficiency and improving the energy efficiency of DAC systems and exploring new technologies that reduce energy consumption. Expanding the geographic scope of LCAs to other regions with similar renewable energy and

storage potential would also provide valuable insights for scaling DAC locally. By addressing these challenges, DAC can become a key player in achieving the global goal of reducing atmospheric CO₂ concentrations and mitigating climate change.

8 Bibliography

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