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A multifunctional rooftop unit for direct air capture†‡

Keju An,§[a] Jamieson Brechtl,[a] Stephen Kowalski,[a] Cheng-Min Yang,[a] Michelle K. Kidder,[b] Costas Tsouris,[b] Christopher Janke,[c] Meghan Lamm,[b] Katie Copenhaver,[b] Josh Thompson,¶[b] Tugba Turnaoglu,[a] Brian Fricke,[a] Kai Li,*[a] Xin Sun[d] and Kashif Nawaz*[a]

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Direct air capture (DAC), which captures CO_2 from ambient air, is a critical technology to reduce greenhouse gases in the atmosphere in order to avoid climate disasters. Due to the relatively low concentration of CO_2 (400 ppm), a large amount of air needs to be moved through DAC devices, which requires lots of energy. Currently, DAC technologies are deployed mainly in centralized systems and require extensive infrastructure and initial capital cost. A potential solution is to utilize existing infrastructure for DAC. In this study, we propose a distributed DAC system that utilizes existing commercial rooftop heating and air conditioning (HVAC) units to capture CO_2 from the air. There are approximately 15 million such units already installed on commercial buildings in the United States, and they move a large amount of air every day. Adding DAC functionality to these units will significantly reduce the cost of infrastructure and operation. A modular approach was used to introduce DAC into a rooftop unit. Modules filled with triethylenetetramine-functionalized polyacrylonitrile sheets were developed and installed on the condenser coil side of the rooftop unit. The rooftop unit with DAC functions effectively captured CO_2 from the air, and the addition of the DAC modules had little effect on the unit's original functionality. A preliminary techno-economic analysis was also conducted, and the results potentially suggest that utilizing existing commercial rooftop units for carbon capture is a feasible approach to reducing greenhouse gases.

Environmental significance

Direct air capture (DAC) is a promising and widely accepted technology that can address climate challenges by directly capturing CO_2 from the atmosphere through chemical or physical absorption processes. Currently, DAC technologies are deployed mainly in centralized systems and require extensive infrastructure and initial capital cost. One strategy to reduce the infrastructure cost of DAC systems is to use existing infrastructure. In the DAC process, a large quantity of air is moved through the contactor (sorbent system) to be captured by the sorbents, which need high energy input. The cost and energy input for DAC systems could be reduced by combining the DAC function with other existing infrastructure/equipment. Rooftop units (RTUs) move ambient air across a heat exchanger to transfer heat between the refrigerant and the ambient air. The addition of DAC modules to the RTU would make use of the already existing equipment and multi-functionalize the device. Utilizing existing commercial rooftop units for carbon capture is a feasible approach to reducing greenhouse gases.

1. Introduction

Since the industrial revolution, gigatons of CO_2 have been emitted into the atmosphere.[1] This greenhouse gas is released primarily through the burning of fossil fuels and other industrial activities. If the release of CO_2 is not carefully abated, the global mean temperature is projected to increase 2–4 °C over the last second half of this century.[2] Several previous reports[3] have suggested that such changes to the climate can cause sea level rise induced by glacier recession,[4,5] increased sedimentation in river basins,[6] changes in vegetation patterns and agricultural yield,[7–9] reduced food security,[10] and other deleterious outcomes. Thus, the reduction and mitigation of CO_2 in the atmosphere is one major challenge for humanity as a whole.

One method to reduce the amount of atmospheric CO_2 is *via* direct air capture (DAC) technologies, which use sorption processes to extract CO_2 directly from the atmosphere. Because of

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[a] *Buildings and Transportation Science Division, Oak Ridge National Laboratory, Oak Ridge, TN 37830, USA. E-mail: lik1@ornl.gov; nawazk@ornl.gov*

[b] *Manufacturing Science Division, Oak Ridge National Laboratory, Oak Ridge, TN, 37830, USA*

[c] *Chemical Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37830, USA*

[d] *Energy Science & Technology Directorate, Oak Ridge National Laboratory, Oak Ridge, TN 37830, USA*

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§ National Renewable Energy Laboratory, Golden, CO 80401, USA.

¶ Chevron Richmond Technical Center, Richmond, VA 23230, USA.

the relatively low concentration of CO_2 in the air, chemical absorption dominates the DAC development, even though some physical adsorbents such as metal–organic frameworks (MOFs) can be used. Both solid and liquid sorbents have been developed and show promising results in various DAC systems. Examples of solid and liquid sorbents include amine-functionalized silica and aqueous NaOH, respectively.[11] However, the adsorption mechanisms of these two types of sorbents differ significantly.[12] Carbon dioxide is adsorbed on the surfaces of solid sorbents and can subsequently be released when the material is subjected to relatively low temperatures of 80 to 120 °C. As for liquid sorbents, CO_2 typically binds with a liquid flowing through a packed bed-air contactor; the CO_2 can then be released using regeneration methods that involve heating the solution.[13,14] One disadvantage of liquid sorbents is that regeneration usually requires heating temperatures significantly higher (approximately 900 °C[14]) than those required by solid sorbents. Because they consume less energy than liquid processes during regeneration, solid DAC processes have been extensively adopted by several companies, such as Climeworks and Global Thermostat.

Table 1 provides selected CO_2 capture data of reported adsorbent materials under different experimental conditions.[11] The amines had the highest CO_2 uptakes (1.09–6.85 mmol g^{-1}), whereas the zeolites had the lowest uptakes (0.15–0.87 mmol g^{-1}). The Mg-MOF-74 sorbent had the lowest CO_2 uptake of the listed materials. Lastly—unlike the MOFs, zeolites, and alkali carbonates—the amines exhibited higher CO_2 uptake values at levels typical of ambient air environments.

Most current state-of-the-art DAC technology is deployed in centralized systems, where large carbon capture apparatus can directly inject captured carbon dioxide into geologic formations. Though they have the advantage of being co-located with the eventual storage site, they require extensive infrastructure to be built, which could lead to various associated challenges.[25] For example, the overall energy demand may limit the system's scalability if it is too great relative to the regional or global power supply.[26] Additionally, because stand-alone DAC systems are in the early stages of development, an exhaustive techno-economic assessment of these technologies has yet to be performed.[27] The sorbents used in DAC systems also present a challenge because they must demonstrate that they can efficiently react with air mixtures containing low concentrations of CO_2.[25] An issue for the current DAC systems is the relatively high cost of CO_2 storage, which can be in excess of $600/ton with current carbon capture technologies.[28] New technologies are needed to reduce this cost to below $100/ton,[29] which would help fulfill a goal of the Carbon Negative Shot (one of the US Department of Energy's (DOE's) Energy Earthshot Initiatives) and make DAC a potentially viable technology.[30–32]

The requirement of equipment and electricity of these synthetic DAC technologies puts them at a major disadvantage as compared to natural DAC systems such as trees. In the former, electricity is needed not only to capture CO_2 but also to compress it for transfer in the pipeline. One must then ask the question as to whether renewable electricity used for synthetic DAC would be more effective in reducing atmospheric CO_2 than using the same quantity of renewable electricity to replace a fossil source of CO_2 such as that from coal or natural gas power plants. A study by Jacobson[33] suggested that this would not be the case. For example, they found that using wind to run a DAC system reduced less CO_2 than using it to replace a coal electricity generating plant. Furthermore, replacing a coal power plant with renewable energy sources reduces air pollution, coal mining, and coal infrastructure as well, whereas using a synthetic DAC system does not. As a result, the social cost benefit of using wind to replace coal was 5–10 times that of using it to run DAC in that study. In summary, the use of wind and solar in place of coal for electricity production is more effective in reducing atmospheric CO_2 than using synthetic DAC technologies that are powered by renewable energy sources.

One strategy to reduce the infrastructure cost of DAC systems is to use existing infrastructure. In the DAC process, a large quantity of air is moved through the contactor (sorbent system) to be captured by the sorbents, which need high energy input. The cost and energy input for DAC systems could be reduced by combining the DAC function with other existing infrastructure/equipment. Rooftop units (RTUs) move ambient air across a heat exchanger to transfer heat between the refrigerant and the ambient air. The addition of DAC modules to the RTU would make use of the already existing pressure drop in the RTU and multi-functionalize the device. Fig. 1 illustrates a possible RTU configuration with DAC integrated, wherein the DAC modules are upstream of the outdoor

Table 1 Carbon dioxide capture data of some sorbent materials reported in the literature (adapted from Wang *et al.*[11])

Type	Sorbents	CO_2 uptake (mmol g^{-1})	Absorption conditions	Ref.
Amine-functionalized	Pentaethylenehexamine	6.85	25 °C, 400 ppm	15
	Monoethanolamine	1.92	25 °C, 440 ppm	16
	2-Amino-2-methyl-1-propanol	1.09	25 °C, 440 ppm	16
Metal–organic framework (MOF)	Mg-MOF-74	0.05	25 °C, 500 ppm	17
	ZU-16-Co	1.05	25 °C, 400 ppm	18
	mmen-Mg_2 (dobpdc)	2.05	25 °C, 390 ppm	19
Zeolite	Low-silica type X (LSX)	0.87	25 °C, 395 ppm	20
	Zeolite 13X	0.40	25 °C, 500 ppm	17
	Tetraethylenepentamine-ZSM-5	0.15	25 °C, 5000 ppm	21
Alkali carbonate	$K_2CO_3/\gamma\text{-}Al_2O_3$	1.15	Ambient air	22
	K_2CO_3/ZrO_2	1.04	25 °C, 450 ppm	23
	K_2CO_3/Y_2O_3	0.64	Ambient air	24

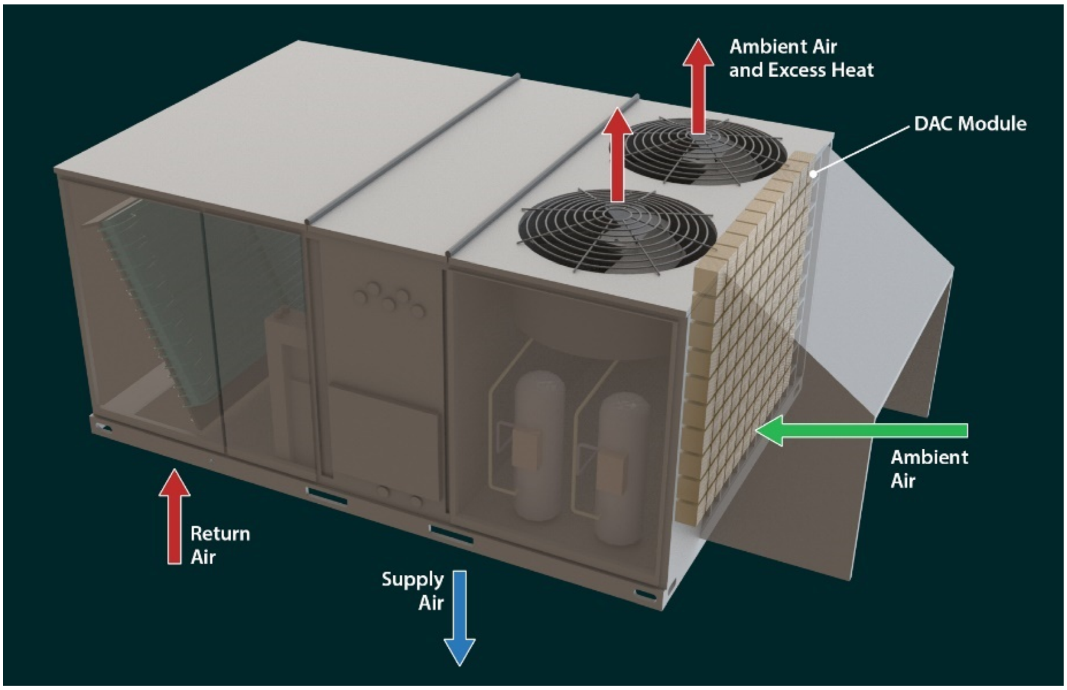


Fig. 1 Schematic of a typical unitary roof top unit with the DAC module.

coil. Currently, RTUs are the dominant method of heating and cooling for light commercial applications. The major advantages of RTUs include no working parts in the living space, lower construction costs, and efficient design modification.[34] An estimated 15 million RTUs are currently on commercial buildings in the United States.[35] Because RTUs already move ambient air as part of their primary function, the additional energy necessary to overcome the added pressure drop required by DAC may be less than the energy required for current dedicated DAC technologies such as the Climeworks Capricorn system,[36] though methods to transport the captured CO_2 to storage sites would require further energy usage for compression and pumping. Recently, Climeworks employed DAC technology on a system with overall dimensions similar to an RTU that was also mounted on a rooftop[37] and was reportedly able to adsorb the carbon equivalent of 36 000 trees.[38] The CO_2 collected can be stored as carbonate minerals by exposing it to alkaline-rich minerals or recycled into chemicals and fuels including, methanol, dimethyl ether, and hydrocarbons.[26,39,40]

Currently, very few studies on distributed DAC technology exist. Recently, Sadiq *et al.* reported a mobile DAC system that employed a MOF-based nanocomposite[41] and was incorporated within a mobile unit for distributed deployment. Their results showed an operational cost of \$35–\$350 ton-CO_2^{-1} depending on the source of regeneration energy. Li *et al.*[42] reviewed mobile DAC as a route to scaling up DAC through distributed deployment on vehicles, ships, low- and high-altitude aircraft, and others. So far, however, there have been no studies that comprehensively examine both the costs of and the effects of an outdoor environment on a distributed RTU DAC system. Therefore, the purpose of this study is to provide a novel investigation on how weather, airflow, and fan power affect the CO_2 capture performance and cost of a distributed DAC RTU system to assess the feasibility of this technology. It is understood that the use of DAC with an RTU will inevitably increase the power usage of the RTU. With much of today's electricity supplied by sources that increase atmospheric CO_2, the increased power usage of the RTU could result in a net increase in atmospheric CO_2 rather than a decrease as was observed by Jacobson.[33] However, in the future when fossil fuel combustion is no longer used to produce power, but further atmospheric CO_2 removal is still necessary, many methods of DAC may be deployed to reduce atmospheric CO_2 levels.[30]

2. Methodology

2.1 Preparation of the RTU for CO_2 capture

A unitary rooftop heat pump or air conditioner, either of which may be called an RTU, contains a mechanical refrigeration system and an indoor and outdoor fan along with their associated controls. The RTU is usually installed on the roof or beside a building to provide heating or cooling for occupant comfort. Fig. 1 shows the schematic of a typical RTU installed on the rooftop. The conditioned air moves through the roof and enters the bottom of the unit as return air. After being heated or cooled and dehumidified, it returns to the conditioned space as supply air, again through the bottom of the unit and the roof. The supply air is distributed through ductwork to different areas of the conditioned space. The mechanical refrigeration system requires that heat transferred from the conditioned space to the refrigerant during cooling mode be rejected to the ambient. This heat transfer takes place in the outdoor coil, through which ambient air is moved by the outdoor fan (or fans). Similarly, in heating mode, the heat that is added to the conditioned space by the refrigeration system is extracted from the ambient through the outdoor coil, through which ambient air is moved by the outdoor fan(s). Generally speaking, commercial RTUs move air using two major subsystems, namely the indoor and outdoor fans, during normal operation.

A significant amount of air moves through RTUs. Since ambient air with a low concentration of CO_2 (approximately 415 ppm) is already moving through the outdoor coil and fan subsystem; this work focuses on extracting CO_2 from the ambient air using this readily available source. A significant amount of air moves through RTUs. For instance, the ASHRAE HVAC Systems and Equipment handbook notes that airflow can be 80–160 L s^{-1} (170–340 cfm) for a heat transfer of 4.2 kW (14 330 Btu h^{-1}) and a 17 K temperature difference.[43] However, airflow values are higher in the available literature for commercial RTUs. For example, Trane Technologies and Johnson Controls provide data on commercial packaged RTUs. Data for the Trane Precedent[44] and Voyager 2 (ref. 45) product lines show an average airflow of 834 cfm/ton. Data for the Johnson Controls Series 5,[46] ZX/ZY/ZQ/ZL,[47] and Series 20[48] product lines show an average condenser airflow of 941 cfm/ton. Both airflow rates are significantly higher than those listed in the ASHRAE handbook.[43]

2.2 Adsorbent synthesis

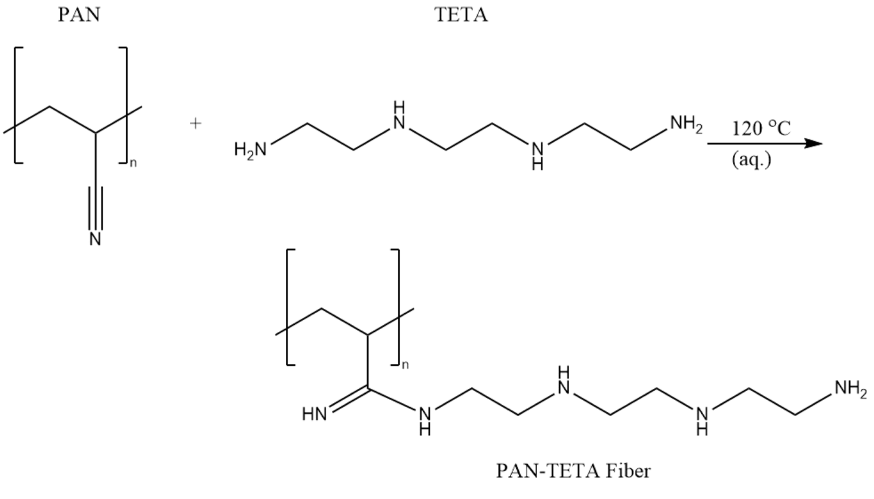
In this study, triethylenetetramine-functionalized polyacrylonitrile (PAN-TETA) was used as the adsorbent to capture CO_2 from the air. PAN-TETA was prepared by reacting polyacrylonitrile (PAN) with triethylenetetramine (TETA) (Scheme 1). The reaction was conducted in 19 L of solution containing PAN and TETA using water as the reaction medium. The amine groups on PAN-TETA can chemically bond with CO_2 and achieve a CO_2 absorption capacity of 0.33 mmol g^{-1} in 400 ppm CO_2 (in N_2). PAN-TETA also adsorbs 55–60% of its weight in moisture at 25–40 °C.

2.3 Execution of rooftop experiments

For this investigation, modules were designed to orient PAN-TETA optimally for using the airflow through the outdoor coil of a typical RTU. The material was fabricated as sheets with thicknesses of approximately 3 mm. After the sheets were dried, they were cut into rectangles with approximate dimensions of 50 mm (2 in., width) × 100 mm (4 in., length). Groups of these rectangles were stacked into extruded 6063 Al tubes with square cross sections. The extruded Al tube sections, which had dimensions of 110 mm (4.5 in.) × 110 mm, were cut to a depth of approximately 100 mm (4 in.). A UV-resistant polymer mesh with an opening size of 1.59 mm (0.0625 in.) was glued onto the aluminum square containers. Twenty-five rectangles of PAN-TETA were installed into each extruded Al tube section such that the 3 mm × 100 mm (0.25 in. × 4 in.) edge of each rectangle was facing the incoming airflow. This was intended to allow the longest residence time of airflow against PAN-TETA to optimize the absorption process. A photograph of one filled tube module and its corresponding schematic are shown in Fig. 2a and b. Ninety of these modules were assembled, and each module was numbered so its performance could be monitored throughout the experiments. The RTU that was used for testing is shown in Fig. 2c. This unit has been used on a prototype commercial building and has been fully instrumented. The prototype commercial building is located at the Oak Ridge National Laboratory and is referred to as the Flexible Research Platform 2 (FRP 2). This building was documented by Im *et al.*[49]

Prior to any experiments with PAN-TETA, the face velocity on the outdoor coils was characterized using a hot-wire anemometer (Alnor® Velometer® Thermal Anemometer AVM440) to investigate the effect of face velocity on the absorption rate of PAN-TETA. The face velocity averaged approximately 1 m s^{-1}. Further information about the air face velocities and the measured data are given in the appendix.‡

The modules were installed on a rack system with the end covered by the mesh material directly against the fins of the outdoor coil on the two exposed faces, north and west. Fig. 2d and e show the modules assembled on the north face and its corresponding schematic, respectively. A map of the modules was maintained such that the same module was placed in the same location for each test. Another layer of the same mesh material was installed on the outside of the modules to limit the amount of foreign material that PAN-TETA would be exposed to. Furthermore, a sun and rain shield was installed over the DAC



Scheme 1 Synthesis of the PAN-TETA.

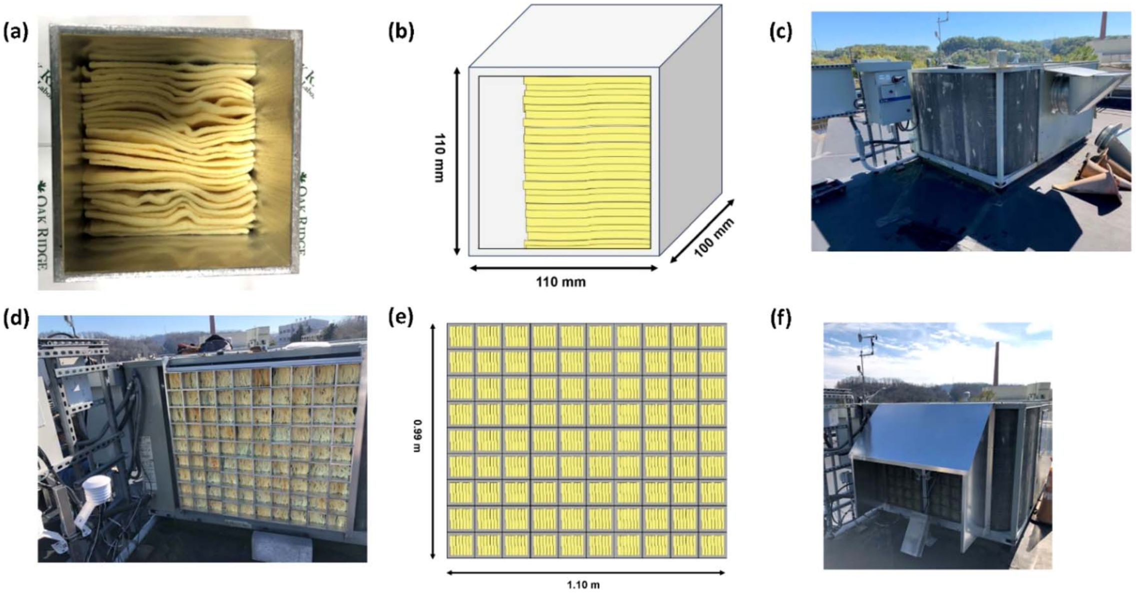


Fig. 2 (a) Photograph of a PAN-TETA module, (b) corresponding schematic of the PAN-TETA module, (c) RTU already in service, (d) photograph of the modules installed on the north face of the RTU outdoor coil, (e) corresponding schematic of the installed module system, and (f) rain and sun shield and upstream instrument assembly installed on the north face of the RTU outdoor coil.

modules, as shown in Fig. 2f. Instruments to measure CO_2 concentration (Vaisala GMP252), humidity (Vaissala HMP7), and temperature (Vaisala TMP1) were installed both upstream and downstream of the filter modules, and a differential pressure transducer (Omega PX277-05D5V) was installed to measure the pressure drop across the modules and outdoor coil. The upstream instruments were installed in a weatherproof housing. The downstream instruments were installed in a similar housing inside the cabinet between the outdoor coil and the outdoor coil fans. The instruments were added to the existing FRP 2 data acquisition system that was already used to collect data from the RTU. The intention of installing the upstream and downstream CO_2 concentration measurement instruments was to determine if the change in CO_2 concentration through the modules would be detectable. The CO_2 was not detectable using these instruments (Fig. S1‡); however, later analysis with thermogravimetric analysis-mass spectrometry (TGA-MS) showed the amount of CO_2 collected (Fig. S2‡). A further discussion of the upstream and downstream CO_2 measurements is provided in the appendix.‡

2.4 Sampling and data acquisition

The process used to conduct the experiments with the modules was as follows. The modules were first weighed using an electronic scale (Sartorius MCA20201S). They were then fully dried in an oven at 55 °C until a constant final weight of the material was achieved. The modules were then weighed again before being installed in the RTU. The RTU was typically operated based on the cooling needs of the building only during daytime hours but was occasionally operated based on the 24 h cooling needs of the building. The experiments were conducted from February to October in 2022 to study the adsorption capacity under varying ambient temperature and humidity conditions. The absorbent modules were installed in the RTU and operated for 7 days at a time each month because this was hypothesized to be a suitable amount of time for PAN-TETA to absorb the maximum amount of CO_2. The measured ambient air conditions were collected over the operating time of the RTU. At the end of the 7 days, the modules were again weighed to determine the increase in mass from the week's exposure to ambient air. After weighing, small samples were extracted from specific modules to be further analyzed using TGA-MS to identify the amount of CO_2 adsorbed. TGA measures weight change (loss or gain) and the rate of weight change of a material as a function of temperature or time under a controlled atmosphere. It has been widely used to evaluate the CO_2 absorption and desorption behavior of a sorbent.[50,51] The TGA and evolved gas analyses were performed using a TGA 5500 (TA Instruments) coupled with a quadrupole mass spectrometer with a Faraday detector (Cirrus 3, MKS Instruments). Samples between 3 and 7 mg were loaded into Pt pans and allowed to equilibrate at 30 °C. The samples were then heated to 250 °C at 10 °C min^{-1} under an Ar flow while mass spectrometry (MS) spectra were collected every 3.03 s in an *m*/*z* range of 1 to 50. The TGA and MS data were analyzed using Trios software (TA Instruments). Specifically, samples were extracted for TGA from modules installed in areas with higher face velocities as shown in Table 2 to check if the amount of CO_2 absorbed was higher because of the higher face velocity, as would be expected.

3. Results and discussion

3.1 RTU performance

In this section, the field test results are presented to demonstrate the implementation of PAN-TETA absorbent modules in an RTU to understand the potential effects of DAC in an existing

Table 2 RTU parametric data set for February and June 2022

	February 2022			June 2022		
	Mean	Max	Min	Mean	Max	Min
CO_2 upstream with RTU module (ppm)	417.27	443.42	398.11	416.29	498.12	363.02
CO_2 downstream with RTU module (ppm)	425.15	451.10	406.91	415.46	495.85	359.13
CO_2 upstream without RTU module (ppm)	416.76	495.23	397.62	400.39	455.95	367.85
CO_2 downstream without RTU module (ppm)	420.07	495.07	400.71	402.50	451.29	365.56
Outdoor temperature with RTU module (°C)	3.15	12.16	−4.14	26.89	35.93	22.23
Indoor temperature with RTU module (°C)	19.31	20.13	18.56	21.73	23.09	20.44
Outdoor temperature without RTU module (°C)	2.88	12.98	−4.51	26.60	35.67	18.12
Indoor temperature without RTU module (°C)	20.05	20.93	19.08	21.62	23.84	20.04
Outdoor relative humidity with RTU module (%)	64.10	94.42	31.86	71.67	91.99	44.61
Indoor relative humidity with RTU module (%)	27.80	30.81	25.08	50.49	57.37	43.85
Outdoor relative humidity without RTU module (%)	55.93	91.30	18.22	59.90	83.49	34.78
Indoor relative humidity without RTU module (%)	22.98	25.60	20.34	52.53	57.63	48.49

system. To ensure that the results were reported consistently, they were averaged over 5 min periods. The typical indoor and outdoor conditions (*i.e.*, temperature and relative humidity profiles) in February and June are shown in Fig. 3 and Table 2. February and June were selected to represent cold and dry conditions and hot and humid conditions, respectively. In both months, relatively constant temperatures and relative humidity levels were maintained inside the building, although the

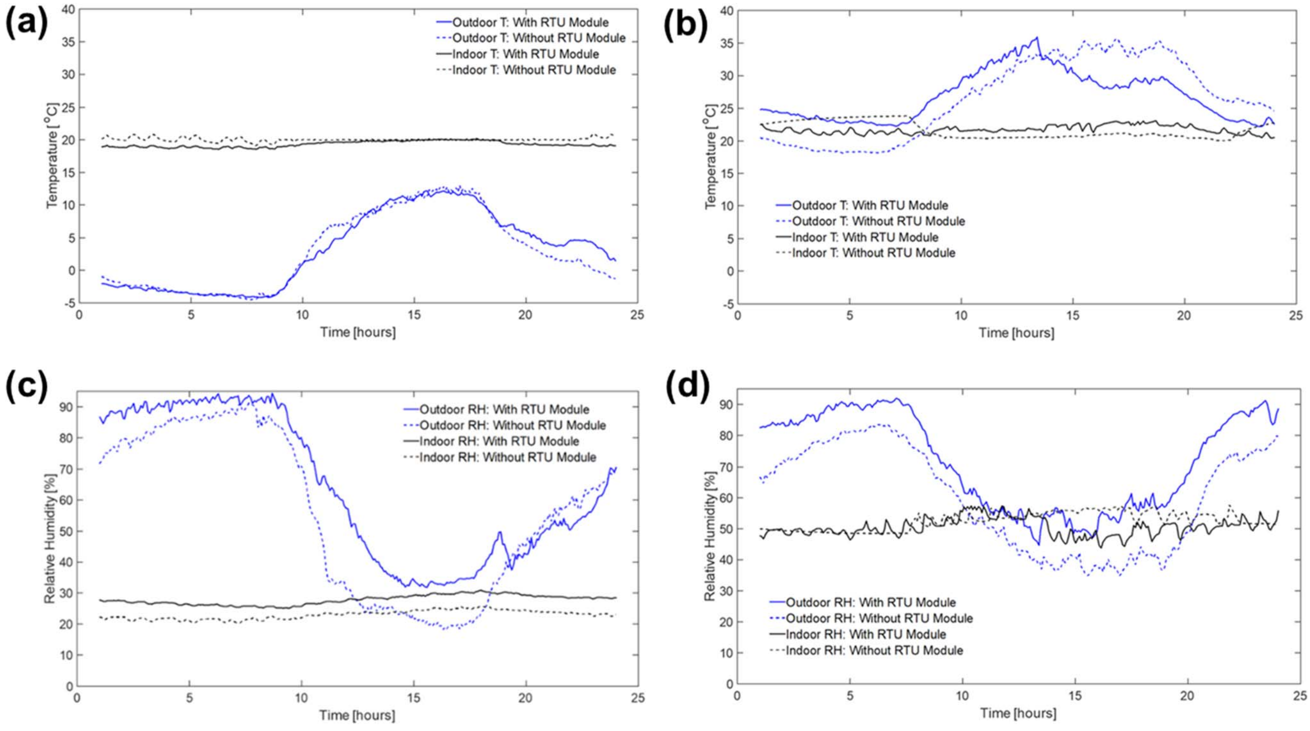


Fig. 3 Typical outdoor and indoor temperature (T) and relative humidity (RH) profiles with and without the RTU modules during February (a and c) and June (b and d) 2022.

outside conditions changed greatly during the 24 hour period, as shown in Fig. 3. On average, the indoor conditions were controlled at 20.1 °C and 22.9% relative humidity during February with the RTU modules. In June, indoor conditions were controlled at 21.7 °C and 50.5% relative humidity. The PAN-TETA amine-based adsorbent, which generally exhibits slow kinetics and therefore is unlikely to approach equilibrium CO_2 capacity in a short adsorption time, was used for the RTU application.[52] The rate of CO_2 loading can be highly dependent on ambient temperature, CO_2 partial pressure, and the amine content, shape, and pore size of the sorbent.[53] The slow kinetics could be caused by CO_2 taking longer to enter the amine sites that are more difficult to reach within the adsorbent pores, which become more difficult to access as more CO_2 is adsorbed.[54] In addition, the formation of bicarbonate allows for a high equilibrium capacity, but the kinetics are still very slow, especially at lower temperatures.[55] Fig. 4 shows the upstream and downstream CO_2 concentrations for February and June, respectively. Under the cold and dry February conditions, the difference between the CO_2 upstream and downstream concentrations was 7.8 ppm on average and 14.2 ppm maximum with the RTU modules (Fig. 4a), whereas the difference was 3.3 ppm on average and 19.8 ppm maximum without the RTU modules, as shown in Fig. 4b. During June, the difference between the CO_2 upstream and downstream concentrations was 0.82 ppm on average and 14.7 ppm maximum with the RTU modules (Fig. 4c), whereas the difference was −2.1 ppm on average and 9.6 ppm maximum without the RTU modules (Fig. 4d). For February and June, the difference between the upstream and downstream CO_2 concentrations was less than 20 ppm mainly because of the slow kinetics of the amine-functionalized sorbent and high face velocity (0.34 to 1.35 m s^{-1}, Table S1‡). The mean and maximum values for the indoor and outdoor conditions and upstream and downstream CO_2 concentration data are presented in Table 2. Importantly, the CO_2 concentrations reported in Fig. 4 and Table 2 are the averages of the direct measurements from the CO_2 sensors. The total amount of CO_2 captured by the absorbent modules was quantified through TGA-MS, and the monthly CO_2 loadings for the 7 day duration are discussed in detail in the next section.

3.2 CO_2 and moisture loading

The RTU DAC performance was comprehensively analyzed for different climatic conditions using high-resolution daily weather data. The CO_2 and H_2O loadings were collected from the sampled PAN-TETA adsorbents. Between five and eight samples were collected from the randomly dispersed positions in the RTU module for each month. For each sample, the cyclic period was 7 days, and the samples were analyzed using TGA. Fig. 5a shows the monthly H_2O loadings for different temperature ranges, in which the upstream temperature data were collected from temperature sensors in real time. In 2022, the temperature in Oak Ridge, Tennessee, varied from −10 °C to 35 °C. The highest H_2O loadings, 182 and 184 mg g^{-1}, occurred during warm and humid months (May and June), and the lowest H_2O loadings, 97 and 107 mg g^{-1}, occurred during dry and cold months (February and March). During February, the average H_2O loading was 15.9% lower than the overall duration average of 151.4 mg g^{-1} but was 13.6% higher than the average

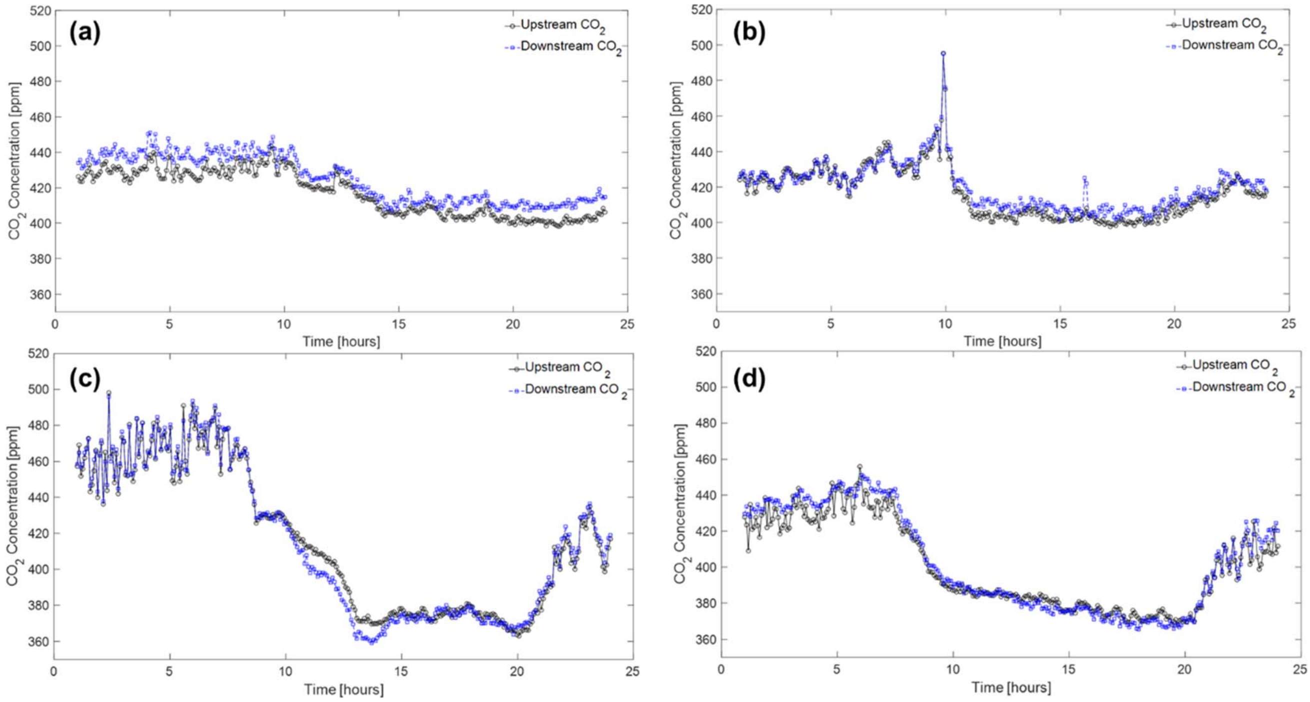


Fig. 4 Profiles of CO_2 upstream and downstream concentrations with (a and c) and without (b and d) the RTU modules during February (a and b) and June (c and d) 2022.

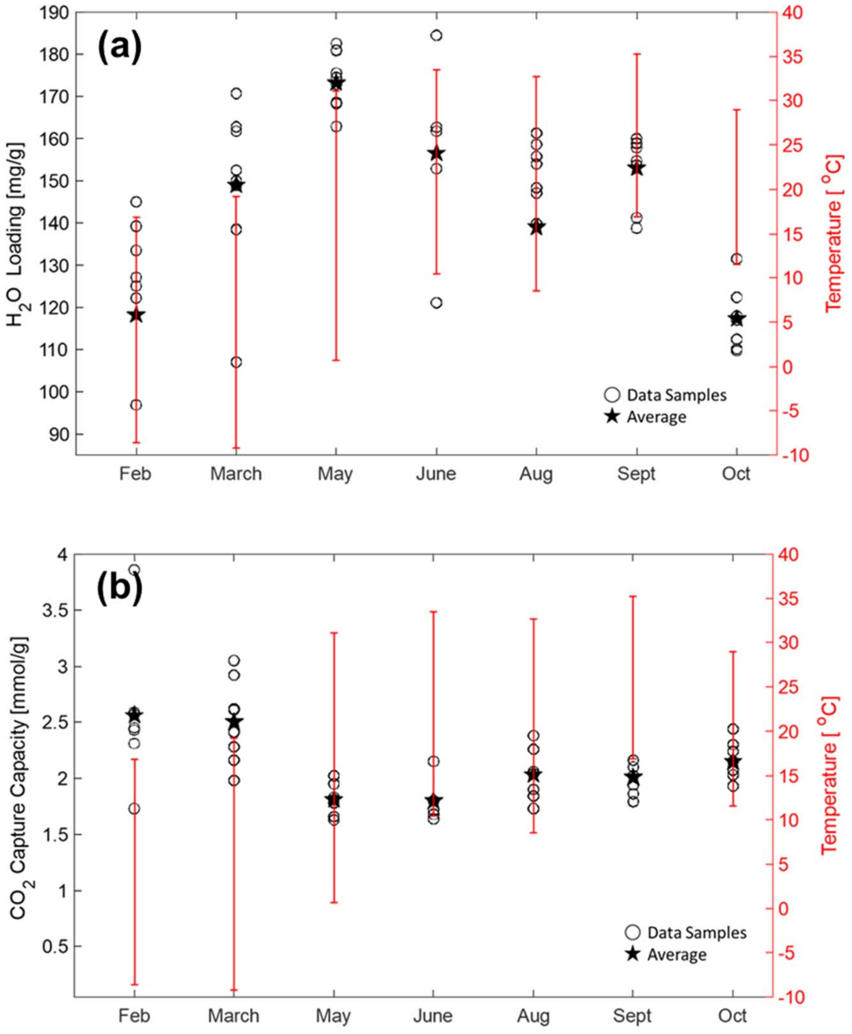


Fig. 5 Results of monthly RTU H_2O (a) and CO_2 loading (b) from February to October 2022.

H_2O loading observed during May. As expected, the moisture content in the PAN-TETA adsorbent was higher during the hot and moist month of May compared with dry periods typically observed during February. Lastly, the moisture content between February and May varied from 10.7% to 46.8%.

For the CO_2 loading test, the behavior of the PAN-TETA adsorbent during the CO_2 loading test was the opposite of its behavior during the H_2O loading test under the atmospheric temperature conditions, as shown in Fig. 5b. The lowest CO_2 loadings of 1.62 and 1.64 mmol g^{-1} were observed during warm and humid times in May and June, whereas the highest CO_2 loadings of 3.86 and 3.04 mmol g^{-1} occurred during the dry and cold conditions in February and March. The average CO_2 loading during February was 20.1% higher than the annual average of 2.11 mmol g^{-1}. Furthermore, the average CO_2 loading for May was 14.7% lower than the annual average CO_2 loading. The CO_2 loading here is much higher than the loading tested in dry N_2 (0.33 mmol g^{-1}). The high loading is caused by the presence of moisture. It is reported that CO_2 reacts with immobilized amine in the dry state and forms carbamate, whereas it forms bicarbonate in the presence of moisture.[56] Therefore, the loading of CO_2 in PAN-TETA increases in the presence of moisture.

The colder and drier geospatial regions were found to be favorable for PAN-TETA DAC performance in RTUs. These results agree with previous studies on amine-based sorbents used in DAC applications.[12,57,58] However, ambient air conditions vary widely by location because of daily and seasonal variations. Consequently, the performance of RTU DAC applications is highly dependent on the weather fluctuations in their respective locations. One limitation of this research was that all collected data were for a fixed RTU location in Oak Ridge, Tennessee.

3.3 Techno-economic analysis on RTU applications

The final cost includes the cost of raw materials, chemical wastes, maintenance, outer structure, additional energy demand, and labor. The estimated cost of PAN-TETA material development for DAC is given in Table 3. The cost of producing 7.2 kg of adsorbent materials for RTU demonstration on a pilot scale was $3221.20 for 900 PAN-TETA sheets. The reactants, PAN and TETA, are commercially available in large quantities. Currently, the costs of

PAN and TETA are respectively \$15.89 per sheet and \$8.65 per lb in small quantities, but these costs can be reduced when PAN and TETA are purchased in larger quantities for larger-scale production. The deionized water used in the reaction was produced in the lab using a deionized water system, and the cost was estimated. A large portion of the total cost (49.4%) came from the labor cost. However, labor costs will be much lower on a commercial scale; the scale for this study was quite small. Table 4 shows the estimated cost of the structural materials used for the RTU for CO_2 capture. Table 5 displays the estimated cost of electricity, Table 6 displays the estimated cost of transportation, and Table 7 displays the estimated cost of material regeneration. Fig. 6 presents the hourly power consumption for the RTU with and without the modules over the year. The annual average power consumption for the RTU modules was 8.2 kW h, which is 24.9% higher than the annual average power consumption without the RTU modules (6.15 kW h). In this work, the following assumptions for the economic analysis were used:

- A 10 day cycle was assumed for the adsorbent PAN-TETA material.
- The CO_2 loading was assumed to be 3 mmol g^{-1}.
- The electricity usage was assumed to be 2.07 kW h during the 10 day cycle (Table 5).

Table 3 Estimated cost for the adsorbent materials

	Material cost per unit (\$)	Unit	Unit usage	Cost (\$)	Reference
Polyacrylonitrile (PAN)	15.89	Sheet	13.5	214.50	—
Triethylenetetramine (TETA)	8.65	lb	84.8	733.50	—
Water	3.61	L	104	375.40	60
Waste	2.12	kg	144	305.20	61
Labor	33.18	h	48	1592.60	62
Total	—	—	—	3221.20	—

Table 4 Estimated cost for the structural materials

	Material cost per unit (\$)	Unit	Unit usage	Cost (\$)	Reference
Top roof	0.095	in.^{2}	672	63.84	1/16 in. 6061 Al sheets (McMaster)
Side wall	0.095	in.^{2}	748	71.06	1/16 in. 6061 Al sheets (McMaster)
Mesh	0.089	in.^{2}	3645	324.40	Chemical-resistant polypropylene plastic mesh (McMaster)
Container	4.875	in.^{2}	360	1755.00	6063 Al rectangular tube, 3/16 in. wall thickness, $4\frac{1}{2}$ in. high × $4\frac{1}{2}$ in. wide (McMaster)
L-shape sheet	0.21	in.^{2}	152	31.92	1/4 in. 6061 Al sheets (McMaster)
T-slot frame	1.05	in.^{2}	35	36.96	T-slotted framing (McMaster)
Total	—	—	—	2283.18	—

Table 5 Estimated cost for additional electricity usage

	Unit component	Unit	Unit usage	Cost (\$)	Reference/notes
Energy usage	0.74 (min) 4.57 (max)	kW h	—	—	2.07 (average)
Electricity cost for 5 years	—	\$	—	6763.30	Energy Outlook, EIA[59]
Electricity cost for 10 years	—	\$	—	12 729.50	Energy Outlook, EIA[59]

Table 6 Estimated cost for transportation and filter exchange

	Unit component	Unit	Unit usage	Cost (\$)	Reference/notes
Transportation	0.52	\$ per mi	2 mi	1.04	63
HVAC labor cost	23.38	\$ per h	0.5 h	11.69	For filter exchange[64]
Total cost for 5 years	—	—	—	2323.20	For a 10 day cycle
Total cost for 10 years	—	—	—	4646.50	For a 10 day cycle

Table 7 Estimated cost for material regeneration

	Unit component	Unit	Unit usage	Cost ($)	Reference/note
Mechanical convection oven	—	—	—	1270.60	81 L capacity[65]
Power consumption	1.6	kW h per cycle	—	—	For a 10 day cycle
Total cost for 5 years	—	—	—	24.09	For a 10 day cycle
Total cost for 10 years	—	—	—	48.18	For a 10 day cycle

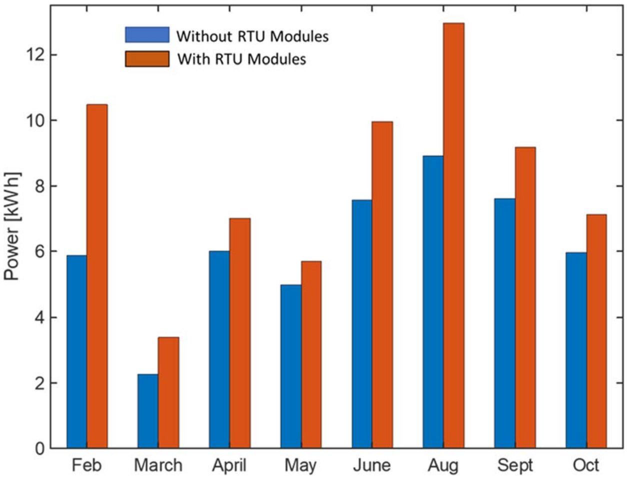


Fig. 6 Comparison of additional hourly power consumption with and without RTU modules for February to October 2022.

- The electricity cost was estimated using the Annual Energy Outlook 2023 by the US Energy Information Administration (EIA).[59]
- The transportation distance was assumed to be 2 mi (Table 6).
- A convection oven was used for the sorbent regeneration (Table 7).

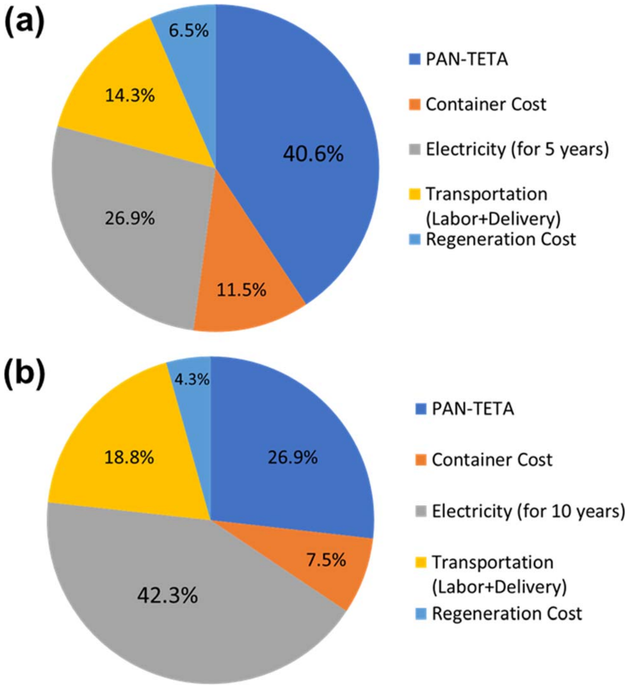


Fig. 7 Breakdown of the cost of the RTU DAC modules for (a) 5 year analysis and (b) 10 year operation.

Diagrams of the levelized costs of CO_2 capture for 5 and 10 years of operation are shown in Fig. 7. For 5 years of operation, the PAN-TETA material production is 40.6% of the total cost. To achieve \$100/ton of CO_2 levelized cost of CO_2 capture, 42.5 t of CO_2 per year is required for 5 years of analysis, and 30.5 t of CO_2 per year is required for 10 years of operation, as shown in Fig. 8. These capacities are 265 times (5 years) and 190 times (10 years) greater, respectively, than the current CO_2 capacity for a single RTU (0.16 t of CO_2 per year). To achieve this capacity and cost target from the Carbon Negative Shot, increasing the number of installations of the RTU DAC is needed. In addition, the RTU analysis result was compared with the reported learning rates of Li-ion batteries (30%) and solar PV (23%).[25,31] The one-factor learning curve is modeled as

$$C(x) = ax^b, \tag{1}$$

$$\mathrm{LR} = 1 - 2^b, \tag{2}$$

where $C(x)$ is the price of producing the next unit of the technology after a total cumulative production of x, a represents the initial cost of the technology, and b is the rate of cost reduction. The learning rate, LR, is defined as eqn (2). The learning curves for the 5 and 10 year analyses both indicate that a learning rate of 50% for the RTU application, which is a much faster learning rate compared to those of solar or even Li-ion battery technologies, satisfies the \$100/ton of CO_2 levelized cost of CO_2 capture.

4. Summary and conclusions

DAC is a promising technology for reducing atmospheric CO_2 levels. Many scientific groups recognize DAC as an essential and scalable approach that complements other climate actions. However, several challenges must be overcome to make DAC technology economically viable and environmentally sustainable. This study explored the potential of utilizing existing RTU technologies for DAC. Our research found that DAC technology integrated into an existing RTU could effectively capture atmospheric CO_2 under varying weather conditions. However, the loading capacity of CO_2 was significantly influenced by fluctuations in the atmospheric temperature and relative humidity throughout the year. The use of packed adsorbent

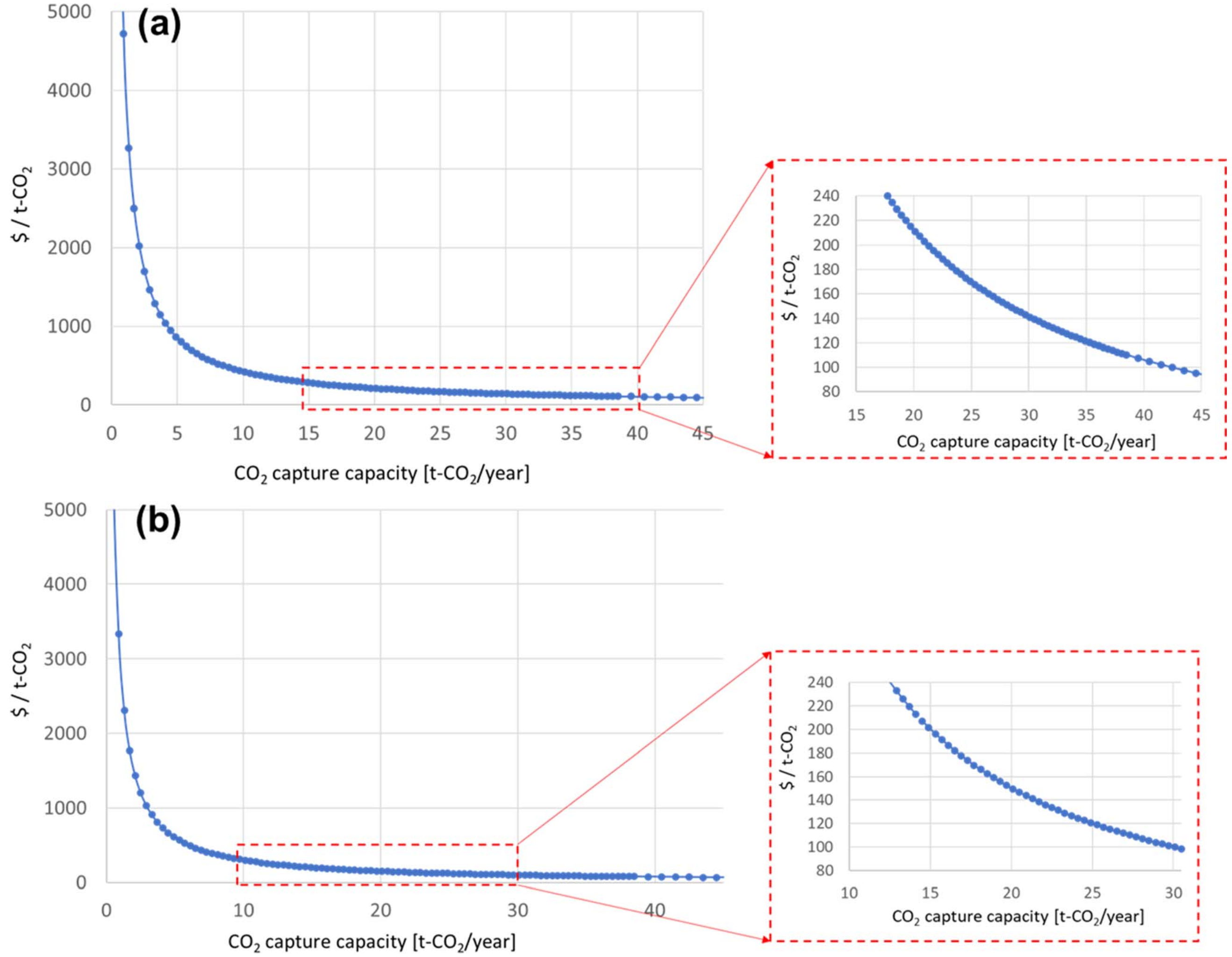


Fig. 8 Levelized cost of CO_2 capture based on assumed CO_2 capture capacity: (a) 5 year analysis and (b) 10 year analysis.

materials increased power consumption by creating an added pressure drop, which could significantly affect the overall cost of RTU DAC. Additionally, how to utilize the captured CO_2 is also an important factor in evaluating the DAC technology. We are working on utilizing the captured CO_2 from RTU, including greenhouse enrichment, storage, and CO_2 conversion into building materials.

The cost of adsorbent materials was a major limitation during the 5 year analysis, accounting for 40.6% of the total cost. In the 10 year analysis, electricity cost occupied a larger proportion of the total cost (42.3%) due to the added pressure drop across the adsorbent structure. Thus, it is essential to optimize operational processes and develop efficient adsorbent materials to reduce the application cost of RTU DAC. The challenges and benefits of RTU DAC operation are highly specific and require further analysis to verify its CO_2 capture performance and identify the most cost-effective solutions. In addition to the current removal cost, further research is needed on the capture of CO_2 from RTU DAC after the regeneration step. The small-scale, dispersedly captured CO_2 from the RTU DAC necessitates a smart network for collection, storage, or utilization. Encouragingly, scientists specializing in CO_2 capture and utilization can contribute their expertise to this field. By advancing the use of materials for CO_2 capture and utilization, the urgent need to mitigate climate change and its effects can be properly addressed.

In conclusion, the findings of our study suggest that RTU DAC technology has the potential of becoming an effective and scalable solution for capturing atmospheric CO_2. However, the future development and co-optimization of efficient adsorbent materials and RTU operational control will be essential to reduce the deployment cost of RTU DAC. Further research is required to evaluate RTU DAC locations and their impact on cost-effectiveness. This analysis should consider multiple parameters, including climatic conditions, energy costs, storage/utilization expenses, and political incentives for building owners to adopt RTU DAC technology. Evaluating these dimensions will provide valuable insights into optimizing the deployment of RTU DAC systems and enhancing their economic viability. Additionally, a full boundary cradle-to-grave life cycle assessment that tracks all installation and manufacturing-related emissions is needed to evaluate the feasibility and sustainability. The current techno-economic analysis framework presented here can also be used to back-calculate the target adsorbent material cost/performance and the levelized cost of electricity to achieve the cost target of DAC identified in the Carbon Negative Shot with this approach. More importantly, this project demonstrated the importance of multi-disciplinary collaborations in addressing the pressing global issue, and hopefully it will inspire further adsorbent material innovation, RTU system design/control and more refined techno-economic analysis in a holistic and iterative manner to advance the field of DAC technologies.

Conflicts of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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