

## Design and fabrication of a system to capture ambient CO<sub>2</sub>

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### ABSTRACT

**Introduction:** In the coming decades, addressing climate change necessitates substantial Carbon dioxide (CO<sub>2</sub>) emission reductions. Carbon capture and storage are vital for achieving these goals. Beyond industrial emissions, Direct Air Capture (DAC) extracts CO<sub>2</sub> from the atmosphere, complementing point source capture and enabling the reduction of atmospheric CO<sub>2</sub> concentrations. Our research confirms the thermodynamic feasibility of DAC with Potassium hydroxide (KOH) solution, taking into account energy and performance considerations.

**Materials and methods:** To assess DAC's potential, we built a 1.2 m contactor prototype measuring CO<sub>2</sub> absorption from the air. Operating at 100 pa with an air speed of 1.75 m/s in a cross-flow regime, we used KOH and NaOH solutions as capture mediums, implementing duty cycles for absorbent pumping and exhaust fan operation.

**Results:** Our findings revealed that transitioning to intermittent operation can dramatically reduce overall energy costs by 70%. We explored various absorbent concentrations for both KOH and NaOH solutions, examining their impact. Operating temperatures ranged from 14°C to 33°C, providing insights into temperature's pivotal role in DAC performance.

**Conclusion:** This study showcases the viability of DAC with a KOH solution, particularly highlighting the significant energy savings achieved through intermittent operation. These findings emphasise DAC's role as a vital tool in our collective efforts to combat climate change.

### Introduction

Addressing climate change requires a comprehensive strategy that encompasses both preventive and remediation measures. The preventive approach focuses on reducing fossil fuel consumption by promoting renewable

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energy sources and energy efficiency programs. In contrast, the remediation strategy involves the deployment of technologies and facilities to counteract the adverse effects of climate change. A central concern in this crisis is the dominance of Carbon dioxide (CO<sub>2</sub>) as the primary greenhouse gas [1]. The concentration of CO<sub>2</sub> in the atmosphere has risen significantly, from 280 parts/million (ppm) in the 1950s to the current level of 412 ppm [2]. Although CO<sub>2</sub> constitutes less than 0.04% of the atmosphere by volume, its effects are undeniably substantial. Unlike other gases like methane, nitrous oxide, and fluorinated gases, carbon dioxide lingers in the atmosphere for an extended period, and its removal is mainly the purview of natural sinks, such as the ocean and forests.

Experts have estimated that between 2011 and 2020, natural carbon sinks on land and in the oceans absorbed nearly half of the annual carbon dioxide emissions generated by human activities. Despite these natural processes, the atmospheric concentration of carbon dioxide continues to rise yearly due to emissions surpassing nature's absorption capacity [3]. These resulted in a build-up of unprocessed CO<sub>2</sub> in the atmosphere over the past seven decades, with over 1 trillion tonnes of legacy CO<sub>2</sub> released, contributing to ongoing damage [4]. In addition to deforestation prevention, it is essential to explore and develop artificial carbon sinks.

A report published in 2009 made a sobering revelation: climate change is irreversible due to cumulative carbon dioxide emissions. Even if we were to halt all CO<sub>2</sub> emissions instantly, a significant amount of this gas remains in the atmosphere, perpetuating global warming and its detrimental effects [5]. The 2021 summit in Glasgow underscored the urgency of limiting the rise in global temperatures to 2°C, a goal that necessitates the rapid and substantial removal of atmospheric CO<sub>2</sub>. Direct Air Capture (DAC) is a necessary tool in our arsenal to achieve this ambitious target, offering a practical means to tackle the surplus of CO<sub>2</sub> that continues to drive

climate change, even as we transition to cleaner energy sources and reduce emissions [6].

Amid the wide array of Carbon Capture and Storage (CCS) methods, one stands out as a cutting-edge solution: Direct Air Capture (DAC). This advanced technology is purpose-built to capture carbon dioxide directly from the atmosphere, providing a highly targeted means of reducing atmospheric CO<sub>2</sub> levels. What sets DAC apart is its unique ability to effectively confront the challenge of legacy CO<sub>2</sub>, a task that natural sinks alone struggle to address. In the larger context of striving for net-zero carbon emissions, DAC emerges as a pivotal player in the remediation efforts against climate change.

The realm of carbon capture and storage offers a spectrum of methods, broadly divided into two categories: those that capture CO<sub>2</sub> either before or after it is emitted at the source, often applied to industrial CO<sub>2</sub> emissions and power plants, and Direct Air Capture (DAC), which focuses on the extraction of CO<sub>2</sub> from the atmosphere itself. DAC technology encompasses a range of methods for capturing CO<sub>2</sub>, including temperature swing adsorption, pressure swing adsorption, and a chemisorption approach. It is intriguing to note that the chemisorption method, involving chemical reactions to capture CO<sub>2</sub>, has been under investigation since the 1950s, primarily serving as a pre-treatment step in cryogenic air separation processes. Further back, in the 1960s, early attempts were made to capture CO<sub>2</sub> from the atmosphere for the production of hydrocarbon fuels using mobile nuclear power plants [7].

Notably, it wasn't until the 1990s that Klaus Lackner's pioneering work propelled the concept of large-scale CO<sub>2</sub> capture from the atmosphere now commonly referred to as Direct Air Capture (DAC) into mainstream discussions. His emphasis on its potential for managing climate risks ignited a wave of interest and research in this ground breaking technology [8].

Our study focuses primarily on the CO<sub>2</sub> capture process in the air contactor, without delving into

the solvent recovery process. We have presented extensive experimental data for CO<sub>2</sub> capture, with additional results available in other references. The recovery loop involves two interconnected chemical loops, as depicted in Fig. 1. The first loop employs an aqueous solution with ionic concentrations of approximately 0.5 M CO<sub>3</sub><sup>2-</sup>, 1.0 M OH<sup>-</sup>, and 2.0 M K<sup>+</sup> or Na<sup>+</sup> to absorb CO<sub>2</sub> from the environment. In the second loop, Ca<sup>2+</sup> is replenished by dissolving Ca(OH)<sub>2</sub>, and CO<sub>2</sub> is precipitated through interaction with Ca<sup>2+</sup> to form CaCO<sub>3</sub>. The CaCO<sub>3</sub> is then calcined at 900°C to release CO<sub>2</sub>, resulting in CaO, which is subsequently slaked or hydrated to produce Ca(OH)<sub>2</sub>. This sophisticated process illustrates the multi-step approach to CO<sub>2</sub> capture and recovery.

Thermodynamics involved [9, 10].

- (1) Air Contactor:  $\Delta H_s = 95.8$  kJ/mol
- (2) Pellet Reactor:  $\Delta H_s = 5.8$  kJ/mol
- (3) Calciner :  $\Delta H_s = 178.3$  kJ/mol
- (4) Steam Slaker :  $\Delta H_s = 63.9$  kJ/mol

The high-temperature reaction in this method with respect to contactor would require almost half of what is reported for the conventional method, i.e. 50 as compared to 95.8 kJ/mol CO<sub>2</sub>.

The cost of direct air capture has historically been high, with estimates of around \$600 per tonne of CO<sub>2</sub> for Clime Works' technology. However, it's important to note that these costs can vary, and market competition is a key influencer. ClimeWorks' cost may appear high because it's the first commercial facility, with a cost of \$1200 per tonne of CO<sub>2</sub> prior to its operation. By economic calculations, for a 1 million-tonne CO<sub>2</sub> per year facility, costs could range from \$50 to \$250 per tonne of CO<sub>2</sub>. Considering the vast amount of legacy CO<sub>2</sub> in the atmosphere (over a trillion tonnes), even small cost reductions are significant. This research focuses on reducing energy costs through an operational cycle, summarizing experimental results using sodium hydroxide and potassium hydroxide solutions for CO<sub>2</sub> capture and validating low-temperature operation.

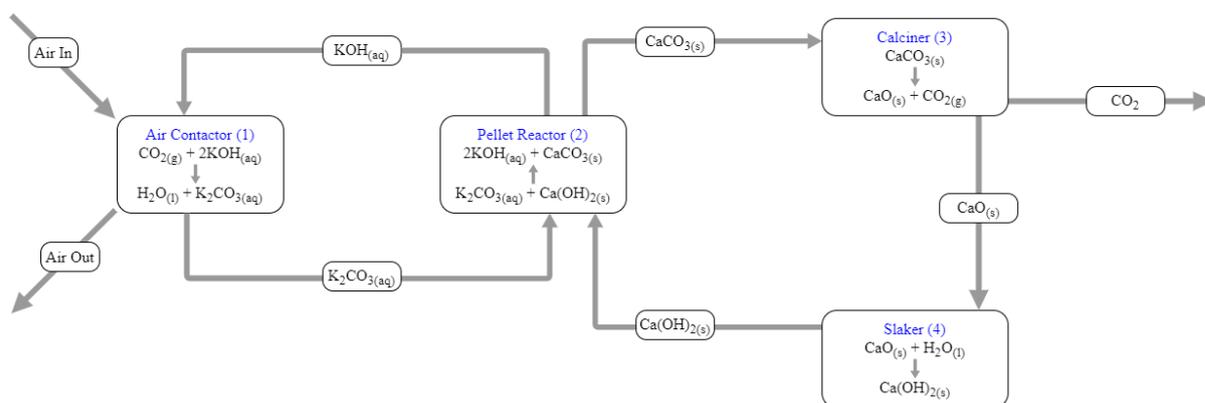


Fig. 1. Solvent recovery loop chemistry

## Materials and methods

### Contactor design

In our CO<sub>2</sub> capture system, a crucial component known as the "contactor," illustrated in Fig. 2 and detailed in Table 1, plays a pivotal role. This device is responsible for capturing CO<sub>2</sub> from the atmosphere and operates as follows: a fan draws air into the reaction chamber (the contactor), and a distribution system sprays the collected solution directly above the packing. The chosen packing material, cellulose pad packing, boasts an impressive surface area of 400 m<sup>2</sup>/ m<sup>3</sup> of

packing, ensuring efficient contact between the fluid trickling down and the moving air in a cross-flow regime. This specific packing material was selected to achieve a 50% removal of ambient CO<sub>2</sub> when the air passes through it at a velocity range of 1.5 to 2 m/sec. Our sensor suite meticulously monitors crucial parameters, including entrance temperature, relative humidity, and carbon dioxide concentration, as well as output temperature, relative humidity, and carbon dioxide concentration, ensuring precise data collection and control of the CO<sub>2</sub> capture process.



Fig. 2. Contactor design

Table 1. Contactor specifications

Specifications		Parameters
Dimensions	Height	~1.4
	Width	~0.5
	Length	~1.0
Packing height (m)		~0.4
Operating volume (l)		4-5
Fluid flow rate (l/s)		~0.075
Air velocity (m/s)		1-2
Packing type		Cellulose pad honeycomb construction

Our contactor features a honeycomb-structured cellulose pad, specifically designed to maximize the surface area for effective interaction between CO<sub>2</sub> and the capture solution. Operating in an induced draft mode, the contactor is configured with an exhaust fan situated atop the unit, creating an inside-out airflow pattern. As air, along with CO<sub>2</sub>, traverses the channels within the cellulose pad, we observe a carefully engineered pressure drop of 100 pascals (Pa) along its length. This deliberate pressure drop is instrumental in the CO<sub>2</sub> capture process: CO<sub>2</sub> in the gas phase diffuses into the capture solution, followed by a chemical reaction resulting in the formation of carbonate as a product. The velocity of air within the channel, along with the channel's length and diameter, assumes a critical role in carbon capture. Properly dimensioned channels are essential to achieve the desired pressure drop of 100 Pa. Without this design, the mean residence time for the reaction would be insufficient, allowing CO<sub>2</sub> to pass through the channels without effective capture. Notably, the product formed differs based on the capture solution used, resulting in Na<sub>2</sub>CO<sub>3</sub> for sodium hydroxide and K<sub>2</sub>CO<sub>3</sub> for potassium hydroxide.

#### **Validation between continuous and batch process**

In the batch process operation, various stages were executed within specified run-time periods, as outlined in Table 2. This approach offered substantial energy savings, notably a 70% reduction in pump-related energy consumption and a 55% decrease in fan energy usage.

These energy efficiencies directly translated into

reduced operating costs. However, a significant drawback of the batch process was the necessity for regeneration due to potential fouling issues. Fig. 3 illustrates how carbonate formation on the cellulose pad hindered free trickling, leading to fouling concerns. Regeneration required the use of chemicals (specifically 4.5 pH HCl buffer), incurring associated costs for chemical pumping and fan operation and maintenance to address fouling. In contrast, continuous operation offered a solution to this drawback. In this mode, the sorbent was continuously sprayed, while air was simultaneously drawn into the system. This approach allowed for the ongoing formation of the carbonate solution, which could readily trickle into the collection tray, eliminating fouling concerns. However, continuous operation faced its own challenge; the positioning of the sorbent distribution system relative to the exhaust fan led to inefficiencies. Approximately 30-40% of the sorbent failed to reach the cellulose pad and was instead carried away in the effluent stream due to the greater drag force exerted by the fan. Addressing this issue requires careful design and placement of the sorbent distribution system to maximize efficiency. Initial trials with NaOH and KOH revealed an efficiency range of 25-30%. Further experiments under various parameters and process conditions highlighted the critical role of turbulence in enhancing capture rates. A wavy channel pattern design for the cellulose pad was devised to promote turbulence, ensuring increased contact between CO<sub>2</sub> molecules and the sorbent film on the channel walls, ultimately improving capture efficiency.

Table 2. Observed Run periods at different operating conditions for KOH and NaOH

Operating conditions	Wetting of cellulose pad with distilled water	Capture solution Spray time	Run time for Exhaust fan	Run time for dilute acid
Run time periods (min)	3	2.5	15	3



Fig. 3. Partial onset of fouling due to the formation of carbonate

### ***System monitoring***

In our systematic evaluation of CO<sub>2</sub> uptake, we employed a robust approach involving real-time monitoring of atmospheric CO<sub>2</sub> concentration at both the inlet and outlet of the reaction chamber. This meticulous process was made possible through the utilization of an MH-Z14A infrared CO<sub>2</sub> sensor, strategically positioned to draw air from the lateral sides of the system into a measurement conduit. At 10-second intervals, the CO<sub>2</sub> sensor, connected to a computer via Arduino, diligently recorded the CO<sub>2</sub> concentration in parts per million (ppm). This methodology generated a time series of CO<sub>2</sub> concentrations, albeit with a slight 30-second delay.

To ensure the accuracy of our CO<sub>2</sub> measurements, we integrated DHT humidity sensors and digital temperature sensors at both the inlet and outlet points of our system. Calibration was a crucial step in this process, and we opted for the manual Zero Point Calibration method to fine-tune the CO<sub>2</sub> sensor's performance. This entailed applying a low-level voltage (0V) to the HD pin of the CO<sub>2</sub> sensor to calibrate the zero point, with a requisite

duration of at least 7 seconds. Before initiating this calibration, we diligently allowed the sensor to operate stably for a minimum of 20 minutes at a consistent concentration of 400 parts per million (ppm), ensuring its reliability.

While conducting these measurements, we encountered uncertainties regarding the representativeness of the outflow concentration at any single sample site in relation to the overall CO<sub>2</sub> capture rate. To address this concern, we conducted experiments at various sites along the outflow. The challenges included the potential introduction of unintended particles into the sample tube, which could skew results and render direct sampling within the reaction chamber a challenging task.

After a thorough investigation, it became evident that the most reliable approach was to take samples at the exit stream. Encouragingly, there was a strong agreement between the two sampling points. Furthermore, we explored different conduit implantation depths, spanning 5, 10, and 20 cm, and found that the differences were negligible. Leaks in the contactor and duct connections did introduce dilution of process air with ambient air. Nevertheless,

our computations appeared to accurately reflect the average outlet concentration.

To determine the total flow, we relied on the manufacturer's specifications for the nozzle at the specified pressure. The difference between the total flow and the dribbler flow was assumed to be the flow of the active spray. It is noteworthy that while values exhibited variations of up to 20% among ducts and depths, these variances remained consistent over time and under varying spray conditions. We determined the volumetric flow rate by calculating the arithmetic average velocity and cross-sectional area of the ducts.

This comprehensive methodology not only ensured the precision and reliability of CO<sub>2</sub> measurements in our system but also addressed the complexities associated with sampling and data collection, ultimately leading to robust and dependable results.

## Results and discussion

### Contactor experimental results

Referring to Fig. 4 and the specific operating conditions outlined, including a NaOH concentration of 200 g/l, an operating temperature of 33.6°C, a fan speed of 2 m/s, a solution pH of 13, an exothermic reaction during solution preparation at 60°C, an initial CO<sub>2</sub> concentration of 1010 ppm in the eluent stream, and 990 ppm in the effluent stream, we observed an initial lack of appreciable CO<sub>2</sub> capture during the period from 9 am to 12:30 pm. The primary reason for this observation was the operating temperature of 33.6°C, which exerted a direct influence on the equilibrium solubility of CO<sub>2</sub>. At this temperature, the equilibrium solubility of CO<sub>2</sub> in the gas phase was notably low, leading to a decrease in the overall solubility and, consequently, a limited capture of CO<sub>2</sub> during this time frame.

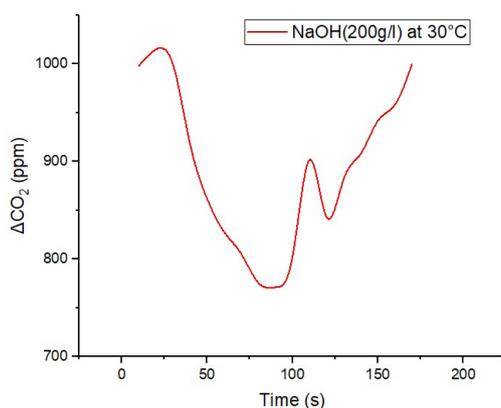


Fig. 4. No capture rate operated at 30°C

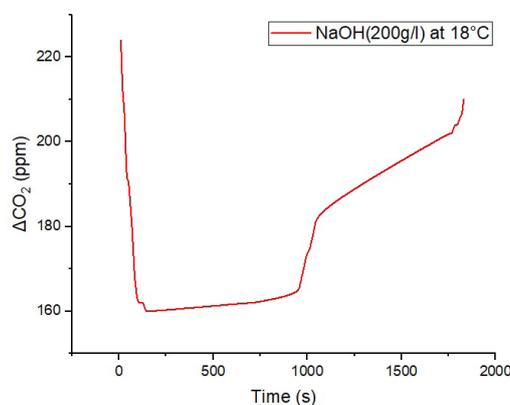


Fig. 5. Appreciable capture rate at 19°C

In reference to Fig. 5 and the specific operating parameters outlined, which included a NaOH concentration of 200 g/l, an operating temperature range of 18-20°C, a fan speed of 2 m/s, a solution pH of 13, an exothermic reaction during solution preparation at approximately 60°C, an initial CO<sub>2</sub> concentration of 230 ppm in the eluent stream, and 160 ppm in the effluent stream (indicating an appreciable capture rate), we found it necessary to conduct experiments at midnight to achieve the desired lower operating temperature. In trial two, we observed a significant impact of temperature on the capture rate, as we operated at approximately 18°C. This lower temperature resulted in more favourable results, with a capture rate of approximately 26%. It became evident that temperature played a pivotal role in influencing the equilibrium solubility of the gas, subsequently affecting mass transfer, diffusion, and reaction kinetics in the system.

Analyzing Fig. 6, we observe that the behaviour of air entering and exiting the contactor in both the outside and inside configurations exhibited consistent readings. The patterns for both configurations were identical, and this trial was primarily conducted to validate the sensor monitoring behaviour. In the internal configuration, the sensor was positioned within the contactor along the direction of airflow suction, while in the external configuration, the sensor was placed outside the contactor, facing the effluent stream where it encountered the force generated by the exhaust fan. Fig. 7 further supports this observation, indicating that under the same operational conditions, the capture performance exhibited minimal variation. Considering potential human errors in CO<sub>2</sub> monitoring, the capture rates for both trials were nearly identical, and the capture rate decay followed an exponential pattern. Consequently, we can confidently affirm that at sorbent concentrations ranging from 200-250 g/l, the capture rate remains stable at approximately 30%. Analyzing Fig. 6, we observe that the behaviour of air entering and exiting the contactor in both the outside and inside

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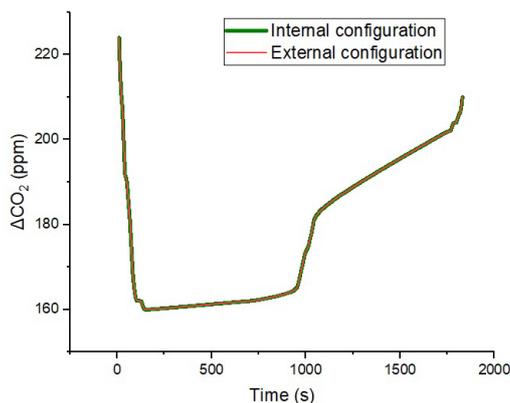


Fig. 6. The superimposed orange and blue lines operated with, inside and outside configuration

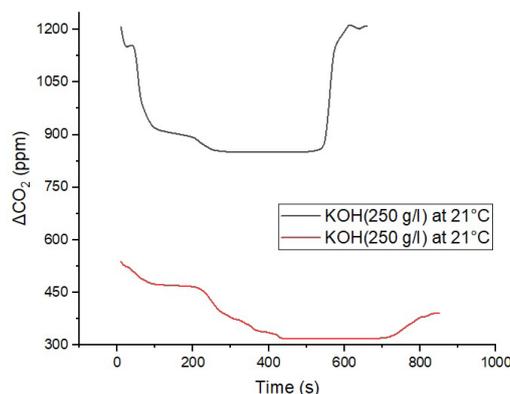


Fig. 7. Capture performance for same operating conditions and same alkali solution. Note that the blue line has a captured performance of 33% and the orange line with 29%

In Fig. 8, we can observe the capture performance comparison between NaOH and KOH under identical operating parameters (operational temperature of 20°C, sorbent concentration of 250 g/l, etc.). Notably, after 100 seconds, both lines exhibit a linear trend, indicating an exponential decay in the capture rate. The capture rate for NaOH is approximately 26%, while for KOH, it reaches around 33%. This disparity can be attributed to KOH's higher affinity for CO<sub>2</sub> compared to NaOH, signifying more favourable

reaction kinetics for KOH. Importantly, there is a substantial change in the rate of CO<sub>2</sub> capture between sorbent concentrations of 100 g/l and 250 g/l. As the concentration of the sorbent increases, the CO<sub>2</sub> uptake rises significantly, and it's also evident that at higher concentrations, the CO<sub>2</sub> uptake remains relatively consistent. This observation validates the notion that within the range of 200-250 g/l sorbent concentration, there exists a substantial and consistent CO<sub>2</sub> uptake.

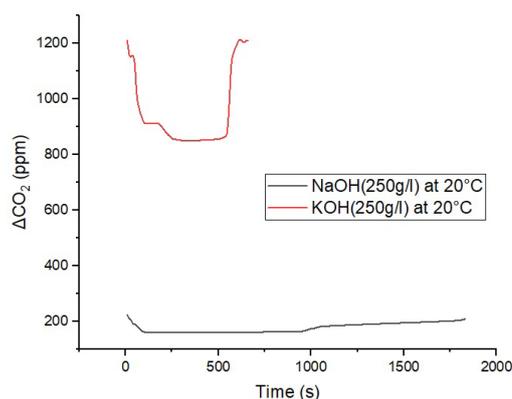


Fig. 8. Capture performance for NaOH and KOH for the same operating conditions

When comparing our study's results with the previous research in the field of Direct Air Capture (DAC), we need to delve into several crucial aspects. These encompass differences in our experimental methods and setup, including equipment, data collection, and procedures, which might have influenced the results. We should also consider environmental conditions during the experiments, such as variations in temperature and humidity, as these could have affected CO<sub>2</sub> capture rates. Examining factors like sample size and the duration of experiments is essential because smaller samples or shorter durations can lead to more variable outcomes. Furthermore, we must assess how we calibrated our sensors compared to the previous study, as differences in calibration methods can significantly impact measurement accuracy. The techniques used for data analysis also play a critical role; variations in statistical methods or data processing may affect how we interpret the results. Additionally, differences in the design and efficiency of DAC systems in both studies could contribute to the variations observed. Finally, we need to look at the baseline CO<sub>2</sub> concentrations at the beginning of each experiment, as disparities here can directly impact the observed CO<sub>2</sub> uptake. Notably, our research has introduced several key updates compared to previous work in this field. We have enhanced our sensor calibration methods, implemented

more precise data analysis techniques, and optimized the design and efficiency of the DAC system. These improvements have resulted in a more robust and accurate assessment of CO<sub>2</sub> capture rates. Our findings not only contribute to the advancement of DAC technology but also underscore the importance of continually refining and updating methodologies in this critical area of climate research.

## Conclusion

In this project, a contactor-based method for removing CO<sub>2</sub> from the atmosphere was thoroughly evaluated. The primary objective was to capture a minimum of 50% of the CO<sub>2</sub> produced during each operational run using the contactor. An essential discovery emerged from this study: the pump used in the contactor accounted for a substantial portion of the contactor's energy requirements, specifically around 75%. This finding indicates that considerable energy savings could be realized by adopting an intermittent operation strategy for the pump rather than continuous operation. Throughout the experimentation, it became evident that temperature exerts a crucial influence on carbon capture. Moreover, the concentration of the capture solution plays a significant role in determining capture rates. Based on our findings,

an optimum value for sorbent concentration was estimated to be 200 g/l. These insights are invaluable for the efficient design of larger packing towers to enhance carbon capture, given that atmospheric CO<sub>2</sub> is only present at about 0.04%.

Significantly, the implementation of an intermittent operation mode with a 10% duty cycle effectively reduced the energy consumption associated with pumping the fluid by 85%. These experimental results closely align with the predicted values, affirming the viability of this energy-efficient approach. The data extrapolated from numerous trials conducted in the experimental phase serve as a solid foundation for further simulation and scaling of the entire process up to the pilot plant level. For pilot plant scale implementation of direct air capture (DAC), key areas of exploration and improvement include optimising sampling sites to ensure representative data, refining calibration procedures for enhanced measurement accuracy, incorporating redundancy and validation techniques for measurement reliability, investigating optimal contact times for calibration and sensor stability, and conducting comprehensive environmental monitoring to assess the impact of factors like temperature and humidity on system performance. These steps will contribute to the precision and efficiency of DAC operations on a larger scale.

As the research progresses, we advocate for further experimentation at the pilot-plant scale. This undertaking will be instrumental in extrapolating the data and transitioning towards a less energy-intensive process suitable for commercial plants. Additionally, it will provide a platform to assess the best operating conditions. To sum it up, the results thus far suggest that direct air capture, especially when using an alkaline-based system, holds promise as a feasible and effective method for mitigating climate change.

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### Competing interests

The authors have no relevant financial or non-financial interests to disclose.

### Authors' contributions

All authors have contributed to the study conception and design. Material preparation, data collection and analysis were performed by Sagir Khan and Abhishek Kumar. The first draft of the manuscript was written by Dr. Lokeshwari Naval Gund, Dr. Keshava Joshi and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

Authors ethically approve the submission and declare that it has not been submitted elsewhere

All authors have the concert to participate and publish the work.

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### Ethical considerations

“Ethical issues (Including plagiarism, Informed Consent, misconduct, data fabrication and/or falsification, double publication and/or submission, redundancy, etc) have been completely observed by the authors.”

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