

# A pilot-scale prototype contactor for CO<sub>2</sub> capture from ambient air: cost and energy requirements

Joshuah Stolaroff<sup>1</sup>, David Keith<sup>2</sup>, Greg Lowry<sup>1</sup>

<sup>1</sup>Carnegie Mellon University,  
Pittsburgh, Pennsylvania, USA

<sup>2</sup>University of Calgary,  
Calgary, Alberta, Canada

## Abstract

Systems for capturing carbon dioxide (CO<sub>2</sub>) from ambient air for sequestration (“air capture”) have recently been proposed, but the feasibility of such systems has been disputed. Most components of these systems operate in existing industries, but the component making contact with air for initial extraction of CO<sub>2</sub> (the “contactor”) is not well-understood. We build a prototype contactor which uses sprayed sodium hydroxide (NaOH) solution to absorb atmospheric CO<sub>2</sub>. A range of pressures, NaOH concentrations, and nozzles are tested, yielding relatively low pumping energy requirements ranging 6-15 kJ/mol-CO<sub>2</sub>. The authors conclude the proposed design is feasible for use in an air capture system.

**Keywords:** carbon capture, sequestration, air capture, sodium hydroxide

## Introduction

Global climate change is one of the most serious environmental problems we now face. In order to mitigate climate change, deep reductions in carbon dioxide (CO<sub>2</sub>) emissions will be required in the coming decades. Given the extent of emissions reductions needed to stabilize atmospheric CO<sub>2</sub> concentrations and the inertia involved in shifting the world's primary energy sources, carbon capture and storage will likely constitute a substantial share of emissions reduction in the next several decades.

Nearly all current research on carbon capture focuses on capturing CO<sub>2</sub> from large, stationary sources, such as power plants, as it is produced. Such plans usually entail capturing CO<sub>2</sub> from flue gas, compressing it, and transporting it via pipeline to a sequestration site, where it is injected under ground or otherwise stored. In contrast, the system described in this paper captures CO<sub>2</sub> directly from ambient air (“air capture”).

This strategy is not meant as a replacement for capture from point sources, but may act as an important complement, since CO<sub>2</sub> emissions from any sector can be captured, including emissions from diffuse sources such as automobiles, where on-board carbon capture is very difficult. Additionally, in a future economy with low carbon emissions, air capture can be deployed to generate negative net emissions. This ability to reduce atmospheric CO<sub>2</sub> concentrations faster than natural cycles allow would be particularly desirable in scenarios where climate sensitivity is on the high end of what we expect, resulting in unacceptable shifts in land usability and stress to ecosystems.

Previous research has shown that air capture is theoretically feasible. Fundamentally, the process is one of concentrating CO<sub>2</sub> from atmospheric levels to those required for compression (~90%). In the thermodynamic limit, the energy requirement of this process (i.e., the change in free energy of mixing) is only about 20 kJ/mol, or 3% of the energy released in combustion of gasoline[1]. If an

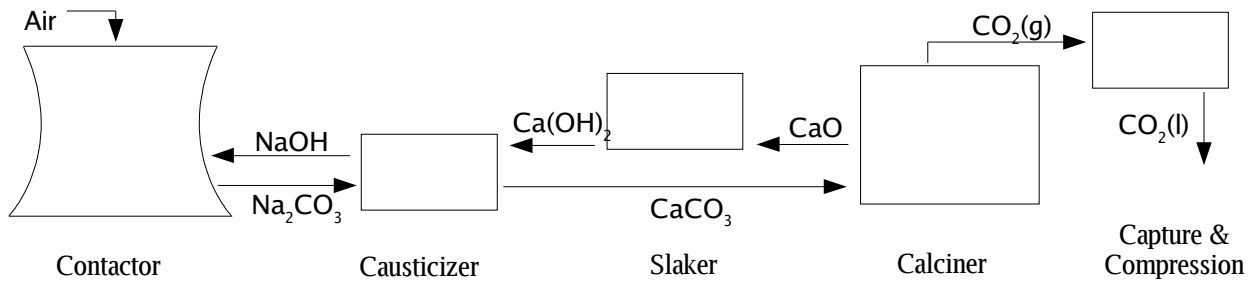


Figure 1: Overview of example air capture system. CO<sub>2</sub> is captured from the atmosphere and compressed for sequestration. NaOH and CaO are used and regenerated in closed chemical loops.

efficient mechanism for this process can be found, it has also been argued that land-use requirements would be modest[ibid.] and that local atmospheric transport of CO<sub>2</sub> would not be limiting[2].

In order to support the feasibility of air capture, example systems using current technology have been proposed[1,3,4,5]. The proposals generally include a sodium hydroxide (NaOH) or calcium hydroxide (Ca(OH)<sub>2</sub>) solution which absorbs CO<sub>2</sub> and is regenerated by "calcination" (see next section), though the specific form has remained vague.

The cost of operating an alkali-metal capture system has been claimed as low as 25-75 \$/t-C [4], though it has been disputed whether the system could be operated at any cost and energy requirement low enough to make it feasible[6]. The component of the system which makes contact with air to extract CO<sub>2</sub> (which we term the "contactor") remains a particular point of contention, with suggested forms including large convection towers[3], open, stagnant pools[1], and packed scrubbing towers[6].

### Example system

We develop an example air capture system based on those proposed previously in the literature. The top-level components of the example system are diagrammed Figure 1. It is a reproduction of the example system proposed by Keith et al.[5]. In the contactor, CO<sub>2</sub> is absorbed into NaOH solution forming sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>), which is then sent to the "causticizer", where the NaOH is regenerated by addition of lime (CaO) in a batch process. The resulting CaCO<sub>3</sub> solid is sent to the calciner where it is heated in a kiln to regenerate the CaO, driving off the CO<sub>2</sub> in the process known as calcination. The CO<sub>2</sub> is then captured from the flue gas by conventional means (such as an amine system), compressed, and sequestered for long term storage. The net result is that CO<sub>2</sub> is concentrated from atmospheric levels to those required for compression and storage. The primary inputs are energy, water, and small amounts of Na<sub>2</sub>CO<sub>3</sub> and CaCO<sub>3</sub> to make up for losses in the regeneration process.

The appeal of the system is that the chemicals involved are all inexpensive, abundant, and relatively benign, and that the processes are well-understood as current industrial-scale practices. This is a system which could be engineered today with existing technology and which can scale to capture a significant fraction of CO<sub>2</sub> emissions without depleting scarce resources.

The causticizer and calciner together constitute the "caustic recovery process", practiced for decades at large scale in the pulp and paper and aluminum manufacturing industries. For purposes of estimating the costs and energy requirements of our example system, we can draw directly on the experience of these industries. Likewise, CO<sub>2</sub> capture from flue gas is widely studied and practiced at power plants, and we can draw directly on existing data.

Aside from CaO-addition and calcination, there are other ways to regenerate Na<sub>2</sub>CO<sub>3</sub> to NaOH

which may be applicable to a NaOH-based contactor[7]. In particular, sodium borates can be reacted with  $\text{Na}_2\text{CO}_3$  at high temperature to release  $\text{CO}_2$  as a gas and regenerate NaOH. The process, known as autocaustization, has been demonstrated in the pulp and paper industry[8]. It has not been significantly analyzed for use in an air capture system, but it appears that it has much lower theoretical energy requirements and can be executed with fewer process units than a traditional caustic recovery system. Additionally, it may be possible to extract  $\text{CO}_2$  from the reacting solution at high pressure and purity, saving energy in the final compression step. Industry experience demonstrates that this is possible, though cost and energy requirements are not as readily available.

In contrast to the caustic recovery system, the contactor, while not without precedent, does not have an obvious industrial analogy from which we can estimate cost and energy requirements. We propose a specific form of the contactor modeled after a power plant cooling tower and functioning similarly to a power plant sulfur-scrubbing tower. We built a prototype of the contactor which demonstrates feasibility of the process and assists estimation of the costs and energy requirements of the full-scale analogue.

$\text{CO}_2$  extraction from air with NaOH solution has been a well-known process for many decades [9,10]. The most common industrial method of absorbing a gas into solution is to drip the solution through a tower filled with packing material while blowing the gas up through the tower. However, "packed tower" designs such as these are suited for high removal efficiency (generating carbon-free air) rather than high throughput and low energy use per unit of  $\text{CO}_2$  captured. An alternate strategy is to use an empty tower with the solution sprayed through, much like a power plant evaporative cooling tower or a sulfur-scrubbing tower for combustion flue gas. This design lends itself to a large throughput of air with a small pressure drop. Additionally, the simplicity of this design and the analogy with industrial cooling towers allow for easier estimation of the costs.

## Methods

We sought to construct a prototype contactor that would demonstrate the feasibility of  $\text{CO}_2$  capture by NaOH spray and allow us to measure the energy requirements in a way such that the results could be scaled up to a full-sized contactor. We required (1) a tower diameter large enough to accommodate a spray nozzle such that  $\text{CO}_2$  absorption by the spray can be isolated from  $\text{CO}_2$  absorption by the wetted wall, (2) a tower tall enough such that drops travel at steady-state velocity for most of the fall, and (3) solution loss by entrainment is minimal and small caustic droplets are filtered from the outlet air for safety.

The prototype contactor is diagrammed in Figure 2. NaOH solution is sprayed through the tower, collected, and recirculated from a 15-L reservoir while air is blown through the tower co-currently at approximately 0.35 m/s. The co-current design allowed for simpler construction and maintenance of the particle trap system, though it may sacrifice some efficiency over an alternative counter-current design. Additionally, temperature, relative humidity, and pressure drops are recorded.

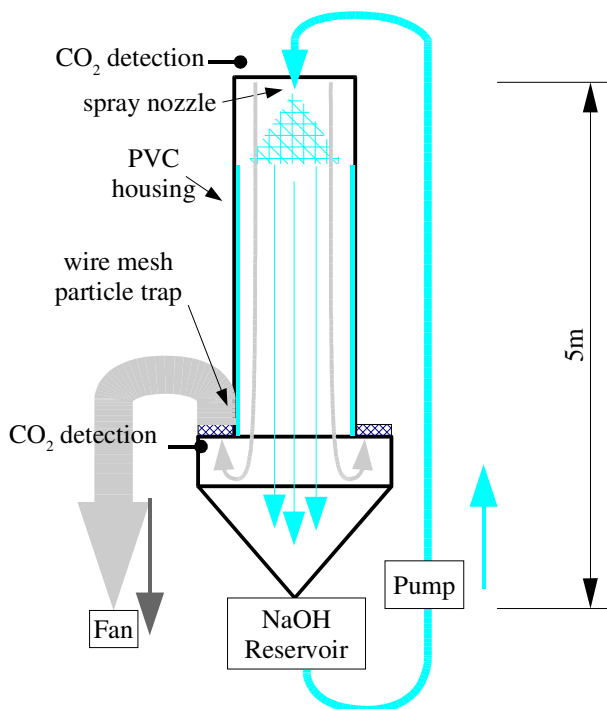


Figure 2: Prototype design. NaOH spray absorbs  $\text{CO}_2$  from air pulled through by a blower while  $\text{CO}_2$  concentration is measured at the inlet and outlet.

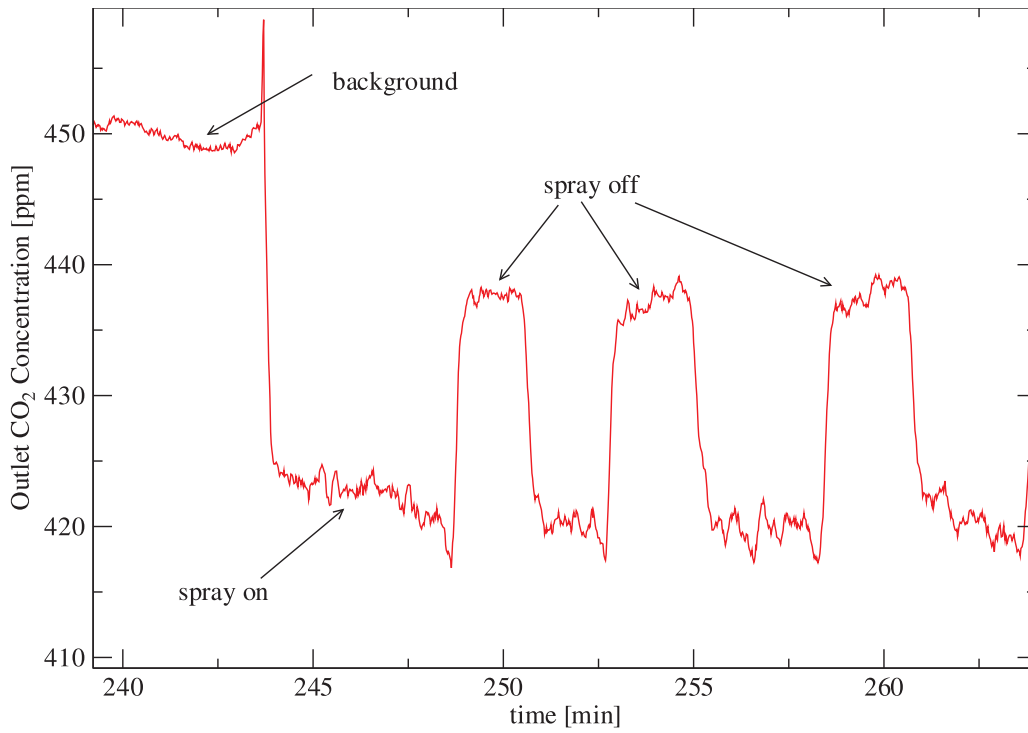


Figure 3: Outlet CO<sub>2</sub> concentration during a typical trial. Turning the spray on and off allows separation absorption due to spray and due to the wetted wall.

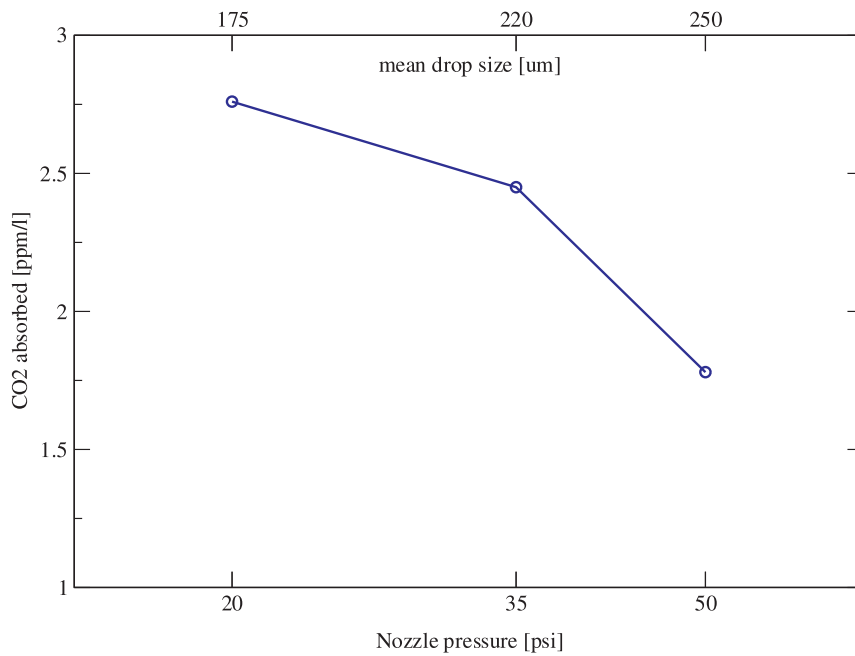
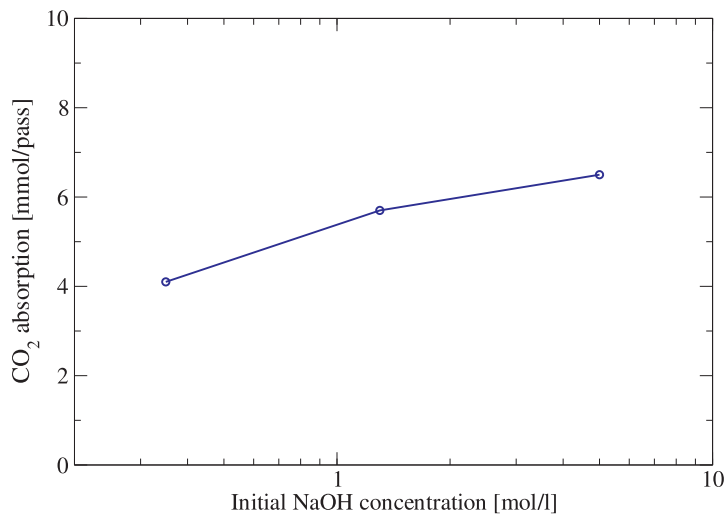


Figure 4: CO<sub>2</sub> absorption for several nozzle pressures. The top axis shows drop size corresponding to each nozzle pressure according to manufacturer data. Smaller drops absorb more efficiently.

Two different spray nozzles, an Allspray spiral full cone and Delevan narrow full cone, were used. While seven nozzles from three manufacturers were initially tested, these two were observed to produce the smallest drops at flow rates appropriate to the system, 1-6 L/min. Most nozzles have flow rate and some have drop size specifications provided by the manufacturer. We measured drop size distributions on all nozzles except the Delevan (which was not available at the time of testing) using a laser diffraction spray particle analyzer and found general agreement with manufacturer claims. Drop size is only used in calculations for Figure 4, and there we use manufacturer data for water.

Figure 5: CO<sub>2</sub> absorption for several concentrations of NaOH. Absorption increases with concentration.



In the experiments we used nozzle pressures ranging from 15 to 90 psi. Higher pressures tend to produce smaller drops for a given nozzle, but of course require more energy. However this is only a trade-off in operation, not in design, since some types of nozzles produce very small drops at low pressures. While the nozzles chosen were most suitable among those tested, they are not optimal, and we expect that in a real system smaller drops would be produced at similar or lower pressures by careful choice of nozzle.

In order to investigate the effect of NaOH concentration on CO<sub>2</sub> absorption, we tested three concentrations. A 0.35 M solution represented a dilute state, with viscosity and vapor pressure about the same as water. A 5 M solution, about 20% NaOH by mass, represented a high-concentration solution which has a viscosity about 4 times that of water and is hydroscopic in most climatic conditions. 1.33 M served as an intermediate concentration (each concentration is separated by about a factor of 4).

## Results

Figure 3 shows the outlet CO<sub>2</sub> concentration during a typical trial. CO<sub>2</sub> is absorbed by NaOH spray and by NaOH solution on the walls, reducing the outlet CO<sub>2</sub> concentration compared with the background (inlet). By running the system to steady state, then suddenly turning the spray off we can separate the absorption of the wall from the spray. Figure 3 shows several on-off cycles. The absorption rate is calculated from the average difference in peak heights. Under the conditions in this trial, 17 ppm of CO<sub>2</sub> is absorbed by the spray and 17 ppm by the wetted wall from the air passing through the system. Considering just the spray effect, this is an absorption rate of 3.7 mM CO<sub>2</sub> per pass.

By adjusting nozzle pressure with other parameters held constant, we indirectly measure the effect of drop size. We combined our measurements with manufacturer-reported data for flow rate and mean drop size as a function of nozzle pressure[11] to produce Figure 4.

With other parameters fixed, changing the solution concentration had competing effects. Higher concentration solutions should absorb better due to a faster reaction rate[12] but the higher viscosity

solution breaks into larger drops. Figure 5 shows absorption for 3 different NaOH concentrations. While absorption increases with higher NaOH concentration, it does not increase as fast as theory would suggest if drop size were constant.

By dividing the energy to lift the solution by the CO<sub>2</sub> absorbed per pass, we find the pumping energy requirements for various conditions range from 6 to 15 kJ/mol-C (neglecting pump inefficiency). This is relatively small compared with the heat of combustion of, e.g., gasoline, 600 kJ/mol-C, and with the energy requirements of the overall system as estimated by Keith et al.[5]. There is additional energy required to generate pressure at the nozzle head, though in a real system with 120m height, this becomes relatively small.

## Discussion

Our development and demonstration of a NaOH-based contactor addresses previous criticism of example air capture systems. Our findings demonstrate that a NaOH spray tower is a feasible and relatively low-cost contactor for a calcination-based air capture system. We do not claim the design presented is optimal; there are still many aspects of the contactor and overall system that may be improved. However, the contactor completes the set of components which are demonstrated to operate; each of the others has analogies in currently-operating industries. The existence of a feasible contactor supports previous proposals for a calcination-based air capture system, in particular it supports the estimates by Keith et al.[5], who argue that air capture can be achieved at costs relevant to current climate policy. Forthcoming research by the authors will estimate the cost and energy requirements of the complete system more precisely.

## References

- [1] Dubey M. K., Ziock H., Rueff G., Elliott S., Smith W. S.. Extraction of carbon dioxide from the atmosphere through engineered chemical sinkage. ACS – Division of Fuel Chemistry Reprints. 2002;47:81–84.
- [2] Johnston N.A.C., Blake D.R., Rowland F.S., et al. Chemical transport modeling of potential atmospheric CO<sub>2</sub> sinks. Energy Conversion and Management. 2003;44:681–689.
- [3] Lackner K. S., Grimes P., Ziock H. J.. Capturing carbon dioxide from air. Proceedings of the First National Conference on Carbon Sequestration (Washington, DC). 2001.
- [4] Zeman, Frank S., Lackner, Klaus S.. Capturing carbon dioxide directly from the atmosphere. World Resources Review. 2004;16:62–68.
- [5] Keith, David W., Ha-Duong, Minh, Stolaroff, Joshua K.. Climate strategy with CO<sub>2</sub> capture from the air. Climatic Change. 2005. (Available online.)
- [6] Herzog, Howard. Assessing the feasibility of capturing CO<sub>2</sub> from the air. (tech. rep.) MIT Laboratory for Energy and the Environment. 2003.
- [7] Tran H., Mao X., Cameron J., Bair C.M.. Autocausticizing of smelt with sodium borates. Pulp and Paper Canada. 1999;100:35–40.
- [8] Lindberg, Daniel K., Backman, Rainer V.. Effect of Temperature and Boron Contents on the Autocausticizing Reactions in Sodium Carbonate/Borate Mixtures. Industrial Engineering Chemical Research. 2004;8:6285–6291.
- [9] Greenwood, K., Pearce, M.. The removal of carbon dioxide from atmospheric air by scrubbing with caustic soda in packed towers. Transactions of the Institution of Chemical Engineers. 1953;31:201–207.
- [10] Hoftyzer, P.J., Krevelen, D.W.. Applicability of the results of small-scale experiments to the design of technical apparatus for gas absorption. Transactions of the Institution of Chemical Engineers, Supplement (Proceedings of the Symposium on Gas Absorption). 1954;32:S60–S67.
- [11] Allsray Inc.. Product Specifications. (tech. rep.) Allsray Inc. 2002.
- [12] Danckwertz P. V.. Gas-Liquid Reactions. New York: McGraw-Hill 1970.