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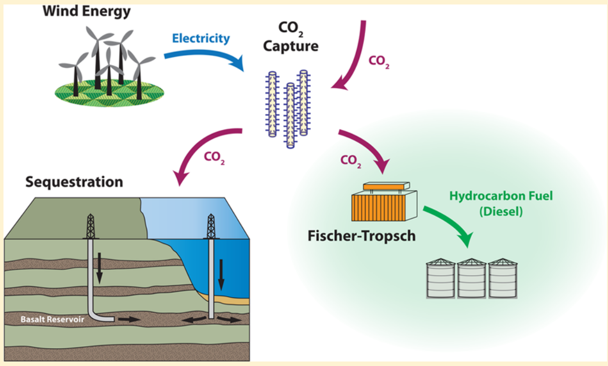
Co-Location of Air Capture, Subseafloor CO_2 Sequestration, and Energy Production on the Kerguelen Plateau

David S. Goldberg,[†,*] Klaus S. Lackner,[‡] Patrick Han,[†,∥] Angela L. Slagle,[†] and Tao Wang[‡,§]

[†]Lamont-Doherty Earth Observatory of Columbia University, Palisades, New York 10964, United States
[‡]Department of Earth and Environmental Engineering, Columbia University, New York, New York 10027, United States

Ⓢ *Supporting Information*

ABSTRACT: Reducing atmospheric CO_2 using a combination of air capture and offshore geological storage can address technical and policy concerns with climate mitigation. Because CO_2 mixes rapidly in the atmosphere, air capture could operate anywhere and in principle reduce CO_2 to preindustrial levels. We investigate the Kerguelen plateau in the Indian Ocean, which offers steady wind resources, vast subseafloor storage capacities, and minimal risk of economic damages or human inconvenience and harm. The efficiency of humidity swing driven air capture under humid and windy conditions is tested in the laboratory. Powered by wind, we estimate ~75 Mt CO_2/yr could be collected using air capture and sequestered below seafloor or partially used for synfuel. Our analysis suggests that Kerguelen offers a remote and environmentally secure location for CO_2 sequestration using renewable energy. Regional reservoirs could hold over 1500 Gt CO_2, sequestering a large fraction of 21st century emissions.



INTRODUCTION

Strategies for stabilizing atmospheric greenhouse gas concentrations will need to consider future CO_2 emissions from an enormous resource of worldwide fossil fuel supplies and a diverse range of mitigation technologies.[1−4] Globally, manmade sources emit ~30 Gt CO_2/yr. If all potential resources of conventional fossil fuels (oil, gas, and coal) were entirely combusted, total atmospheric emissions may exceed 5500 Gt CO_2 or 1500 Gt C.[5,6] Exploitation of unconventional fossil fuels (tar sands, methane hydrates) and new extraction technologies could double this amount.[7] Even if all emissions from large fixed sources could be captured, the roughly 30−50% of global emissions due to transportation and mobile sources would still be released into the atmosphere. The likelihood is that fossil fuel emissions will increase for decades, and thus, not allow for stabilization of atmospheric CO_2 below current levels of ~400 ppm.

In this paper, we study a combined and novel approach for CO_2 capture and energy production in a remote environment—on the Kerguelen plateau in the southern Indian Ocean—where we propose that long-term CO_2 capture, sequestration, and energy production infrastructure could be developed and implemented with minimal risk of postinjection leakage or environmental damage. One approach for carbon capture proposes new technologies to remove CO_2 from ambient air flowing over chemical sorbents, such as strong alkali elements, to produce a CO_2 offstream.[8−11] Because CO_2 mixes rapidly in the atmosphere, such air capture systems may be sited without regard to their distance from CO_2 sources, avoiding the major technical challenges and risks of transport and eliminating the requirement of proximity of sources to potential reservoirs. Air capture could ultimately enable atmospheric CO_2 to be reduced below current levels. The economics of CO_2 air capture, alternative fuel sources, and geo-sequestration depend on many independent cost elements, which may or may not prove to be commercially sustainable over the long-term or publically acceptable for climate mitigation. However, the cost of developing new technologies is often unpredictable,[12] and with the potentially irreversible and damaging accumulation of CO_2 in the atmosphere at current emission levels, the cost of inaction with respect to full investigation of all feasible mitigation strategies is incalculable.[13] Our primary goal in this paper is to present an approach to reduce net atmospheric CO_2 accumulations in a technologically feasible, environmentally secure, and publically acceptable manner.

MATERIALS AND METHODS

Our approach involves combining three colocated methodologies to address this goal, each at a different stage of technological development and commercial maturity. These are (1) CO_2 capture from ambient air, (2) energy production from remote wind resources and in part for synfuel conversion of CO_2, and (3) environmentally secure CO_2 geo-sequestration in

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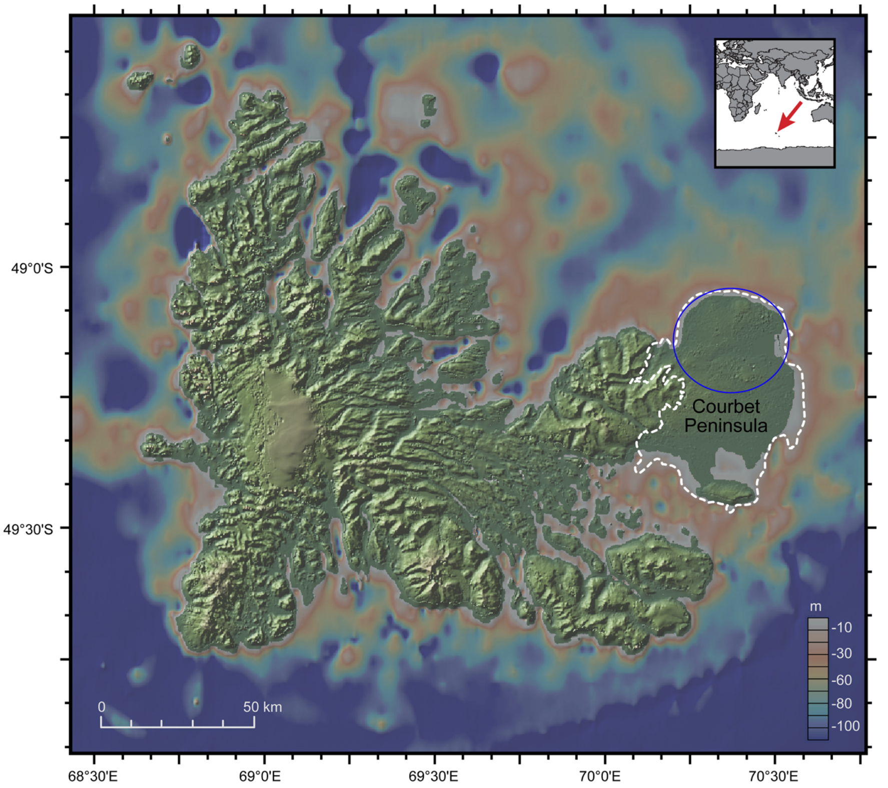

Figure 1. Topography and bathymetry[28] of Kerguelen Island in the southern Indian Ocean. Contoured region (dashed line) outlines ~2600 km^2 with elevation <100 m on the Courbet Peninsula and the adjacent offshore region with <20 m water depth. A representative 1000 km^2 area (oval) within this region is considered for potential air capture and wind turbine infrastructure.

oceanic basalt formations. In the following section, we discuss each of these methods and how they could be combined to assess the suitability of the Kerguelen plateau as a remote and self-sustaining location for CO_2 capture and sequestration.

1. Ambient Air Capture of CO_2. Lackner et al[8] first suggested direct capture of CO_2 from ambient air as an energetically and economically viable climate mitigation technology. Air capture is akin to flue gas scrubbing in power plants but because of the low concentration of CO_2 in air, sorbents for air capture must be stronger than those for flue gases. Several approaches for CO_2 capture have been proposed that use different collector surfaces to adsorb or absorb CO_2,[9−11,14−17] using various methods to regenerate the sorbent material and collect CO_2 in continuous cycles. For all approaches, energy is used in the regeneration of the sorbent to collect and compress CO_2 in each cycle. The moisture swing approach,[10,18] in which the evaporative drying of water from a solid sorbent material provides the energy to drive the cycle, has particularly low energy consumption. For this study, we consider whether a moisture swing is feasible under cold and humid conditions, such as those found in Kerguelen, and whether the resin can indeed dry in the wind in this environment. While we focus on the moisture swing approach, other separation technologies can and should also be considered for viable air capture methods. Indeed, the high relative humidity at Kerguelen renders the moisture swing technology less efficient than it would be under drier ambient conditions.

We used a sample of resin-based sorbent composed of a polystyrene backbone with quaternary ammonium ligands attached to the polymer.[10] The quaternary amine groups carry a permanent positive charge balanced by exchangeable Cl^- anions that for CO_2 sorption are replaced by carbonate or hydroxide ions. In the carbonate form the resin captures CO_2 with the low binding energy of the carbonate to bicarbonate reaction but with reaction kinetics faster than that of sodium hydroxide solutions.[10] This process is governed by the following reaction:

$$CO_3^{2-} + CO_2 + H_2O \leftrightarrow 2HCO_3^-$$

Wang et al[18] show that for this solid-resin sorbent the Langmuir isotherm equation describes the CO_2 loading as a function of the partial pressure of CO_2. The isotherm shifts to much higher CO_2 pressures in the presence of water, and therefore, CO_2 loading of the resin strongly depends on the partial pressure of H_2O in air. Thus, a process cycle with CO_2 loaded onto dry resin and then driven off by moisture is created. Once sufficiently dried, the resin is able to absorb CO_2 again. As part of a broader study,[19] we have measured CO_2 saturation for this resin at low temperatures and high relative humidity (RH). To simulate the drying of the resin in the wind, air was blown over the resin in our tests while continuously monitoring CO_2 and H_2O gas content in a closed chamber with an infrared gas analyzer (see Supporting Information (SI)).

Once captured by the resin, CO_2 may be recovered and collected by a number of different processes (regeneration)[18,19] that all require the use of energy. For Kerguelen, the basic design of the hypothetical air capture process relies on the availability of renewable wind energy resources and passive collectors that stand in the wind and take advantage of the high air flow for drying wet resin and for letting their CO_2 load equilibrate with ambient conditions. In order to achieve a significant swing in CO_2 saturation compared to full capacity of the resin and at the same time maintain a substantial CO_2 pressure over the resin during unloading, we utilize a hybrid thermal/moisture swing process where moist air is the sweep gas that carries CO_2 away. Heat is required to raise the temperature of the resin to 45°C. Subsequently, the sweep gas carrying CO_2 is cooled to condense out water, and further cooled until CO_2 precipitates as dry ice. With warming, the CO_2 will convert from dry ice to a pressurized liquid of equal volume. Heat exchange between cooling and warming streams can provide a large part of the necessary heat transfer, and electrically driven heat pumps (powered by other energy sources) will make up any short falls.

Wang et al[19] show that the partial pressure over the wet loaded resin at 45°C is 2 kPa, and a saturation swing from 0.8 to 0.5 of the maximum saturation allowed by the stoichiometry of the resin would reduce the partial pressure below 0.5 kPa. Using the hybrid swing process between ambient conditions and 100% relative humidity at 45°C, a significant fraction of the resin capacity can be regenerated.[10] Based on these conditions, the size of the saturation swing and the partial pressure of CO_2 in the outflow largely determine the total energy requirements of this process.

2. Renewable Wind and Energy Resources. In our approach, we suggest using renewable sources to meet the energy requirements for air capture and consider potential regions around the globe with substantial wind resources. Wind resources generally vary over time and location,[20,21] and in general, offshore average wind speeds on average are 90% greater than speeds over land.[21] This resource is so large that offshore wind alone, if captured, could provide a large fraction of the estimated global electrical energy consumption in 2030.[22] In the Kerguelen plateau region, winds are relatively steady and constant, averaging 4−5 Beauforts (~8.1−14 m/s, 18−30 mph) from the west-northwest,[23] even with seasonal changes. Temperature and relative humidity are also relatively constant throughout the year, ranging between 0 and 10°C and 80−90%, respectively.[23]

On Kerguelen Island itself, the flatter topography on the Courbet peninsula and adjoining near-offshore shelf areas may be the most accessible for wind farms and other infrastructure (Figure 1). Utilizing the vast wind resources of this region, we calculate that both ambient air capture and geologic CO_2 sequestration (described below) can be sustained with sufficient energy to potentially produce synthetic fuels from water and CO_2 feedstocks. Synfuel production relies on the reduction of CO_2 and water to CO by electrolysis and the subsequent production of long chain hydrocarbons using Fischer−Tropsch processes.[24−27] The energy for CO_2 reduction could come from the carbon-neutral wind resource.

3. Geo-Sequestration in Oceanic Basalt. Geologic storage of captured CO_2 is the final step in this combined approach to reducing net CO_2 emissions. The effectiveness of geological CO_2 sequestration depends strongly on a reservoir's storage capacity, stability, and risk for leakage.[4,29] Recent studies identify igneous rocks as promising sequestration targets.[30−33] Large Igneous Provinces (LIPs) are massive emplacements of intrusive and extrusive rocks that can extend 100s of km's from their volcanic sources and occur all over the globe.[34] When LIPs are extruded subaerially, they cool rapidly, forming porous outer rinds with large voids spaces. High porosity has been measured over thick flow sequences on land, such as Columbia River Plateau, Deccan Traps, and CAMP basalts.[35,36] Pilot injection projects for CO_2 sequestration in basalt flows are underway in Iceland[33] and in the Columbia River plateau.[36] These projects anticipate that CO_2 injected in basalt rocks will ultimately be sequestered in the form of thermodynamically stable and environmentally benign minerals. Basalt acts as a natural, in situ weathering reactor both on land and below the seafloor. Buried over time by impermeable marine sediments, submarine LIPs are further sealed while such chemical weathering proceeds.[37−39]

The Kerguelen plateau LIP was formed by a series of Mesozoic volcanic eruptions, creating an elongated basement high extending over more than 3×10^6 km^2 in the southern Indian Ocean and rising more than 1 km above the surrounding seafloor, with small subaerial exposures forming the Kerguelen archipelago and Heard Island.[34,40−42] Kerguelen Island has ~7200 km^2 of exposed basalt rising in rough topography, widely eroded into canyons and runoff valleys with the exception of the high glacial regions in the west and the Courbet Peninsula in the east (Figure 1). Bathymetry around the Courbet Peninsula is relatively shallow with flat sediment cover for 2−3 km offshore.

We assess the storage potential of geological reservoirs on the Kerguelen plateau for injection and sequestration of CO_2. Three criteria are used in this assessment: (1) the presence of a basalt flow with enhanced pore space as a geo-sequestration reservoir; (2) sediment thickness of ≥200 m covering subseafloor basalt; and (3) water depths between 600 and 3000 m. These criteria ensure the physical trapping of injected CO_2 and allow for estimation of the total reservoir capacity.[43] The overlying sediment acts as an impermeable cap to isolate reservoirs from potential upward leakage of injected CO_2. The 600 m minimum water depth ensures sufficient hydrostatic pressure of ocean and sediments to support injection of CO_2 in supercritical state. The 3000 m maximum depth meets the practical limit of deep-water drilling technology and generally falls <500 km from Kerguelen Island. Using data from drilling studies, we interpret the occurrence of pore space and interflow voids within basalt layers on the northern Kerguelen plateau (see SI) and apply the above criteria to assess its potential reservoir storage capacity.

■ RESULTS

In the following section, we present the results of bench-scale laboratory experiments on capture resin drying at high relative humidity, low temperature, and high wind speed—typical atmospheric conditions in Kerguelen. We also present wind and energy production estimates, basic process models and thermodynamic calculations, and geological analysis of storage capacity on the Kerguelen plateau. Together, these results demonstrate the feasibility of our combined approach to reduce net atmospheric CO_2 concentration with their colocation in a remote oceanic location.

Laboratory Experiments. Our laboratory experiments show that a sufficient moisture swing is possible for the solid-resin sorbent under high RH conditions and that high winds dry the resin effectively. Wang et al[19] have conducted a larger thermodynamic study of the equilibrium CO_2 loading of the resin as a function of temperature and relative humidity. Figure 2 shows the equilibrium CO_2 loading of the resin at ambient CO_2 concentrations for 0 and 10°C as a function of relative humidity. Between 0 and 10°C and at RH = ~80%, the maximum achievable resin load (saturation) is reduced by about 22% relative to the loading achievable at RH = ~40% and 25% relative to fully saturated resin.

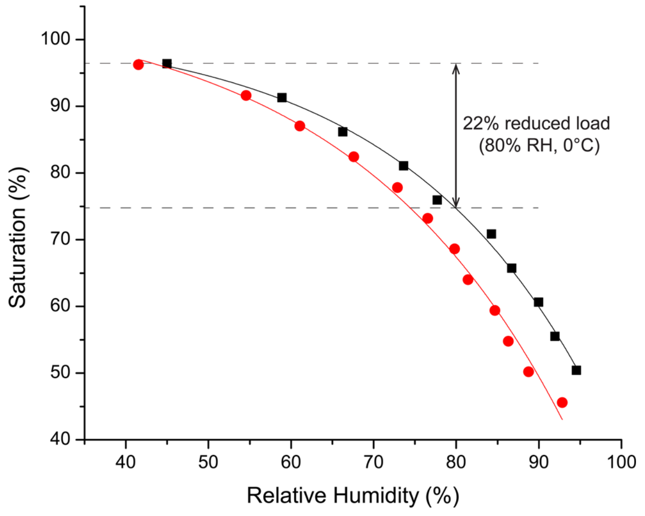

Figure 2. Change in resin CO_2 saturation versus relative humidity for T = 0°C (squares) and 10°C (circles), illustrating a 22% reduction at RH = 80% relative to RH = 40% and a 25% reduction relative to complete saturation. Curves represent exponential equation fits to the data in the form $y = A_1 \exp(-x/\tau_1) + y_o$.

Drying experiments were conducted at T = 0, 10, and 25°C, and at relative humidity RH = 60 to 90%, and nominal wind speeds of 5 m/s, 10 m/s, and 15 m/s. Results are shown in the SI. At low wind speeds (5 m/s), 60 min of drying in air with RH = 90% and T = 0°C can remove ~77% of the moisture added by wetting dry resin. As wind speed increases, more water is removed. At 15 m/s under the same RH and T conditions, ~87% of the moisture is removed. Somewhat surprisingly, less moisture is removed from the resin under higher temperature but otherwise similar conditions. A resin tested at 25°C dried less completely within the same time. As a result, the resin capture capacity is greater at lower temperatures. The drying experiments show that even at high RH and low temperature, high wind speed allows a wet sorbent that has released its CO_2 to dry to the point that it can again equilibrate to the CO_2 at ambient partial pressure.

Computation of Energy Balance. To provide energy for CO_2 capture, we consider the wind energy potential on Kerguelen Island utilizing 1000 km^2 of its flatter topography on the Courbet Peninsula and near-offshore shelf in relatively shallow (<20 m) water to install wind turbines (see Figure 1). Absent a detailed site-specific study and localized wind assessment, we estimate the wind potential on Kerguelen by using a simple Raleigh distribution of wind speed[21] with an average speed v_{mean} = 10 m/s and air density ρ = 1.2 kg/m^3. With these assumptions, the average kinetic energy flux[44] per unit area is given by $J = \rho\ /2 \cdot 6/\pi \cdot v_{\mathrm{mean}}^3$, or 1.1 kW/m^2. Windmill spacing conventionally ranges from four rotor diameters in the wind-facing direction and seven rotor diameters in the wind direction (i.e., 6 per km^2 for 77 m rotors)[21] to as high as 10−15 rotor diameters in the wind direction, to minimize boundary layer disturbances and optimize physical limitations and costs.[45] Assuming larger turbines with 4 × 12 diameter spacing, we compute the total wind power potential for the mean wind speed in the study area (1000 km^2) to be ~18 GW, which is independent of the windmill rotor diameter. Aerodynamics limits the conversion of wind energy flux to mechanical energy (Betz limit).[44] Hence, the net capacity of each turbine is assumed to be about 37%, including the Betz limitation, interferences between wind mills, and transmission losses.[46] All other things being equal however, steady winds and higher average wind speed yield more energy. Operating with 80% turbine availability (accounting for maintenance downtime, low/peak cutout times, and more limited turbine accessibility), the annual wind potential of the study area is about 47 TWh. Expanding a greater number of turbines across a larger area around Kerguelen, and optimizing the wind farm density, could reasonably triple wind energy output in the region.

To predict the energy required for CO_2 capture from colocated air collectors, we consider that the CO_2 capture potential will be reduced in this high humidity environment. We assume the resin properties are those of the material tested and that practical trade-offs will reduce the maximum available CO_2 partial pressure in the regeneration chamber (<2 kPa) and the size of the saturation swing (<0.3) for the sorbent. Figure 3 shows the calculated energy consumption per mole of CO_2 as a function of these two parameters. As shown in Table 1, the total energy consumption for CO_2 collection can reasonably be maintained <100 kJ/mol CO_2 (630 kWh/ton CO_2) with this process. Translating 47 TWh of available wind energy into a nominal annual capture capacity, Kerguelen could support capturing 75 Mt of CO_2 or more per year (Figure 3).

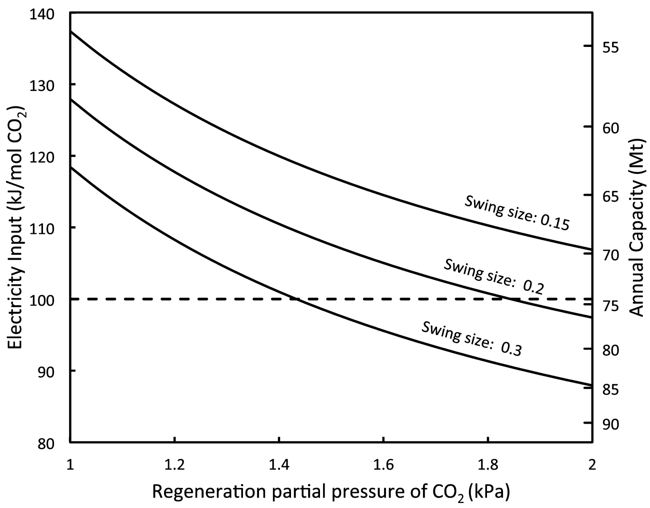

Figure 3. Energy consumption per mol CO_2 (kJ/mol) as function of the CO_2 partial pressure in a combined thermal/moisture swing process for resin regeneration. Reasonable swings (0.2−0.3 in saturation) can be maintained at 100 kJ/mol (dashed line), corresponding to ~75 Mt CO_2 capture per year for an installed power capacity of 47 TWh/yr.

To estimate the net efficiency of the hybrid thermal/moisture swing process outlined above, we assume that the fraction of the heat recovered in the heat exchange between warming and cooling streams is β. A coefficient of performance (COP) of a refrigeration unit or a heat pump can then be estimated following Cheng et al.[47] if we assume that the mechanical work applied is a fraction η of its electric input and that the hot and cold sides are indeed hotter and colder than their adjoining reservoirs by a temperature differential δ. The resulting coefficients of performance for heating (COP_1) and cooling (COP_2) are given by

Table 1. Total Energy Requirements for Resin Regeneration Utilizing the Hybrid Thermal/Moisture Swing Process and Thermodynamic Assumptions[a]

	enthalpy (kJ/mol)	thermal recovery	enthalpy input	heat pump COP	electric input (kJ/mol)
wet resin heating	855	0.8	171	7.5	23
sweep air heating	82	0.8	16	7.5	2
water added to sweep air	263	0.8	53	4.7	11
desorbing CO_2	32	0	32	4.7	7
sweep gas refrigeration	236	0.8	47	1.9	25
freezing CO_2	25	0	25	0.8	32
total electricity consumption					99

[a] Thermodynamic assumptions as follows: Make-up water and resin enter the cycle at 0°C, on the Regenerator side; sweep gas, sorbent, and water are heated to 45°C. The recovered gas and sorbent are subsequently cooled to 0°C and water condenses out, recovering as much heat as possible. A heat recovery efficiency, $\beta = 0.8$, is assumed. Gas is then cryogenically cooled to precipitate CO_2 as dry ice (T_{sublim} = $^{-}130$°C). Heat pumps and refrigeration units operate with electrical efficiency, η. In order to transfer heat, we assume a temperature differential of $\delta = 5$ K, decreasing the efficiency of the heat pump. Additional assumptions for this calculation include: water loading of the resin (swing between wet and dry), $W = 0.7$ kg/kg; Heat capacities: $C_{\text{air}} = 1$ J/g/K, $C_{\text{resin}} = 1.1$ J/g/K, $C_{\text{water}} = 4.2$ J/g/K; Heat of evaporation, $H_{\text{evap}} = 40.7$ kJ/mol; Maximum loading of the resin per swing, as fraction of the stoichiometric maximum, $S_{\text{resin}} = 0.8$; Size of the saturation swing, $S = 0.25$; Cationic charge on the resin, $E_{\text{resin}} = 1.7$ mol/kg; Partial pressure of CO_2 released, $pCO_2 = 1.6$ kPa.

$$\mathrm{COP}_1(t_1, t_2) = \eta \frac{t_1 + \delta}{t_1 - t_2 + 2\delta}$$

$$\mathrm{COP}_2(t_1, t_2) = \eta \frac{t_2 - \delta}{t_1 - t_2 + 2\delta}$$

During heating the upper temperature increases steadily and in refrigeration the lower temperature decreases. With this assumption, we calculate average COPs for T_1 and T_2, the upper and lower temperature limits, respectively.

$$\frac{1}{\langle \mathrm{COP}_1 \rangle} = \frac{1}{T_1 - T_2} \int_{T_2}^{T_1} \frac{\mathrm{d}t}{\mathrm{COP}_1(t, T_2)}$$

$$\frac{1}{\langle \mathrm{COP}_2 \rangle} = \frac{1}{T_1 - T_2} \int_{T_2}^{T_1} \frac{\mathrm{d}t}{\mathrm{COP}_2(T_1, t)}$$

Table 1 accounts for the energy requirements for these process steps, assuming the heat exchange efficiency between heating and cooling streams is $\beta = 0.8$. The effective efficiency assumed here is comparable to those obtained for other heat pump designs.[48] This calculation does not optimize the system but provides reasonable estimates of energy consumption that could be achieved with this hybrid process.

Computation of Reservoir Capacity. To determine how much captured CO_2 could be sequestered in this location, we use the known bathymetric,[28] sediment thickness and geological data[49] in the northern, central, and Elan Bank regions of the largely submarine Kerguelan plateau LIP and calculate a potential reservoir area covering as much as ~1.6 × 10^6 km^2. Drilling studies across the northern Kerguelen plateau LIP identified a series of thin high-porosity basalt layers, totalling 10−20 m in net thickness.[50] They are characterized by fractured intervals with porosity values of 7−12% separated by lower porosity layers. These higher porosity intervals are analogous to subaerial basalt flow tops and provide injection reservoirs similar to continental basalt flows.[30,39] The existence of such high-porosity layers at different locations across the Kerguelen plateau LIP, emplaced over a period of more than 30 Ma, suggests that they are pervasive and recurring over its volcanic history, but not synchronous. Porous flows are thus unlikely to be contiguous across the entire plateau, even though flow tops may be continuous for 100s km distance from their volcanic source.[51] Nevertheless, total reservoir volume can be estimated for representative flows observed at existing drill sites and at different depths (see SI). Because drilling data and observations are sparse, we use conservative values for a net 10 m thick basalt reservoir with average porosity of 10% that could be available for CO_2 injection, albeit at different depths across the northern, central and Elan Bank regions of the Kerguelen plateau LIP (Figure 4). Assuming that liquefied CO_2 (CO_2 density ~1 g/cm^3) is injected to fill these reservoirs, we estimate a potential storage capacity of ~1500 Gt CO_2. Even with low or moderate permeability, injection could be accomplished with multiple wells and using current lateral drilling technology[52] to accommodate ~75 Mt CO_2 per year. This is an enormous volume even with conservative assumptions about the reservoir characteristics (i.e., thickness, porosity, and injectability). With our estimated CO_2 collection rates from ambient air around Kerguelen Island, even 1% of this reservoir volume would be sufficient for hundreds of years of CO_2 injection and storage.

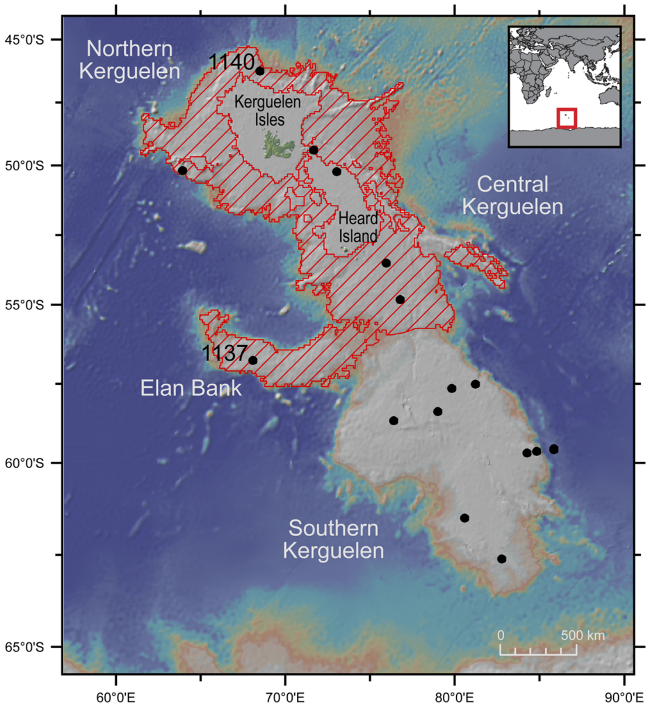

Figure 4. Map of Kerguelen plateau LIP in the southern Indian Ocean, with seafloor bathymetry and location of drill sites. Hatched outline shows potentially suitable regions for CO_2 sequestration using methodology from Goldberg and Slagle[43] in the northern, central and Elan Bank regions.

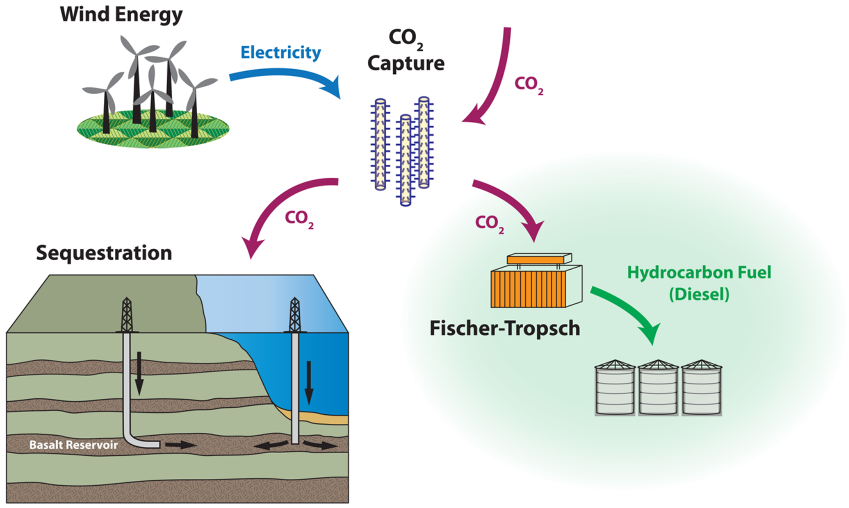

Figure 5. Schematic of potential wind energy resource use on Kerguelen. With ambient air capture, sufficient energy could be collected to sequester 75 Mt of CO_2 or more in subseafloor basalt reservoirs or produce ~770 million gal of diesel fuel annually using electrolysis and Fischer−Tropsch processes.

■ DISCUSSION

The reduction of atmospheric CO_2 concentrations by ambient air capture combined with geo-sequestration in subseafloor formations offers a powerful tool for carbon management. Potential sites differ in terms of formation characteristics, technical capacity, and economic potential, as well as human impact, but this option can focus decision-making on optimizing storage locations with respect to renewable energy resources, human and environmental risks, and public acceptance. Ambient air capture also offers a mechanism to measure and quantify output volumes of CO_2.

An important consideration for implementation of this approach is the cost of establishing and operating colocated infrastructure at remote sites. In any scenario, the costs for carbon capture and storage are very large.[4] Infrastructure costs would be considerable in the case of Kerguelen. Although there is limited existing infrastructure at the present, large industrial activities have been staged there in the past. Kerguelen produced and exported seal oil for most of the 19th century under British authority[53] and the island has since been used for French military and scientific activities.[54] For the proposed activities, however, major infrastructural requirements would include wind turbine farms, transmission lines, power stations and substations for which installed capital costs are >$190/MWh,[46] totalling $9−10B for the scale of operations proposed here. Additionally, drilling, pipeline, and storage infrastructure could easily reach $100−200M per well[55] for multiple offshore platforms, totalling again to ~$10B or greater levels of investment.

The operating costs of the proposed system are perhaps most important in assessing its viability, but also the most difficult to estimate. The cost of various air capture approaches, in particular, has been quantified to be as high as $600−1000 per tCO_2[56,57] or as low as $25−30 per tCO_2.[12] Lackner et al.[12] and others argue that such long-term costs are often difficult to estimate at the early stages of new technology development, and in Kerguelen, the net cost and efficiency of local renewable electricity will in large part determine the operating cost of air capture. Thus, if air capture efficiency could be achieved for 100 kJ/mol CO_2 (i.e., Table 1) at a net cost of $50/$tCO_2$ or less (electricity cost <$0.08/kWh), then the ~75Mt CO_2/year captured would cost ~$3.5B per annum of operation. This is equivalent to the amount of CO_2 produced by twenty-five 500 MW coal-fired power plants and would relieve the build-up of that amount of atmospheric CO_2. The operating costs for sequestration, including gas compression, pipeline costs, and injection have been estimated to be $1−9/$tCO_2$.[52,58] The volume produced by air capture in Kerguelen would therefore cost an additional $0.5−0.7B per annum to sequester, assuming the higher cost range for offshore and remote operations. Greeshem et al.[59] note that sequestration costs could double in some (continental) locations due to the difficulty and expense of obtaining rights to subsurface pore space, making long pipeline transport economical. The cost penalties for remote operations and maintenance in Kerguelen must be reconciled with its infrastructure costs, but air capture and offshore storage would likely avoid such large add-ons.

One potential advantage of the proximity of colocating renewable wind resources and captured CO_2 on Kerguelen is that they could also be used as chemical feedstock to produce long chain synthetic hydrocarbon fuels, such as methanol and diesel. Using electrolysis and Fischer−Tropsch processes,[24−27] we estimate that the available 47 TWh of wind energy could be converted annually into ~770 million gallons of diesel using ~8 Mt of collected CO_2. This amounts to only 10% utilization of the installed air capture capacity, assuming all of the wind energy is used for fuel production. Synfuel could offer a substitute commodity for sequestered CO_2 in the event of economic fluctuations, or be used as a resource to support the local infrastructure. If all of the captured CO_2 were indeed converted, the commercial value of 770 M gal of produced fuel[60] would be on the order of $3B, similar to the annual cost of air capture if realized at $50/$tCO_2$. Balancing this net value for potential synfuel production with the cost of air captured CO_2 is perhaps a practical and objective measure of when this approach could become economically feasible and justified. The proportion of energy used for CO_2 collection and sequestration versus fuel production could be scaled to balance the fuel needs and short-term economics of the proposed operation. Over the long-term, wind resources could be increased across the region to allow for greater energy production or a different product balance. Even without synfuel production, our primary goal remains to provide an environmentally secure and sustainable location for CO_2 sequestration using renewable energy in Kerguelen. With the combined use of wind resources, CO_2 air capture, and geo-sequestration with synfuel production, Kerguelen could indeed function as an energetically self-sustainable carbon collection point (Figure 5).

Numerous studies have explored the technical and environmental risks and public issues involved with geological CO_2 sequestration.[61,62] Specifically, risks associated with leakage, groundwater safety, land access, storage permanence, and long-term liability remain outstanding issues of major concern for on−land CO_2 sequestration. Locating CO_2 storage reservoirs in the subseafloor setting, however, offers long-term risk benefits such as permanent and safe sequestration, minimal environmental risks from leakage, distance from populated areas, and negligible expected damages.[63] Offshore sequestration mitigates risk of damages from induced earthquakes and concerns of harm from produced/expelled fluids for potable aquifers after CO_2 injection.[64,65] Below 100 m water depth in the ocean, CO_2 dissolved in seawater will remain in solution due to the confining hydrostatic pressure (>700 μatm) except where pCO_2 levels are anomalously high in ocean upwelling zones.[66] Thus, if porous flow of injected CO_2 were to migrate through a basalt reservoir, through low-permeability sediment caprock and into the ocean, deep seawater will ultimately provide a secure reservoir with virtually infinite storage capacity. Flow through porous rock is unlikely to be abrupt[67] and seawater displaced by CO_2 injection will be benign in the ocean. Also, because subseafloor basalts are laterally extensive and saturated with seawater, the consequences of small pressure increases due to injection are unlikely to cause faulting.

In the absence of such risks and concerns, the regulations governing sequestration in a remote offshore location may be simplified and implemented with minimal human inconvenience. Kerguelen Island and surrounding seas are indeed remote and uninhabited, but remain a territory of France. Implementing the activities proposed here would certainly require appropriate access agreements, international monitoring protocols, and more fully developed regulations for carbon sequestration. Collective regulation could be most effective through public-private partnerships, including energy, resource, environmental, and intergovernmental expertise.

In summary, the potential benefits of long-term carbon management at a remote ocean site such as Kerguelen are large. Choosing offshore sites for CO_2 air capture using carbon-neutral energy sources and in close proximity to large and secure reservoirs for sequestration allows for optimization of the energy resources, minimum human and environmental risks, measurable CO_2 mitigation, and a greater likelihood for public acceptance. Specific challenges for CO_2 capture in the Kerguelen environment are its constant high humidity, low temperatures, and remote location. Other locations with carbon-neutral energy sources and in close proximity to large and secure reservoirs for geological sequestration of captured CO_2 may be viable as well. Some other possible locations with large wind resources[21] and potential basalt reservoirs[43] include Iceland and Greenland in the north Atlantic, and Chile and Argentina in the south Atlantic oceans. Mobilizing the industrial infrastructure in these areas would be costly, but could ultimately provide sufficient air capture and sequestration capacity for the reduction of atmospheric carbon to preindustrial levels. A cost/benefit analysis of such remote installations must incorporate all technical and societal factors, although many costs remain difficult to estimate at the present time. Nevertheless, CO_2 is accumulating daily in the atmosphere and new scientific research, geophysical and hydrological surveying, technological site assessment, and economic evaluation should be energized in order to explore and evaluate the feasibility of air capture and sequestration in remote locations as soon as possible. Considering remote locations for global CO_2 management is clearly one possible solution that will require considerable investment and long-term commitments to research. Site-specific studies must be conducted at any potential target location. Establishment of a viable pilot program in the next few years would allow assessment of scaling up these technologies and sustaining combined solutions that address the global climate change issue for the long-term.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Experimental laboratory data measured in this study and field data collected during previous ocean drilling expeditions are presented. This material is available free of charge via the Internet at http://pubs.acs.org.

■ AUTHOR INFORMATION

Corresponding Author

*Phone: 845-365-8674; fax: 845-365-8777; e-mail: goldberg@ldeo.columbia.edu.

Present Addresses

§T.W.: State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou, Zhejiang Province, China.
‖P.H.: Department of Chemical and Environmental Engineering, Yale University, New Haven, Connecticut 06520.

Notes

The authors declare the following competing financial interest(s): Columbia University has submitted patent applications for aspects of this work that could potentially result in commercial use of the research. K.S.L. is also a shareholder and board member in Kilimanjaro Energy, a company that is commercializing air capture technology.

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