

The urgency of the development of CO₂ capture from ambient air

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CO₂ capture and storage (CCS) has the potential to develop into an important tool to address climate change. Given society's present reliance on fossil fuels, widespread adoption of CCS appears indispensable for meeting stringent climate targets. We argue that for conventional CCS to become a successful climate mitigation technology—which by necessity has to operate on a large scale—it may need to be complemented with air capture, removing CO₂ directly from the atmosphere. Air capture of CO₂ could act as insurance against CO₂ leaking from storage and furthermore may provide an option for dealing with emissions from mobile dispersed sources such as automobiles and airplanes.

carbon dioxide | negative emissions

Stabilizing atmospheric CO₂ will require drastic emission reductions. Nearly half of all CO₂ emitted will stay in the atmosphere for centuries (1, 2). According to the Intergovernmental Panel on Climate Change (IPCC), CO₂ emissions must be reduced by 30–85% by 2050 to be on track for stabilizing atmospheric CO₂ between 350 and 440 parts per million by volume (ppmv) (3). Not only would emissions from coal have to essentially stop by 2050 (4), but also emissions from other fossil fuels would have to be reduced. Beyond 2050, CO₂ emissions would have to continue to fall to levels approaching zero to achieve a full stabilization of the atmospheric CO₂ concentration (5, 6). It may even prove necessary to reduce excess CO₂ in the atmosphere below current levels or below future stabilization levels (7).

Carbon-free renewable and nuclear energy resources are theoretically sufficient for humankind's energy needs, especially if combined with significant increases in energy efficiency (8). It is unclear, however, whether these resources can be deployed rapidly and widely enough and overcome socio-political obstacles related to cost, environmental impacts, and public acceptance. In a world that strives for continued economic growth, moving the energy infrastructure away from fossil fuels is a challenging task.

Point-source CO₂ capture and storage (CCS) allows for the continued use of fossil fuels in power plants and in steel and cement production while largely eliminating their CO₂ emissions. Point-source CCS could be implemented without significant changes for consumers, which would be beneficial given the societal inertia in dealing with climate change. With only point-source CCS available, however, consumers would have to drastically

change their demand for liquid hydrocarbon fuels (5, 6).

Air capture could address automobile and airplane emissions inaccessible to point-source CCS. It could also remove residual emissions from point-source capture. Stabilization of atmospheric CO₂ at 450 ppmv cannot be accomplished this century if point sources equipped with CCS keep on emitting at just 10% of their current rates (9). Air capture could also deal with fugitive emissions from the transport and storage stages of CCS and thereby manage the risk of CO₂ leakage from geological storage (10, 11).

Point-source CCS and air capture are not the only options for reducing CO₂ emissions. Stationary power plants could be replaced with ones based on nonfossil resources. Mobile emitters could be eliminated by transitioning to biofuel, hydrogen, or electric vehicles. Most of these technologies, however, are not yet in an advanced stage of deployment. Air capture technology may prove superfluous if switching to new carbon-free energy systems proceeds quickly. Should this transition prove difficult, however, air capture could be an alternative that can also reduce the atmospheric CO₂ concentration. In addition, air capture may provide a new policy route for facilitating an international agreement on climate mitigation (12, 13). As Weitzman (14) and others have convincingly argued, there are significant uncertainties regarding the costs and benefits associated with climate change as well as with climate change mitigation. The development of air capture, even though itself uncertain, could be an insurance policy against low-probability high-impact events.

Current State of Air Capture Technology

Capturing CO₂ from air is technically feasible and has been practiced for decades to maintain safe levels of CO₂ in submarines (15) and spaceships (16). In addition, many processes for liquefying air require removal of H₂O and CO₂ before or during cooling (17). Complete CO₂ scrubbing from air at small scales has been studied for many decades (18–20).

Large volumes of air need to be processed to collect meaningful amounts of CO₂. Because of the ratio of air to CO₂ molecules (2,500:1), air capture systems cannot afford the effort to prepare or modify air (21), which eliminates capture technologies that put energy into the air, such as heating, cooling, or pressurizing air. The only feasible techniques involve either absorption or adsorption on a sorbent. With such techniques, energy is required only to regenerate the sorbent. This regeneration process operates on the sorbent mass, which scales with the mass of the CO₂ captured rather than the much larger mass of the air.

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For air capture combined with CO₂ storage, energy limitations are particularly stringent because CO₂ emissions associated with the use of energy could partially or completely cancel out air capture. There are several ways of gauging the relevant energy scale. For example, the heat of combustion of gasoline resulting in 1 mol of CO₂ is about 700 kJ, whereas the electricity produced at a coal plant is about 150 kJ/mol of CO₂. Another set of scales comes from the requirements in the capture process. The free energy of mixing CO₂ in air at 300 K is 22 kJ/mol. The mechanical work required to compress 1 mol of CO₂ isothermally from 0.1 to 6 MPa is about 11 kJ. Energy investments will have to exceed these thermodynamic requirements.

All currently discussed methods of air capture use absorption or adsorption on collector surfaces. In some cases, the sorbent is a liquid (21–25); in others, it is a solid (26–28). Most methods rely on batch processes that separate collection and regeneration. Liquid sorbents flowing over surfaces allow for a continuous process that keeps capture and sorbent regeneration separate. Membrane processes like the electrochemical system by Eisaman (29) and the partial pressure-driven technique by Trachtenberg's group that releases CO₂ to a vacuum (30) work continuously. Rau (31) suggests continuously operating electrolytic water-splitting systems that absorb CO₂ near one electrode.

A different approach to capturing CO₂ from air enhances natural processes, e.g., biomass growth on land (32) with subsequent formation of recalcitrant carbon (33), or biomass growth in the ocean via ocean fertilization (34), or dispersion of alkaline mineral bases such as olivines and serpentines (35). We distinguish these methods from the direct capture methods discussed above.

One can break the air capture process into three steps: (i) contacting the air, (ii) absorption or adsorption on a sorbent, and (iii) recovery of the sorbent. For membranes operating continuously, steps ii and iii are tightly integrated. For batch-type operations, they are distinct units of operation.

Contacting ambient air requires a physical structure channeling it to sorbent surfaces. Air can be driven by machinery or flow due to ambient conditions, e.g., by natural wind, thermal convection, or wind-driven pressure gradients. Fanning air is possible but energetically limited to low velocities or, equivalently, low pressure drops. Maintaining a pressure drop of 100 Pa requires 6.2 kJ/mol of CO₂ in the air. Air moving at 10 m/s has an embedded kinetic energy of 3.6 kJ/mol of CO₂. Considering allowances for the capture

efficiency of the collector and energy efficiency of the fans, 100 Pa or equivalent flow velocities of about 15 m/s define an upper limit of practical pressure drops.

Low-pressure drops or low-flow speeds are not necessarily detrimental to efficient air capture designs. Transport of momentum and CO₂ follow similar laws, and a design that dissipates an amount of momentum comparable to the initial momentum flux can also capture a significant fraction of the CO₂ present in air (26, 36). As a result, even systems with a low flow speed can efficiently capture CO₂.

Inside the contactor, the transfer rate of CO₂ is affected by airside transport limitations and by the uptake rate of the sorbent, which in turn is shaped by transport and chemical reaction rates of the material. The effective sorbent area is determined by the boundary layer surface. Smaller substructures that enhance the overall kinetics are subsumed in an effective uptake rate. The airside transfer coefficient scales with the boundary layer thickness and can be engineered to match the transfer coefficient on the sorbent side. In a honeycomb of long channels with laminar flow, the channel radius sets the air layer thickness. One can control airside transfer coefficients over a particular sorbent by choosing the channel diameter.

In most systems with air moving over sorbent surfaces, the transport laws for momentum dissipation and CO₂ diffusion are very similar. In the laminar limit, both follow the same diffusion law with similar diffusion constants. Details of the design will matter, as the density is a scalar quantity, whereas momentum involves three vector quantities. Momentum is also transported via the stress tensors, e.g., pressure gradients. As a result, it is possible to dissipate momentum while maintaining a constant convective momentum flow.

A major difference between transport of momentum and of CO₂ is the boundary condition on the sorbent surface. Momentum flow at a wall always approaches zero whereas the CO₂ concentration on the same surface can vary between zero and ambient partial pressure. A sorbent geometry that is airside limited in CO₂ transport will have a low concentration of CO₂ on the sorbent surface; a system that is surface-uptake limited will have a high partial pressure. An airside-limited system that dissipates all of the momentum initially available will also transfer a large fraction of its CO₂ to the sorbent surface because the transport follows similar equations. Such a system will have a pressure drop $\Delta P = \rho v^2$, with ρ the density of air and v the flow velocity through the channels of the sorbent system.

If, for a fixed geometry, the sorbent strength is decreased until the system becomes surface-uptake limited, the momentum transfer to the sorbent surface remains unchanged while CO₂ transfer is reduced. Thus, sorbent-side-limited geometries lead to a higher pressure drop per unit of CO₂ captured. However, for a fixed sorbent, reducing the boundary layer thickness and thereby reducing the airside transfer coefficient will increase the uptake rate per unit of sorbent surface and thus reduce the requirement for material. Optimization balances the need for more sorbent against an energy penalty due to excessive pressure drops. In the optimal system, the partial pressure of CO₂ near the surface is much smaller than ambient but much larger than zero with the system operating in the transition between sorbent and airside limitations. At present, most sorbents have uptake rates per unit area that are comparable to those of films of strong NaOH solutions. The choice of such sorbents results in a design with a boundary layer of a few millimeters and a flow-path length of a few tens of centimeters, assuming low flow on the order of a few meters per second (26).

Sorbent materials span a range of options. Most physisorption is too weak to operate at 400 ppmv, and therefore nearly all systems discussed take advantage of acid–base chemistry. Early attempts used sodium or calcium hydroxide (19, 36–38). Liquid hydroxide solutions can operate in a continuous mode; Keith and his group follow this approach (24). Eisenberger and Jones use a solid tertiary amine, a weak base analogous to an ammonia solution (28). Lackner et al. (39) use a strong-base quaternary ammonium ion, a typical anionic exchange resin. However, this sorbent is always partially carbonated, thereby reducing the effective strength of the base. Eisaman et al. (29) use a weak-base amine sorbent in membrane form, and they use electric currents to maintain a high concentration of base on the airside contact area. Rau (31) suggests a similar concept for electrolysis systems where the electrolysis that produces hydrogen and oxygen also creates an acid on one electrode and a base on the other. The base then reacts with CO₂ from the air to form carbonates. Because this system also produces hydrogen, it will require large energy inputs.

An important design goal is to achieve a high uptake rate of CO₂, reducing the sorbent surface requirement. A high intrinsic uptake rate also requires a high airside transfer rate, which implies a thin boundary layer thickness. Different materials will result in different design strategies that are affected by the intrinsic uptake rate but also by available technologies to adequately shape these surfaces.

For now, the uptake rate on the surface of a 1-M NaOH solution sets a benchmark to which all other sorbents can be compared.

Economics of Future Technologies Is Not Predictable

For air capture to play a substantial role in managing CO₂ in the atmosphere, it needs to become economically feasible on a large scale. Estimates of future costs for a fully established technology are by their nature highly uncertain and thus vary significantly, with estimates ranging from as low as \$30 per metric ton of CO₂ (t CO₂) to \$1,000/t CO₂ (40–43). Within this wide range, however, it is important to distinguish the estimates for ultimately achievable costs from those for initial implementation based on the current technology.

The American Physical Society (APS) argues that air capture is unlikely to play an important role because cost estimates of a particular air capture technology are approximately \$600/t CO₂ (40). The APS estimate pertains to a technique (38) not necessarily representative of the actual cost of future deployment. Early cost estimates 10–20 times larger than what would be competitive do not rule out economically viable implementation in the future.

In estimating the costs of a new device or plant, there are three cases to consider: an existing system built already, a one-of-a-kind fully developed but never built system, and a new untested technology. This approach to estimation simplifies the approach of Shenhar (44), who distinguishes two different categories of untested high-technology innovations. The most straightforward cost estimate is that of a device that has been built before. Uncertainties are quite small, but even in this simple case, conventional cost estimation adds contingencies that can easily be 15% or more (45).

The second class of estimates aims at the cost of a system that has been completely specified in all of its parts but has never been built. For such a system, contingencies are much larger. Some components may not work together as anticipated, necessitating costly redesigns. Lack of experience increases uncertainty, and thus contingencies routinely reach 100% (46).

The third class of estimates attempts to establish the cost of a system that is still subject to research and development (R&D) and has not yet been fully designed. Here, one would have to calculate the cost of a future project including the impact of learning achieved in projects that will lead to second- and subsequently first-class cost estimates. This third category applies to air capture technology. An accurate estimate today of future costs is simply impossible; a system that can be built now should be seen as a straw man to

be replaced with improved designs. Confusing an estimate of this third type with an estimate of the second type is likely to lead to an overestimate.

Not surprisingly, cost estimates of novel technologies have often been wrong. The costs of new technologies can drop by orders of magnitude as they develop and mass production ensues, and examples are plentiful. The cost of a central processing-unit cycle has changed by about six orders of magnitude (47). The cost of solar panels has dropped almost 100-fold since the 1950s (48, 49). Efficiency improvements in gas turbines have moved them from a scientific curiosity in the 1930s to a mainstay in power generation and aviation today (49). Once sulfur emission trading was enacted, sulfur reductions at power plants proved within 4 y to be 10 times cheaper than experts predicted shortly before the start of trading (50). Observed cost reductions of existing technologies pose a conundrum for estimating future costs. Clearly, if one could design the lower-cost version at the outset and thus correctly assess its cost from the beginning, one would not be stuck with the high cost of early implementation.

In many engineering communities, the existence of such improvements is acknowledged and even expected. The computer industry relies on it. There is a strong expectation that next year's computers will be better and/or cheaper than those of today. This trend (Moore's law) has held for several decades (47). Policies toward wind and solar energy are based on the assumption that R&D and learning by doing will continue to drive prices down. It is recognized that when these were nascent technologies, meaningful cost estimates would have been exceedingly difficult.

Outside the CCS community, promising technologies are developed for CO₂ emission reductions without excessive concern over initially high costs. Electric cars provide a path toward moving CO₂ emissions from the transportation sector to power plants, where options exist to eliminate them. We estimate in *SI Text A* that the Tesla Roadster, a fully electric, high-performance car, has an effective cost for avoiding tailpipe emissions of \$600/t CO₂, equal to the APS cost estimate for air capture. The Tesla Roadster is a first-of-its-kind luxury car. There is every expectation that prices will come down and indeed, the battery cost of a Nissan Leaf appears significantly lower. However, despite the high initial cost, the technology was successfully brought to market. We do not argue that electric cars are undesirable. Instead, we argue that the Tesla—undoubtedly much more mature than air capture—provided a start for electric vehicle technology at a cost as high as the

upper range of present cost estimates for air capture. Costs for both technologies are likely to come down, but it is difficult to predict where they will ultimately settle (e.g., ref. 51).

The CCS research community today seems more focused on incremental improvements that allow for defensible cost estimates of a technique than on experiments with more innovative concepts as seen in the car industry. This philosophy, which is exemplified by the APS study, goes back to the early days of CCS when the US Department of Energy used \$10/t C (\$3/t CO₂) as an unreasonable short-term target for carbon management costs (National Energy Technology Laboratory News Release, "Energy Department Launches Thirteen New Research Projects to Capture and Store Greenhouse Gases," July 21, 2000) (52).

It is also embedded in the IPCC Special Report on CCS, which spends much effort on estimating costs of various CCS alternatives while limiting itself mostly to currently available technology (53).

Reliance on inherently inaccurate cost estimates for new technology when formulating research policy is likely to be counterproductive. Demanding an assurance of economic viability at the outset stifles innovation, favors incrementalism, and keeps game-changing ideas from consideration.

What Prices Are Affordable?

Because it is impossible to predict the cost of an undeveloped technology, it is instructive to ask instead what cost targets must be met to make air capture a useful technology for climate change mitigation. Even this question is difficult to answer because the price paid for 1 t CO₂ depends on the application and on the available alternatives. Currently, the market price of CO₂ as a chemical commodity varies dramatically as transport by truck is a significant cost. In many locations, the price of truck-delivered CO₂ exceeds \$100/t CO₂, even reaching \$300/t CO₂ (54).

Enhanced oil recovery (EOR) represents a market for CO₂ large enough to impact climate change. Most of the used CO₂ is thought to stay underground if one factors in the reuse of the CO₂ that is returned with the oil (55). Typically, CO₂ is delivered to the EOR site by pipeline at competitive prices (56). (Average prices during the 1990s were around \$11/t CO₂.)

The availability of CO₂ is limited, however, at many remote sites. Approximately for every metric ton of CO₂ permanently pushed into the ground, 1 t of oil (about seven barrels) can be recovered. Hence, \$70/t CO₂ for EOR would raise the price of oil by only \$10/barrel (bbl).

These commercial prices set cost targets for air captured CO₂ that would ensure a certain market size for captured CO₂. The lower the cost of this CO₂, the larger the potential market. By contrast, for CCS at power plants the amounts of CO₂ captured would have to be immediately very large and thus rapidly overwhelm local markets for commercial CO₂.

The scale of CO₂ fluxes in the energy sector is so large that eventually managing CO₂ has to stand on its own and cannot be handled as a by-product of other enterprises. If climate change were universally perceived as a serious calamity, air capture as an emergency measure might be valuable at costs much higher than \$100/t CO₂ (57). It is more likely that if air capture were more than \$100/t CO₂ (\$0.85/gallon of gasoline) and there was no credible path for cost reductions, alternatives to fossil fuels would be developed (e.g., biofuels) and eventually displace them.

If realizable below \$50/t CO₂, air capture would be a strong contender among the various options and would not necessarily be tied to fossil fuels. For example, the availability of CO₂ from the air would open the door to algae-based fuel production schemes that require CO₂ as input. Synthetic fuels could be made from wind or solar energy. A carbon price of \$30/t CO₂ would add \$0.25/gallon of gasoline (\$0.07/L). High gasoline prices in Europe and large price fluctuations in the United States have shown that such a change in price would not seriously challenge the competitiveness of liquid hydrocarbon fuels. {The spread of US fuel prices during the last decade is equivalent to \$183/t CO₂. Gasoline averaged \$1.16/gallon in 1998 and \$2.71/gallon in 2008 [in (2000)\$, US Energy Information Administration (EIA), www.eia.doe.gov/emeu/aer/txt/ptb0524.html]}.}

Lower Bounds on the Cost

The question remains whether air capture can achieve the necessary cost reductions. If we take \$600/t CO₂ as a baseline (40), the challenge seems large but no larger than the corresponding challenges in other climate mitigation technologies. Necessary improvements are comparable to those that were initially required for solar and wind energy. According to the APS study, there are no obvious limits from a thermodynamics or materials perspective that would make further cost reductions impossible. Heuristics based on some industrially practiced separation processes (58) may lead one to observe that air capture technology must be very innovative to succeed, but correlations between the ratio of practically achieved efficiency and thermodynamically allowed efficiency and the dilution of the extracted

compound hardly constitute an immutable law of thermodynamics. Applying the same logic to the extraction of uranium from seawater would suggest that such efforts could not possibly succeed, considering the fact that the concentration of uranium in seawater is about 3 parts per billion. However, there have been largely successful efforts both at Massachusetts Institute of Technology (Cambridge, MA) and in Japan (59–61).

The low end of the range of cost estimates of air capture rests on two observations. First, the concentration of CO₂ in air is high enough to allow for small collector devices. Second, the binding energy required from an air capture sorbent is only slightly larger than that required for scrubbing CO₂ from the flue stack of a coal-fired power plant. Because there is nearly 0.4 L of CO₂ in every cubic meter of air, it requires little air movement for a collector to contact a large amount of CO₂.

For CO₂ collectors standing passively in the air, the cost of sorbent regeneration dominates the cost of contacting CO₂. The cost of regenerating a fully loaded sorbent depends on its mass and volume and on the binding energy that must be overcome. For a chemical sorbent, the volume or mass per unit of CO₂ bound does not depend on the initial concentration in the gas stream. The minimum required binding strength of the sorbent, however, depends on the concentration of the CO₂ in the gas stream. The relationship between the Gibbs free energy of sorption and minimum concentration is logarithmic (40), resulting in a relatively small difference between the binding energy required for a flue gas scrubber and an air scrubber. Because the energy difference is small and the amount of sorbent that needs to be processed is approximately the same, we expect that the best flue gas scrubbers will have slightly lower CO₂ regeneration costs than air capture devices. Flue gas scrubbers could therefore serve as an approximate lower bound on the cost of air capture. However, this bound is highly uncertain. We argue that a long-term lower bound for sorbent regeneration at \$25/t CO₂ may be plausible, because some estimates for flue gas scrubbers with all costs (not just regeneration) included are already below \$30/t CO₂ (53).

Scaling

Air capture, much like photovoltaic technology, does not necessitate large units of operation. We propose a standardized collector that lends itself to mass manufacturing, resulting in independent units that could be readily transported to their points of use. The economic feasibility of air capture may depend in part on mass manufacturing of small units, e.g., the

size of a shipping container (26). Cost reductions in mass manufacturing of goods and machinery have been far more dramatic than those in the utility sector.

Mass production could allow air capture to become relevant to climate change mitigation. For illustrative purposes, we assume that a mass-produced device could capture 1 t CO₂ per day. In terms of weight and complexity, such a unit would be similar to a car (*SI Text B*). According to the International Association of Motor Vehicle Manufacturers (<http://oica.net/category/production-statistics/>, last visited Oct 2, 2011), the world production of cars and light trucks in 2010 came to 73 million units. A production rate of just 1 million air capture units per year combined with a lifetime of 10 y would result in a steady-state CO₂ capture rate of 3.6 Gt CO₂/y, which is a significant fraction of the world's total output. At a production rate of 10 million units, the asymptotic uptake rate would exceed current CO₂ emissions. None of these production rates would seriously challenge the world's manufacturing capacity, as demonstrated by the magnitude of the automobile industry's output. (It is the interface with the utility sector that could cause severe constraints. On the basis of the current status of the humidity swing resin technology, the electricity demand of capturing 3.6 Gt CO₂/y would require 120 GW of dedicated electric power. Such demand represents about 3 y of growth in China.)

Motivations for Air Capture

i) Compensating for Mobile CO₂ Emissions. Billions of small sources of CO₂ account for between one-third and one-half of society's total CO₂ emissions of ~30 Gt CO₂/y (excluding emissions from deforestation). The emissions associated with the transport sector could be addressed by collecting CO₂ directly from the air while maintaining the current transportation infrastructure. Air capture could provide an alternative or a complement to the electrification of cars and to the exclusive reliance on biofuels in the remaining transportation sectors. We cannot know today which technology will prove the winner, but alternatives are certainly worth investigating.

Without air capture, nonpoint sources of emission will need to be phased out over the next few decades if we want to stabilize the climate (6). Contrary to the point of view expressed in the APS study, we do not consider it sufficient to confine initial efforts to the large point sources. An 80% reduction of CO₂ emissions by 2050 in developed countries cannot be achieved even if all point-source emissions were captured. Reductions outside of concentrated sources are equally important, and air capture provides one option to address

these emissions. The inclusion of aviation emissions in the European cap and trade systems shows that the political debate has already moved past point sources (62).

ii) A Closed Carbon Cycle with Synthetic Fuels.

Liquid hydrocarbon fuels are valuable because of ease of handling and exceptional volumetric energy densities. However, unless air capture is used to close the carbon cycle, the use of carbon-based fuels is not sustainable. In principle, H₂O and CO₂ can provide material feedstock for producing carbonaceous energy carriers such as methanol, synthetic diesel, or gasoline or more exotic alternatives like dimethyl-ether, using energy from renewable or nuclear energy sources.

Liquid carbon-based fuels from sunshine or other forms of renewable energy are technically feasible. At present, it is the cost of renewable electricity that limits their introduction. Unless the cost of air capture remains in the hundreds of dollars per metric ton of CO₂, the cost of electricity will dominate the cost of synthetic fuels. If air capture becomes affordable for CCS applications, it also would be economical for closing the carbon cycle by using synthetic fuels (63).

iii) Reducing the Need for Transporting CO₂.

One of the main challenges of large-scale CCS deployment is the construction of an extensive CO₂ pipeline network, because CO₂ would need to be carried from the place where it is captured to the storage site. Building pipelines would be expensive, necessitate difficult to obtain legal permissions, and face risks and environmental issues as well as public scrutiny where pipelines cross populated or protected areas. International geopolitics may interfere when pipelines cross borders, and physical obstacles may limit transport over mountains or bodies of water.

By contrast, air capture can operate at the storage site and would eliminate the need for transporting CO₂ over long distances. There is no need for a CO₂ pipeline infrastructure to develop and to match sources to sinks of CO₂. Many large point sources are located in places where no safe, large storage options are available. Conversely, some of the most suitable disposal sites are far from sources. Air capture by taking advantage of remote locations can greatly reduce "NIMBY" effects, which have been observed already (64).

iv) Compensating for CO₂ Leakage from Geologic Storage Sites.

CO₂ leakage, even with rates at the percentage level, may not render CCS economically unattractive (10, 65), but it negatively affects the scope of achievable reductions. It also constrains the maximum storage capacity, as the

average leakage rate sets a minimum emission level. Without the option of recapturing leaked CO₂, the cost of irreversible leakage could be very high unless it is discounted. The application of a high discount rate to irreversible leakage is at best questionable (66).

According to the IPCC, there is high confidence that safe storage sites are abundant (52). Good geological storage sites should safely retain CO₂ over thousands of years (53), but probabilities of failure, however small, should never be entirely excluded. Air capture cannot prevent the damages associated with catastrophic gas loss [e.g., Lake Nyos in 1986 (67) or Hutchinson, Kansas in 2001 (68)] but provides a means of recapturing leaked CO₂, thereby insuring against gradual leaks. The ability to recapture makes it possible to hold the operator of the reservoir responsible by effectively monetizing the climate risk of leakage, and thus, forces the operator to consider the economic consequences of ignoring the potential for a leak. The owner of a storage reservoir that leaks CO₂ into the atmosphere should be considered an emitter who has to make compensation for the CO₂ lost. Without a means of recapturing the leaked CO₂, CCS deployment could be hindered as leaks are not entirely preventable and, in the future, may not fit within the remaining CO₂ budget. The price of air capture could thus affect the price of geological storage, perhaps as part of a mandatory leakage insurance policy (11). It must be stressed that, even at low cost, air capture would be much more effective as insurance against an accident with low but nonzero probability than as a built-in component of an inherently leaky storage system.

Reducing the potential cost of leakage also opens the door to more accurate accounting of CO₂ storage (39). Accounting and monitoring of CO₂ may become critical in establishing public acceptance of large-scale deployment of CCS. Accounting methods do slightly increase the risk of leakage, however, due to intrusive sampling of the reservoir. Air capture would provide insurance to manage the risk of sampling-enhanced monitoring of geologic CO₂ storage. Increased accountability of the operator in turn would encourage better reservoir choices.

v) Long-Term Considerations. Air capture on a large scale could create net negative emissions, reducing excess CO₂ stored in the atmosphere, oceans, and terrestrial biomass. Cao and Caldeira's model calculation (69) indicates that the return of most of the excess CO₂ stored in the ocean and on land will occur rapidly. The possibility that CO₂ levels could actively be lowered raises the concern that it might be

used to justify inaction. A wait-and-see attitude is ill-conceived for several reasons. First, one should not rely on a technology that has not been demonstrated at scale and at an affordable price. Second, the impact of excessive greenhouse gas concentrations is not immediate. Thus, it could be too late for action by the time the scope of the damage becomes clear. It is necessary to act, even in the presence of uncertainty. Third, some damage may be irreversible and inaction will increase the risk of such damage. Fourth, the available time is short and actions are necessary on all fronts. Carbon mitigation costs will not come down until action is taken.

Finally, the ability of a technology to support an overshoot scenario provides an immediate advantage because the world is probably already in an overshoot scenario. The optimal CO₂ stabilization point could well be lower than the current CO₂ concentration in the air (70). The CO₂ level that the world will reach with best effort will be higher than can or should be accepted. And even if most of the world agrees upon a comprehensive system of greenhouse gas regulation, rogue nations (i.e., the North Koreans of the future) will always create a risk of unpredictable emissions. Hence, it is important to develop technologies that can reduce the CO₂ concentration in the air.

To quantify the role that air capture could play in atmospheric CO₂ reductions, consider two simple scenarios that represent extreme cases. For the first scenario, average emissions between today and 2050 will have raised atmospheric levels by just 1.5 ppmv/y, bringing the concentration level to ~450 ppmv. In this scenario, energy consumption by 2050 may have doubled, but improved generation efficiency and a shift in energy mix will have reduced fossil fuel consumption by one third, leaving 20 Gt CO₂/y to be dealt with by CCS and air capture which, in this scenario, will remove 10 Gt CO₂/y from the atmosphere. By assumption, the price of captured CO₂ has become affordable, i.e., less than \$50/t CO₂. Worldwide, annual air capture costs would add up to as much as \$500 billion. For comparison, at \$100/bbl, the annual cost of US oil consumption (19.5 million bbls/d) amounts to \$712 billion.

If stabilization at 450 ppmv is sufficient, further ramping up of air capture would not be necessary. However, if moving to lower concentrations were to prove necessary and governments were to become reliable purchasers of additional CO₂ reductions, the air capture industry could grow rapidly. Using an aggressive growth rate of 15%/y, which is still small compared with growth seen in wind [based on

EIA data, wind energy in the United States grew by an average of 39%/y between 2004 and 2008 (http://www.eia.gov/cneaf/solar.renewables/page/trends/table1_12.pdf, last accessed, Oct 2, 2011)] and solar energy [based on EIA data, photovoltaic module production grew an average of 34% per year in the United States between 2000 and 2009 (<http://www.eia.gov/cneaf/solar.renewables/page/solarphotv/solarpv.html>, accessed Oct 2, 2011)], CO₂ capture rates could quadruple in a decade. Thus, a reduction rate of CO₂ in the air comparable to today's emission rate is feasible within a decade provided there is the perception of urgency and the political will to solve the problem. If it is decided that 350 ppmv is the safe target, it would take about five decades to return to those levels.

In a second scenario, we assume that the world ignores CO₂ emissions and fails to develop CO₂ capture technologies, leading to an average increase of 3 ppmv/y through 2050. Such a rate of increase would result in a CO₂ concentration of

510 ppmv and an annual rate of increase in atmospheric CO₂ concentration of 4 ppmv in 2050. If this emission rate persists on average for another 50 y, CO₂ levels would exceed 700 ppmv by 2100. Stabilizing at 350 ppmv by 2150 would require an annual reduction of 7 ppmv. This scale is enormous, and it would be much more difficult for air capture to help solve this problem than the problem in the first scenario.

Although these scenarios are simplistic, the message is clear: A reduction by 100 ppmv appears plausible, whereas a reduction by many hundreds of ppmv is likely to be prohibitively expensive, even if one assumes cost-effective implementations of air capture technology. This example demonstrates that the possibility of affordable air capture technology does not provide any justification for a delay-and-overshoot global strategy.

Conclusions

Air capture research is still in its infancy and the practicality of large-scale de-

ployment needs to be further explored. The inability to produce accurate cost estimates for a nascent technology, however, should not be considered a reason for withholding support. Indeed, air capture is clearly feasible, and there are several lines of argument that suggest that its cost could well come down to a level that would make air capture economically interesting. Air capture would provide a different approach to reducing CO₂ concentrations in the atmosphere. There is abundant R&D to be undertaken with regard to the various possible materials, components, and workings of air capture technology. Given the enormity of the global climate challenge, we think this R&D needs to be scaled up urgently.

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- Matthews HD (2006) Emissions targets for CO₂ stabilization as modified by carbon cycle feedbacks. *Tellus: Ser B* 58:591–602.
- Archer D (2009) *The Long Thaw: How Humans Are Changing the Next 100,000 Years of Earth's Climate* (Princeton Univ Press, Princeton, Oxford).
- IPCC (2007) *Intergovernmental Panel on Climate Change. Climate Change 2007, Fourth Assessment Report, Synthesis Report* (Cambridge Univ Press, New York).
- Kharecha PA, Hansen JE (2008) Implications of 'peak oil' for atmospheric CO₂ and climate. *Global Biogeochem Cycles* 22:GB3012.
- Lackner KS (2006) Carbon sequestration. *The Future of Oil in the 21st Century*, ed Mabro R (Oxford Univ Press, New York) pp 241–281.
- Matthews HD, Caldeira K (2008) Stabilizing climate requires near-zero emissions. *Geophys Res Lett* 35:1.
- Solomon S, Plattner GK, Knutti R, Friedlingstein P (2009) Irreversible climate change due to carbon dioxide emissions. *Proc Natl Acad Sci USA* 106:1704–1709.
- Lackner KS, Sachs JD (2005) A robust strategy for sustainable energy. *Brookings Pap Econ Act* 2:215–284.
- Tavoni M, van der Zwaan BCC (2011) Nuclear versus coal plus CCS: A comparison of two competitive base-load climate control options. *Environ Model Assess* 16: 431–440.
- Ha-Duong M, Keith DW (2003) Carbon storage: The economic efficiency of storing CO₂ in leaky reservoirs. *Clean Technol Environ Policy* 5:181–189.
- Lackner KS, Brennan S (2009) Envisioning carbon capture and storage: Expanded possibilities due to air capture, leakage insurance, and C-14 monitoring. *Clim Change* 96:357–378.
- Sarewitz D, Nelson R (2008) Three rules for technological fixes. *Nature* 456:871–872.
- Barrett S (2010) *Climate Treaties and Backstop Technologies* (Center for Economic Studies and Ifo Institute for Economic Research, Munich), Working Paper No. 3003.
- Weitzman M (2009) On modeling and interpreting the economics of climate change. *Rev Econ Stat* 91:1–19.
- Carey R, Gomezplata A, Sarich A (1983) An overview into submarine CO₂ scrubber development. *Ocean Eng* 10:227–233.
- Dallbauman L, Finn J (1999) Adsorption processes in spacecraft environmental control and life support systems. *Adsorption and Its Applications in Industry and Environmental Protection: Applications in Environ-*
- mental Protection*, ed Dabrowski A (Elsevier, New York), Vol II, pp 455–471.
- Castle WF (2007) Fifty-years' development of cryogenic liquefaction processes. *Cryogenic Engineering: Fifty years of progress*, eds Timmerhaus KD, Reed RP, Rizzuto C (Springer, New York), pp 146–160.
- Payne JW, Dodge BF (1932) Rate of absorption of carbon dioxide in water and in alkaline media. *Ind Eng Chem* 24:630–637.
- Spector NA, Dodge BF (1946) Removal of carbon dioxide from atmospheric air. *Trans Am Inst Chem Eng* 42: 827–848.
- Astarita G (1967) *Mass Transfer with Chemical Reactions* (Elsevier, New York).
- Lackner KS, Ziock HJ, Grimes P (1999b) Carbon dioxide extraction from air: Is it an option? *Proceedings of the 24th International Conference on Coal Utilization & Fuel Systems* (Clearwater, FL), ed Sakkestad BA (Coal & Slurry Technology Association, Washington, DC), 885–896.
- Lackner KS, Grimes P, Ziock HK (2001) Capturing carbon dioxide from air. *Proceedings of the First International Sequestration Conference* (Department of Energy National Energy Technology Laboratory, Alexandria, VA), Session 7B: Capture, V: Adsorption Studies, pp 1–15.
- Stolaroff JK, Keith DW, Lowry GV (2008) Carbon dioxide capture from atmospheric air using sodium hydroxide spray. *Environ Sci Technol* 42:2728–2735.
- Mahmoudkhani M, Heidel K, Ferreira J, Keith D, Cherry R (2009) Low energy packed tower and caustic recovery for direct capture of CO₂ from air. *Energy Procedia* 1:1535–1542.
- Zeman FS, Lackner KS (2004) Capturing carbon dioxide directly from the atmosphere. *World Resour Rev* 16(2): 157–172.
- Lackner KS (2009) Capture of carbon dioxide from ambient air. *Eur Phys J Spec Top* 176:93–106.
- Li W, et al. (2010) Steam-stripping for regeneration of supported amine-based CO₂ adsorbents. *ChemSusChem* 3:899–903.
- Choi S, Drese JH, Eisenberger PM, Jones CW (2011) Application of amine-tethered solid sorbents for direct CO₂ capture from the ambient air. *Environ Sci Technol* 45:2420–2427.
- Eisaman MD, et al. (2011) CO₂ separation using bipolar membrane electrodialysis. *Energy Environ Sci* 4: 1319–1328.
- Ge J, Trachtenberg M, McGregor M, Cowan R (2001) Enzyme-based facilitated transport: Use of vacuum induced sweep for enhanced CO₂ capture. *Proceedings of the 31st International Conference on Environmental Systems*, Orlando, Florida.
- Rau G (2009) Electrochemical CO₂ capture and storage with hydrogen generation. *Energy Procedia* 1:823–828.
- Thomson AM, Izaurralde RC, Smith SJ, Clarke LE (2008) Integrated estimates of global terrestrial carbon sequestration. *Glob Environ Change* 18(1):192–203.
- Lehmann J, Gaunt J, Rondon M (2006) Bio-char sequestration in terrestrial ecosystems—a review. *Mitigation and Adaptation Strategies for Global Change* (Springer, Dordrecht), Vol 11, pp 395–419.
- Coale KH, et al. (2004) Southern Ocean iron enrichment experiment: Carbon cycling in high- and low-Si waters. *Science* 304:408–414.
- Schuiling R, Krijgsman P (2006) Enhanced weathering: An effective and cheap tool to sequester CO₂. *Clim Change* 74:349–354.
- Lackner KS, Grimes P, Ziock HJ (1999a) "Carbon Dioxide Extraction from Air?", LAUR-99-5113 (Los Alamos National Laboratory, Los Alamos, NM).
- Stucki S, Schuler A, Constantinescu M (1995) Coupled CO₂ recovery from the atmosphere and water electrolysis: Feasibility of a new process for hydrogen storage. *Int J Hydrogen Energy* 20:653–663.
- Baciocchi R, Storti G, Mazzotti M (2006) Process design and energy requirements for the capture of carbon dioxide from air. *Chem Eng Process* 45:1047–1058.
- Lackner KS, Park A-H, Matter JM (2009) Methods and devices for tagging large industrial CO₂ streams of fossil carbon with parts per trillion carbon-14 for accurate inventory accounting in geological storage of carbon dioxide. Invention Report, Patent Appl 61/177,145, submitted to Columbia Patent Office on May 11.
- American Physical Society (2011) *Direct Air Capture of CO₂ with Chemicals: A Technology Assessment for the APS Panel on Public Affairs* (American Physical Society, Washington, DC).
- Lackner KS (2010) Washing carbon out of the air. *Sci Am* 302:66–71.
- Keith DW (2009) Why capture CO₂ from the atmosphere? *Science* 325:1654–1655.
- The Royal Society (2009) *Geoengineering the Climate: Science, Governance, and Uncertainty*. (The Royal Society, London).
- Shenhar AJ (2001) One size does not fit all projects: Exploring classical contingency domains. *Manage Sci* 47:394–414.

45. Odeck J (2004) Cost overruns in road construction: What are their sizes and determinants? *Transp Policy* 11(1):43–53.
46. Peck M, Scherer F (1962) *The Weapons Acquisition Process: An Economic Analysis* (Division of Research, Graduate School of Business Administration, Harvard University, Cambridge, MA).
47. Lundstrom M (2003) Applied physics. Moore's law forever? *Science* 299:210–211.
48. Nemet G (2006) Beyond the learning curve: Factors influencing cost reductions in photovoltaics. *Energy Policy* 34:3218–3232.
49. Ferioli F, Schoots K, van der Zwaan BCC (2009) Use and limitations of learning curves for energy technology policy: A component-learning hypothesis. *Energy Policy* 37:2525–2535.
50. Rubin ES, Yeh S, Hounshell DA, Taylor MR (2004) Experience curves for power plant emission control technologies. *Int J Energy Technol Policy* 2(1–2):52–69.
51. Ferioli F, van der Zwaan BCC (2009) Learning in times of change: A dynamic explanation for technological progress. *Environ Sci Technol* 43:4002–4008.
52. U.S. Department of Energy National Energy Technology Laboratory (NETL) News Release (July 21, 2000) Energy department launches thirteen new research projects to capture and store greenhouse gases. (US Department of Energy NETL, Washington, DC).
53. IPCC (2005) *Intergovernmental Panel on Climate Change, Special Report on Carbon Dioxide Capture and Storage* (Cambridge Univ Press, New York).
54. Suresh B, Schlag S, Ishikawa Y (2007) *CEH Marketing Research Report: Carbon Dioxide*, Chemical Economics Handbook (SRI Consulting, Menlo Park, CA).
55. Ravagnani AG, Ligerio E, Suslick S (2009) CO₂ sequestration through enhanced oil recovery in a mature oil field. *J Petrol Sci Eng* 65(3–4):129–138.
56. Steeneveldt R, Berger B, Torp T (2006) CO₂ capture and storage: Closing the knowing-doing gap. *Chem Eng Res Des* 84:739–763.
57. Keith DW, Ha-Duong M, Stolaroff JK (2006) Climate strategy with CO₂ capture from air. *Clim Change* 74(1–3):17–45.
58. House K, et al. (2011) Economic and energetic analysis of capture CO₂ from ambient air. *Proc Natl Acad Sci USA* 108:20428–20433.
59. Driscoll MJ, et al. (1981) *Systems Studies on the Extraction of Uranium from Seawater* (Massachusetts Institute of Technology, Energy Laboratory, Cambridge, MA).
60. Kabay N, Egawa H (1994) Chelating polymers for recovery of uranium from seawater. *Sep Sci Technol* 29(1):135–150.
61. Rao L (2011) *Recent International R&D Activities in the Extraction from Uranium from Seawater* (Lawrence Berkeley National Laboratory, Berkeley, CA).
62. Graves C, Ebbesen SD, Mogensen M, Lackner KS (2011) Sustainable hydrocarbon fuels by recycling CO₂ and H₂O with renewable or nuclear energy. *Renew Sustain Energy Rev* 15(1):1–23.
63. Meltzer J (2012) Climate change and trade: The EU aviation directive and the WTO. *J Int Econ Law* 15(1):111–156.
64. Van Noorden R (2010) Carbon sequestration: Buried trouble. *Nature* 463:871–873.
65. van der Zwaan BCC, Gerlagh R (2009) Economics of geological CO₂ storage and leakage. *Clim Change* 93: 285–309.
66. Stern N (2006) *Stern Review on the Economics of Climate Change*. Cabinet Office–HMS Treasury (Cambridge Univ Press, Cambridge, MA).
67. Kling GW, et al. (1987) The 1986 Lake Nyos gas disaster in Cameroon, West Africa. *Science* 236:169–175.
68. Allison ML (2001) Hutchinson, Kansas: A geologic detective story. *Geotimes* 46(10):14–18.
69. Cao L, Caldeira K (2010) Atmospheric carbon dioxide removal: Long term consequences and commitment. *Environ Res Lett* 5(2).
70. Hansen J, et al. (2008) Target atmospheric CO₂: Where should humanity aim? *Open Atmos Sci J* 2:217–231.