

Geological Carbon Dioxide Sequestration

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A key means of reducing greenhouse gas emissions from fossil fuels is to separate and concentrate CO₂ from large point sources and inject it underground. The injection process, so-called “geological carbon sequestration”, uses off-the-shelf technology from the hydrocarbon industry and can be deployed at a useful scale. Widespread deployment will require a greater understanding of processes that trap CO₂ underground, improved means of monitoring the injection stream, and a small number of large-scale experiments in settings with the most important representative geology. If successful, geological sequestration could greatly reduce greenhouse gas emissions while we continue to benefit from fossil fuels until true alternatives emerge.

KEYWORDS: carbon sequestration, carbon management, carbon storage, carbon dioxide, greenhouse gas abatement

INTRODUCTION

In the global energy system, fossil fuels currently account for 85% of all energy supply and more than 98% of transportation fuel. Given the expectations of enormous energy demand growth (a doubling or tripling in 50 years) and the low cost and high availability of fossil fuels, it is widely believed that fossil fuel consumption will continue at a large scale through the next century. However, combustion of these fuels releases enormous volumes of greenhouse gases, chiefly CO₂. Every year, 25 gigatonnes (Gt) of man-made CO₂ enter the atmosphere, nearly all of it from fossil fuel combustion. Concerns about the risks of greenhouse gas emissions and attendant climate change impacts are increasing, and it appears that these risks are substantial. This has prompted renewed focus on reducing emissions through improved efficiency, renewable energy supplies, and nuclear fission. Even if future demand growth was met by carbon-free sources of energy, there would be no reduction in emissions because fossil energy use will continue, especially in developing nations with rapid economic growth.

In this context, carbon dioxide sequestration has emerged as a key technology pathway to reducing greenhouse gas (GHG) emissions (IPCC 2005). Sequestration is the long-term isolation of carbon dioxide from the atmosphere through physical, chemical, biological, or engineered processes. Geological carbon sequestration (GCS) appears to be among the most promising large-scale approaches to major emission reduction in the next 20–50 years (MIT 2007) for several reasons:

1. The potential storage capacities are enormous. Formal estimates of global storage potential vary substantially but are likely to be between 3000 and 10,000 gigatonnes of

CO₂. This can be compared with the annual global anthropogenic flux of 25 gigatonnes.

2. GCS uses existing technology. Carbon dioxide has been separated from large point sources for nearly 100 years and has been injected underground for over 30 years.
3. GCS is actionable. In the U.S., Canada, and many other industrial countries, large CO₂ sources, like power plants and refineries, lie near prospective storage sites. These plants could be retrofitted today and injection begun.

However, in order to achieve substantial GHG reductions, geological storage needs to be deployed at a large scale. For example, a 1 GtC/yr (3.6 GtCO₂/yr) abatement requires GCS from 600 large pulverized-coal plants (~1000 MW each) or 3600 injection projects at the scale of Statoil’s Sleipner project (Pacala and Socolow 2004), during which one million tonnes of CO₂ have been injected every year for ten years (Arts et al. 2004).

STORAGE MECHANISMS

A number of geological reservoirs appear to have the potential to store many hundreds to thousands of gigatonnes of CO₂ (IPCC 2005). The most promising reservoirs are porous and permeable rock bodies, generally at depths of 800 m or greater. Given conventional geotherms, the pressure–temperature constraints would make CO₂ a supercritical phase at these depths (Bachu 2000). In this state, CO₂ acts like a liquid, with a density less than that of brine, a fairly low solubility in water, and a viscosity less than that of oil (Fig. 1). The high densities are critical to successful storage such that a large volume of CO₂ can be injected into a limited pore volume.

Three classes of target reservoirs are capable of sequestering large volumes of CO₂:

- *Saline formations.* These contain brine in their pore volumes, commonly with a salinity greater than 10,000 ppm.
- *Depleted oil and gas fields.* A combination of water and hydrocarbons fills their pore volumes, and in some cases, economic gains can be achieved through enhanced oil or gas recovery. Injection of substantial quantities of both natural and anthropogenic CO₂ already occurs in the U.S.
- *Deep coal seams.* Often called unmineable coal seams, these are composed of organic minerals and contain brines and gases in their pore and fracture volumes.

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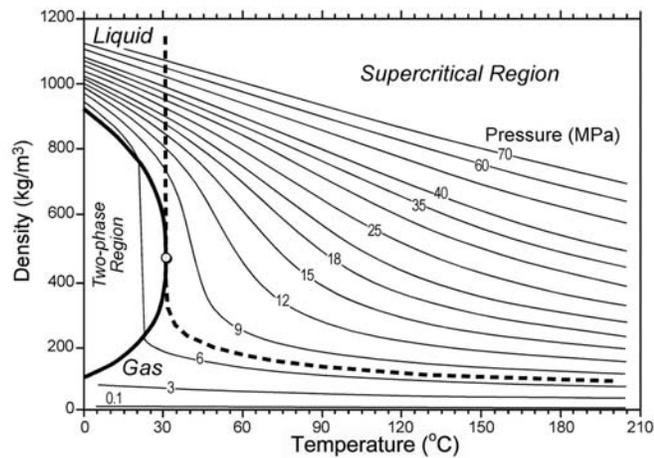
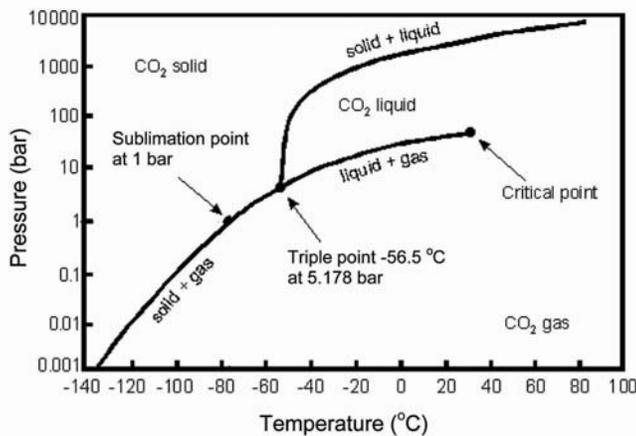


FIGURE 1 Physical properties of CO₂ with respect to temperature and pressure. (Left) Phase diagram for CO₂. Note that the pressure axis is logarithmic. In most cases, geological sequestration will take place at temperatures and pressures above the critical point. (Right) Graph of CO₂ density versus temperature at given pressures. For most geological settings, CO₂ would be sequestered at densities of 600–800 kg/m³. AFTER IPCC (2005) AND BACHU (2003)

Because of their large storage potential and broad distribution, it is likely that most geological sequestration will occur in saline formations. However, initial projects will probably occur in depleted oil and gas fields, accompanying enhanced oil recovery, due to the great abundance and quality of subsurface data and the potential for economic return. Although there remains some economic potential for enhanced coal bed methane recovery, initial economic assessments do not appear promising, and substantial technical hurdles remain in the way of obtaining the desired benefits (IPCC 2005).

For these three reservoir classes, CO₂ storage mechanisms are reasonably well defined and understood. CO₂ sequestration targets will require physical barriers to CO₂ migration towards the surface. These barriers will commonly take the form of impermeable layers (e.g. shales, evaporites) overlying the reservoir, although they may also be dynamic in the form of regional hydrodynamic flow. Physical trapping allows for very high CO₂ pore volumes, in excess of 80%, and acts immediately to limit CO₂ vertical flow. At the pore scale, capillary forces will immobilize a substantial fraction of a CO₂ bubble, commonly measured to be between 5 and 25% of the CO₂-bearing pore volume. The fraction is determined by the interfacial tension angle, the wettability angle of the pore surfaces, and the effective diameter of the pore throat (e.g. Gladkikh and Bryant 2003). This process acts immediately to trap CO₂ as a residual phase in the pores and continues to act during and after plume flow. Once in the pores, over a period of tens to hundreds of years, the CO₂ will dissolve in other pore fluids, including hydrocarbon species (oil and gas) and brines. The CO₂ is fixed indefinitely in the pores, unless other processes intervene. Over longer time scales (hundreds to thousands of years), the dissolved CO₂ may react with minerals in the rock volume, precipitating the CO₂ as new carbonate minerals. In the case of organic mineral frameworks such as coals, the CO₂ will physically adsorb onto the rock surface, sometimes displacing other gases (e.g. methane, nitrogen).

Although substantial work remains to characterize and quantify these mechanisms (see below), they are understood well enough today to develop reliable preliminary

estimates of the percentage of CO₂ that can be stored over some period of time. These estimates are based in part on decades of studies in analogous systems, including hydrocarbon systems, natural gas storage operations, hazardous waste injection, and CO₂-enhanced oil recovery. In the case of enhanced oil recovery, CO₂ has been injected underground for over 30 years, with no demonstrated examples of substantial leakage. In addition, numerous CO₂ accumulations occur naturally around the world, including the CO₂ domes of the central Rocky Mountains (Stevens et al. 2001), high-CO₂ gas fields (Brennan et al. 2004), and the carbogaseous (charged with carbon dioxide) provinces of Europe (e.g. Czernichowski-Lauriol et al. 2003). In these regions, large volumes of CO₂ have been stored safely for tens of millions of years.

This information provides the basis for qualitative risk estimation. Specifically, it is very likely that more than 99.9% of injected CO₂ will be retained for 100 years, and likely more than 99% for 1000 years (IPCC 2005). In other words, the crust is generally well configured to store CO₂.

POTENTIAL RISKS

Since supercritical CO₂ is buoyant at the relevant crustal pressures and temperatures, it will seek the Earth's surface in most settings. A large CO₂ accumulation would exert forces on the reservoir, cap rock, faults, and wells. CO₂ must also be injected at pressures above reservoir pressures, creating a pressure transient during and after injection. In addition, dissolved CO₂ forms carbonic acid, which can alter rock and well-bore properties and composition. Therefore, despite confidence in the storage mechanisms discussed above, the possibility of leakage from storage sites remains.

These risks were recently highlighted by geochemical analysis and laboratory experiments carried out at a pilot injection in South Liberty, Texas (Hovorka et al. 2006). Kharaka et al. (2006) observed rapid dissolution of some minerals, chiefly carbonate, oxide, and hydroxide minerals. Although this population represented a small fraction of the rock volume (<2%), the rapid kinetics of dissolution liberated iron, manganese, calcium, and organic carbon molecules into the brine. This raised questions about the possible environmental effects of GCS and about the long-term fate of injected CO₂.

In order to evaluate the possibility of catastrophic leakage, Pruess et al. (2006) attempted to determine the conditions for positive feedbacks that might lead to runaway CO₂ release. Importantly, they found none. Rather, CO₂ interac-

tion with the crust generally produces negative feedbacks; for example, acidic dissolution of carbonate minerals increases pH and reduces reactivity of brines. In some cases, it appears that storage mechanisms are self-reinforcing. The geochemical modeling of Johnson et al. (2004) suggests that CO₂ reactions with some minerals (e.g. chlorite) reduce the porosity and permeability of shale, making it a better physical seal. This behavior was observed by Watson et al. (2005) in shales overlying natural CO₂ accumulations. These studies suggest that while Kharaka et al. (2006) may have discovered a new element of risk, that element does not appear to represent a major concern to CO₂ storage.

The geomechanical response to CO₂ injection may still cause concerns. In a parallel set of studies, Johnson et al. (2005) simulated large pressure excursions from CO₂ injection. They concluded that under certain conditions, such excursions lead to fracture dilation, with some seepage of CO₂ into overlying units. In the case of most cap rocks, which have both fractures and reactive minerals (e.g. chlorite), this creates a competing rates problem between dilation of fracture and precipitation of reactive minerals in fracture voids. In this system, fracture closure or dilation is sensitive to CO₂ diffusion distance and reaction rate. In addition, the pressure transient from injection could lead to fault-slip-induced fluid migration (e.g. Wiprut and Zoback 2002). While it is generally possible to predict the conditions under which this might occur (e.g. Chiaramonte et al. 2006), effective storage will require proper system calibration and injection management.

Additional work will reduce the uncertainties associated with long-term sequestration. Nonetheless, it should be noted that today's knowledge gaps do not cast doubt on the fundamental likelihood that GCS can be deployed both widely and successfully. Rather, they suggest a science program that can quickly constrain uncertainties and point to the aspects of risk that require mitigation or technical development.

MONITORING AND VERIFICATION

Once injection begins, a program for monitoring and verification of CO₂ distribution is required in order to:

- Understand key features, effects, and processes needed for risk assessment
- Manage the injection process
- Delineate and identify leakage risk and surface escape
- Provide early warnings of failure near the reservoir
- Verify storage for accounting and crediting

For these reasons, monitoring and verification is a chief focus of many research efforts, including programs by the U.S. Department of Energy (US DOE 2006) and the European Union (CO₂ ReMoVe), and the International Energy Agency's Greenhouse R&D Programme.

Early research and demonstration projects will require more involved monitoring systems than future commercial projects because they will be attempting to establish the scientific basis for geological sequestration. Today there are three well-established large-scale injection projects with ambitious scientific programs that include monitoring: Sleipner in the Norwegian North Sea (Arts et al. 2004), Weyburn in Saskatchewan (Wilson and Monea 2004), and In Salah in Algeria (Riddiford et al. 2005).

These projects are being carried out in substantively different geological settings, and in each project, roughly 1 million tonnes CO₂ are injected per year (FIG. 2). At Sleipner,

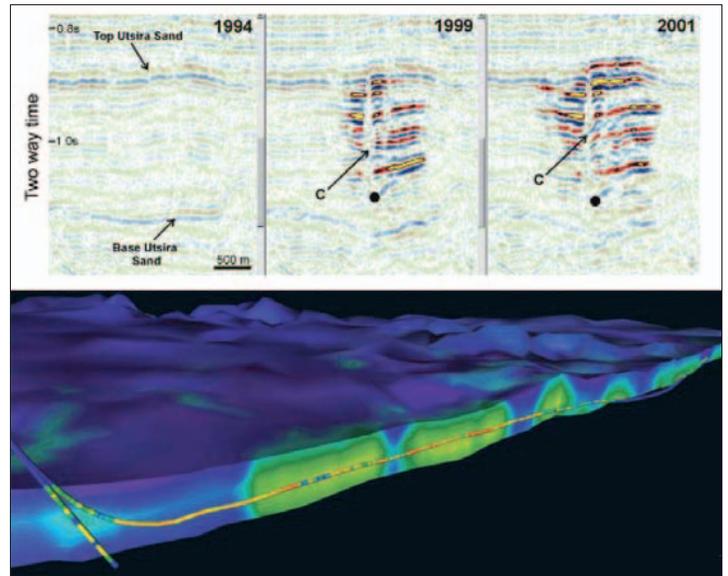


FIGURE 2 Data from two large injection projects. **(Top)** Time-lapse reflection seismic images from Sleipner collected before injection (1994) and afterwards. The bright colors show impedance contrasts generated by free-phase CO₂. **(Bottom)** Simulation of injection of CO₂ in a reservoir at the Kretchba field, Algeria. The thin lines are wells that penetrate the target formation; the colors show relative porosity. Images courtesy of Statoil and BP

CO₂ enters a 100 m thick, very porous and permeable, fairly homogeneous, brine-bearing, deep-water sandstone 800 m below the North Sea's floor (Arts et al. 2004). At Weyburn, CO₂ is injected into a 10 m thick, highly heterogeneous, fractured, oil-bearing carbonate shelf succession ~1300 m below southern Saskatchewan (Wilson and Monea 2004). At In Salah, CO₂ enters a 10 m thick, low-porosity and low-permeability, fractured, heterogeneous, brine-bearing fluvial-tidal network ~2100 m below central Algeria. This range in geological setting, pore structure, crustal velocity, and depth provides insight into the utility of various monitoring approaches in the field (MIT 2007).

Perhaps surprisingly, in the context of these and other research efforts, there has been little discussion about what are the most important parameters to measure and in what context (research/pilot versus commercial). Rather, the literature has focused on the current ensemble of tools and their costs. In part due to the success at Sleipner, 4-D seismic (space plus time-lapse) surveys have emerged as the standard for comparison, with 4-D surveys deployed at Weyburn and likely to be deployed at In Salah. This technology excels at delineating the boundaries of a free-phase CO₂ plume and can detect low saturation of conjoined free-phase bubbles that might be an indicator of leakage. Results from these 4-D seismic surveys establish the basis for long-term effectiveness of geological sequestration.

However, time-lapse seismic surveys do not measure all the relevant parameters, and the technique has limits in some geological settings. Key parameters for research and validation of CO₂ behavior and fate involve both direct detection of CO₂ and detection through proxy data sets. Temperature, pressure, pH, resistivity, EM potential, crustal deformation, and changes to stress azimuth and magnitude all provide insight into the distribution, saturation, and mechanical response to CO₂ injection (MIT 2007). Interpretation of these data requires greater empirical and experimental control, including petrophysical studies of CO₂-rock-brine-hydrocarbon systems.

LARGE-VOLUME EXPERIMENTS

As mentioned previously, achievement of substantial CO₂ emissions reductions through GCS will require hundreds to thousands of large-volume injection facilities distributed around the world. Each existing large project and some small projects (e.g. the Frio Brine Pilot; Hovorka et al. 2006) have provided some demonstration of effectiveness, monitoring technologies, and operational economics. Importantly, each existing large project also has revealed an important aspect of the geology that was not previously known or in some cases incorrectly characterized. For example, at Sleipner, the importance of small flow heterogeneities was not anticipated but was clearly seen (Arts et al. 2004). At Weyburn, CO₂ migrated in unexpected ways along secondary fractures (Wilson and Monea 2004).

Such features would not have been revealed through a small-scale (<100,000 tonnes CO₂/yr) experiment because geological thresholds in the Earth's crust are sensitive to the magnitude and rate of excursions (e.g. pressure build-up, changes in pH). Unless those thresholds are reached, no effect occurs. For example, an attempt to induce earthquakes in the Rangely field of northwestern Colorado required massive injection of fluids to overcome the fracture yield criteria of the system (Raleigh et al. 1976). Because the Earth's crust is a complex, heterogeneous, nonlinear system, field-based demonstrations are required to understand the likely range of crustal responses, including those that might allow CO₂ to escape from reservoirs. In addition, many important effects (e.g. cement or casing corrosion, induced seismicity, mechanical failure of wells and cap rocks) can only be understood in the context of large demonstrations and large experiments. Despite a substantial scientific effort at Weyburn and Sleipner, many parameters that could have been measured to circumscribe the most compelling scientific questions have not yet been collected. These include distribution of CO₂ saturation, stress changes, and well-bore leakage detection.

Substantial GHG abatement requires a geographically diverse portfolio of large injections. Importantly, the three largest demonstrations are in geologically distinct settings, which helps to demonstrate that GCS may have broad application. However, key potential sites have not yet been tested at scale (e.g. Cenozoic basins of eastern China, Himalayan foreland, U.S. Gulf Coast). In each active demonstration project to date, current measurement, monitoring, and verification technologies are not formally integrated and may not be sufficient to document either injection volumes or leakage risk and activity within the necessary full range of important geological or surface geographic conditions.

SCIENTIFIC GAPS

The current set of large-scale experiments, studies of natural and man-made analogs, and integrated laboratory and numerical experiments provide confidence that we will soon be able to deploy GCS at scale. That is *not* to say that the scientific work is complete. There remain questions about the long-term fate of CO₂, the cost of operation, and storage effectiveness. While there are many topics of interest, a few of high importance and scientific value are discussed below.

CO₂ Dissolution and Precipitation Kinetics

The rate at which CO₂ dissolves in brines of varying composition, temperature, pressure, and mixing degree greatly affects the long-term trapping mechanisms (i.e. the formation of carbonic acid, bicarbonate, and new minerals). These issues in turn affect other important concerns, such as the configuration and infrastructure of storage reservoir engineering (e.g. Keith et al. 2005) or the long-term fate of CO₂

(Ennis-King and Paterson 2003). Although there has been some work on the controls of dissolution rate (e.g. interfacial effects; Yang et al. 2005) more could be done.

Similarly, knowledge of CO₂-brine-mineral dissolution and precipitation kinetics is limited. Recent years have seen many experimental studies on individual minerals or classes of minerals (e.g. Carroll and Knauss 2005 and references therein). Still, much remains to be learned about rock systems, including true multiphase chemistry, mineral-CO₂ equations of state, and minerals that may represent only a small volume of the rock but have rapid dissolution kinetics (e.g. metal oxides or hydroxides).

As a subtopic, most GCS work on mineral reaction kinetics has focused on pure CO₂-rock-brine systems. Very little work has been done on gas streams with small concentrations of other gases, in particular SO_x, NO_x, and H₂S. These co-contaminant gases have the potential to dramatically alter the chemical response of a gas-brine-rock system, even if very small amounts of these gases are present (Knauss et al. 2005). Because the capture and separation of trace gases with CO₂ may save capital and operating expenses, investigation of reactions in such systems may prove useful in the near future to determine if mixed-gas systems present additional concerns or risks.

Groundwater

The majority of sequestered CO₂ will be stored in saline formations at depth. CO₂ stored in these formations should have little or no effect on groundwater. However, if CO₂ were to reach the surface along some fast pathway, then CO₂ might enter fresh groundwater systems. It is highly unlikely that the rate or volume of CO₂ would present a problem. However, the results of Kharaka et al. (2006) have raised the possibility that rapid local reactions could release unwanted elements and compounds into groundwater. Again, for most reservoirs this may have no effect. However, some reservoirs have elevated levels of natural arsenic. In such a system, even a small release of CO₂ might result in an increase in local arsenic concentrations that could bring a municipal water supply out of compliance with U.S. Environmental Protection Agency regulations. Similarly, widespread deployment of GCS could potentially displace enough water to create saline groundwater intrusions in contiguous formations.

Wells

Wells almost certainly present the greatest risk to leakage because they are drilled to bring large volumes of fluid quickly to the Earth's surface. In addition, they remove the aspects of the rock volume that prevent buoyant migration. Well casings and cements are susceptible to corrosion from carbonic acid. When wells are adequately plugged and completed, they are likely to trap CO₂ at depth effectively. However, large numbers of orphaned or abandoned wells may not be adequately plugged, completed, or cemented (Ide et al. 2006), and such wells represent potential leak points for CO₂.

One analog site is particularly well suited for study. Crystal Geyser, Utah (Shipton et al. 2005), is a well that penetrated a natural CO₂ accumulation in 1936, was poorly completed, and has erupted CO₂ ever since (Fig. 3). Eruptions are episodic and vary in size. Measurements of individual and sequential eruptions suggest that large events bring tens of tonnes of CO₂ to the surface, with an average daily flux of 40–50 tonnes (Gouveia et al. 2005). During the eruptions, atmospheric concentrations of CO₂ were not recorded at or above dangerous levels. While this style of eruption does not appear to present a substantial risk (Bogen et al. 2006), more study is needed to understand how representative of well leakage this site may be.

Little is known about the probability of escape from a given well, the likelihood of such a well existing within a potential site, or the risk such a well presents in terms of potential leakage volume or consequence. Current approaches involve statistical characterization of many wells and semi-quantitative analysis (Celia et al. 2006), or modeling and simulation of features and processes in well-bore environments (Gerard et al. 2006). Work in understanding the key features of wells (e.g. fracture geometry and character), the chemical response of well components to CO₂ systems, and the evolution of natural and engineered interfaces could provide both better estimates of well-bore integrity risk and potential mitigation and remediation strategies (IPCC 2005).

Mitigation, Mineralization, and Alternative Schema

In general, it is believed that the risks posed by GCS are extremely small and readily managed (IPCC 2005). However, there is room for scientists and engineers to investigate related technologies and engineering schemes to further reduce or altogether eliminate such risks.

The primary risk of CO₂ leakage stems from the forces buoyant CO₂ exerts on the storage environment. However, once CO₂ dissolves into the surrounding fluid, this fluid becomes more dense than one without CO₂, greatly reducing or eliminating such risks. Keith et al. (2005) proposed a method to rapidly mix brine and CO₂ by inducing reservoir circulation. In one simple configuration, the majority of CO₂ would dissolve into the formation brine within 50 years. It is likely that other drilling configurations could work even more rapidly, and the injected brine could be spiked with metal oxides to increase the rate of mineral precipitation in the reservoir, further reducing leakage risk.

Finally, if the risks become truly problematic, one could directly react CO₂ with metal oxide-bearing minerals (IPCC 2005). In most cases, such reactions are exothermic, but the kinetics are very slow. Engineering approaches to this problem (e.g. Lackner 2002) remain expensive, with costs of \$50–100/tonne CO₂ (substantially higher than conventional storage costs). However, improved understanding of mineral reaction kinetics, interfacial reactions, and novel materials could provide insight into ways to reduce these costs. Ultimately, if metal or biological processes could catalyze direct mineralization reactions, they might provide an alternative to conventional storage.

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FIGURE 3 An eruption from Crystal Geyser, Utah. Eruptions commonly reach 5–10 m height. Note: the individuals at the site do not appear at risk. Courtesy of Z. Shipton. Inset: Children playing at Crystal Geyser. Note the original casing sticking out of the active well.

CONCLUSION

Geological carbon dioxide storage appears to be a safe, effective means of achieving a substantial reduction of atmospheric carbon dioxide emissions while maintaining the benefits of fossil fuel use. The present knowledge base suggests that billions of tonnes can be stored safely each year through injection into conventional reservoirs such as saline formations or depleted oil and gas fields. However, accelerated deployment of this technology could greatly benefit from additional basic geoscience research. Many GCS uncertainties involve current understanding of structural geology, stratigraphic heterogeneity, and CO₂-brine mineral kinetics.

In this context, the greatest priority in managing the CO₂ from continued fossil fuel use should be the initiation of commercial-scale experiments that include a substantial science program. Questions that regulators, financiers, operators, and public stakeholders might have about performance, safety, effectiveness, and risk require investigations at commercial scale. Thus, the geoscience community has a unique opportunity to be part of the solution by presenting to the world a major technology pathway to reducing the impacts of global climate change. ■

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