THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

Exploring Alternative Opportunities for Direct Air Capture Technologies

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CHALMERS UNIVERSITY OF TECHNOLOGY

Gothenburg, Sweden 2025

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Abstract

The 2050 net-zero emissions target envisions in part compensating for emissions through the use of carbon dioxide removal (CDR) technologies. Among these, direct air capture (DAC), which captures CO_2 directly from the air rather than from point sources, has gained attention due to its scalability and potential for placement near storage sites. However, as a novel energy-intensive technology requiring large-scale deployment, DAC remains a costly option for CO_2 removal. Non-classical deployment opportunities for DAC technologies could lower costs.

While the technologies developed for DAC are different from those developed for capturing carbon at point sources, one possibility would be to deploy the former at point sources with dilute flue gas streams. Another option is to consider deploying DAC technologies near energy sources. This study aims to evaluate two such alternative deployment opportunities for two commercialized DAC technologies: Temperature Vacuum Swing Adsorption (TVSA) and Alkaline Absorption with subsequent calcium looping (ALK-ABS). The analysis includes a cost assessment of using DAC for capturing CO_2 from low-concentration flue gas streams and for integrating DAC into combined heat and power (CHP) plants to use their excess heat.

We find that, for capturing CO_2 from dilute flue gas streams (0.5-4%), monoethanolamine (MEA)-scrubbing, developed for capturing carbon from point sources, demonstrates better economic performance than TVSA and ALK-ABS. Based on the levelized cost of CO_2 avoidance, at CDR price of 400\$/tCO_2 purchase of CDR credits is more economical than onsite capture for flow rates below 500t/h for CO_2 concentrations under 2.6%.

The integration of DAC into CHP plants presents a viable business opportunity, particularly in the emerging CDR market. Our findings indicate that at CDR price of 615 €/tCO_2, CDR could contribute up to 80% of the net cash flow for CHP plants in the future, with DAC alone accounting for 12% of the total net cash flow. Estimates suggest that integrating DAC into CHP plants across Sweden could achieve approximately 33% of the country's national CDR target.

Keywords: *Direct air capture, Technoeconomic, Negative emissions, Carbon dioxide removal*

List of publications

The thesis is based on the following appended papers, which are referred to in the text by their assigned Roman numerals:

I. Hoseinpoori S, Pallarès D, Johnsson F, Thunman H. A comparative exergy-based assessment of direct air capture technologies. Mitigation and Adaptation Strategies for Global Change. 2023 Oct;28(7):39.

II. Hoseinpoori S, Johnsson F, Pallarès D. Capture or offset: Techno-economics of decarbonizing of industries with flue gas streams containing low CO_2 Concentrations. Manuscript under internal review

III. Hoseinpoori S, Roshan Kumar T, Beiron J, Johnsson F, Svensson E, Pallarès D. Integration of Sorbent-Based Direct Air Capture into Combined Heat and Power Plants with Post-Combustion Carbon Capture. Submitted to Journal of Energy.

Sina Hoseinpoori is the principal author of the papers listed. Professor David Pallarès contributed with discussions, methodology development, and editing of **Papers I-III**. Professor Filip Johnsson, contributed with discussion and editing of **Papers I-III**. Professor Henrik Thunman, contributed with discussion of **Paper I**. Tharun Roshan Kumar contributed with discussions, methodology development, and editing of the **Paper III**. Dr. Johanna Beiron contributed with discussions, editing of **Paper III**. Associate Professor Elin Svensson contributed with discussion and editing of **Paper III**.

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Sina Hoseinpoori

Gothenburg, March 2025

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1 Introduction

The carbon dioxide concentration in the atmosphere has been increasing steadily since the beginning of the industrial revolution due to an increase in anthropogenic emissions [1]. As seen in Figure 1, the concentration of CO_2 in the atmosphere has increased from 315 ppm in 1960 to 424 ppm in 2024 [2]. As a result, the mean global temperature has risen by 1.5 °C in comparison to pre-industrial level[3]. The Paris agreement was signed in 2015 to limit the temperature rise to well below 2 °C before the end of century. This corresponds to an atmospheric carbon budget that approximately requires reaching net-zero CO_2 emissions by 2050, which is the target for the EU [4].

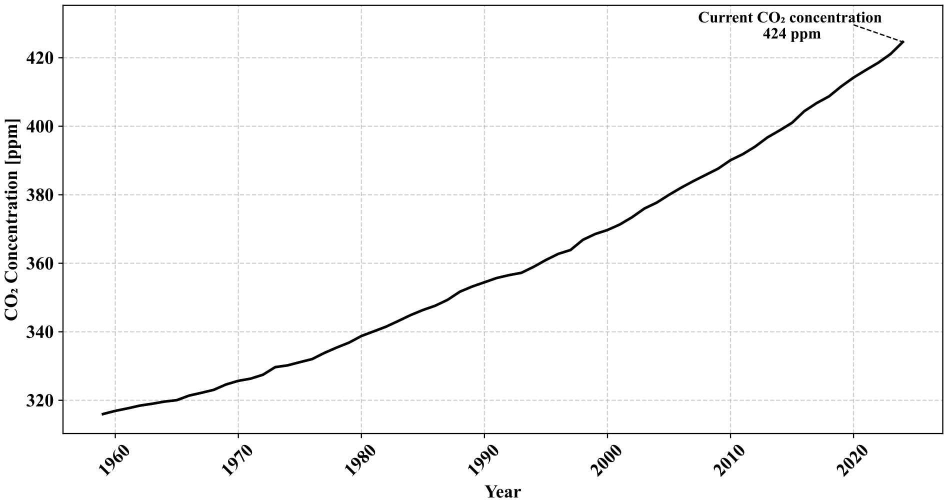


Figure 1:Historical CO_2 concentration in the atmosphere. Data retrieved from [2]

Limiting the increase of CO_2 in the atmosphere can be achieved by transitioning to renewable energy sources and reducing energy use; capturing and storing emissions; and removing CO_2 from the atmosphere, listed in decreasing urgency and increasing cost, see Figure 2. Each layer in the pyramid in that figure facilitates the employment of the measures in the next layer up.

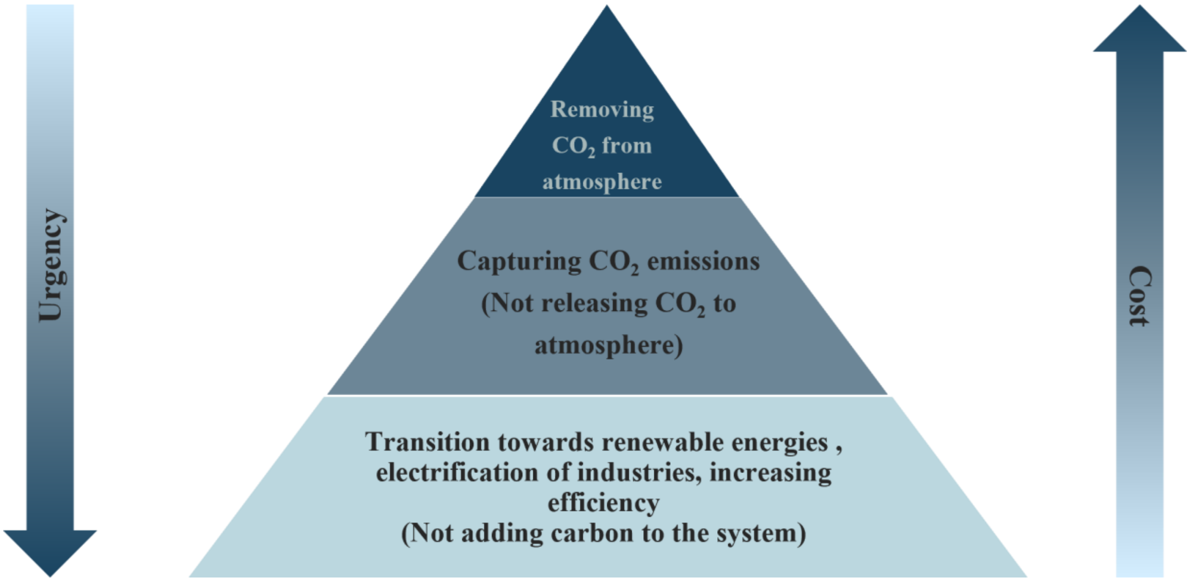


Figure 2: The pyramid of mitigation action for achieving net-zero from a system (not single-plant) perspective based on cost and urgency. The arrows show the direction of increase in urgency and cost, respectively.

A transition toward renewable energies would allow higher efficiencies for carbon capture processes as less indirect CO_2 is emitted during the capture process leading to higher CO_2 avoidance rates. Furthermore, by generating less CO_2 the demand for carbon dioxide removal (CDR) could potentially be limited to the emissions that cannot be cut in other ways or those associated with high mitigation costs.

Currently annual emissions to the atmosphere are 37.4 Gt of CO_2; these are required to be net-zero by 2050. Without significant and immediate cuts in these emissions, the relevant carbon budget is unlikely to be met, resulting in an emissions overshoot needing to be compensated by CDR methods [1]. Various CDR methods have been proposed in literature.

Figure 3 shows the main CDR methods available today[5]:

- **Afforestation/Reforestation:** Removal of CO_2 through either restoring previously deforested areas or planting trees in previously unforested areas.
- **Biochar:** Storage of carbon in the form of charcoal through pyrolysis of biomass.
- **Enhanced weathering:** An artificial stimulation of natural processes of minerals reacting with CO_2 in the air forming carbonate minerals on land or in oceans.
- **Bioenergy with carbon capture and storage (BECCS/BioCCS):** A group of technologies that capture CO_2 from stacks on large point sources such as power plants and industrial processes (e.g. pulp and paper mills) using biomass as feedstock and then store the CO_2 permanently in geological formations deep underground.
- **Direct air capture (DAC) and storage:** Technologies that capture CO_2 from the air through a chemical process and then store it permanently.
- **Ocean Fertilization:** Ocean alkalinization is an approach to carbon removal that involves adding alkaline substances to seawater to enhance the ocean's natural carbon sink.

DAC and BioCCS output a high-purity stream of CO_2 that can be stored to create CDR value (top part of the pyramid in Figure 2) or converted to fuels/materials to contribute to limiting the increase in carbon mass above the ground (bottom part of the pyramid in Figure 2). This thesis focuses on DAC technology.

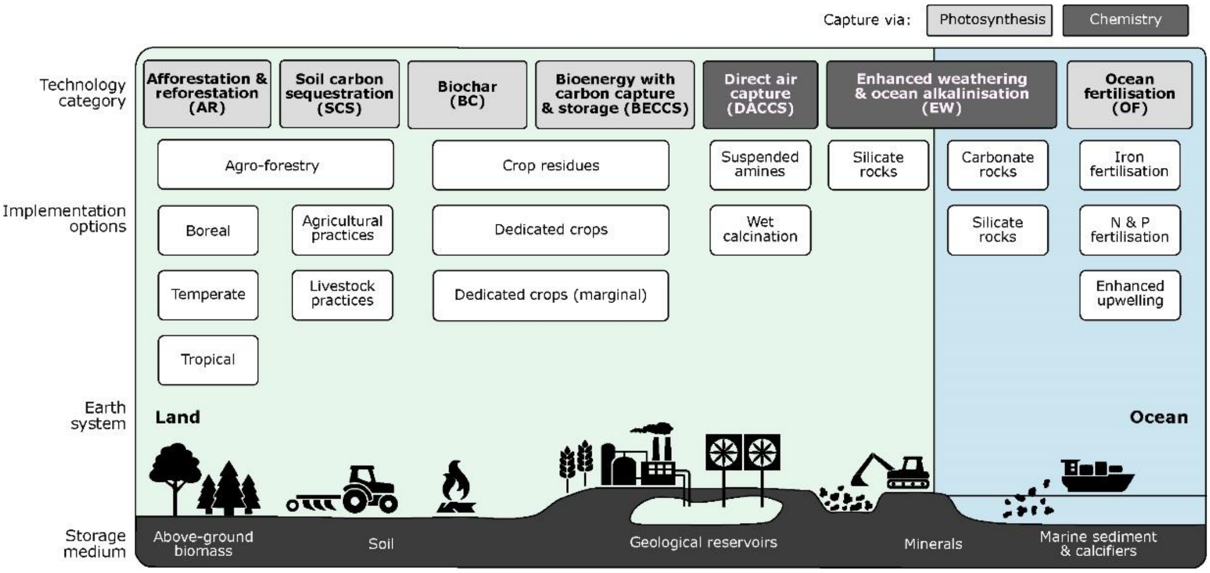


Figure 3: Different methods of carbon dioxide removal. Taken from [5]

1.1 Structure of the thesis

The thesis consists of this introductory essay and three appended papers.

Paper I provides a review of different DAC technologies and compares them from an exergy point of view to evaluate the impact of material consumption on their overall system performance.

Paper II evaluates the application of DAC technologies (developed for carbon capture from atmospheric air) as a means for point source capture for industries emitting flue gas streams with low CO_2 concentration.

Paper III evaluates the plant-scale economics of integrating DAC into a CHP plant providing district heating.

This introductory essay is structured as follows:

Chapter 2 presents the technological background based on Paper I, along with a summary of previous works and the statement of aim and research questions.

Chapter 3 summarizes the methodology used to answer the identified research questions (based on the detailed descriptions provided in Papers II and III).

Chapter 4 summarizes and discusses the main results related to the research questions addressed, based on Papers II and III.

Chapters 5 and 6 conclude and elaborate on future work, respectively.

2 Technological background

Direct air carbon capture and storage (DACCS) processes comprise the following four main steps [6], as depicted in Figure 4:

1. Direct Air Capture by circulation of air through a sorbent for the removal of CO_2, followed by regeneration of the sorbent and release of CO_2.
2. CO_2 conditioning (e.g., liquefaction for transport).
3. Transportation of CO_2 to the sequestration location.
4. Storage of CO_2.

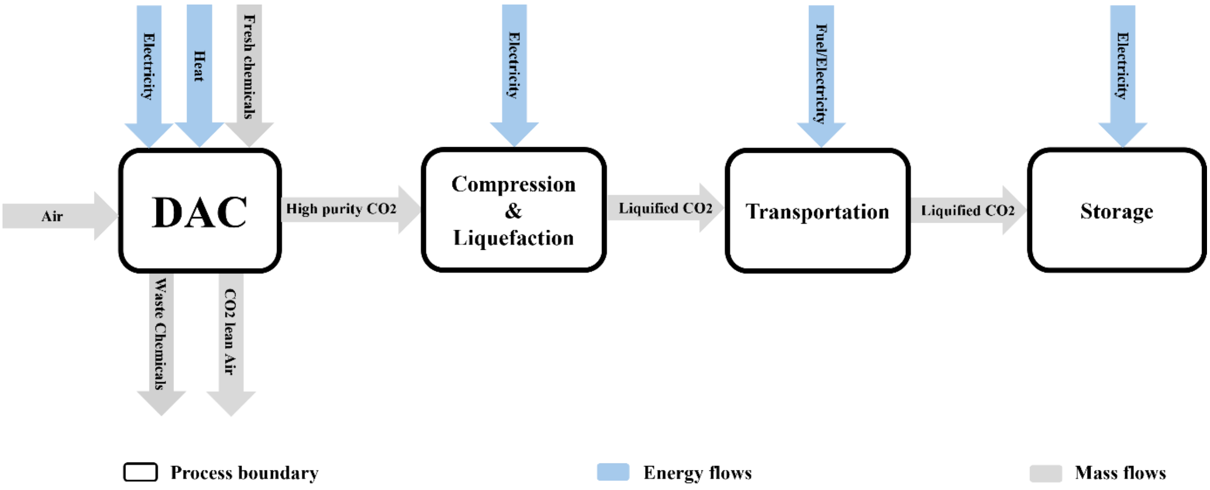


Figure 4: Flowchart for CDR via DACCS.

Since DAC is a gas separation technology, it is commonly compared to carbon capture and storage (CCS) via CO_2 absorption. Table 1 outlines the differences between CCS and DAC, the most important difference being that CCS captures CO_2 from flue gas streams (therefore generating carbon avoidance and the possibility to contribute to CDR (a different category in the hierarchy in Figure 2 if the carbon dioxide in the flue gas stream is biogenic in origin), while DAC removes CO_2 from the atmosphere (hence generating CDR). Further, sizing and location of CCS units is governed by the emission source, while for DAC the location and sizing can be optimized according to different parameters, e.g. weather, access to energy, infrastructure, carbon storage, etc.

Because the concentration of CO_2 in flue gas streams is 2-3 orders of magnitude greater than in atmospheric air, the minimum thermodynamic work for separation of CO_2 in a CCS application is 2-8 times less than for DAC. Thus, the predicted cost for DAC (225-835 $/t$CO_2$) is a few times higher than for the corresponding stage of CCS (40-120 $/t$CO_2$). The current costs for DAC and CCS are higher than these Nth-of-a-kind figures, since only a few plants have been taken into operation. Finally, note that since flue gases usually are at high temperature and contain sulphur oxides and nitrogen oxides, CCS requires cooling and gas cleaning prior to the capture unit.

Table 1: Comparison of main design parameters of CCS (point source capture) and DACCS

	CCS	**DACCS**
Technology readiness level (TRL)	TRL 8-9[7]	TRL 6-7[8]
Inlet CO_2 concentration	4-20%$_{vol}$	420 ppm (0.042%$_{vol}$)
Product	CO_2 emission avoidance (for fossil CO_2) or CDR (for biogenic CO_2)	CDR
Location	Bound to emission source	Result of cost optimization
Scale	Dependent on emission source	Dependent on access to energy and infrastructure
Operation	Dependent on flue gas conditions and composition	Dependent on climate conditions
Prior gas cleaning	Required	Not required
Capture rate	90-99%[9]	Strongly varying, result of the cost optimization
Minimum thermodynamic work requirement	0.45 GJ/tCO_2 [10]	0.052-0.225 GJ/tCO_2
Current cost of avoidance/removal	Approx 470 \$/tCO_2[11], [12]	450-1200 \$/tCO_2[13], [14]
Predicted cost	40-120 \$/tCO_2[15] (without transport and storage)	110-835 \$/tCO_2[16][17]

2.1 Main DAC processes

Absorption

Absorption is a process where a gas molecule enters a liquid or solid sorbent material [17]. Absorption-based DAC uses an aqueous hydroxide solution as sorbent and is therefore commonly referred to as liquid DAC (L-DAC). The process is derived from the pulp and paper industry[18]. Therefore, many components of this process have reached maturity. The process is comprised of two cycles: capture and regeneration [19]; and four main components: air contactor, causticizer, calciner, and slaker which is later complemented with a compression cycle and natural gas combined cycle as power island, see Figure 5.

I. **Air contactor:**

In the air contactor, where the absorption process occurs, air is forced by a fan through a packing with potassium hydroxide (KOH) (or sodium hydroxide (NAOH), as alternative) solution [20]. The CO_2 is dissolved in the liquid phase to form carbonate ions which then react with potassium ions according to Eq.1 to form potassium carbonate.

$$2KOH_{(aq)} + CO_{2(g)} \rightarrow K_2CO_{3(aq)} + H_2O_{(l)} \qquad \Delta H = -95.8\,kJ/mol \qquad (1)$$

II. Causticizer:

Since potassium carbonate is highly soluble in water, the liquid carbonate solution is sent to a causticizer where it reacts with calcium hydroxide fines. During the reaction, the hydroxide solution is regenerated, and calcium carbonate crystals start to form around the calcium fine according to Eq.2. The causticizer process traditionally consists of precipitator, clarification unit, thickener, and mechanical filtration[21]. Recently, this process was replaced by a solid-liquid fluidized bed where a crystallization reaction occurs[22]. This technology was first developed as a water softening process. The outlet carbonates from the fluidized bed are in the form of pellets. Hence the component is commonly referred to as pellet reactor.

$$\mathrm{K_2CO_{3(aq)}} + \mathrm{Ca(OH)_{2(s)}} \rightarrow \mathrm{CaCO_{3(s)}} + \mathrm{2KOH_{(aq)}} \qquad \Delta H = -5.8\ kJ/mol \qquad (2)$$

III. Calciner:

The calcium carbonate pellets from the pellet reactor are first dried using the slaker (see next) as preheater and then sent to the calciner, which is a circulating fluidized bed heated up to 900 °C[19]. There, the calcium carbonate decomposes into calcium oxide and CO_2. The heat for the calcination process is produced through oxy-firing natural gas, which outputs a CO_2/H_2O mixture as flue gas sent to water removal and CO_2 compression. The use of natural gas results in an increased amount of CO_2 to be transported and stored and constitutes one of the main drawbacks of absorption-based DAC. Absorption-based DAC would require storing 1.48 tonnes of CO_2 per tonne of CO_2 removed from air [23]. This highlights the importance of electrification and component development for absorption-based technologies.

$$\mathrm{CaCO_{3(s)}} \rightarrow \mathrm{CaO_{(s)}} + \mathrm{CO_{2(g)}} \qquad \Delta H = 178.3\ kJ/mol \qquad (3)$$

IV. Slaker:

The slaker is a bubbling fluidized bed where calcium oxide from calciner reacts with steam to regenerate calcium hydroxide.

$$\mathrm{CaO_{(s)}} + \mathrm{H_2O_{(l)}} \rightarrow \mathrm{Ca(OH)_{2(s)}} \qquad \Delta H = -63.9\ kJ/mol \qquad (4)$$

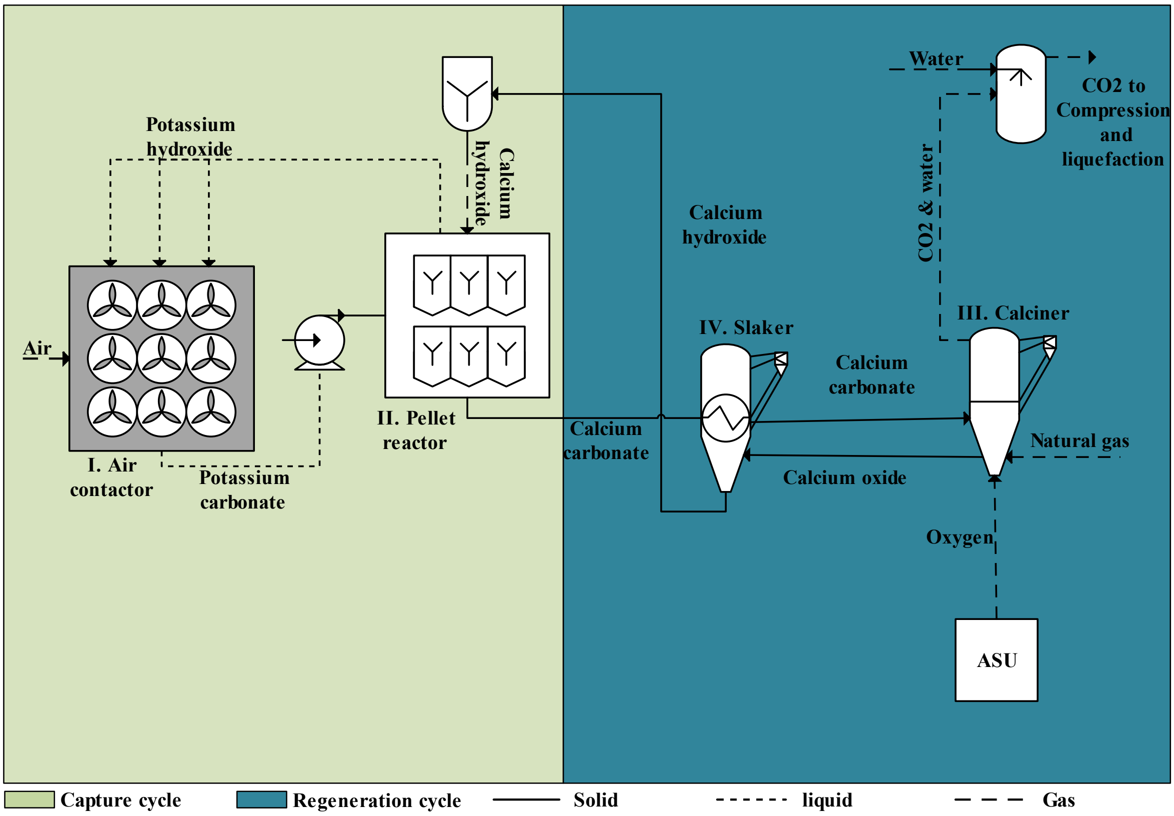


Figure 5: Process scheme for absorption-based DAC

Adsorption

Adsorption-based DAC has received significant attention in the literature, mostly due to the relatively low temperature level (around 80-120 °C) of the required heat input. This makes it possible to integrate the process at limited scale into industrial processes with available waste heat [24], [25], [26]. The adsorption process works based on the affinities of the sorbent towards certain gaseous species under different conditions [27]. For DAC purposes, the sorbents researched are commonly in the solid state. Therefore, the term solid-DAC (s-DAC) is a common alternative name.

Adsorption-based processes can be characterized according to:

- Gas-solid contactor design: The first step of every adsorption process is the actual adsorption step, in which air is brought into contact with the sorbent material. Due to the low concentration of CO_2 in the atmosphere, a large amount of air needs to pass through the sorbent material in order to saturate it. Therefore, designing an air contactor with a trade-off between a small pressure drop and a large gas-to-solids surface is essential for reducing the power consumption of the fan/blower. Various contact methods and contactor designs, such as fluidized bed [28], packed bed [29], and monoliths [30], [31] have been proposed for DAC application. However, only the packed bed design has shown promising results.
- Regeneration method: The amount of gas adsorbed in an adsorption process is a function of temperature, pressure, and composition of the inlet stream [32]. Hence different

adsorption processes are identified based on the adsorption and desorption/regeneration conditions. Three main sorbent regeneration methods for DAC purposes are:

1. Temperature swing adsorption (TSA): the sorbent is heated up for desorption.

2. Steam-assisted temperature swing adsorption: the sorbent is contacted with steam to increase its temperature and keep a low partial pressure of CO_2 during desorption. Such steam-assisted desorption results in faster regeneration of the sorbent and larger adsorbing capacities of the sorbent regenerated. On the other hand, the regenerated sorbent will contain residual water, and the product stream will have a high moisture content requiring condensation [33].

3. Temperature vacuum swing adsorption: desorption happens at a higher temperature and lower pressure than ambient conditions, by means of a vacuum pump (extra electricity requirement[34], [35]) and temperature control. This cycle allows for the utilization of heat at lower temperatures.

- Sorbent material: The main attributes of interest are the internal mass transfer, heat conductivity, and adsorption kinetics [36]. Amine-functionalized sorbents are the most common materials for DAC adsorption processes, mostly due to their adsorption kinetics and performance in the presence of moisture [34]. Their adsorption of CO_2 is enhanced through adsorption of water, although the level of enhancement reduces as the level of water adsorption increases. Adsorption-based DAC has a stream of water as byproduct. Climeworks (the commercial technology provider for adsorption-based DAC) reports 0.8-2 tH_2O/tCO_2 because of co-adsorption of water and CO_2 on their sorbent. This would allow co-location of DAC with an electrolyser and methanation unit for carbon capture and utilization purposes[37].

Unlike absorption processes in which each process step occurs in a dedicated component, most of the adsorption processes are modular, with the entire process happening in one module. Figure 6 shows the six main steps of the temperature-vacuum swing adsorption commercialized DAC process by Climeworks [38]. This process uses an amine-functionalized sorbent encapsulated in metal frames and arranged in a zigzag pattern inside the contactor frame [29]. The process consists of the following steps:

1. Adsorption: Inlet and outlet are open for air to pass through the reactor where water and CO_2 are adsorbed, and the CO_2-lean gas is vented.
2. Blowdown: The inlet is closed, and a vacuum pump reduces the pressure to 0.2 bar by venting the gas remaining in the column (waste gas) via a vacuum pump.
3. Preheating: The bed is heated while the vacuum pump maintains the desorption pressure. The desorbed gases (mostly water at this stage) are vented.

4. Desorption: The bed is heated to the desorption temperature of 100 °C (80 °C-120 °C) and the CO_2-rich desorbed gas is collected for further processing.
5. Cooling: The inlet and outlet of the column are closed, and the bed is allowed to cool down to 90 °C (to avoid sorbent oxidation) [34]through heat exchange with the ambient air.
6. Pressurization: The inlet of the column is opened, and a new batch of air is allowed to enter the reactor, thus increasing the pressure to the adsorption pressure and re-starting the cycle.

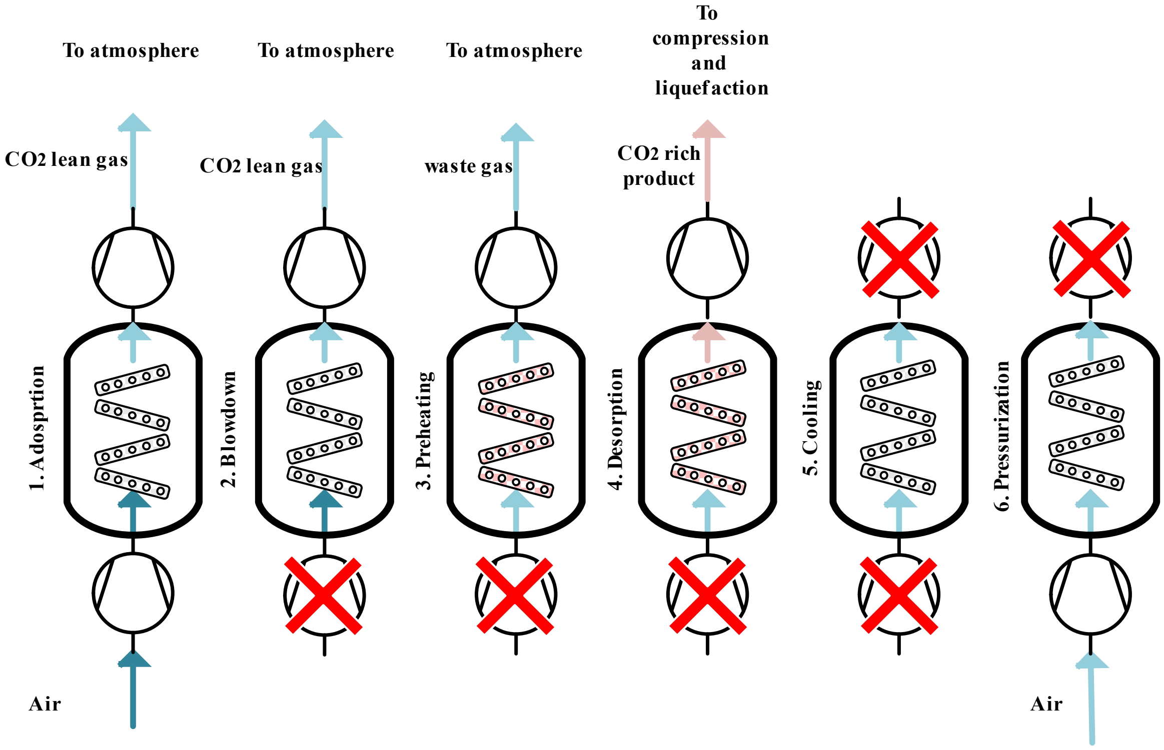


Figure 6: Process steps for temperature vacuum swing adsorption.

Ion exchange

The ion exchange (also called 'moisture swing') process was introduced by Klaus Lackner in 2009 [39]. This process removes CO_2 through bringing open air into contact with a membrane composed of the sorbent, which is a type of anionic exchange resin. The material contains chloride ions, which are replaced by hydroxide and carbonate ions when the material is washed in solutions that contain these ions. These ions bind to CO_2 in the bicarbonate state according to Eq. 5:

$$CO_3^{2-}(R^+)_2.xH_2O + CO_{2(g)} \leftrightarrow 2[HCO_3^- R^+.yH_2O] + (x-2y-1)H_2O_{(g)} \qquad (5)$$

In order to release CO_2, the equilibrium can be pushed backwards by exposing the saturated material to water vapor. Thereafter, the material is placed back in contact with ambient air for natural drying. The sorbent will cycle between the carbonate and bicarbonate forms. Since each bicarbonate needs one positive charge and each carbonate needs two positive charges, the maximum loading of the material is equal to half of the positive charge concentration.

Electrochemical

Electrochemical processes have come into focus recently for the purpose of electrifying DAC processes. Electrodialysis is one of the main electrochemical DAC systems. In this process, hydrogen is sent to an anode to produce ionic hydrogen, while air is sent to a cathode to generate hydroxide. CO_2 is absorbed by the hydroxide at the cathode, yielding carbonate or bicarbonate, which reacts with hydrogen at the anode to release CO_2.

Electro-swing adsorption is another electrochemical DAC process, in which air passes through a nanotube made of sorbent. The affinity of the specific sorbent for this process towards CO_2 can be increased by positively charging the material. When the material is saturated, the process is reversed by changing the polarization to release CO_2.

The main advantage of electrochemical processes is that there is no need for many auxiliary components, such as heat exchangers, or for heating up large volumes of material, thereby increasing the thermodynamic efficiency of the system. Moreover, in electrochemical processes, unlike adsorption processes, CO_2 is directly targeted and removed without the involvement of any secondary output streams. On the other hand, conveying air through the fuel cell requires high fan power. Electrochemical processes for DAC are still in the early stages of development, and the stabilities of the sorbent materials remain to be proven. Therefore, they are not considered in this work.

Passive DAC

Passive DAC generally refers to processes where the wind naturally moves air through the sorbent instead of the air being moved by a fan, reducing the energy requirement per tonne of CO_2 removed. Carbonation of natural hydroxides is the most studied passive DAC process. Given that natural carbonation is a slow process, passive DAC systems are generally very slow and therefore require a large area for sorbent distribution. As in the case of absorption-based DAC, once calcium hydroxide is converted to calcium carbonate, it is sent to a calciner. Then the outlet calcium oxide reacts with steam to regenerate the calcium hydroxide. The company Heirloom has commercialized passive DAC technology that uses calcium hydroxide as the sorbent. Passive DAC processes are not included in this work due to lack of data availability.

2.2 Comparison of different technologies

Table 2 presents energy and material requirements for some of the DAC technologies studied in the literature. The technology readiness level and cost of capture for each process is also added where available.

Table 2: energy and material requirements for some of the DAC technologies studied in the literature

Technology	**Material**	**Heat (kWh/tCO_2)**	**Electricity, (kWh/tCO_2)**	**Capacity loss/recovery (%/cycle)**	**Material consumption (kg/t_{CO_2})**	**Cost reported ($/$tCO_2$)**	**TRL**
Adsorption TVS	Amine functionalized [40], [41]	1,500–3,305	500–700	0.05 [42], [43]	13.87	280-580 [17]	7-8
	MOF(Cr) [44]	2,688	220	1.5 [42]	531.51		3-4
	MOF(Mg) [44]	1,319	201	0.005 [42]	0.73		3-4
	PEI on alumina [30]	1,656	218	0.05 [42]	7.39		5-6
	K2CO3/AL2O3 [45]	889	945	0.21 [45]	75.26		3-4
	Hydrated K2CO3 [46]	1,170–1,410	150–260	0.05	88.77	220-270 [17]	3-4
Adsorption TSA	TRI-PE-MCM-41 [47]	2102	76	6.25 [42]	3435		3-4
	PEI on Silica [28]	2083	694	0.05 [42]	7.39		3-4
Absorption	KOH (NG + grid) [48], [49]	1458.	366.00	1	25.45 KOH, 30.86 $CaCO_3$	225-545 [17]	6-7
	KOH (NG only) [48], [49]	2450.	0	1	25.45 KOH, 30.86 $CaCO_3$		6-7
	NaOH [21]	1678	440	1	18.18 NaOH, 228 CaCO3 [21], [50]		
	NaOH [51]	2250	494	1	18.18 NaOH, 3.5 $CaCO_3$ [21], [50], [52]		
Ion exchange	Anionic exchange resin [39], [53], [54]	0	378	Max 100,000 cycles [55]	1.515–2.02 [55]	110 [17]	3-5

To compare the DAC technologies with respect to materials, electricity, and heat, the efficiency of each DAC technology has been calculated from three perspectives.

First law perspective: Provides an overview of total energy demand per tonne of CO_2 captured.

$$\eta_I = \frac{W_{min,CO2}}{EL_{in}+Q_{in}} \tag{6}$$

Second law perspective: Takes into account the equivalent work value of the heat supplied by considering its relative temperature level.

$$\eta_{II} = \frac{W_{min,CO2}}{EL_{in} + Q_{in} * \left(1 - \frac{\mathrm{T_{env}}}{\mathrm{T_{bf}}}\right)} \tag{7}$$

Exergy perspective: As illustrated in Figure 4, DAC uses heat, electricity, and fresh chemical materials to remove CO_2 from the atmosphere. While the second law perspective includes electricity and the work equivalent value of heat, it neglects the material flows. The exergy perspective takes into consideration the work equivalent value of the material flows through their chemical exergy, thereby accounting for the impact of material degradation and the chemical material thermodynamic value.

$$\eta_{ex} = \frac{Ex_{ch,CO_2}}{\sum Ex_{ch,in} - \sum Ex_{ch,byproducts}} \tag{8}$$

Figure 7 shows the values of these three efficiencies for the DAC processes considered. Ion exchange process presents a very high first-law efficiency (Eq. 6), relative to the others, due to recovery with moisture at very low temperatures. For all other processes, the first law efficiency is below 10%, due to the low concentration of CO_2 in the atmosphere. However, the efficiency based on the second law perspective (Eq. 7) shows an increase for all technologies except absorption-based DAC. This highlights the drawback of high-temperature processes and their need for fuel combustion.

Finally, when applying the exergy-based efficiency (Eq.8), efficiency values drop for all processes due to the consideration of chemical degradation of the material. The adsorption processes and ion exchange process suffer the most from material degradation, positioning the development of these processes as a research target.

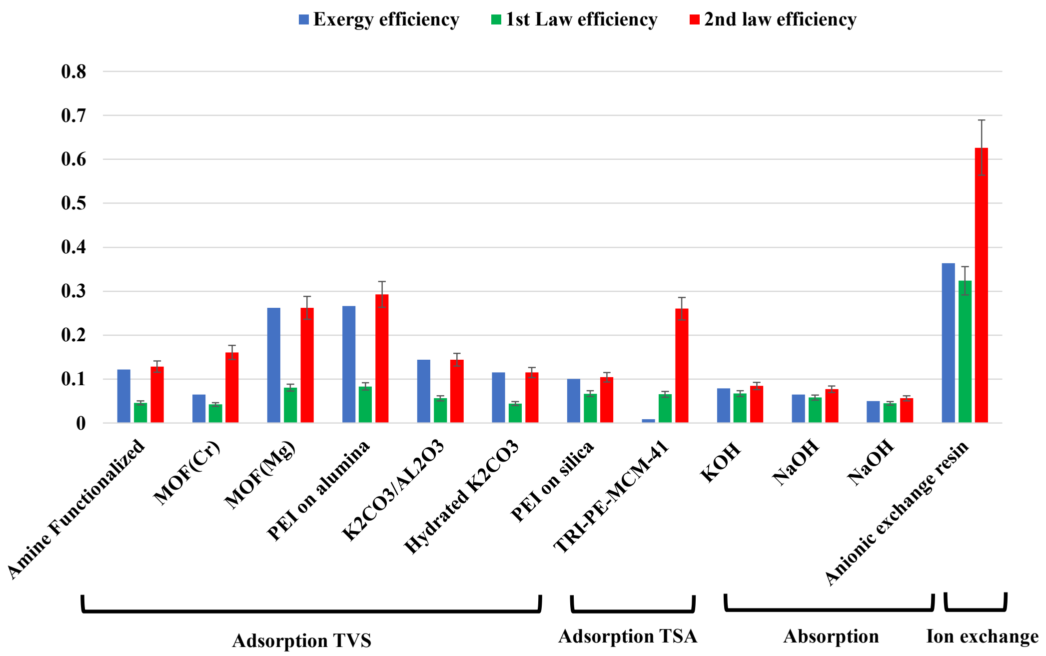


Figure 7: DAC system average efficiency values (based on first law (Eq. 6), second law (Eq. 7), and exergy (Eq. 8)). The black bars show value ranges whenever possible based on available literature data. Source: **Paper I.**

2.3 Challenges for DAC

The primary challenge to large-scale deployment of direct air capture is its high net removal cost. The cost picture is dominated by three key factors:

- high energy demand: Energy requirements can only be reduced through technological advancements and process integration. This requires research on process optimization and/or integration with other industrial processes.
- indirect emissions from the DAC chain: The impact of indirect emissions can be mitigated by using renewable energy sources to power DAC operations. As a result, the recent literature has increasingly focused on evaluating DAC performance under different energy supply scenarios.
- significant capital investment: The high capital investment required for DAC technologies can be reduced through further deployment, allowing the technology to progress along the learning curve.

This underscores the importance of strategic DAC deployment, which involves two key considerations:

1. **Geographical Location**
   The location of DAC deployment influences several factors that impact net removal cost:

  - **Climate conditions**, which affect the energy requirements of the process [56], [57].
  - **The availability and carbon footprint of different energy sources**, which influence DAC's operational expenditure[41], [58].
  - **Access to infrastructure for CO_2 transport and storage**, which can increase the cost based on distance and type of transport[59].

2. **Application of the Technology**
   While DAC was originally developed for CO_2 removal from ambient air, it can also be considered a gas separation technology optimized for low CO_2 concentrations in the inlet stream. Moving beyond the traditional DAC application could unlock new use cases based on the technology's core attributes rather than just its input source. This broader application potential could drive additional demand, further accelerating deployment and helping to lower capital costs along the learning curve.

2.4 Aim

The overarching aim of this thesis is to explore two alternative opportunities for the deployment of DAC technologies, namely an alternative cost-effective location and an alternative cost-effective application. This thesis focuses on adsorption-based and absorption-based DAC, which are the most developed DAC technologies, and which have reached commercial deployment. We use established methodologies from the literature to answer the following three research questions (RQ):

RQ I: Do DAC technologies offer better economic performance than conventional amine flue-gas scrubbing capture (developed for CCS) when used as point source capture technology for industries with flue gas streams with a low CO_2 concentration? (investigated in Paper II)

RQ II: What is the most economic option between CO_2 capture and CO_2 offset for industries with flue gas streams with a low CO_2 concentration considering? (investigated in Paper II)

RQ III: Is integration of DAC into combined heat and power plants that provide district heating a cost-efficient deployment opportunity for adsorption-based DAC? (investigated in Paper III)

3 Methodology

This chapter briefly presents the overall methodology used in this thesis to address the three research questions stated in section 2.3. For more detailed descriptions, the reader is directed to the appended papers.

Figure 8 shows the assessment framework of this thesis with the models used to address each research question. Aligning with the common practice to use cost metrics as the primary decision-making criterion, the assessment framework for this thesis is based on comparing net removal costs or carbon avoidance costs with assumed prices on the voluntary carbon offset markets, here referred to as CDR prices. Thus, if the estimated costs for a given case are lower than the assumed CDR prices, the option is considered economically viable.
A technoeconomic model is employed to estimate net removal costs or carbon avoidance costs, based on flow and size data from energy and mass balances, which are closed through a process or plant model.

For **RQ I** and RQ **II** (investigated in **Paper II),** a process model is used to close heat and mass balances and determine the sizing of components. Then a technoeconomic model post processes these results to assess the carbon removal/avoidance costs which are compared with CDR prices to assess economic viability. In contrast, for **RQ III (**investigated in **Paper III)**, the technoeconomic assessment and comparison with CDR prices are integrated directly into the plant model. The following sections provide summaries for each of the models used.

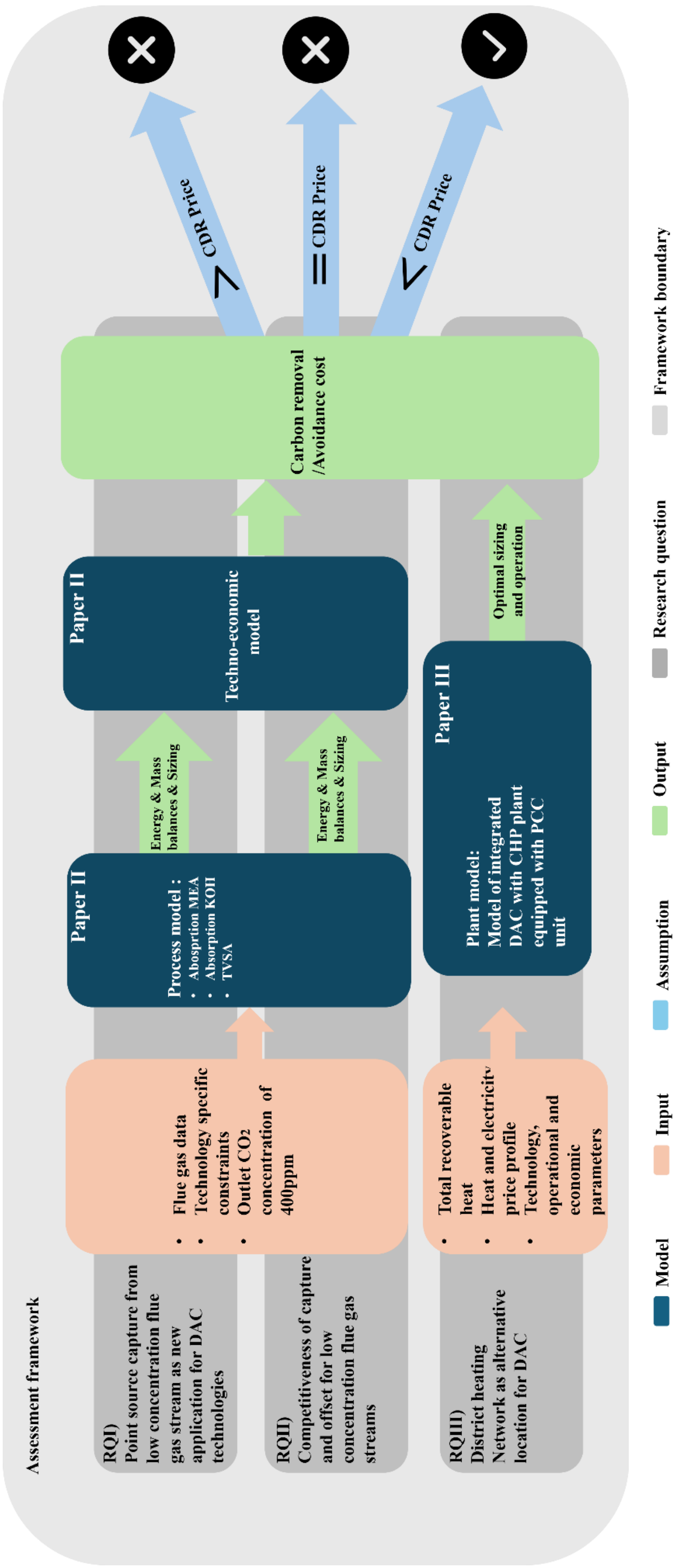


Figure 8: A graphical representation of the methodology used in this thesis.

3.1 Process model

The process models that close the mass and energy balances are summarized below. By using these process models, the equipment for each process was sized to achieve an outlet CO_2 concentration of 400 ppm (effectively yielding net zero direct CO_2 emissions). The assessment is carried out for a range of inlet flow rates (475-2700 t/h) and CO_2 concentrations (0.5-4%). Additional specific process descriptions and constraint details are given in **Paper II.**

- Absorption MEA (MEA-ABS)

  A steady-state process model for MEA-ABS is constructed in Aspen Plus v14.0. The model also includes downstream compression and liquefaction. The MEA-ABS model is based on the work of Biermann et al. [60], while the compression and liquefaction model is based on work of Deng et.al. [61]. The size of the direct contact cooler, absorber, and striper are determined by changing the diameter of the column while keeping the same height in order to achieve flooding limit at each column (here 80%). The height of the washer is changed to limit MEA in emissions to the atmosphere to 0.1 ppm. Finally, the compression and liquefaction process is dimensioned so that the outlet CO_2 stream composition meets the specifications for medium pressure ship transport required by the Northern Lights project [62].
- Absorption via KOH (ALK-ABS):

  A steady-state process model for absorption via KOH with subsequent calcium looping is developed in Aspen Plus v14.0. The model follows the general description given by Keith et al. [19] albeit using electricity from the grid instead of from a dedicated power island. The air contactor component is modeled using a rate-based model built in absorption column in Aspen Plus and is sized by varying the gas flow rate to achieve the desired capture rate. The rest of the components and flows are also sized to maintain 900 ℃ calcination temperature in the calciner. 90% retention in pellet reactor and a 30% wt concentration of calcium hydroxide after slaker.
- Temperature vacuum swing adsorption (TVSA):

  A 1-D transient model of a six-step TVSA column was developed in gPROMS using its built-in adsorption column model. The design of the column is changed from the zigzag plates to series of stacked plates which is closer to common adsorption columns. Customized codes for adsorption isotherms [34] and a co-adsorption model were added to account for enhancement of CO_2 adsorption through co-adsorption of water[34]. The model was run in a Monte Carlo analysis framework with varying time of adsorption, time of heating, time of desorption, column height, and molar fraction of water at inlet, in which the result of each run case was later used as an input to the technoeconomic model [63], thus assessing the cost of the multiple configurations simulated and making it possible to find the one with the lowest cost.

3.2 Technoeconomic model

Figure 9 shows the boundaries for the cost assessment used, with two regions representing levelized costs of carbon capture (LCOC) (Eq. 9) and carbon avoidance (LCOA) (Eq. 10).

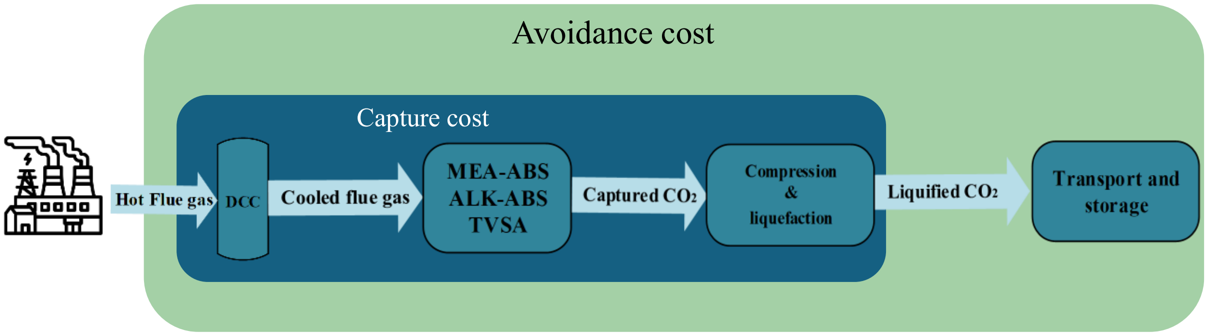


Figure 9: System boundaries for calculating levelized cost of CO_2 capture (blue frame) and levelized cost of CO_2 avoidance cost (green frame). Source: Paper II

The levelized cost of capture is used to compare the performance of carbon capture processes, while the levelized cost of avoidance includes the transportation and storage costs to enable comparison of a given process with offsetting via purchase of CDR credits.

$$\text{Levelized cost of capture} = \frac{C_{\text{inv}} + C_{\text{fixed O\&M}} + C_{\text{variable O\&M}}}{m_{CO_2,\text{captured}}} \tag{9}$$

$$\text{Levelized cost of avoidance} = \frac{C_{\text{inv}} + C_{\text{fixed O\&M}} + C_{\text{variable O\&M}} + C_{\text{transportation \& storage}}}{m_{CO_2,\text{avoided}}} \tag{10}$$

The capital investment (C_{inv}) of each component is determined using the bottom-up/top-down cost estimation method shown in Figure 10. Economic parameters, assumptions, and cost escalation factors are chosen based on the case studies. The top-down approach includes extracting data reported in the literature or using vendor data for a whole unit (including all associated equipment), typically reported as engineering procurement and construction costs. This approach is used for equipment that is either commercially available or for which data is available in the literature for entire subsystems (e.g. for most of absorption-based DAC components). The bottom-up approach uses energy and material flow data from the developed process models to size and assess the cost of each piece of equipment. The direct cost of each piece of equipment was obtained from direct cost data or regressed direct cost functions available in the literature (e.g. MEA-absorption and TVSA). The total capital requirement (TCR) estimated from the hybrid top-down/bottom-up capital cost estimation method is annualized over the assumed plant lifetime or the design lifetime of the process technology. The fixed operation and maintenance ($C_{\text{fixed,O\&M}}$) is estimated based on total plant cost and labour cost required adopting the method from[64]. The variable operation and maintenance cost, $C_{\text{variable,O\&M}}$, is dominated by the costs of electricity and heat, which in this work are based on assumed values. The cost for transportation and storage ($C_{\text{transportation \&storage}}$) varies strongly based on location and are therefore used as a sensitivity parameter in our work.

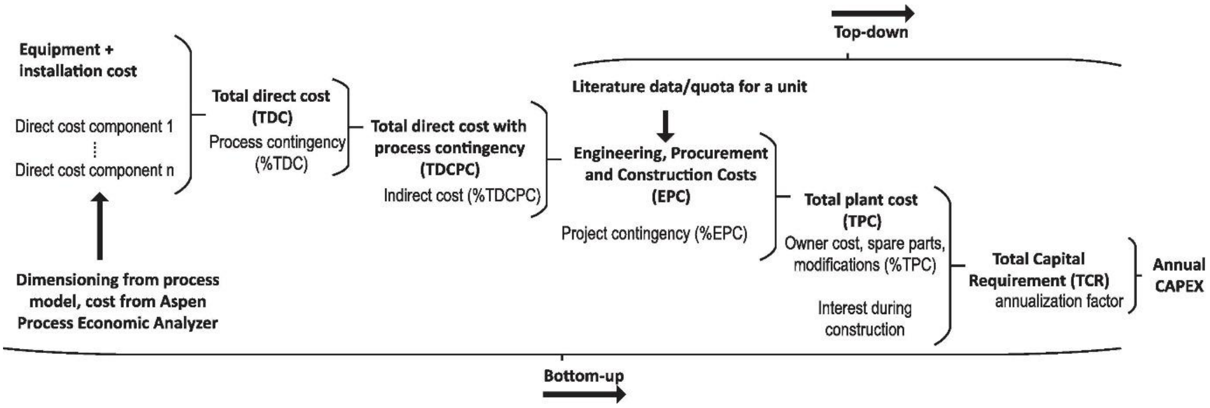


Figure 10: Capital cost estimation method using bottom up or top-down method. The arrows indicate the entry point and direction of cost scaling factors.[61], [64], [65], [66]

3.3 Plant model

The plant model presented in **Paper III** (see Figure 8) uses linear optimization to evaluate the performance of DAC when optimally integrated with a CHP plant and a Post-Combustion Capture (PCC) unit.

Figure 11 shows the overall framework for the analysis, which begins with running an existing process model for a CHP plant with PCC and using the result to create the Actual Cooling Load Curve (ACLC), which provides the recoverable heat available at low (30–50 °C) and mid (50–90 °C) temperature levels.

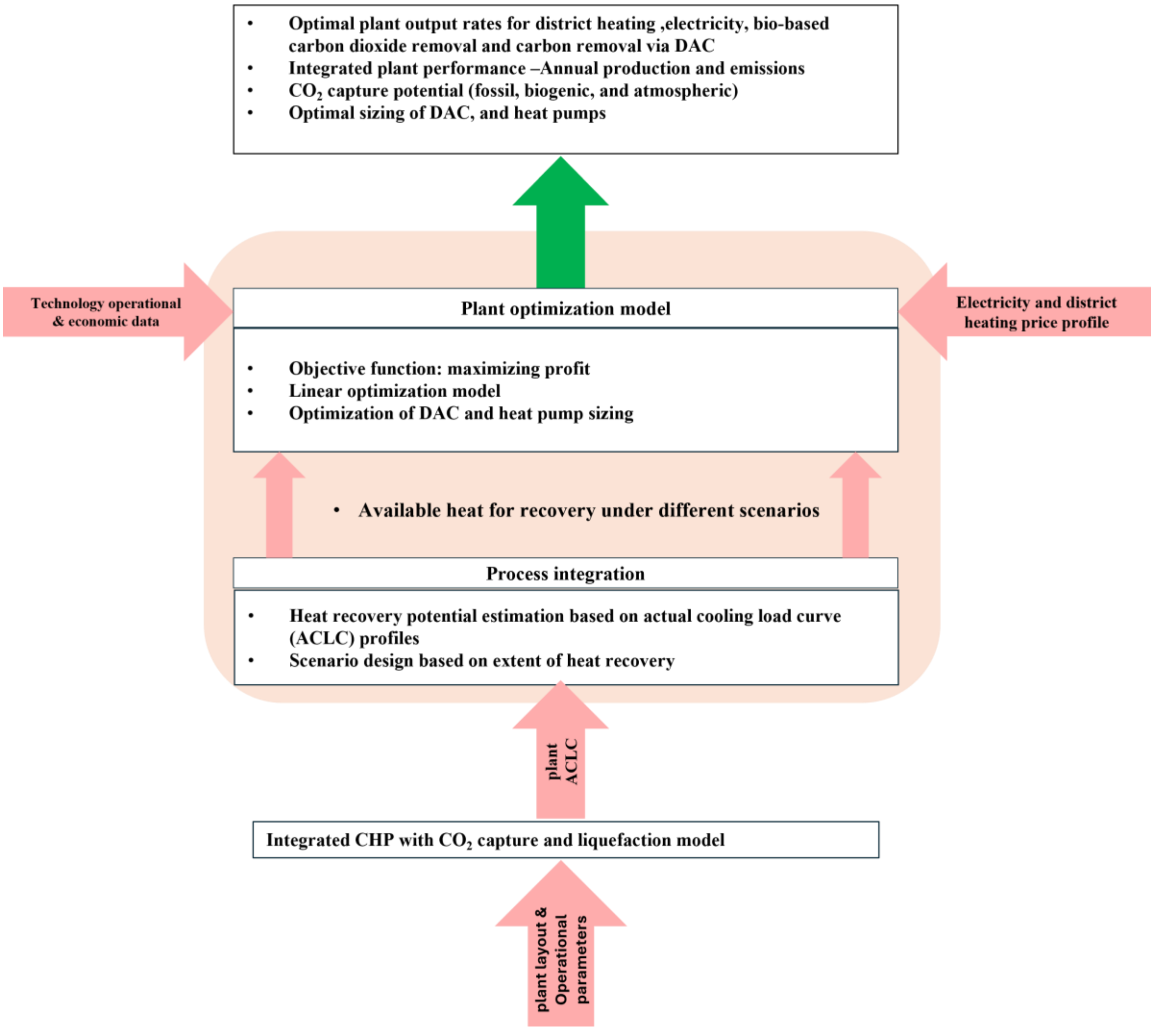


Figure 11: Overall framework for assessing the integration of DAC into CHP plants. Source: **Paper III**

A linear optimization framework is then developed to assess the interactions between CHP, DAC, PCC, and the district heating network. This model incorporates the technological and operational parameters of each component, along with heat and electricity price curves. The model sets as objective to maximize the plant's annual net cash flow while maintaining energy and mass balances within the system. For this, the model is allowed to adjust sizing of the DAC process and operation of the system (dispatch strategy for heat, electricity, and CDR). The net cash flow includes revenues from electricity and district heating sales, carbon dioxide removal (CDR) credits (from capturing both biogenic and atmospheric CO_2), and avoided CO_2 taxes (from capturing the fossil carbon share of the fuel). On the cost side, fuel and investments in new equipment are considered.

Furthermore, a sensitivity analysis studying the impact of five main parameters (heat price, electricity price, electricity price volatility, operational hours, fuel type) on the optimal amount of CDR is conducted. Table 3 presents the values of these parameters for the base case as well as for a sensitivity analysis.

Table 3: Sensitivity analysis parameters.

	Base case	**Sensitivity analysis**
Heat price	Profile in southern Sweden (average, 10.26 €/MWh)-	50%, +50%, +100%, +900%
Electricity price value	Profile in southern Sweden (SE3) in 2022 (average, 129 €/MWh)	±30%
Electricity price volatility	Profile in southern Sweden (SE3) in 2022	Low: the predicted electricity price profile in 2050 in northern Sweden (SE1) High: the predicted electricity price profile in 2050 in southern Sweden (SE4)
Fuel type	Municipal solid waste	Biomass
Operational hours /heat profile	Waste-fired CHP 8,000 full-load hours/year	Bio-CHP plant 4,500 full-load hours/year

4 Results and discussion

This chapter presents the main results from **Papers II** and **III** appended to this thesis. These selected results are presented in two sections:

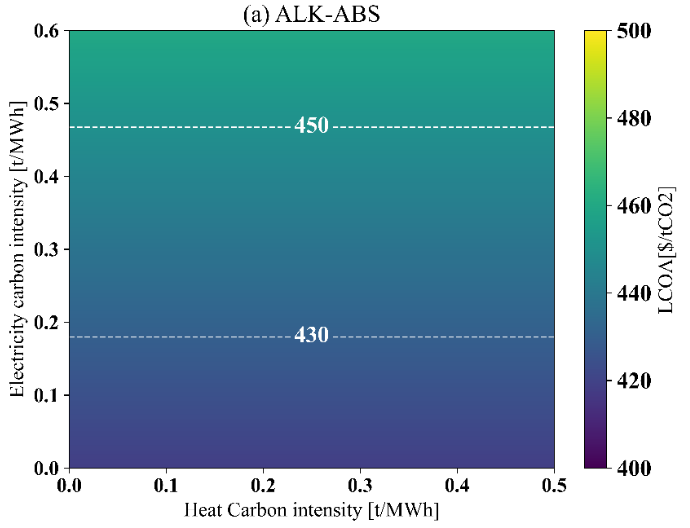
i) techno-economics of CO_2 capture from flue gas stream with CO_2 concentration (seeking to answer RQ I & II),

ii) integration of DAC into combined heat and power plants (seeking to answer RQ III).

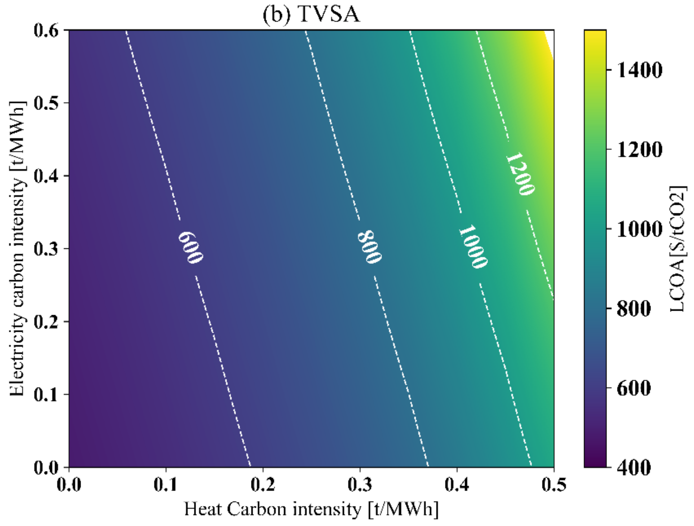
4.1 Capturing CO_2 from low-concentration flue gas streams

The results presented in this section are based on the work included in Paper II. Figure 12 presents the levelized cost of capture for the three technologies considered (ALK-ABS, MEA-ABS, TVSA) for different flue gas flow rates (475-2700 t/h) and CO_2 concentrations (0.5-4%). We find that MEA-ABS tends toward having the lowest cost among the three technologies as the absolute mass rate of capture increases (by means of higher CO_2 concentration and/or greater flue gas flow rate), yielding the lowest cost for flow rates above 500 t/hr. The LCOC for both ALK-ABS and MEA-ABS grows exponentially as the absolute mass rate of capture decreases, with a steeper increase for MEA-ABS. Hence, at flow rates below 500 t/hr and concentrations below 0.5%, ALK-ABS has better economic performance compared to MEA-ABS. The TVSA process has the worst economic performance among the three for the flue gas ranges studied. This is due to the limitation of packed bed columns in handling large gas flow rates, which results in a higher number of columns required for handling a flow rate, leading to a high capital cost. In contrast to MEA-ABS and ALK-ABS, the LCOC for TVSA almost stays constant with flue gas flow rate. Hence, at small enough flow rates we expect TVSA to have the best economic performance among the three.

(a) ALK-ABS



(b) TVSA



(c) MEA-ABS

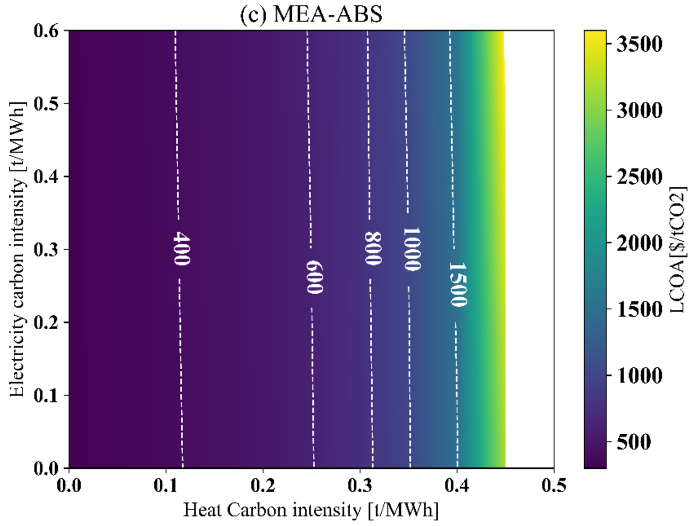


Figure 12: Levelized cost of capture as a function of CO_2 concentration in dry flue gas and flue gas flow rate, for a) MEA-ABS, b) ALK-ABS, c) TVSA. Source: **Paper II**

Figure 13 shows the cost break down of LCOC for each carbon capture technology considered, for cases with a flow rate of 500 t/h at varying CO_2 concentrations. Please note that Fig.13 complements the corresponding figure in **Paper II**, which represents cases with a flow rate of 1000 t/h. The costs are divided into capital expenses, fixed operation and maintenance, and the consumption of heat, electricity, and materials. The results indicate that for most cases, capital expenses constitute the largest cost component, largely due to the low CO_2 concentration in the flue gas. However, as the CO_2 concentration increases, the share of capital costs declines, while operational costs gain significance.

Figure 13 underscores the high material consumption costs for TVSA, primarily driven by the expensive sorbents required for the process, in agreement with conclusions from the comparison of efficiencies in Figure 7. These elevated costs are more justifiable in direct air capture (DAC) applications, where higher carbon dioxide removal (CDR) credit prices are anticipated. However, in the case of CO_2 capture from flue gases, these costs significantly impact economic feasibility. For MEA-ABS and ALK-ABS, material costs represent a minor share of total expenses, attributed to low sorbent replacement rates and lower-cost materials, respectively.

While the absolute electricity demand differs across the three technologies, the relative share of electricity costs is similar. In contrast, the cost share for heat supply varies significantly, with MEA-ABS showing the highest proportion of heat-related costs due to its lower LCOC value.

The results for the cost break down of the LCOC at 1000t/h flue gas flow rates presented in **Paper II** show the same trends as in Figure 13.

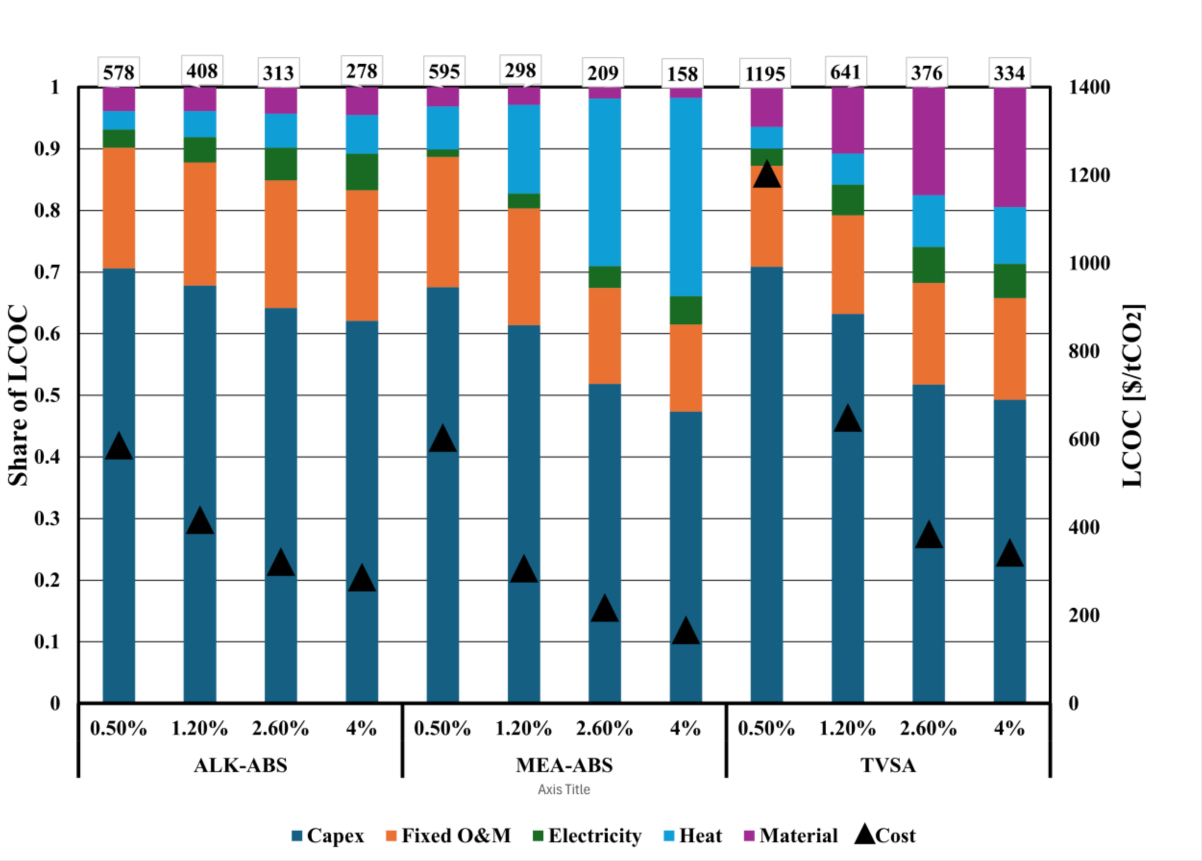
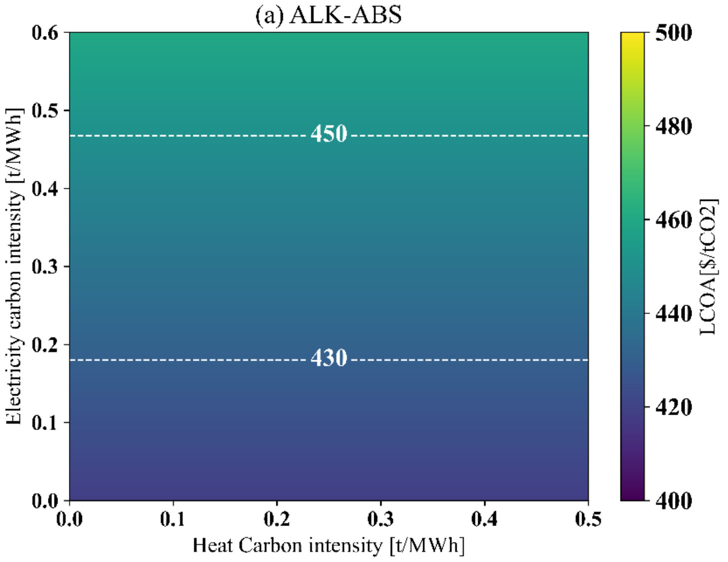
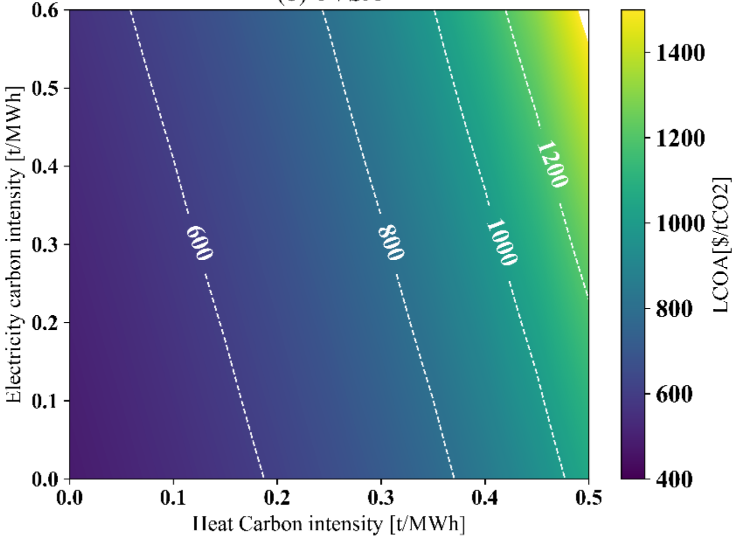


Figure 13: The cos breakdown of LCOC for each capture technology.
The data tags (up top) and triangles (in bars) show the LCOC for each case.

Figure 14 illustrates the LCOA for the three different carbon capture technologies as a function of the carbon footprint of the heat and electricity consumed. This analysis is based on cases representative of the hydrogen direct reduction (HDR) steel production process, i.e., with a flue gas CO_2 concentration of 2.6% and a flue gas flow rate of 500 t/h. To account for the full carbon capture chain, a transport and storage cost of $105 per tonne of CO_2 is assumed.


(a) ALK-ABS


(b) TVSA

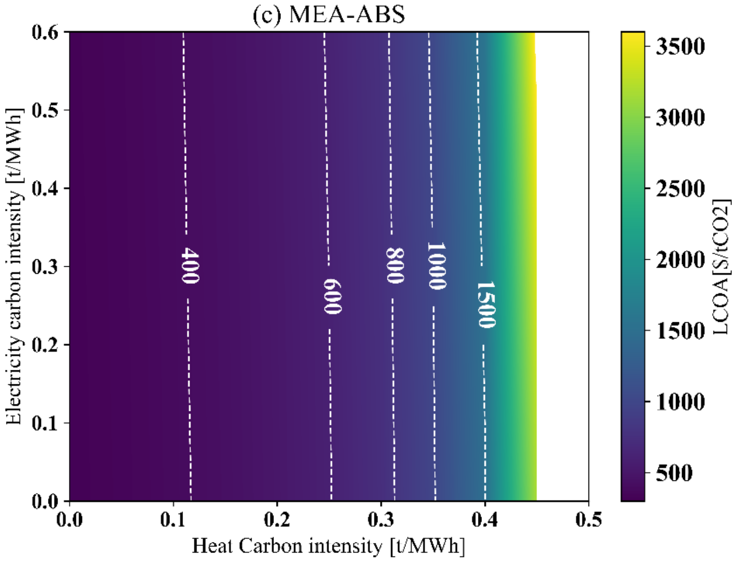

(c) MEA-ABS

Figure 14: LCOA as function of the carbon footprints of the heat and electricity supply, for the case of [CO2] =2.6% and 500 t/h (representative for hydrogen direct reduction in steel industry). a) ALK-ABS, b) MEA-ABS, c) TVSA.

The results indicate that the LCOA of ALK-ABS remains relatively stable regardless of the carbon footprint of the energy input, whereas MEA-ABS and TVSA are significantly affected. The key factor behind ALK-ABS's cost stability is its use of oxy-fuel combustion, which provides heat while integrating carbon capture. In theory, this could allow for a negative carbon footprint through oxy-fuel combustion of bio-based fuel.

For MEA-ABS, the LCOA is highly sensitive to the carbon footprint of heat supply but less dependent on the carbon intensity of the power supply. When low-carbon heat is available, MEA-ABS achieves the lowest LCOA, making it the most cost-effective option in such scenarios. However, if the carbon intensity of heat exceeds 0.15 tCO_2/MWh, ALK-ABS becomes the more economical choice.

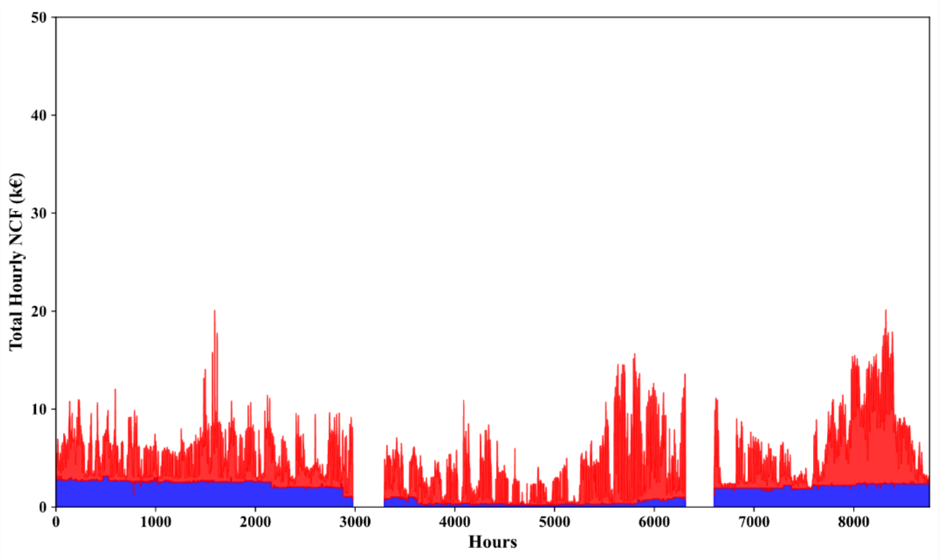
TVSA exhibits the highest LCOC among the three technologies, with its LCOA being strongly influenced by the carbon footprint of heat (to a greater extent) and electricity (to a lesser extent).

Thus, the results shown in Figure 14 indicate that, at CDR credit prices below 400 $/$tCO_2$, capturing CO_2 would become less favourable compared to emission and offset through purchase of CDR credits. Note that this applies not only to the flue gas conditions studied (500t/hr of flue gas with a CO_2 concentration of 2.6%), but also to those with smaller flow rates and lower CO_2 concentrations (as they yield increased costs).

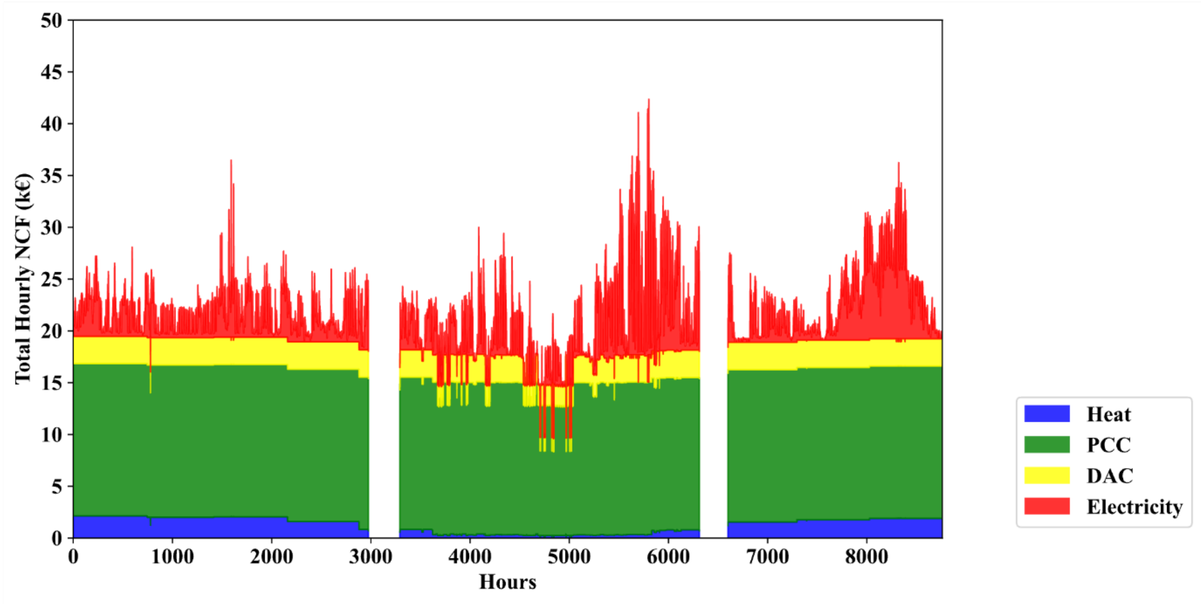
Compared to the case for aluminium production industry (studied in **Paper II**), the HDR-steel case presented here shows that ALK-ABS becomes more cost-effective than MEA-ABS at a lower heat carbon intensity. Moreover, for the aluminium production case the MEA-ABS is economically competitive within larger ranges of carbon footprint of the heat. Finally, it can be observed that higher inlet CO_2 concentrations reduce the LCOC gap between TVSA and the other two processes studied.

4.2 Integration of DAC into combined heat and power plants

The results presented in this section are based on the work included in **Paper III**. Figure 15 shows the hourly net cash flow (NCF) of the reference CHP plant (167MW waste-fired CHP), for both business as usual (Figure 15a) and the retrofit after addition PCC and DAC (Fig. 15b). As illustrated in Figure 15a electricity sales constitute the largest share (68.3%) of net cash flow for the assumed business-as-usual conditions (which include an electricity price curve averaging 129€/MWh). According to Figure 15b, for a CDR credit selling price of 615€/tCO_2 (assumed based on DAC cost estimation from [16] and 65€/tCO_2 for transportation and storage for Sweden), the integration of PCC and DAC yields a large increase in the net cash flow with contributions from DAC (11.8%), capture of biogenic CO_2 via PCC (67.1%), electricity sales (14.9%), and heat sales (6%). The results show that entering the CDR market offers a profitable business case for CHP plants where CDR becomes the plant's main product. In the case of CDR via capturing biogenic CO_2, this will raise concerns regarding the efficient use of biomass, which need to be addressed at the policy level. Further, CDR via DAC also implies a reduction in the net outputs of heat (13%) and electricity (37%) from the CHP plant, which might lead to shortages if such a system is widely implemented by CHP plants.



a) Business as usual



b) Integrated system

Figure 15:Hourly NCF levels from sales of heat, electricity, CDR via DAC and CO_2 captured from the flue gas, for a) the reference CHP plant before PCC and DAC integration, b) reference CHP plant after PCC and DAC integration. note that the two time periods without production correspond to maintenance shutdowns. Source: **Paper III**

The optimization results for the base case indicate that a total 237 kg CO_2 per MWh of fuel combusted can be removed from the atmosphere, where DAC removes 82 kg. At a CDR credit price of 615 €/tCO_2, this corresponds to a NCF value of €145 (with DAC contributing € 54.4) to be compared with € 5 for heat and € 55.8 for electricity. This positions CDR as the most profitable product from an NCF perspective. Given full operational flexibility, the plant would therefore prioritize operation for CDR generation.

In Nordic countries where a high number of CHP plants deliver heat for district heating, grid-wide implementation of such a system can contribute a large share of the CDR demand for the country to reach its climate target. In Sweden, for example, 82 kg CO_2 per MWh of fuel combusted would result in 3.3 Mtonne of CDR via DAC if this system were implemented in all CHP plants with thermal capacities above 50MW.

Figure 16 shows the results of the sensitivity analysis for different parameters (fuel type, operational hours, electricity price volatility, electricity price, heat price – see Table 3) on the total CDR achieved by the integrated system. According to the figure, the total CDR achieved by the PCC unit only varies with the parameters that change the total biogenic CO_2 generated by the plant (i.e., operational hours and fuel type) since the model is constrained to capture 90% of all CO_2 emissions. The total CDR achieved via DAC instead responds positively to heat availability, and negatively to electricity and heat prices. This can be interpreted as the integration of DAC to CHP plants being a less favourable case for demand-following plants in comparison to base-load plants. The electricity price presents a higher impact on total CDR cost via DAC than the heat price, mainly due to the fact that the electricity prices used here are much higher than heat prices. The electricity price volatility does not appear to strongly impact the results.

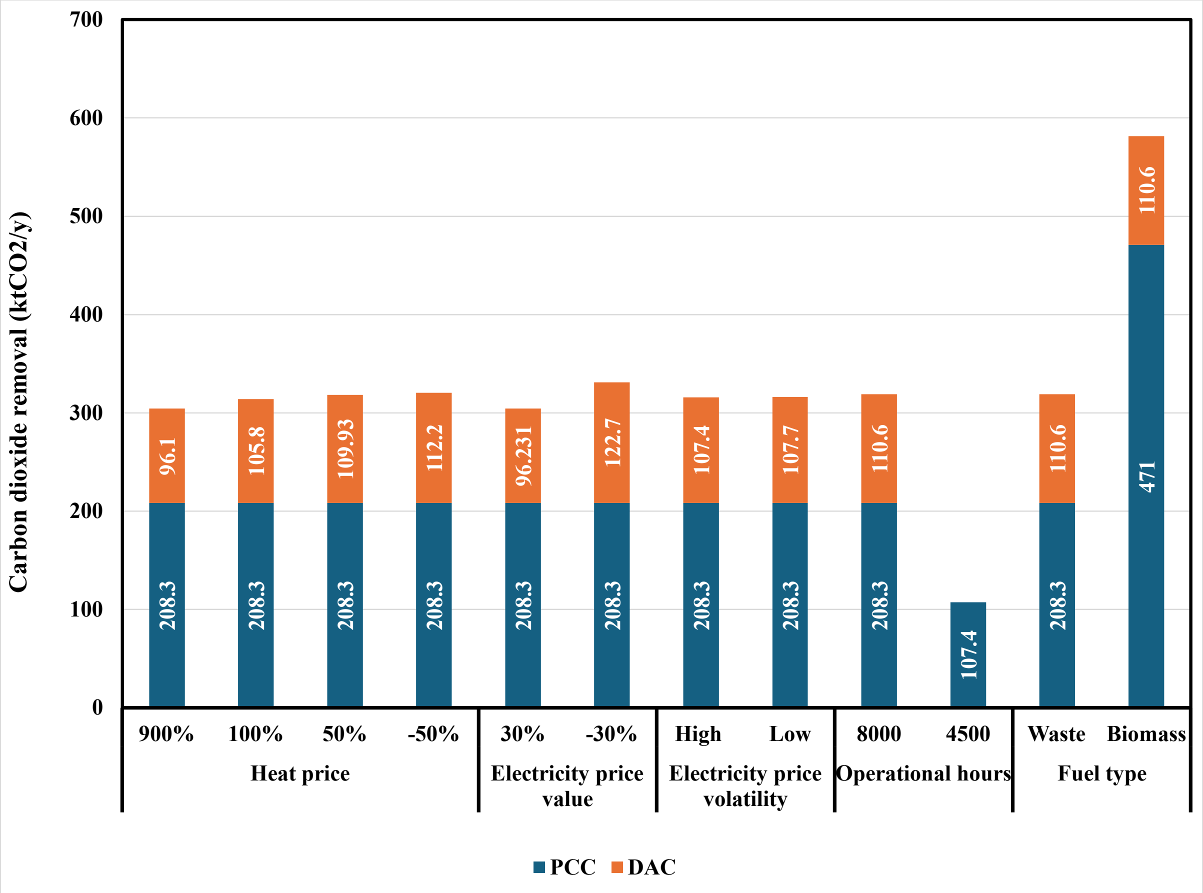


Figure 16: The sensitivity of the CDR to five key parameters for the CHP-PCC-DAC system. Source: **Paper III**

The total operational hours of the plant have a strong impact as DAC is a capital-intensive technology and thus requires a high-capacity usage to become cost competitive. Therefore, in the case of low operational hours the resulting optimal DAC capacity is equal to zero.

Finally, regarding a fuel shift (biomass and waste are considered) results do not show any strong impact on the total CDR via DAC since they both present the same amount of heat available for DAC. This depends on the use of operational parameters that are close in value.

5 Conclusions

Direct air capture (DAC) technologies are deemed necessary to reduce the impact of anthropogenic CO_2 emissions from hard-to-abate industries and to contribute to net negative emissions to compensate for a likely overshoot in emissions. The present work focuses on evaluating two alternative opportunities for DAC deployment to foster cost reduction.

i. **New cost-effective application**: Utilizing DAC technologies (originally developed for removing CO_2 from the ambient air) as point-source capture units for industries with flue gas streams containing low CO_2 concentrations.
ii. **New cost-effective location**: Integrating DAC into district heating CHP (combined heat and power) plants.

Regarding the new application as point-source capture technology, we conduct a technoeconomic assessment of three DAC technologies—MEA-ABS, ALK-ABS, and TVSA—for flue gas streams with low CO_2 concentrations (Paper II). For flue gas flow rates above 500 t/h, MEA-ABS offers the lowest cost among the three technologies investigated. The results show that for a 0.5% CO_2 concentration, the breakeven point between MEA-ABS and ALK-ABS occurs at flue gas flow rates below 500 t/h, with the breakeven flow-rate threshold decreasing at higher CO_2 concentrations. For flue gas flow rates below the breakeven flow rate threshold, ALK-ABS has the lowest cost.

The results for the levelized cost of carbon avoidance (LCOA) indicate that at CDR credit prices below \$400/tCO_2 (a value higher than the estimated price for CDR technologies such as BECCS) CO_2 emission and offset is a more economical option than point-source capture from flue gas streams with CO_2 concentrations below 2.6% and flow rates under 500t/h. The LCOA for the investigated technologies is closely tied to the carbon intensity of energy supply and the cost of CO_2 transport and storage. The carbon footprint of energy supply, particularly heat, has a significant impact on LCOA. For the case studied (representative for the HDR steel industry) the threshold value enabling competitive use of point-source capture is 0.15–0.2 t/MWh. The cost of CO_2 transport and storage impose the highest level of uncertainty on LCOA values, highlighting the need for initial deployments to improve cost estimates.

Regarding the integration of DAC with CHP plants providing district heating and equipped with CCS, results under conservative assumptions for DAC and electricity costs show that 0.82 kg of CO_2 can be removed per MWh of fuel combusted at a cost of 615€/tCO_2. Large-scale deployment could facilitate achieving 33% of Sweden's national CDR target. This confirms that using low-temperature heat for DAC is economically advantageous, even when heat has a market price. Therefore, locations where heat is currently wasted by being released into the environment offer a strong opportunity for DAC deployment.

6 Future work

The scope of the research work framing this thesis can be divided into two main areas: i) identifying opportunities for supplying low-cost carbon dioxide removal (CDR) via direct air capture (DAC), and ii) estimating the demand for CDR from various industries. In this context, integration of adsorption DAC into combined heat and power (CHP) plants connected to district heating network has been evaluated to therefore presenting vicinity of these CHP plants as an optimal location for DAC deployment. Additionally, industries with flue gas streams containing low CO_2 concentrations have been identified as potential buyers of CDR, as purchasing CDR credits may be more cost-effective for them than capturing CO_2 directly.

The next phase of this research could follow a similar approach. A geospatial analysis of the net removal cost of the DACCS chain could be conducted to identify locations that offer opportunities for low-cost CDR via DAC. This would involve evaluating different DAC technology configurations, energy sources, and geospatially resolved costs of CO_2 transport and storage. Combined with a life cycle assessment of DAC technology, this analysis would provide insights into the net removal cost of CDR via DAC.

Additionally, future work could assess the demand for CDR from carbon-intensive industries by comparing the marginal cost of capture for capture rates above 90% with various CDR price points. This will help determine what is the most economical level for residual direct emissions for these industries, emissions that require offsetting through the purchase of CDR credits.

The results of both current and future research are highly dependent on assumed CDR prices, and therefore uncertain, as the only existing market for CDR is voluntary, and its future structure remains uncertain. To address this uncertainty, we plan to conduct a survey among industries to assess their willingness to pay for CDR credits in the voluntary market. This would enable more concrete conclusions to be drawn from the research findings.

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