Chapter 13
Carbon Capture and Storage in Geologic Formations

Abstract Carbon dioxide (CO_2) emissions, the most important anthropogenic greenhouse gas (GHG), can be reduced by CO_2 capture and storage (CCS). This strategy is applicable to many large stationary sources including power generation plants, oil and gas refinery, cement production and other industrial sectors generating large point source of CO_2. While the technology for CCS is currently available, significant improvements are needed to enhance confidence in storage security. In 1996, the first CCS project established for the purpose of mitigation of CO_2 emission began injecting CO_2 into deep geological formation in offshore aquifer in the North Sea, Norway. Since that time, science has advanced in areas such as geophysics, chemical engineering, monitoring and verification, and other areas, while also governments have funded demonstration projects at various sizes ranging from small-scale proof of concept to industrial-scale demonstration projects. Five industrial-scale CCS projects are currently operational globally with more than 0.035 Pg of CO_2 captured and stored since 1996. Observations from these industrial scale projects and commercial CO_2 enhanced oil recovery (CO_2-EOR), engineered natural analogues as well as theoretical consideration, models and laboratory studies have suggested that appropriately selected and well managed CCS sites are likely to retain almost all of injected CO_2 for long time and provide the benefits for the intended purpose of CCS. However, CCS is still struggling to gain foothold as one of the main options for mitigating climate change due to high costs, advances in other options including renewable energy, as well as discovery of shale natural gas and the associated hydrological fracturing extraction techniques, absence of international action by governments and private sectors on climate change, economic crisis-induced low carbon (C) prices, and public skepticism. The estimated costs for CCS varies widely depending on the application—such as gas clean-up versus electricity generation, type of fuel, capture technology, and assumptions about the baseline technology. Generally, for current technology, CCS would increase cost of generating electricity by 50–100%, and parasitic energy requirement of 15–30%. In this case, capital costs and energy requirements are the major cost drivers. In addition, significant scale-up compared to existing CCS activities will be needed to achieve intended large reductions of CO_2 emissions. For example, a 5- to 10-fold scale-up in the size of individual projects is needed to

capture and store emissions from a typical coal-fired power plant of 500–1000 MW, while a thousand-fold scale-up in size of current CCS enterprise would be needed to reduce emissions by 1 Pg C yr^{-1}. The estimated global oil and gas reservoirs are 1000 Pg CO_2, saline aquifers global potential capacity ranges from 4000 to 23,000 Pg CO_2. However, there is considerable debate about how much storage capacity actually exists and is available for CCS, particularly in saline aquifers. Research, improved geological assessments and commercial scale demonstration projects will be needed to verify the estimated capacity and improve confidence in storage capacity estimates.

Keywords Carbon capture technologies · Coal-fired plants · Geological carbon sequestration · Saline aquifers · Enhanced oil recovery · Seismicity

Contents

13.1   Introduction

A range of technologies which are collectively termed carbon dioxide (CO_2) capture and storage (CCS) are suggested as one of critical options in the portfolio of solutions available for combating climate change, allowing the emission reduction, and stabilization of CO_2 emissions, while also acting as a bridge to low C and C-neutral energy sources (IPCC 2005, 2014). CCS describes a process that

separates a relatively pure stream of CO_2 from industrial or power plants and stores it in suitable geological formations after conditioning and compression (IPCC 2014). It is a process which involves the integration of four main steps: (i) the CO_2 capture, i.e., the separation of CO_2 from large point source such as large fossil fuel energy-related facilities and major CO_2 emitting industrial sources, (ii) compression of CO_2 from gas to liquid or denser gas to reduce the volume, (iii) transportation (i.e., transportation of pressurized CO_2 from point of capture and compression to the storage location, either by pipelines or other means), and (iv) storage (i.e., long-term storage of CO_2 by isolating it from the atmosphere), commonly by sequestering it in the geologic formations underground (IPCC 2005; Benson et al. 2012). Although it is applicable to other point sources, CCS research has mostly focused on capturing CO_2 from power generation fossil fuels—such as coal, or gas-fired power plants. In the short term, CCS could offer benefits of slowing growth of CO_2 emissions while other lower GHG emission energy technologies are maturing and deploy widely. Additionally, over the longer term, CCS could be used to reduce emissions from sources that are difficult to eliminate in any other way such as energy intensive industrial processes, natural gas clean-up, hydrogen production, fossil fuel refining, petrochemical industries, and cement production (Benson et al. 2012). The International Energy Agency (IEA) estimates that when fully implemented, CCS could contribute about 20% of CO_2 reduction by the end of this century (IEA 2008, 2013).

Technologies are currently available to carry out all four steps of CCS, but implementation of CCS remains a challenge because the cost of capturing CO_2 is too high for economical industrial scale deployment. Also, integration of CCS with electricity production or industrial processes has not been demonstrated at a large-scale, because of high costs of capturing CO_2 from the flue gas, and also, confidence needed that the storage of CO_2 in geological formation can be safe and effective over time periods of 1000 years and longer is still lacking. While in principle, CCS could be deployed on a much wider basis today, the challenges of deploying CCS, especially that of retrofitting pre-existing large point sources with CCS designed without the consideration of CCS as the future option should not be underestimated. Integration of CCS into existing power generation facilities and other industrial operations that demand highly reliable performance is destined to result into technological challenges, on top of large capital investments and significant operating costs required for CO_2 capture and storage. With todays' CO_2 capture technology, 10–30% of the energy output may be consumed by CO_2 capture unit alone depending on the age and type of power plant, and the degree of systems integration. The dated plants tend to be less efficient compared to the modern plants. Efficiency in both CO_2 capture technology and power generation will be needed to offset the energy penalty of CO_2 capture. An enormous effort has been devoted to advancing CCS technology, with 15 large-scale projects actively in operation globally, 7 more under construction, and 21 more in planning phase globally, although the final financial commitment decision remains pending for majority of them (GCCSI 2016). The CO_2 capture and storage capacity of these 22 projects in

operation or under construction is estimated at 0.04 Pg CO_2 yr^{-1} (GCCSI 2016). In 2016, five industrial-scale demonstration projects, capturing 0.0005–0.002 Pg CO_2 yr^{-1} from different industrial sources and store it in deep geological formation, have been operational for several years around the world, demonstrating that, at least on a limited scale and size, CCS can be safe and contribute to reducing CO_2 emissions. Four of these capture CO_2 from natural gas clean-up, while in the fifth, CO_2 is captured from coal-to-synthetic natural gas plant. None of the operating industrial-scale projects takes CO_2 from coal-fired power plants, however, and only two of the current projects under construction will capture CO_2 from power plant.

The captured CO_2 is generally purified and compressed before being transported to the storage sites. The majority of the ongoing research and demonstration efforts have been devoted to CO_2 removal from stationary large CO_2 emission sources which include power production sources, mostly because: (i) these sources are by far the dominant contributors of total anthropogenic CO_2 emissions accounting for nearly 60% of GHG emissions (Victor et al. 2014), (ii) the technologies of capturing CO_2 from the stationary sources is more mature than those for capturing CO_2 from mobile sources such as transportation devices, (iii) based on economies of scale—it is more attractive to implement CCS technologies on large-scale power plants for some foreseeable future (IPCC 2005). Fossil fuel power plants are the most susceptible to CO_2 reduction mandates, due to large point source. Therefore, CCS may be strategic for their future CO_2 mitigation options. Furthermore, unlike other energy intensive industries, power plants cannot move to other countries with fewer CO_2 emission restrictions.

Fossil fuel derived energy presently dominates most aspects of modern human activities and the modern day way of life. Fossil fuels are also projected to remain as the main source of energy for the foreseeable future. Although United Nations Framework Convention on Climate Change (UNFCCC) and Kyoto Protocol cover a wide array of CO_2 sources and other greenhouse gases (GHGs) responsible for climate change, the combustion of fossil fuels in both stationary and mobile sources remains the largest source of GHG emissions, including CO_2 which accounts for 35% of total anthropogenic GHG emissions in 2010 (Bruckner et al. 2014). Fossil fuels are currently supplying more than 80% of total primary energy demand. Among the many anthropogenic GHG sources, the energy is by far the largest source releasing 68% of anthropogenic GHGs in 2010, of which, about 90% of energy-related emission is CO_2 originating mostly from fossil fuel combustion while CH_4 and N_2O account for 9 and 1%, respectively (Fig. 13.1; IEA 2015a). Energy sector CH_4 emissions originate from oil and gas extraction, transmission, and distribution, and N_2O from energy transformation, industry, transport and buildings (IEA 2015a). Other sectoral emissions include agriculture (11%), industrial processes (7%) and other sources (14%) (IEA 2016; IPCC 2013). The share of energy-related CO_2 emissions from coal increased from 38 to 44% between 2000 and 2014, while that of natural gas remained at 20%, and that of oil declined from 42% in 2000 to 35% in 2014 (IEA 2015a).
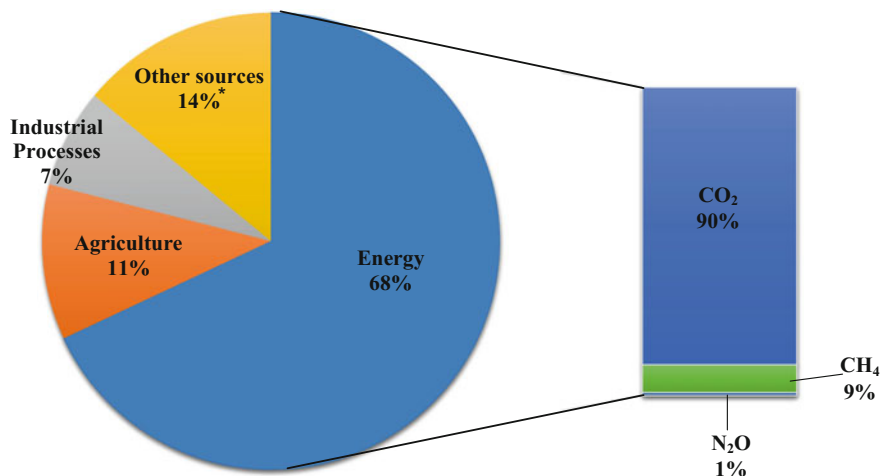
*Other sources include: biomass burning, peat decay, indirect N_2O emissions from non-agricultural emissions of NO_x and NH_3, waste and solvent use.

Fig. 13.1 Shares of global anthropogenic greenhouse gases emission in 2010. *Data source* IEA (2016)

Suitable techniques for capturing CO_2 were originally developed circa 1950 for use in food processing, chemical manufacturing and in connection with the production and utilization of natural gas as the energy source as well as H_2 purification (Zaman and Lee 2013). The technology involved scrubbing the gas stream with chemical solvent, producing pure CO_2 as a valuable product for use in food processing and chemical manufacturing. The capture process itself was a relatively small scale compared to those needed for power plants (Zaman and Lee 2013). Subsequently these techniques were adapted for capturing CO_2 from flue gas streams of coal or natural gas combustion systems for carbonation of drinks and CO_2-enhanced oil recovery (CO_2-EOR). In these processes, CO_2 is obtained as a valuable product also, and the capture process is relatively small as compared to those needed for power plants to mitigate CO_2 emissions. Therefore, when existing CO_2 separation techniques are implemented on large-scale power plants they are not cost-effective (DOE/NETL 2010) due to much higher volumetric rate of low pressure flue gas that is needed to be processed and the presence of other impurities such as SO_2. As a result, when CCS is implemented in a typical pulverized coal power plant the cost of electricity is estimated to increase by up to 80% and the generating capacity decrease by as high as 30% (Zaman and Lee 2013). Horn and Steinberg (1982) are among the first to suggest the process used to separate CO_2 from natural gas and utilization of separated CO_2 for EOR as a method to control CO_2 emission.

Even though the industry has been pushing for the adoption of CCS as the significant breakthrough mitigation option since its inception, the Intergovernmental Panel on Climate Change (IPCC) was hesitant to recognize it as

one of an appropriate CO_2 mitigation pathway earlier, and it was not listed among the recognized mitigation options in the IPCC Second Assessment Report published in 1995 (IPCC 1995), and in its Third Assessment Report of 2001, CCS also received much less attention (IPCC 2001), probably due to relatively lack of in-depth risk analysis. In 2002, the 7th Conference of Parties to the UNFCCC invited the IPCC to conduct a scientific assessment of potential for CCS as a CO_2 emission mitigation option, which resulted into publication of a special report on CCS in 2005. Publication of the Special Report on CCS by the IPCC in 2005 (IPCC 2005) and the support by influential bodies such as the International Energy Agency (IEA) (IEA 2004, 2009) gave CCS the much needed credibility. However, the climate change summit of Copenhagen Denmark in 2009 appears to be the turning point in the perception of CCS, due to its lack of global signal that climate change mitigation must be taken seriously in investment decision, with industries finding less emphasis in climate change mitigation and lack of reasons to invest in deployment of CCS on a large-scale since it adds significantly to the cost of power generation and other products that involve the use of fossil fuels.

There has been a wide divergence of opinion about the feasibility, long-term risks, and even the very need for CCS. Whereas, CCS has been offered as a vision of future in which the impacts of growing fossil fuel reliance are minimized by capturing and storing CO_2 instead of allowing it to accumulate in the atmosphere (Jaccard 2005; IPCC 2005), and some have projected that CCS is a technology critical to solve the climate change while continuing the reliance on fossil fuels, and conclude that CCS is an essential tool for reducing CO_2 emission to the atmosphere sufficiently quick to avoid worse consequences of climate change (IPCC 2005; IEA 2008, 2013; Gibbins and Chalmers 2008b), others believe it will not make significant contribution to solving the climate change problems, and still others believe CCS to distract from making needed decisions to begin phasing out fossil fuels immediately in favor of renewable energy sources and C-free or C-negative energy sources. Some researchers have argued that investing in CCS is not money well spent, as the global climate-energy situation are becoming increasingly dire, bold measures with the near-term influence are needed to reduce, rather than sustain fossil fuel reliance (Stephenson 2013). It has been suggested that the vision of CCS has enabled complacency about the growing dangers of fossil fuel dependence by providing a false sense of optimism, and therefore, investments in CCS need to be halted (Stephenson 2013).

Regardless of ongoing debate in scientific community, knowing whether CCS is part of climate change solution or not will likely take many decades, and the answer will most likely be heterogeneous, with some regions and industries adopting it as part of the preferred sets of options for reducing emissions and others finding other mitigation alternatives. Only when the decision makers widely recognize that slow and incremental change will not solve the climate change problem will it be clear that every possible option is needed to address GHGs emission and climate change. The objectives of this chapter are to present the global narrative of CCS in terms of some current technological and scientific understanding and developments.

13.2   What Is Carbon Dioxide Capture and Storage?

The CO_2 capture and storage involves capturing CO_2 arising from large point sources such as combustion of fossil fuels in power generation, preparation of fossil fuels—including natural gas processing, biomass based fuels, certain industrial processes such as production of H_2, ammonia, iron and steel, cement, and ethanol fermentation (Benson et al. 2012). Capturing of CO_2 involves separating the CO_2 from other gases in flue gas stream—including N_2 and water vapor. The captured and purified CO_2 is compressed into liquid or high density gas and then transported to the storage site where it can be stored and separated from the atmosphere for long time or permanently for the explicit purposes of avoiding atmospheric GHG emissions. The potential sequestration sites include depleted natural gas and oil reservoirs, underground sedimentary basins and saline aquifers, and coal seams that cannot be mined. A number of studies have explored technological and scientific dimensions of CCS. The greatest focus of ongoing research and demonstration of CCS has been on techniques for capturing CO_2 from coal-based electric power generation, logically so, because nearly 40% of total anthropogenic fossil fuels CO_2 emission is from the application of coal as a feedstock for power generation. Coal-fired power plants also dominate the lists as the biggest stationary CO_2 point sources. In addition, the global resources of coal are significantly large compared to other fossil fuels (Fig. 4.3). A typical coal power plant emits 0.8–1.0 Mg CO_2 per Terawatts hour (TWh = 10^{12} W h) of net electricity generated, and a 1000 MW electrical coal power plant at a 75% annual load factor emits about 6000 gigagrams (Gg = 10^{12} g) CO_2 yr^{-1} (Benson et al. 2012). Much of the ongoing research has focused on scientific understanding of CO_2 capture, transport, and storage processes, but relatively less attention has been given to regulatory, legal, long-term permanence, public perceptions, economic, and future liability considerations, even though these elements are as important for any successful implementation of industrial-scale CCS projects.

The goal of CCS is to reduce emissions from large stationary sources such as power generation, natural gas processing, hydrogen (H_2) production from coal or gas, cement manufacturing or steel making. A full lifecycle emission using CCS technology on individual facility can reduce 65–85% of CO_2 emission from fossil fuel (Benson et al. 2012). Some of the remaining emissions involves activities beyond the plant boundary. Assessments under a range of stabilization scenarios suggest that the contribution of CCS is anticipated to be 20% of needed reduction over the next century—which is similar to that of renewable energy and use efficiency gains (IPCC 2005; IEA 2008). In the future, CCS may also contribute to emission reduction from transportation sector through H_2 production and use for light and heavy duty vehicles, electrification of vehicles, and production of synthetic fuel using captured CO_2.

Although the major focus of capturing CO_2 for climate change mitigation has been coal-fired power generation plants, there are several important industrial applications that produce nearly pure CO_2 vents. These include raw natural gas and

synthesis gas (syngas) purification, production of high value products (e.g., ammonia, other synthesis gas products such as H_2, synthetic natural gas (SNG) from coal, coal-to-liquids, ethanol fermentation, or methanol production). These have been the major focus of the ongoing small scale technology demonstration projects due to relatively higher CO_2 concentration and lower costs of purification. Most of H_2 is made from natural gas via CH_4 reforming process which does not produce pure CO_2 stream however, (Shah et al. 2007), but H_2 made from heavy oil, petcoke or coal gasification produces large pure CO_2 stream. Other industrial point sources of CO_2 include cement kilns, iron/steel manufacturing, oil refinery and bulk chemicals. The large industrial point CO_2 sources have typically released less than 10^6 Mg CO_2 yr^{-1} which are relatively small sources compared to large power generation plants. Nevertheless, the higher purity of industrial CO_2 vents makes them targets for CCS due to lower costs of CO_2 capture and purification. Countries such as China with many large-scale coal gasification plants and large synthesis gas capacity for ammonia, methanol, and H_2 have large pure CO_2 vents for lower cost CCS (Simbeck and Roekpooritat 2009). Key determinants of the extent of CCS deployment include: (i) capacity for storage in appropriate sites in geological formations, (ii) policy framework to encourage emission reduction, (iii) lack of low-cost opportunities for reducing emissions such as renewable energy or nuclear power, (iv) pace of technological progress to lower the cost of geological storage, (v) interest in deployment of CO_2-EOR at large-scale, (vi) public acceptance, and (viii) access to large-scale capital investment needed for CCS projects.

13.3 Energy Consumption and Carbon Dioxide Emissions

Fossil fuels are the predominant primary energy present in the world and accounted for nearly 87% of commercial energy use carriers in 2014 (BP 2015). Fossil fuels are also the dominant fuel for power generation, producing about two thirds of electricity currently, and projected to provide a similar fraction in 2035 (IEA 2015c). However, they also account for most of local conventional pollution and global CO_2 emissions. Due to advantages in cost, technological maturity, and established infrastructure, fossil fuels are likely going to remain as a major component of global energy supply for a foreseeable future. However, relying on fossil fuels has posed major challenges to the world which need to be addressed, including: (i) climate change, (ii) air pollution, (iii) lack of affordable efficient alternative energy carriers, and (iv) energy insecurity.

Electricity demand has been increasing with economic growth. Due to high CO_2 emission from power generation sources, majority of current GHG mitigation research effort are devoted to CO_2 removal from the stationary power production sources. In addition, these sources share many similarities with the existing processes where CO_2 capture has been used for several decades, and therefore, technologies for CO_2 capturing from stationary sources are more mature than those

from the mobile sources such as transport devices. Economies of scale also make it economically attractive to implement CCS technologies on large power plants, at least within some foreseeable future. The CO_2 emissions from fossil fuel energy sources represent about 60% of the global anthropogenic GHG emissions. In 2013, fossil fuels accounted for 82% of the global total primary energy supply (TPES) (IEA 2016) and contributing 70% of CO_2 emissions (IEA 2014). Oil, coal, and natural gas represented 31, 29 and 21% of the global TPES, respectively in 2013, but coal accounted for 46% of the global CO_2 emissions due to its heavy C content per unit of energy released, while oil and natural gas contributed 33 and 20%, respectively (IEA 2016). In 2013, 163.6 Exajoules (EJ) (28.9%) of coal, 179.9 EJ (31.7%) of oil, and 121.5 EJ (21.4%) of natural gas were consumed by the world economies (IEA 2015b; IPCC 2011). Since the industrial revolution, annual CO_2 emissions from fossil fuel combustion dramatically increased from near zero in 1750 to 10.1 Pg C in 2014 (Fig. 13.2; Le Quéré et al. 2015, 2016). Growing global energy demands from fossil fuels play a key role in observed increasing trend in fossil fuel CO_2 emissions. Average decadal CO_2 emission growth since 2000 is estimated at 2.5% yr^{-1} (Le Quéré et al. 2015, 2016).

The global TPES increased by nearly 150% between 1971 and 2013 (Fig. 13.2), mainly relying on fossil fuel, and fossil fuel use continues to grow worldwide, especially in countries with rapidly developing economies. The growing world
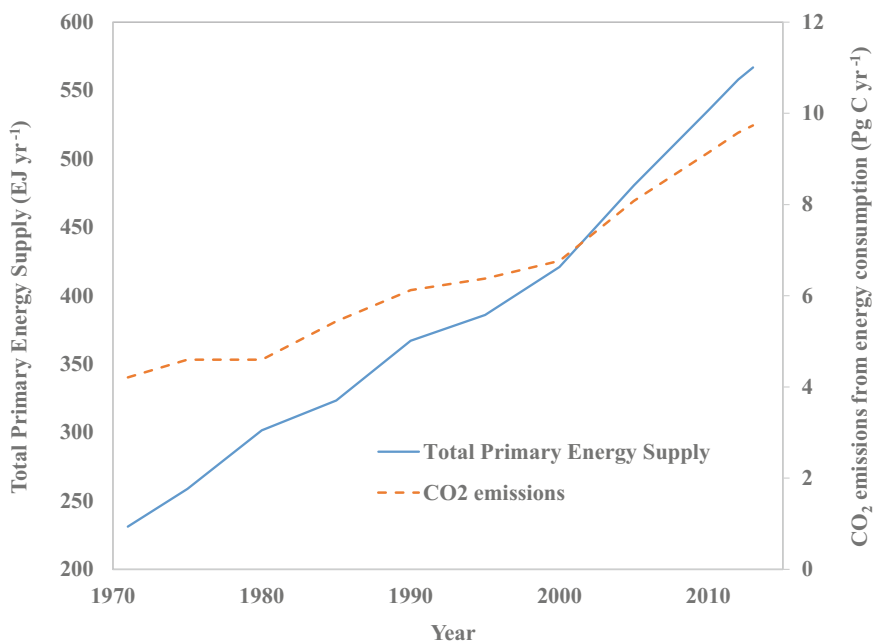

Fig. 13.2 Primary energy supply changes and CO_2 emissions from fossil fuel use from 1971 to 2013. *Data source* IEA (2016), Le Quéré et al. (2015, 2016)

energy demand from fossil fuels plays a key role in increasing trends of CO_2 emissions. The global primary energy consumption grew at an average rate of 1.4% yr^{-1} between 1990 and 1995; 1.6% yr^{-1} between 1995 and 2001, 2.6% yr^{-1} between 2001 and 2015, and more than doubled from 231.44 EJ in 1971 to 567.0 EJ in 2013 (Fig. 13.2; IEA 2015b, 2016). The contribution of global CO_2 total emissions from industrialized economies decreased from 66.5% in 1971 to 38.3% in 2013, but this decline is mostly attributed to increased emissions from developing economies, dominantly China and India rather than decline in emissions from industrialized economies (IEA 2016). In 2013, power and heat generation was by far the largest CO_2 emission sources, accounting for 42% while the transport sector accounted for 23% of CO_2 emissions (Fig. 13.3), implying that targeting CO_2 emissions from stationary energy generation sector by CCS could have greater impact in climate change mitigation.
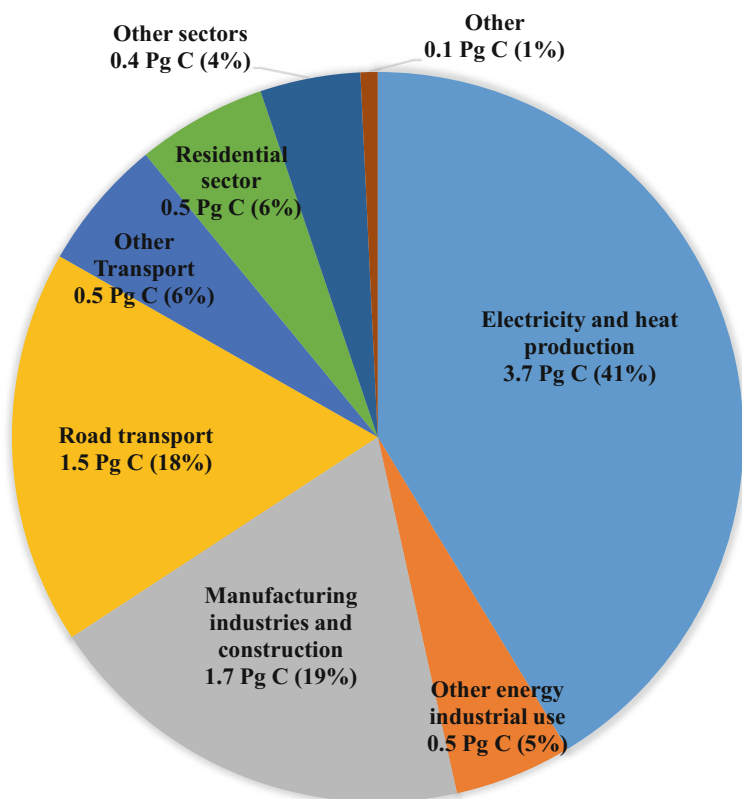

Fig. 13.3 The global CO_2 emissions by sector in 2013. *Data source* IEA (2016)

13.4   History of Carbon Capture and Storage

The injection of CO_2 underground was not totally new technology when it was first suggested for climate change mitigation. The suggestion of capturing CO_2 derived from anthropogenic large point sources and disposing it in the ocean as a way to control the atmospheric CO_2 increase and climate change mitigation was first proposed in 1970s by Marchetti (1977) who suggested that the Mediterranean undercurrent entering the Atlantic at Gibraltar had sufficient capacity to store all of CO_2 produced in Europe until the year 2100 and beyond. In addition, Horn and Steinberg (1982) were among the first to suggest a process for separating CO_2 from the natural gas purification and use it in EOR as a way of controlling atmospheric emissions of CO_2. In the 1970s and 1980s, as the production from oil fields in the USA was declining, oil companies started injecting water, natural gas, and CO_2 to recover more oil and extend the production lifetime of oil reservoirs. However, the CO_2-EOR was done almost exclusively using CO_2 from natural underground CO_2 reservoirs, so it was not leading to climate change mitigation. Nevertheless, CO_2-EOR provided the practical experience and understanding of the behavior of CO_2 underground, the knowledge of suitable cap rock that can sustain CO_2 best, and also the best injection pressures for CO_2, as well as wells placement and pipeline transportation of CO_2 (Dooley et al. 2010). In 1990s and 2000s, climate change mitigation emerged to become a global policy agenda, and resulting to UNFCCC by 1992, and the Kyoto Protocol in 1997 which included commitments of all developed countries to reduce their GHGs emission. However, not all developed countries ratified or complied with the Kyoto Protocol provisions. Despite Kyoto commitments and other subsequent international agreements on climate change mitigation, addressing the seemingly unstoppable CO_2 emissions from fossil fuel combustion remained an urgently challenging problem without a viable solution agreeable to major stakeholders. Today, CO_2-EOR remains a main driver of CCS, and many demonstration projects have used CO_2 for EOR as a way to minimize the costs for CCS projects.

In 1996, Startoil began injecting 0.001 Pg CO_2 yr^{-1} into an aquifer 800 m beneath the North Sea at Sleipner, Norway (Torp and Gale 2004). The CO_2 for the injection was stripped from Sleipner natural gas to meet the specifications for sale of natural gas in Europe. Startoil was faced with CO_2 emission tax of $50.00 ton^{-1} of CO_2 emitted, but opted to inject CO_2 into a seabed aquifer to avoid paying CO_2 emission tax, and this process began an entirely new approach for controlling CO_2 emission. By the end of the century and beginning of 21st century, the legal and regulatory framework, as well as risks associated with underground CO_2 and societal acceptability was gaining attention within scientific community, as well as national and international agencies. Two other industrial-scale CCS demonstration projects—one in Canada (Weyburn) and one in Algeria (In Salah Gas Project) were established (Riddiford et al. 2003; White et al. 2004; Fig. 13.4). The success of these initial projects led to the optimism that CCS can apparently be a promising option to contribute significantly to climate change mitigation within a short period
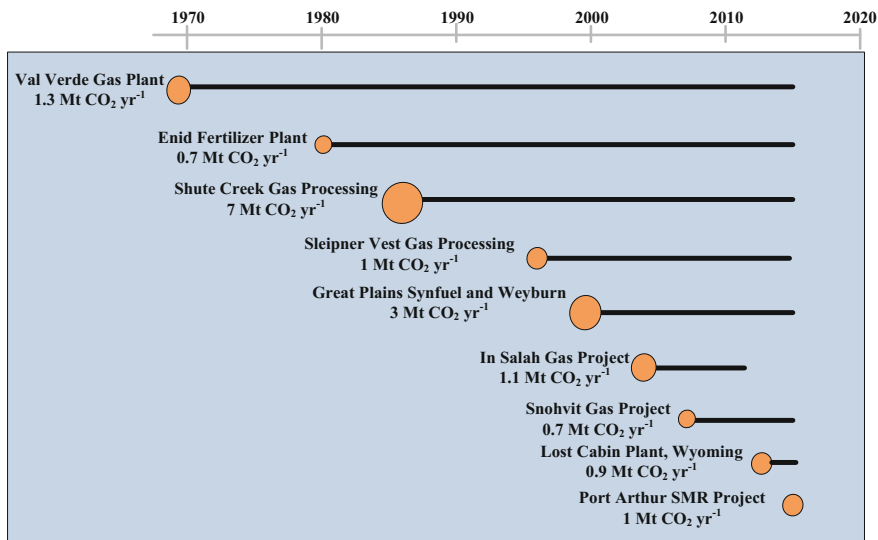
Fig. 13.4 Timeline of starting dates of operational CCS industrial-scale demonstration projects. *Circle* size distinguishes approximate annual mass of CO_2 stored, while *black lines* represents operation timeline

of time since the late 2000s. However, the pace of deployment of new projects has slowed, even though governments and private sector investment in science and technology of CCS has continued to build a strong and broad foundation for it.

The option of storing CO_2 in ocean has largely been abandoned since the publication of the IPCC Special Report on CCS (IPCC 2005), primarily due to: (i) high costs, (ii) low storage permanence, and (iii) significant ecological and environmental impacts. The current discussions and research have revolved around the injection of CO_2 into geologic reservoirs and other storage options including CO_2 mineralization, but techniques involving CO_2 mineralization are expected to play a limited role in climate change mitigation (Wang et al. 2014; Boot-Handford et al. 2014). Some of the full scale operational industrial CCS demonstration projects that store atmospheric CO_2 are presented in Fig. 13.4. Six industrial-scale projects have been operational globally and exclusively for the purpose of demonstrating CO_2 emission mitigation from the inception of CCS in 1996 to present, injecting 0.0007–0.003 Pg CO_2 yr^{-1}, mostly for EOR. Five of the successful commercial-scale CCS projects that have been injecting approximately 0.001 Pg CO_2 yr^{-1} are:

1. Startoil's Sleipner offshore gas platform in the North Sea, Norway, injecting CO_2 from natural gas purification into undersea deep saline formation.
2. Weyburn, Saskatchewan, Canada injecting CO_2 from Dakota Gasification (brown coal (lignite) gasification via synthetic natural gas (SNG) purification) plant from North Dakota, USA. Purified CO_2 is piped to Weyburn and used in CO_2-EOR.

3. Wyoming, USA facility injecting CO_2 from natural gas purification, which is piped to Power River Basin of Wyoming and used for CO_2-EOR.
4. BP's In Salah facility in Algeria, Africa with CO_2 from natural gas purification with injection to the same formation but at a distance from the production well. The injection operation has been terminated.
5. Startoil's Snøhvit Project in Norway, which injects CO_2 from liquefied natural gas facility into a saline formation underneath the Barents Sea.

All the successful commercial-scale demonstration projects have the economic advantage of producing large pure CO_2 vent from raw natural gas purification or other sources. Natural gas from the fields contains about 2% to more than 15% CO_2, and CO_2 removal is required to meet natural gas pipeline energy content specifications regardless of the CO_2 mitigation policy. The early 1970s through early 1990s, projects did not use CCS for the climate change mitigation, nevertheless, they provided considerable CO_2 emission reduction, as well as understanding the injection conditions. The first project implemented solemnly for climate change mitigation purposes was Sleipner project, which is located at offshore Norway. The In Salah gas project in Algeria has ceased to operate. Recovery of hydrocarbon in oil from CO_2-EOR provides economic basis for the CCS project sustainability. The incentive behind Statoil's Sleipner and Snøhvit projects is the European CO_2 emission tax for operations in Norway. Therefore, the overall costs of these CCS projects have been significantly lower than normal CCS costs due to an added economic value of CO_2 capture and/or utilization.

There has been considerable debate whether CO_2 used for EOR should be considered as CO_2 storage for the climate change mitigation purposes? While amount of CO_2 remaining underground varies from site to site, and from oil well to oil well, it is estimated that about 50% of the injected CO_2 never returns to the surface (Stevens et al. 2003). Moreover, in almost all cases, the CO_2 produced with the oil is separated and injected back into the oil reservoir, primarily because CO_2 in this case is a valuable commodity, and avoids the need to purchase more CO_2. It can also be argued that the CO_2 emitted into the atmosphere when oil is used negates the benefits of storage, however. In the US, where most of CO_2-EOR is carried out, CO_2 utilization credits for EOR projects are \$0.53 per 1000 standard cubic feet (28.3 m^3) in EOR terms, which is significantly less than the total cost of CCS, unless the CO_2 is being captured regardless of CO_2 mitigation issues.

About 40 projects of different CO_2 injection scale ranging from 0.0006 to 0.004 Pg CO_2 yr^{-1} are in various stages of development globally (GCCSI 2016). Nevertheless, experience over the past suggests that only few of these are likely to be fully implemented (GCCSI 2016). The proposed projects under development indicate that CO_2-EOR continues to play an important role in demonstrating CCS at industrial scale (GCCSI 2016). Although CCS was designed to capture CO_2 from power plants and large industrial sources, none of the projects currently under operation takes CO_2 from power plant, and only one of the projects under development will utilize CO_2 captured from the power plant (GCCSI 2016). Although there has been large investments by governments and private sector in science and

technology of CCS, similar progress has not been made in social, legal dimensions, and significantly lower private sector financial commitments to CCS have occurred. The economic crisis of 2008 and the related low price of CO_2 emission allowance units in the European Trading Scheme as well as sudden availability of significant quantities of shale natural gas at low prices in the United States of America have resulted in drop in funding for CCS projects in both USA and Europe. Moreover, the worldwide drop in costs of wind and solar energy production is making it a viable option for many households also, while reducing the enthusiasm of CCS.

13.5   Technology for Capturing and Transporting Carbon Dioxide

Coal-fired power plants have been the primary focus for CCS research, mainly due to the magnitude of emissions. Of all stationary CO_2 sources, electricity production is the largest single category accounting for more than two-thirds of global CO_2 emissions from stationary sources. Nearly 40% of human-made fossil fuel CO_2 emissions are from electric power generation (Benson et al. 2012). Although there are other applications, and feedstocks for CCS, coal-fired power plants also dominate the list of biggest stationary CO_2 point sources. However, all existing industrial CCS applications currently in operation are for industrial applications such as natural gas clean-up, biofuel production, and production of synthetic natural gas from coal, mainly due to high costs for capturing CO_2 from power generation. Only one of the commercial scale demonstration project currently captures CO_2 from coal-fired power plant. However, CCS can also be applied to other stationary CO_2 emission sources, including industrial sources such as steel mills, cement plants, ethanol production facilities, and natural gas processing units that remove impurities such as CO_2 and H_2S.

13.5.1   Carbon Dioxide Capture

The CCS requires large CO_2 stationary point sources within reasonable distances of suitable geologic storage locations. It is recommended that the suitable geologic formation should: (i) be deeper than 800 m, (ii) have a thick and extensive cap seal, (iii) have sufficient porosity for large volumes of CO_2 storage, and (iv) sufficiently permeable to permit injection at high rates without requiring overly high pressure (Benson and Cole 2008). Injecting CO_2 below 800 m provides two advantages: (i) CO_2 density is high enough to allow pore filling and decrease buoyancy difference compared with the insitu pore fluids, (ii) to protect underground water sources. The effectiveness of geologic CO_2 sequestration depends on how much CO_2 can be injected into subsurface rock formations, how long it will stay trapped

there, and whether the process of subsurface injection and storage will have negative environmental consequences. All these factors are both technical and socioeconomic constraints that must be evaluated realistically before the CCS project implementation.

For the CCS technology to contribute in reducing CO_2 emission, CO_2 must come from the existing sources through retrofit or replacing the existing power generation plants with new plants with CCS. For a new plant constructed with CCS without replacing the old plant, the captured CO_2 is considered as CO_2 emissions avoided, which will slow the rate of growth of CO_2 emissions with no impact on reducing the existing CO_2 emission levels. Reduction of CO_2 emission requires retrofitting the existing fleet of coal-fired power plants with CCS, or replacing the existing fleet with lower CO_2 emission sources and/or retrofitted with CCS to significantly lower CO_2 emissions from current sources. Also, adding CCS to old existing plants are subject to net capacity and efficiency losses (Simbeck and Roekpooritat 2009). The CCS can also be applied to other types of stationary CO_2 sources including industrial sources such as boilers and blast furnaces, steel mills, cement plants, ethanol production, ammonia production plants, natural gas processing units that remove impurities such as CO_2 and H_2S, and electricity production from both coal and natural gas (IPCC 2005; Benson et al. 2012; IEA 2013) although research on CO_2 capture from these sources has received less attention. CCS in power plants tends to be among the most challenging in terms of financial perspective due to economy-wide increasing costs for power generation. However, electricity production is the largest single source accounting for more than two-thirds of global CO_2 emission from stationary sources. The sources of stationary CO_2 emissions are distributed around the world, but two countries that stands out in coal-fired power generation emissions are China and the United States of America (Yang and Cui 2012).

Technologies for geological storage of CO_2 build on long experience of oil and gas industry in pumping and managing subsurface fluids, including injection of CO_2 to increase oil production (EOR). Capture involves separation of CO_2 from industrial flue gas stream and its concentration. CCS requires concentration of CO_2 to purities of 95% or greater (IPCC 2005). High CO_2 concentration sources minimize purification and compression costs and also makes effective and efficient use of available sequestration resources—i.e., subsurface pore volume. The majority of cost for CCS lies in the CO_2 capture and purification stage. Current rates of geologic CO_2 sequestration are still small (on the order of few million metric tons of CO_2 per year (NETL 2015). To play a significant impact on climate change mitigation, much higher rates of CO_2 injection are needed as envisioned in IPCC Special Report on CCS (IPCC 2005), which has so far not yet been achieved in the ongoing research and demonstration projects. The capture and transport of CO_2 pose some of the principal challenges in the implementation of geologic C sequestration. For example, power plants that utilize CCS technologies are expected to require 10–40% more energy than equivalent plants without CCS. Depending on where in the combustion process the separation of CO_2 occurs, there are three

technological pathways for capturing CO_2 from power generation: (i) pre-combustion capture, (ii) post-combustion capture, (iii) oxyfuel combustion (Fig. 13.5; Benson et al. 2012). The comparative advantages and drawbacks of these steps are summarized in Table 13.1. In the post-combustion CO_2 capture, a separation of CO_2 is added after the boiler without inherently changing the power generation system. Most of the demonstration projects in the power sector aim at post-combustion CO_2 capture. The first step of CO_2 recovery or capture and concentration to high purity CO_2 stream is the most expensive and it can normally be implemented through one of the three above named general processes (IPCC 2005).

13.5.1.1 Pre-combustion Carbon Dioxide Capture

Pre-combustion CO_2 capture involves conversion of fuel feedstock (i.e., coal or biomass or a mixture of coal and biomass) into syngas through gasification, steam reformation, or partial oxidation, and then shifting the syngas chemically to H_2 and CO_2 at elevated pressures, typically 30–70 atmospheres, and then separating H_2 from CO_2. The CO_2 can then be separated to leave H_2-rich fuel gas. Pre-combustion capture allows C to be stripped before the resulting H_2 gas is combusted. This process for power generation requires an integrated gasification combined cycle (IGCC) plant, of which only few are currently operational globally, and retrofitting for this type of plants is therefore, practically impossible. The separation of CO_2
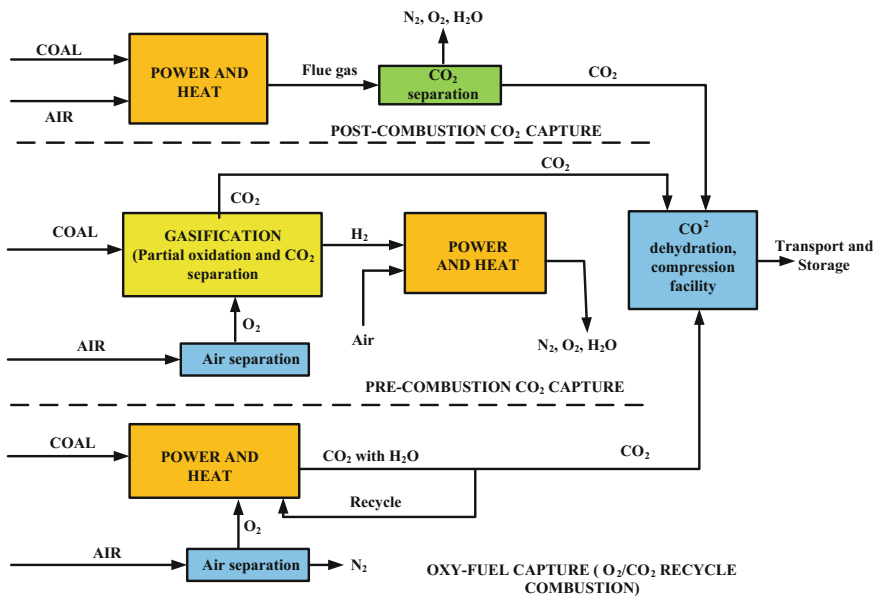

Fig. 13.5 Principles of three main carbon dioxide capture options (after Gibbins and Chalmers 2008a)

Table 13.1 Comparative advantages and disadvantages of post-combustion, pre-combustion and oxy-combustion

Technology	Advantages	Disadvantages
Post-combustion	• Mature technology for other application (e.g., separation of CO_2 from natural gas) • Standard retrofit of generation capability • Technology improvements and cost reductions possible with additional development	• High energy penalty (approximately 30%) • High cost
Pre-combustion	• Lower costs than post-combustion capture • Lower energy penalties than post-combustion capture • High pressure of CO_2 reduces compression costs • Combines with H_2 production for transportation sector • Technology improvements and cost reductions are possible with additional development	• Complex chemical process is required for gasification • Repowering of existing capacity is needed • Large capital investment is needed for repowering
Oxy-combustion	• Avoids the need for complex post-combustion separation • Potentially higher generation efficiencies • Technology improvements and cost reductions is possible with additional development	• New high temperature materials are needed for optimal performance • On-site oxygen separation unit is needed • Repowering of existing capacity is needed

typically uses physical solvent at the higher pressure, and then CO_2 is released as the pressure is reduced to regenerate the solvent. In the pre-combustion capture, CO_2 is not available for capture prior to combustion, but all fossil fuels can be gasified (i.e., partially combusted or reformed) with sub-stoichiometric O_2 and some steam at elevated pressures [typically 30–70 atmospheres (3–7.1 MPa)] to produce synthesis gas mixture of predominantly CO and H_2. Additional steam is then added and the mixture is passed through a series of catalyst beds for the steam-gas shift reaction to approach equilibrium (Eq. 13.1)

$$CO + H_2O \leftrightarrow CO_2 + H_2 \tag{13.1}$$

The CO_2 can be separated to leave a H_2-rich fuel gas. The separation process typically uses a physical solvent, where CO_2 is dissolved at higher pressure and then released as the pressure is reduced (Gibbins and Chalmers 2008a). No heat is required to regenerate the solvent, since CO_2 can be released at above-atmospheric pressure. Therefore, the energy requirement for CO_2 capture and compression in pre-combustion systems may be of the order of half that required for

post-combustion capture. However, pre-combustion capture systems have to pay an efficiency penalty for the CO shift reaction (Eq. 13.1), and even without CO_2 capture, there is lost mass of CO_2 that does not pass through the turbines and generate power. Additionally, the efficiency of H_2 burning gas turbines is lower than conventional natural gas or syngas units, since heat transfer coefficients are higher for combustion products from H_2-rich fuels (Gibbins and Chalmers 2008a).

Pre-combustion Carbon Dioxide Capture in Electric Generation

Pre-combustion CO_2 capture in electric generation is mostly focused on coal through IGCC, and not natural gas combined cycle (NGCC). The NGCC-CO_2 capture is more focused on post-combustion CCS because no major modifications for standard NGCC design is necessary (Benson et al. 2012). In coal-fired power plants, when coal reacts with O_2 and steam at high temperatures and pressure, it produces syngas—a mixture of carbon monoxide (CO) and hydrogen (H_2) in a process of gasification. After gasification step, syngas reacts with steam in a water-shift reactor, which converts CO to CO_2, producing a mixture of CO_2 and H_2. CO_2 is then captured from this fuel gas, leaving just H_2 for the power generation. Generally, CO_2 is isolated along with minor amounts of CO, water, and O_2. This type of power plant is called IGCC power plant using coal, biomass, or their mixture. The sulfur compounds are removed from fuel gas prior to the CO_2 capture. When pre-combustion capture is applied to natural gas power plants, natural gas is first converted to syngas in the presence of O_2 and steam through reforming process. The fuel conversion steps are costly in pre-combustion CO_2 capture (Folger 2013).

Industrial Pre-combustion Carbon Dioxide Capture

The pre-combustion CO_2 capture has the most commercial experience. Most of the existing large-scale CCS projects have been from industrial applications involving CO_2 capture from pre-combustion capture from industrial oil and gas application and not electric power generation. There are natural gas purification and gasification-based synthesis gas (H_2 + CO) purification plants that remove CO_2 at amounts greater than 1 Pg CO_2 yr^{-1}, however, that high purity CO_2 is normally vented. The CO_2 is removed just to meet product gas—generally natural gas or H_2 specifications. Examples of industrial gasification plants with pure CO_2 vents with CCS include Dakota Gasification SNG plant in USA, the Shell Oil Pernis refinery in the Netherlands, and Shenhua Group liquefaction plant in China. Gasification is commonly used for production of H_2, ammonia, synthesis gas for chemicals, and natural gas in industrial settings. Pre-combustion CO_2 capture in electric generation is generally focused on coal through IGCC and not NGCC. The biggest challenge facing pre-combustion CCS currently is the high capital costs.

13.5.2 *Post-combustion Carbon Dioxide Capture*

Post-combustion CO_2 capture involves capture of CO_2 from flue gas after the power generation step, and the separation of CO_2 is mainly from N_2 and relatively little amounts of O_2 and water vapor. Post-combustion CO_2 capture is mainly for pulverized coal, oil-fired or gas-fired power plants, but it can also be applied to IGCC, NGCC and gas capture. Post-combustion separation requires the use of liquid solvents to remove the CO_2 from the flue gas, followed by extraction of the CO_2 from the solvents. The advantages of post-combustion capture include: (i) all existing power plants can be retrofitted with only minor modifications, (ii) energy demand of power plant can be controlled by adjusting the CO_2 capture level or by bypassing the CO_2 capture step at times of peak loads, and (iii) it can be applied to capture CO_2 from industrial manufacturing (e.g., cement production, steel industry (IPCC 2005). Post-combustion capture in electric generation has gained greater interest than industrial sources in recent years due to: (i) decreased interest in pre-combustion CO_2 capture as a result of high capital costs and slow commercial acceptance of IGCC with or without CCS, (ii) improved designs for post-combustion CO_2 capture with more vendor competition and choices of chemical solvents, (iii) minimal impact on traditional NGCC power plant process other than the large need of low pressure steam for CO_2 stripping and CO_2 compressor power, (iv) ability to easily bypass the back-end flue-gas scrubber process when problem with the CO_2 system occur or when there is a need for additional peaking power, and (v) lower capital outlay and ease retrofit to the existing power plant, however, moderately high capacity and efficiency losses and additional space are needed (Benson et al. 2012). Most interest in natural gas based electric generation CCS is with post-combustion CO_2 capture for CO_2 emission avoidance reasons. Lower natural gas prices and improved supplies are making NGCC more competitive with coal-based electric generation for baseload power.

Due to low concentrations of CO_2 (4–15% by volume) and low overall stream pressure in comparison to traditional sources of CO_2 where the feed gas is at high pressure, the partial pressure of CO_2 in flue gas is much lower. In industrial post-combustion, the CO_2 concentration in flue gas varies widely, from 7 to 10% for gas-fired boilers to 14–33% for cement kilns (IPCC 2005). Since CO_2 at such a low concentration cannot be captured effectively by physical adsorption, chemical absorption is likely needed, because physical adsorption depends on partial pressure for adsorption, whereas chemical absorption is less dependent on partial pressure. Reasons for inefficiencies in post-combustion CO_2 capture include the low concentration of CO_2 in the flue gas, large volume of flue gas to be treated, the requirement to compress CO_2 from the atmospheric pressure to storage pressure, and the relatively high temperature of flue gas which needs to be cooled before CO_2 capture. Temperature and pressure swing adsorption and membranes can also be used for separating CO_2 from N_2 in post-combustion capture. However, flue gas contaminants such as SO_2 cause problem to remove and negatively impact the

performance of many technologies such as adsorption. In addition, the gas is almost saturated with water, making selective absorption or adsorption of CO_2 relative to water critical.

13.5.2.1 Physical and Chemical Processes for CO_2 Separation

Several technological options are available for separating CO_2 from a gas stream, and the optimal choice depends on the CO_2 source, the cost, and the ease of deployment. These include: (i) separation of CO_2 using sorbents or chemical solvents, (ii) membrane separation, and (iii) distillation of liquefied gas stream. The choice of technology particularly depends on CO_2 composition of the flue gas. For natural gas turbines the CO_2 concentration ranges from 3 to 4% for natural gas turbines, 10–15% for pulverized coal-fired plants and up to 40–60% for IGCC plants (IPCC 2005). Absorption into physical and chemical solvents, adsorption onto solid substrates, cryogenic separation, transport through CO_2 selective membranes, and mineralization are some of the options used for CO_2 separation (Wilcox 2012; Zaman and Lee 2013). Adsorption process is basically composed of two steps, adsorption and regeneration which operates on a repeated cycle. For CO_2 capture using solvents and sorbents, two-step process is required in which CO_2 is first removed from gas stream using absorption tower, and in second, CO_2 is released from the media in a separate regeneration tower. The low concentration of CO_2 in the flue gas of fossil fuel power plants necessitates large absorption towers for CO_2 separation and the related high costs of CO_2 capture. For natural gas cleanup, cryogenic separation and membrane separation are used, although on a limited basis. Amine solvents (Rochelle 2009), chilled ammonia (Mathias et al. 2010), ionic liquids (Goodrich et al. 2010), polymer membranes (Du et al. 2011) and cryogenic separation are some of potential options for CO_2 capture (Table 13.2). Monoethanolamine solvents are the most mature option and remain the benchmark for cost and technical performance evaluation (Rochelle 2009). Fundamental research in the area of CO_2 separation has grown rapidly and advances have been made across the board (Boot-Handford et al. 2014). The energy required for CO_2 capture is one of the biggest challenges for CCS. From thermodynamic perspective, the minimum energy required depends on CO_2 concentration, and ranges from 3 to 6 kJ mol^{-1} of CO_2 for coal fired plants and 7–9 kJ mol^{-1} of CO_2 for a natural gas plant, which represent only 2–3% of the energy output at the power plant (Wilcox 2012), suggesting that if efficient separation process could be developed, the energy penalty is small. However, in practice, the total energy penalty is significantly greater, about 5–10 times the minimum energy requirement (House et al. 2011). Reducing the energy penalty for CO_2 capture is one of the largest possibilities for lowering the CCS costs.

Table 13.2 Physical and chemical separation approaches for CO_2 capture

Method	Description	Example materials	Advantages	Technological challenges	References
Absorption	CO_2 from gas stream dissolves in fluid and subsequently released from fluid by changing pressure or temperature	Aqueous amine solutions, chilled ammonia, ionic liquids	Established technology with numerous solvent options. Rapid improvements in energy requirements achieved	Reducing energy for regeneration. Solvent degradation	Boot-Handford et al. (2014), Mathias et al. (2010), Rochelle (2009)
Adsorption	CO_2 from the gas stream adsorbs onto solid and subsequently released by changing pressure or temperature	Zeolites, metal organic frameworks, activated C	Potentially lower energy requirement for regeneration	Adsorption capacity and kinetics	Hasan et al. (2013), Banerjee et al. (2008)
Cryogenic	CO_2 is cooled until it becomes solid that separates from the gas stream	None	Avoids the need for solvents or sorbents. Lower energy requirements	Solid separation and handling	Tuinier et al. (2010)
Membranes	CO_2 from pressurized gas stream is preferentially transported through membrane	Polymer membranes, inorganic membranes	Avoids regeneration and energy requirements	Permeability selectivity	Yang et al. (2008), Du et al. (2011)
Mineralization	CO_2 reacts with calcium or magnesium-bearing rocks to form magnesite or calcite	Magnesium silicates, alkali rich waste streams	CO_2 is converted to a solid substance that can be reused as a building material or disposed of surface facilities	Rate of reactions, Mass reactants such as sources of Ca, Mg	Goff and Lackner (1998), IPCC (2005)

13.5.3 Oxy-Fuel Combustion Carbon Dioxide Capture

The third approach for CO_2 capturing is the O_2 combustion, where fuel is combusted under pure O_2 stream or a mixture of CO_2 and O_2 (IPCC 2005). Oxy-fuel combustion CO_2 capture is generally a post-combustion capture, but since the fuel is burnt in pure O_2 instead of air, the exhaust gas contains CO_2 and water. Therefore, CO_2 can be separated by condensation of the water. Coal gasification and oxy-combustion both produce gases that are more enriched in CO_2, thus simplifying or avoiding the need for liquid solvent extraction, but requiring more extensive investment in pre-combustion equipment (Thambimuthu et al. 2005; Rubin 2008). The advantage of oxy-fuel combustion is the lower cost of post-combustion capture. O_2 is generally separated from air (Fig. 13.5). In addition to CO_2 and water vapor, the post-combustion waste may also contain mixtures of trace gases, including O_2 and CO, thus avoiding the need to separate CO_2 from N_2 after combustion. The separation of O_2 from the air is a mature technology, albeit energy-intensive process using cryogenic separation. However, the air separation to generate pure O_2 required for combustion can be large for large scale power plants. Almost three times more O_2 is required for oxy-combustion power plant compared to IGCC power plant of equal size (Folger 2013). Oxy-fuel combustion CO_2 capture is also the least developed technology among the three CO_2 capture processes. However it continues to gain interest and development, likely due to its potential advantage of greatly simplifying the overall CO_2 capture process and avoiding most of the chemical processing associated with pre-combustion and post-combustion CO_2 capture. Oxy fuel combustion also has the potential to increase existing process efficiency in retrofit applications (Benson et al. 2012). The viability and appropriate choice of CO_2 capture depends on the specific power plant. A schematic flow of CO_2 sources, capture techniques, purification and geological storage is presented in Fig. 13.6.

13.5.4 Carbon Dioxide Capture from the Air

Some researchers have been promoting the idea of capturing CO_2 directly from the ambient air using various chemical absorption approaches (APS 2011). This approach will be able to capture non-point CO_2 sources such as mobile transportation fuels from mostly oil. Practical and economically competitive CO_2 capture is a major challenge due to low ambient air pressure and low CO_2 concentration at about 400 ppm. The ultra-low CO_2 partial pressure of just 0.0004 bar (40 Pa) atmospheric pressure likely will require strong bases to capture most of CO_2 from the air as well as very large adsorber or absorber. This could also mean large energy requirements to regenerate the CO_2 from strong basic sorbent into high purity CO_2 stream for compression.
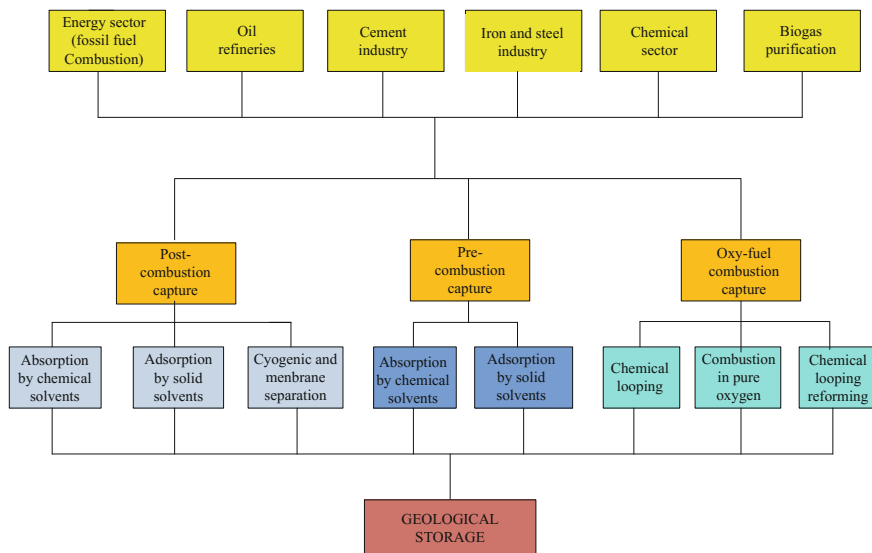
Fig. 13.6 Schematic diagram illustrating different possible sources of carbon dioxide, different capture, separation approaches and storage

13.6   Carbon Dioxide Transport

As outlined previously, CCS is a more distinctly technological approach that begins with the capture and compression of CO_2 from large point sources, followed by purification to obtain nearly pure CO_2. Purified CO_2 is then compressed to liquid for transport and injection. The chemical and physical properties of compressed CO_2 play an important role in the feasibility of geologic CO_2 sequestration. CO_2 becomes a liquid when compressed to high pressures for transport. When CO_2 is subjected to the combination of higher pressures and temperatures that characterize geological CO_2 injection sites, it becomes what is known as a supercritical fluid. The CO_2 is generally transported in three states: gas, liquid, and solid through the land or by the sea. The commercial transport uses tanks, ships and pipelines for gaseous and liquid CO_2. Gas transported at a close to atmospheric pressure occupies large volume and large facilities are also needed for its storage. Therefore, compression allows volume reduction, and it can further be reduced by liquefaction, solidification, or hydration. Liquefaction is an established technology for gas transport by ship—such as liquid petroleum gas (LPG) and liquid natural gas (LNG) which can be transferred to CO_2 transport. Experience of transporting CO_2 by truck or ship is mainly found in the food and brewery industry where CO_2 is generally transported as a compressed liquid (e.g., $-50\ °C$, 0.7–0.8 MPa). Transportation of CO_2 by ship or train requires infrastructure development, including loading and unloading and temporary CO_2 storage, which makes the

option more costly. Transport through ships, trucks, and train is not regarded as preferred option for large-scale systems (IPCC 2005). Pipeline transport is currently the most matured transportation option. In pipeline transportation, the volume is reduced by transporting CO_2 at high pressure which is routinely done in gas pipelines operating at pressures between 10 and 80 MPa. The large-scale storage projects will generally require pipelines transport, and for pipeline transport, CO_2 is compressed to a liquid or dense phase at the ambient temperature (i.e., CO_2 pressure above ~ 8 MPa), with temperature varying with location, but generally lower than 20 °C (Benson et al. 2012). At this temperature and pressure, CO_2 would have a density between 800 and 1200 kg m^{-3}, where larger mass per unit volume can be transported. There are more than 5800 km of CO_2 pipelines in operation in the United States transporting liquid or dense phase CO_2 with pressures above 7.38 MPa and sectioned typically less than 30 km (Parfomak et al. 2009). The CO_2 pipelines are generally made out of carbon-steel, for transporting relatively clean CO_2 without internal coating. Studies have demonstrated that CO_2 transport by pipeline does not pose higher risk of accidental leak than that tolerated for transporting other hydrocarbons by pipelines (Liu et al. 2015; Han et al. 2015; Koornneef et al. 2010). However, transporting CO_2 by pipeline through densely populated area will require modifications to current standards for increased safety, including increased pipeline wall thickness, and protection from damage (e.g., through burying the pipeline) (Koornneef et al. 2010).

13.6.1 Transportation Operational Issues

13.6.1.1 Pressure Drop

Frictional loss of pressure occurs as the CO_2 flows through the pipeline, which is dependent on pipeline diameter, CO_2 flow velocity, viscosity and density, and pipe roughness factor. At constant temperature, pressure drop in a 50.8 cm pipeline transporting CO_2 in dense phase is about 30 kPa km^{-1} (Gale et al. 2009). Pressure drop is generally overcome by increasing the inlet pressure to the pipeline so as to maintain at least 7.38 MPa, or installation of pressure boosters every 100–200 km for longer pipelines (Benson et al. 2012).

13.6.1.2 Corrosion

Components such as SO_2, NO, and H_2S form acid compounds in the presence of water, and it is highly corrosive. In addition, H_2S could react with C steel and form film of iron sulfide (FeS) which may dislodge at times and cause operational problems in the CO_2 compression units. Control of water content by dehydration is therefore essential for safe, cost-effective pipeline operation. Other options include use of protective coating and corrosion inhibitors, but these increases pipeline cost.

13.6.1.3   Hydrate Formation

CO_2 may form hydrate compounds in the presence of water and H_2S, which can block or plug the pipeline and also damage equipment. Hydrate formation can largely be stopped by drying the CO_2 and removing free water. The maximum allowable water content is in the range of 50–500 ppm (Benson et al. 2012).

13.6.1.4   Other Factors

Other important factors for CO_2 transport include operating temperatures and impurities. Operating temperatures are generally dictated by the temperature of the surrounding soil or water, and can range from below zero to as high as 20 °C in the tropics. CO_2 cools dramatically during decompression, as a result, pressure and temperature must be controlled during routine maintenance. Also, depending on the source of the flue gas, and type of CO_2 capture process, CO_2 streams may contain trace concentrations of H_2S, SO_2, NO, O_2, HF, Hg, N_2, and Ar which may impact the physical state of CO_2 stream, CO_2 compressibility, CO_2 density, pipeline integrity, safe exposure limits, and the minimum miscibility pressure of the CO_2 in oil, and impact the use of CO_2 in EOR among other factors (Benson et al. 2012).

13.6.2   Cost of Carbon Dioxide Capture and Transportation

The estimated cost of CO_2 capture in power plants and industrial production processes vary greatly and generally have high uncertainty. Global CCS Institute indicates that CCS would add approximately 40% to the cost of power production in IGCC and natural gas combined cycle plants, and between 50 and 80% for coal-fired power plants with post-combustion or oxy fuel combustion (Irlam 2015). For steel production, CO_2 capture will add 10–15%, while for cement production CO_2 capture will add 39–52% costs. Overall, costs for CO_2 capture will remain highly uncertain until industrial-scale projects are fully implemented. The CO_2 transport costs are a function of pipeline length, diameter, material, route of pipeline, and the safety requirements among other things. Overall, the transport of CO_2 by pipeline benefits from economies of scale, i.e., average transport costs decrease as scale increases. Therefore, larger diameter pipelines are preferred.

13.7   Carbon Dioxide Storage

Storage of CO_2 in deep geological formations has been under development since circa 2000, and is now considered as one of the feasible components in the portfolio of options for reducing GHG emissions. Other options for CO_2 storage which have

been assessed include ex-situ mineralization, ocean storage in dissolved or liquid forms, and CO_2 reuse in the chemical industry. Among the options of storage, only storage in the geological formation is considered to have the capacity, permanence and environmental performance necessary for storage at large scale needed to reduce CO_2 emissions (IPCC 2005; Benson et al. 2012). The geological formations suitable for CO_2 storage occur in sediment basins, and include depleted or depleting oil and gas reservoirs and salt water (saline) filled rocks. The ongoing large-scale industrial scale demonstration projects together with CO_2-EOR, mostly in North America, have provided valuable experience base for assessing the potential for geological storage of CO_2 and efficacy of C storage. The projects have also demonstrated that, for CCS to contribute to the scale needed for large reduction of CO_2 emissions required, a 250-fold increase per year is needed compared to amount currently being sequestered, and effectively sequestering such large volumes requires building a strong scientific foundation for predicting the coupled hydrological-geochemical-geomechanical processes that govern the long-term fate of CO_2 in the deep sub-surface geological strata. Also, additional methods for characterizing and selecting suitable storage sites, subsurface engineering to optimize performance and cost, safe operations, monitoring technologies, remediation methods in case there is a leak, as well as regulatory oversite are required. In addition, the international institutional approaches for managing long-term liability risk are also needed.

Beginning in the early 1990s, there was a great deal of interest in storing CO_2 in the ocean, and two approaches were pursued: (i) biological sequestration through ocean fertilization, and (ii) direct injection of concentrated stream of CO_2 in the ocean. In 2001, the Southern Ocean Iron Experiment was conducted in the Southern Pacific (Buesseler et al. 2004; Coale et al. 2004). Results from this and similar experiments demonstrated rapid increases in biological productivity. However, many questions regarding long-term ecosystem impacts and the effectiveness of this technique in lowering atmospheric CO_2 concentration remain unanswered (Breitbarth et al. 2010). Also, the permanence of stored CO_2 remains debatable. As a result, ocean fertilization is not under serious consideration for large-scale CO_2 storage.

Injection into the mid-depth ocean (1000–3000 m deep) where CO_2 can stay for thousands of years before returning to atmosphere through ocean circulation, or near-bottom of the ocean to create stationary pools of CO_2 have also been proposed. Concerns about biological impacts, high costs, lack of permanence of ocean storage, and also concerns about public acceptance have decreased interest and investment in ocean storage. Although under sufficiently cold ocean at water depths greater than 3000 m, CO_2 transitions from being lighter than water to heavier than water, and remains on the ocean bottom (Schrag 2009), over time the CO_2 will dissolve into ocean water leading to ocean acidification and gradual release back to the atmosphere. It has been proposed to inject the CO_2 under thin layer of ocean bottom sediments therefore, combining some aspects of geological and ocean storage (Schrag 2009). This technique is not well developed, and is a subject of research. Another ocean storage which has been proposed combines CO_2 with sea

production of methane hydrates, such that as CH_4 is released from hydrate structure, CO_2 could replace it (Ohgaki et al. 1996). Of these options, currently only the storage in geological formation is considered to have capacity, permanence, and environmental performance necessary for CO_2 storage at scale needed to materially reduce CO_2 emissions and potentially mitigate climate change (IPCC 2005; Benson et al. 2012).

13.7.1   *Storage of Carbon Dioxide in Deep Geological Formations*

The Earth's crust is made up primarily of three types of rocks: (i) igneous rocks—formed by cooling magma from either volcanic eruptions or magmatic intrusions far beneath the land surface, (ii) sedimentary rocks—formed as thick accumulations of sand, clay, salts, and carbonates over millions of years, and (iii) metamorphic rocks—these can be of either origin that have undergone deep burial with accompanying pressure and thermal alteration. The sedimentary rocks located in the so called sedimentary basins have been the primary focus for ongoing geological storage of CO_2 because the geological storage on geological timescales has already been proven through the presence of oil and gas accumulation in these formations. Sedimentary basins underlie much of the continents, and some of them are co-located with major CO_2 emission sources. In the past few years, there has also been a significant effort to understand the potential of volcanic rocks—primarily basalt, which rely on geochemical reactions between CO_2 and basalt for storage of CO_2 (McGrail et al. 2006). Motivation for evaluating storage in basaltic rocks are: (i) some countries with large CO_2 emissions—such as India, Brazil, and USA are underlain by basaltic rocks, (ii) to test the hypothesis that large fraction of stored CO_2 would be converted to stable minerals such as calcite or magnesite and a assure permanent storage of CO_2 (Aradottir et al. 2011; Oelkers and Cole 2008).

Sedimentary basins often cover thousands of meters of sediments with tiny pore spaces (e.g., 10^{-3}–10^2 µm diameter) in the rocks which are filled with salt water (saline aquifers) and where oil and gas reservoirs are generally found. Sedimentary basins also consist of many layers of sand, silt, clay, carbonates, and evaporate rock formations—composed of salt deposited from evaporating water. The sand layers provide storage of oil, water, and natural gas. The silt, clay, and evaporate layers provide the seal that can trap these fluids underground for millions of years and longer. Geological storage of CO_2 in the sedimentary basins below silt and clay layers aim at taking advantage of the seal much the same way that oil and natural gas are trapped (Gunter et al. 2004). Possible storage formations in sedimentary basins include oil reservoirs, natural gas reservoirs, saline aquifers, and also unmineable coal beds. The overlying thick continuous layer of shale, silt, clay, or evaporate is the single most important feature of geologic formation that is suitable

for geological storage of CO_2 which prevent upward migration of CO_2 by combination of viscous and capillary forces. The presence of oil and natural gas reservoirs is a good demonstration of presence of suitable reservoir seal. For the saline aquifer formations, a significant site characterization effort is required to demonstrate the presence of a satisfactory primary seal. The important attributes of the seal include low permeability (in the order of 10^{-18} m^2 or less) and high capillarity entry pressure (1 MPa or more) (de Coninck and Benson 2014). In addition to CO_2 storage below the seal, CO_2 may also be retained through secondary trapping mechanisms such as solubility (Gilfillan et al. 2009), residual trapping and mineral trapping (Michael et al. 2010; Krevor et al. 2012). In saline aquifers, where pore spaces are initially filled with water, it has been hypothesized that after the CO_2 have been underground for hundreds to thousands of years, chemical reactions will dissolve some or all of CO_2 in the saltwater, with some fraction converted to carbonate minerals, thus becoming part of rock itself (Gunter et al. 2004). These are generally termed as secondary trapping mechanisms that continue to increase storage security over time, and have been the subject of significant research over the years (Benson and Cole 2008).

Another proposed mechanism for trapping CO_2 is the so called solubility trapping in saline aquifers, which is dependent on pressure, temperature, and the salinity of the brine (Koschel et al. 2006). Under typical geological formation storage conditions, the solubility of CO_2 in brine ranges from 2 to 5% by mass, and the laboratory studies have indicated that dissolution of CO_2 tend to be rapid at high pressure when water and CO_2 share the same pore space (Holloway 2008). However, in real injection system, the dissolution of CO_2 could be limited by the variability of contact area between CO_2 and the fluid phase. Solubility trapping decreases the amount of CO_2 subject to buoyant forces that drives CO_2 upwards.

The third type of secondary trapping is the 'mineral trapping' which occur when acidic brines enriched with CO_2 react directly or indirectly with minerals in the geologic formation, leading to precipitation of stable secondary carbonate minerals (Gunter et al. 2004). This mechanism generally traps CO_2 permanently, although significant degree of mineral trapping could take several thousands of years due to silicate mineral dissolution and carbonate mineral precipitation. Hence the impact of mineral trapping may not be realized until far in the future. In addition, the amount of CO_2 trapped by this mechanism is dependent on mineralogical makeup of the storage reservoir rock. For example, rocks with large fraction of feldspar minerals are expected to have significant amount of mineral trapping, while quartz-dominated reservoirs have little or no mineral trapping. Secondary trapping mechanisms are not substitute of high-quality seal, however, they do act over decadal to millennial timescales thereby increase the CO_2 storage security over time, and their relative importance will change over time depending on hydrogeological attributes of the storage site, such as mineralogy multiphase fluid flow properties stratigraphy and structural formation.

13.7.2   Issues Associated with Carbon Dioxide Storage

The ability to scale-up the existing operations is the central among the issues to be resolved before CCS can emerge as a viable option for global CO_2 emission reduction. Therefore, if CCS is to be implemented and have impact on CO_2 emission reduction, at least 20- to 25-fold increase in the amount injected underground will be required annually for CO_2-EOR today. It will also require an infrastructure on the scale of current oil industry. The ability to scale up the existing operations is central among the issues to be resolved before CCS can be considered as one of the viable option for the global CO_2 emission reduction. Achieving such a scale-up relies on several factors, including storage capacity, injectivity, risk management to avoid detrimental environmental impacts such as groundwater pollution, induced seismicity, and ecosystem degradation, as well as availability of intervention methods to remediate unanticipated leakage of CO_2 or other unintended and unplanned events (Table 13.3).

Table 13.3 Risks of CO_2 storage, environmental impacts and risk management approaches

Environmental risk	Impacts	Management
Leakage of CO_2 into the atmosphere	Lack of effectiveness of CCS	Proper site selection, effective monitoring, remediation of leakage pathways
Underground leakage and accumulation of elevated concentrations of CO_2 in ecosystems	Damage to CO_2 sensitive habitats	Proper site selection, monitoring of soil gas, remediation of leakage pathways and ecosystem cleanup
Exposure of high CO_2 to humans	Chronic or acute health concerns from CO_2 exposure	Proper site selection, monitoring, controlled access, remediation of leakage pathways
Leakage of CO_2 into groundwater	Acidification of groundwater and potential dissolution of toxic minerals	Effective site selection, monitoring of groundwater quality, restrict groundwater use if contaminated, remediation of leakage pathways
Leakage of hydrocarbon into groundwater	Contamination of groundwater with hydrocarbon compounds	Proper site selection, monitoring of groundwater quality, restricted use of contaminated water, remediation of leakage pathways, groundwater cleanup
Displacement and leakage of saline brine into drinking water aquifers or surface water	Contamination of groundwater or surface water with dissolved salts	Proper site selection, monitoring of groundwater quality, restricted use of contaminated water, remediation of leakage pathways, groundwater cleanup
Induced seismicity	Potential structural damage as well as felt ground motion	Proper site selection, monitoring of both pressure and seismic activity, limit pressure buildup

13.7.2.1 Permanence of Geologically Stored Carbon Dioxide

One of the key questions for the geologic CO_2 storage is how long the CO_2 will remain trapped underground? Several lines of evidence have suggested that for a well-selected and managed storage formations the retention rates may be high and more than sufficient for the purpose of avoiding CO_2 emissions into the atmosphere. These specifically include: (i) natural oil, gas, and CO_2 reservoirs that have demonstrated the buoyant fluids such as CO_2 can be trapped underground for millions of years, (ii) industrial analogues such as gas storage, CO_2-EOR, acid gas injection, liquid-waste disposal operations that have developed methods for injecting and storing fluids without compromising the integrity of caprock or storage formation, (iii) geochemical processes that can contribute to long-term CO_2 retention—such as physical CO_2 trapping beneath low permeability or impermeable rocks, dissolution of CO_2 in the brine, capillary trapping of CO_2, adsorption on unmineable coal, and mineral trapping. Overall, these trapping mechanisms increase the security of storage over time, and minimizes potential leakage of CO_2 to the land surface, (iv) experiences gained from projects which had large amount of monitoring data, such as Sleipner Project in North Sea and Weyburn Project in Saskatchewan, Canada has demonstrated a high degree of CO_2 containment so far.

Overall, the permanence of geologic sequestration depends on the combined effectiveness of several physical and chemical mechanisms that combine to trap this supercritical CO_2. In general, CO_2 is less dense than the saline groundwater it displaces in the pore space of the rock formations where it is injected. It will therefore, rise buoyantly until trapped beneath an impermeable barrier, or seal formed by a stratigraphic or other structural discontinuity. This physical trapping mechanism is comparable to the natural geologic trapping of oil and gas and can theoretically retain fluids for thousands to millions of years. CO_2 that is not physically trapped in this manner may escape through leakage pathways or it may migrate slowly through the rock pore space and become trapped as a residual fluid held in place by molecular surface tension (Ide et al. 2007). It is expected that some of the injected CO_2 will eventually dissolve in groundwater (brine), and some may be trapped in the form of carbonate minerals formed by chemical reactions with the surrounding rock (Kharaka et al. 2006a). However, all of these processes are susceptible to change over time after CO_2 injection (Hovorka et al. 2006), necessitating long term monitoring during and after the injection has stopped.

In general, the physical trapping mechanisms are viewed as more important over short timescales (a few decades); the relative importance of chemical dissolution and mineral reactions increases over time scales of centuries to thousands of years (Benson et al. 2005; Benson and Cole 2008). Because the permanence of geologic sequestration is one of its principal benefits, there is a critical need to understand the potential for leakage of injected CO_2 back to the atmosphere. Faults, fractures, and stratigraphic discontinuities may offer pathways for CO_2 leakage. Many potential structural traps are known in areas where oil and gas have been extracted, and CO_2 injection can enhance oil recovery, so these areas tend to be preferred sequestration

injection sites. Unfortunately, these areas are also perforated by existing or abandoned wells that may act as conduits for leakage (Bachu and Celia 2009; Gasda et al. 2004).

The injection process itself may affect the geomechanical integrity of trapping structures (Hawkes et al. 2005). Valuable information about potential leakage can and have to be gathered in studies of natural geologic analogs of CO_2 storage and venting (Fessenden et al. 2009; Heath et al. 2009; Evans et al. 2001). However, many aspects of geologic CO_2 sequestration have no typical natural analogs, and therefore, much of the existing understanding has been learned from ongoing commercial-scale demonstrations of CO_2 sequestration operations such as those in Sleipner, Norway and Weyburn, Saskatchewan and In Salah, Algeria (Arts et al. 2008; Mathieson et al. 2011; White 2009), and other smaller pilot projects such as Frio, Texas, USA, Illinois, USA, Japan and others (Hovorka et al. 2006, 2013; Picard et al. 2011). Sequestration field tests at smaller scales have also become a primary venue for learning how to monitor and anticipate the fate of injected CO_2 (McPherson 2009; DOE 2012). Numerical models are an essential tool for understanding the complex interactions among the many factors that control fluid chemistry and transport. Models of geochemical interactions, multiphase fluid transport, and the combined effects of geochemistry and transport have been widely utilized to model the reservoir conditions and reaction properties (Gunter et al. 1997; McPherson and Cole 2000; White et al. 2005; Kaszuba and Janecky 2009). However, the uncertainties in model simulations must be quantified (Zerai et al. 2009) to provide meaningful assessments of risk. A wide range of scientific and engineering expertise is required for understanding the permanence of geologic CO_2 trapping mechanisms over the full range of potential storage time scales (Doughty and Myer 2009; McPherson 2009).

13.7.2.2 Storage Capacity

Just as the potential global amount of terrestrial C sequestration is limited by available land area, the global capacity for geologic carbon sequestration is constrained by the pore volume and distribution of potential storage sites. One frame of reference for geologic storage of injected CO_2 is to compare the volume of extracted fuels to the volume of injected CO_2 produced by combustion of equivalent fuel volumes. Generally, the combustion of oil and bituminous coal produces volumes of compressed supercritical CO_2 that exceed the volume of extracted fuel by factors of three and four, respectively, while the combustion of natural gas produces a nearly equal volume of compressed CO_2. In addition, the storage of CO_2 in dissolved form requires saline groundwater volumes many times larger than the volumes of the extracted fuels. Therefore, the potential underground CO_2 storage capacity needed is much greater than the vast existing operations of the fossil fuel industries. The large difference between the volume requirements of compressed and dissolved CO_2 is one source of uncertainty in estimates of global geologic

sequestration capacity. An analysis (Dooley et al. 2006) suggests that the global storage capacity of physical traps associated with depleted oil and gas reservoirs is about 220 Pg C. The potential global storage capacity of deep porous rock formations that contain saline groundwater is much larger, and estimated to be approximately 3000 Pg C (Dooley et al. 2006). However, these formations are not as well-characterized as oil and gas reservoirs, and less is known about the effectiveness of trapping mechanisms at these sites. Unmineable coal beds have also been proposed for potential CO_2 storage, particularly in conjunction with coalbed methane recovery (Gunter et al. 1997). Overall, the global capacity estimates of geologic CO_2 sequestration remains a challenge because of the many differences in assumptions and the calculation methods. Most recent estimates provide an upper bound on the CO_2 storage capacity of sedimentary basin ranging from 5000 to 25,000 Pg CO_2 (Benson et al. 2012). Oil and gas reservoirs are anticipated to have a storage capacity of an order of 1000 Pg CO_2 (Benson et al. 2012), but, these reservoirs are limited geographically to hydrocarbon-rich regions of the world, and oil and gas reservoirs may not be available until oil and gas are fully depleted or until market conditions favor CO_2-EOR. Saline aquifers are assessed to have largest storage capacity with the global estimates ranging from 4000 to 23,000 Pg CO_2. There is a limited experience for assessing effectiveness and safety of saline aquifers storage, however. Also the uncertainty persists about how much of the estimated large storage potential can actually be utilized (Juanes et al. 2010; Bradshaw et al. 2007; Ehlig-Economides and Economides 2010). A 500-fold scale up of the existing saline aquifer storage project would be required for Pg scale storage. Unmineable coal beds are considered to have low storage potential.

Standardization and transparency of capacity assessment methodologies are needed so that improved estimates of the potential effectiveness of geologic CO_2 sequestration compared to current and future CO_2 emissions can be assessed (Bachu et al. 2007; DOE 2012; Burruss et al. 2009; McPherson 2009). To fully assess the potential for geologic carbon sequestration, economic costs and environmental risks must also be taken into account. Many of the factors affecting geologic CO_2 sequestration depend on local conditions, and will vary according to the type of storage formation. The depleted oil and gas reservoirs are generally well characterized and are less prone to unknown risks. However, they are of limited in capacity and geographic distribution, and may require greater proportional investment in infrastructure. The potential capacity of formations containing saline water is larger and more widely distributed, but few of these formations are well-characterized, leading to large uncertainty in capacity estimates. Unmineable coal beds may have the advantages of proximity to large power plants and CH_4 recovery, but their storage characteristics may be poorly characterized, and also potential future coal and natural gas resources may be rendered unusable by CO_2 storage.

13.7.2.3 Pressure Build up, Injectivity and Induced Seismicity

When CO_2 is injected into a storage reservoir, the pressure increases due to combination of viscous forces associated with multiphase flow of CO_2 within the plume and displacement of in situ fluids (Nordbotten et al. 2005). The magnitude of pressure buildup depends primarily on the permeability and thickness of the storage reservoir and the injection rate. In the case of sealed reservoir (i.e., sealed on the top, bottom, and sides), pressure also increases due to compression of pore—filling fluids (Zhou et al. 2008). Monitoring of pressure buildup and associated geochemical effects are needed for CO_2 storage projects. Concerns over how much excessive pressure buildup limits storage capacity in saline aquifers (Ehlig-Economides and Economides 2010) and induced seismicity (Zoback and Gorelick 2012) have raised concerns among researchers. However, some researchers have argued that these concerns are misplaced for the existing million-tonnes CO_2 yr^{-1} injection projects (Cavanagh et al. 2010). Some techniques for pressure management include (i) injection rate control (Birkholzer et al. 2009), and (ii) brine extraction (Birkholzer et al. 2009). Overall, pressure buildup is a manageable issue (Chadwick et al. 2010).

Injection of CO_2 into saline aquifers could lead to slip along preexisting faults and to associated seismicity (Zoback and Gorelick 2012). In general, induced seismicity is gaining attention with disposal of wastewater from hydraulic fracturing operations for shale gas development in the United States (Frohlich 2012) and natural gas extraction in the Netherlands. Researchers are actively investigating whether and how much induced seismicity is a constraint to CO_2 storage and how to manage injection operations to avoid it. There is a need for research to establish the extent of induced seismicity and its potential limitation for CO_2 injection, as well as how to manage the injection operations to avoid it.

13.7.2.4 Other Issues Associated to Carbon Dioxide Injection

Other risks include deformation of the land surface (Jung et al. 2013), contamination of potable water supplies, and adverse effects on ecosystems and human health. Numerous regulatory issues also affect the implementation of geologic sequestration, including determination of rules affecting injection wells, post-injection ownership, and liability across multiple jurisdictions and even among different countries. As with terrestrial sequestration, geologic sequestration cannot be accomplished in isolation from a broad range of environmental and societal concerns (Sundquist et al. 2008).

13.8   Risks of Geological Storage of CO_2

Two broad categories of risks of CO_2 storage can be identified: (1) risks associated with the release of CO_2 back to the atmosphere, and (2) health, safety, and environmental risks associated with the local impacts of storage operations and potential leakage out of storage reservoir. The consequences of release of CO_2 back to atmosphere are that the CCS may be less effective as a mitigation measure than the anticipated. Also there could be financial liabilities associated in buying credits and also assuming the responsibility for those emissions. Overall, the risks of release of CO_2 to the atmosphere are the greatest during the period of CO_2 injection, which for any particular project are limited to several decades. Legal and administrative mechanisms for managing long-term liabilities beyond the period of operation and post-injection assurance monitoring are currently under development by government agencies worldwide. The health, safety, and environmental concerns of CO_2 storage are similar to those generally associated with oil and gas fields—such as habitat fragmentation, infrequent uncontrolled release from wells, noise, as well as road traffic. Additionally, if CO_2 or brine leaks out of storage reservoir, it could affect groundwater quality and result in locally hazardous concentrations of CO_2 in the air. It can also cause micro-seismicity if injection pressures are very high.

Based on existing scientific understanding of the processes controlling CO_2 migration in the subsurface, CO_2 should remain securely stored in the geological formations for thousands of years or longer if these conditions are met: (i) the seal has low permeability and high capillary pressure to prevent migration into the seal, (ii) CO_2 cannot migrate around the edge of the seal or through breaches in the seal caused by leaking wells, faults, or fractures, (iii) the injection pressure is low enough to avoid fracturing the seal (Benson et al. 2012). Although these principles are clear and straightforward, the main challenge is to identify sites that meet these conditions, since the subsurface geology is naturally complex, and geological storage sites are by necessity large, with CO_2 plumes potentially covering hundreds of km^2 area, and this makes it harder to assess the security of storage.

Although CO_2 is generally non-toxic inert gas, exposure to concentrations in the excess of several percent can lead to adverse consequences, particularly since CO_2 is denser than air, hazardous situations arise when large amount of CO_2 accumulate in low-lying confined or poorly ventilated spaces. If large amounts of injected CO_2 escape from a storage site, it could present risks to health and local environment. However, hazardous conditions would only persist several hundred meters from the site of release (Aines et al. 2009). Such large release could be associated with surface facilities, injection wells, or leakage which remained unchecked, and could impact groundwater and ecosystems. Persistent leaks could suppress respiration in the root zone or result in soil acidification and leading to tree kills. Tree kills have been observed at the soil gas concentration in the range of 20–30% at Mammoth Mountain in California caused by volcanic outgassing of CO_2 which has been occurring for several decades (Martini and Silver 2002).

   Potential local health, safety, and environmental concerns from geological storage include: (i) occupational risks associated with well and field operations, (ii) leakage of CO_2 or brine out of storage and migration to drinking water aquifers, (iii) resource damage near well head due to unwanted CO_2 migration into nearby mineral resources, (iv) ecosystem impacts in the event that CO_2 is released into soil, wetlands or surface waters, (v) public safety risks from exposure to elevated CO_2 concentrations if CO_2 is released at the surface, and (vi) structural damage associated with land surface deformation or microseismicity. Overall, the extensive experience with CO_2-EOR and injection of CO_2 in the ongoing demonstration projects in general indicates that risks from geological CO_2 storage facilities are manageable using standard engineering controls and procedures. The highest risk event that could occur in CCS is loss of well control which is infrequent.

13.9   Monitoring and Risk Management

Monitoring play a key role to observe the behavior of the injected CO_2, calibrate and validate predictive models and provide any early warning that leakage may be imminent, so that remediation measures such as plugging abandoned wells can be implemented. The regulatory oversight capacity is also needed to ensure due diligence for site selection, engineering, operation, monitoring, verification, and necessary remediation for CO_2 storage project if needed. Combination of monitoring techniques—e.g., geophysics, hydrology, and geochemistry that monitor injection rates and pressure, tracks migration of the CO_2 plume, detect leakage out of storage reservoir, and detect microseismic activity are available from a variety of other applications including oil and gas industry natural gas storage disposal of liquid hazardous waste in deep geological formation, groundwater monitoring, food preservation and beverage industries, fire suppression, and ecosystem research. These techniques have been adapted for use in CO_2 sequestration monitoring. Monitoring has been a key element of the industrial scale storage projects (Arts et al. 2008; Mathieson et al. 2011; White 2009). Most of these techniques have also been demonstrated at small-scale pilot projects around the world (Hovorka et al. 2006, 2013; Jenkins et al. 2012; Martens et al. 2012).
   Geophysical monitoring methods can be used to monitor the location of CO_2 plume (Arts et al. 2008; Sato et al. 2011; Couëslan et al. 2013). Seismic imaging can detect compressional wave velocity and attenuation caused by the presence of separate phase CO_2 (Couëslan et al. 2013). Electromagnetic imaging can detect decrease in electrical conductivity in the pore spaces of the rock, while gravity measurements detect decrease in bulk density of the rock caused by presence of CO_2 (Bergmann et al. 2012). Seismic methods for monitoring have been used successfully at Sleipner, Weyburn, the Frio Brine Pilot and Otway Basin Pilot projects and others (Arts et al. 2008; Hovorka et al. 2006; White 2009; Pevzner et al. 2011). Geochemical measurements are generally deployed in two types:

(i) direct techniques—including measurement of brine chemistry and introduced or natural tracers in samples obtained from injection horizons in observation wells, (ii) monitoring the near-surface for possible CO_2 leakage in the immediate vicinity of injection and observation wells, and also from soils and shallow groundwater wells within the injection area. Most of geochemical methods have been used primarily for the pilot scale tests due to the insights they provide, which is important in understanding the interactions between CO_2 and the storage reservoir rocks and fluids (Kharaka et al. 2006a, b). They have also been used extensively at the Weyburn Project (Emberley et al. 2005) and the Illinois Basin Decatur Project (Ussiri, Personal communication). They include: pH, alkalinity, soil gas compositions, and the stable isotope geochemistry, among others. The most diagnostic indicator of brine-CO_2 interactions which indicates breakthrough of CO_2 to monitoring wells is pH. Enrichments of metal constituents compared to pre-injection are indicators of mineral dissolution reactions occurring at depth during brine-CO_2 rock interactions (Emberley et al. 2005; Kharaka et al. 2006a, b). Monitoring of surface fluxes can also directly detect and measure leakage. Surface CO_2 fluxes can be measured directly with eddy covariance towers, flux accumulation chambers and field-portable high-resolution infrared (IR) gas analyzers (Lewicki et al. 2007, 2009; Lewicki and Hilley 2009; Spangler et al. 2010). Much progress has been achieved to quantify and improve detection levels and also increase the number of available surface monitoring of CO_2 leakage techniques (Spangler et al. 2010; Krevor et al. 2010).

13.10   Bioenergy Coupled with Carbon Dioxide Capture and Storage (BECCS)

Bioenergy coupled with CCS (BECCS) (Liu et al. 2011) can mitigate climate change through negative emissions if CCS can be successfully deployed (Cao and Caldeira 2010; Creutzig et al. 2015). BECCS or Bio-CCS is defined as a process in which CO_2 originating from biomass is captured and stored. It can be through energy production process or any other industrial process with CO_2-rich process streams originating from biomass feedstocks. BECCS has a potential for negative emissions which can also compensate for emission from other sectors, especially transport. BECCS is markedly different from fossil CCS because it reduces CO_2 emissions by storing CO_2 in long-term geological sinks while continually sequestering CO_2 from the air through regeneration of biomass resource feedstock. An example of BECCS is the Illinois Basin-Decatur Project, a demonstration project that captures CO_2 from ethanol fermentation and inject it into Mount Simon geological formation—which is projected to store 1 Mt CO_2 yr^{-1} (Gollakota and McDonald 2012). In addition to risks associated with CCS, bioenergy production could reduce C stocks through land use conversion, land disturbance through

tillage. Similarly, use of inorganic fertilizer in bioenergy crop production such as corn production for ethanol causes increased N_2O emissions. Therefore, lifecycle analysis is needed to evaluate the mitigation potential of BECCS.

13.11   Oceanic Sequestration

During 2006–2015, world's oceans accounted for a global net uptake of about 2.6 ± 0.5 Pg C yr^{-1} which occurs naturally through chemical reactions between dissolved inorganic carbon in the ocean surface and the increasing CO_2 concentration in the atmosphere (Le Quéré et al. 2015, 2016). This natural ocean uptake of CO_2 was a primary interest of Roger Revelle when he described the "large-scale geophysical experiment" more than 50 years ago. The oceans will continue to be the primary long-term sink for anthropogenic CO_2 that is not sequestered by other means. The chemical reactions between atmospheric CO_2 and the ocean surface occur rapidly, and over time any absorbed CO_2 is mixed downward throughout the oceans. Over the geological time scales, increases in atmospheric CO_2 concentration has, and will be attenuated in this way by ocean-surface equilibration and mixing with the entire global ocean volume. The global capacity of this natural sequestration mechanism is considered to be large. For example, the oceans could absorb approximately 1000 Pg C while maintaining equilibrium with present-day atmospheric CO_2 levels (Kheshgi et al. 2005; Caldeira et al. 2005). This capacity could also be enhanced by chemical reactions between dissolved CO_2 and marine carbonate sediments. However, the rate of ocean mixing limits natural ocean CO_2 absorption to a pace that cannot match the rate of anthropogenic CO_2 production through fossil fuel combustion and industrial processes.

The long-term mixing and equilibration of ocean and atmospheric CO_2 assures that any proposed deliberate injection of CO_2 into the deep ocean, although potentially sequestering the CO_2 for a period of perhaps centuries, would eventually equilibrate with the atmospheric CO_2. Therefore, permanence of ocean CO_2 sequestration remains questionable. Nevertheless, injection of CO_2 into the oceans requires capture technologies and infrastructure similar to those described above for geologic CO_2 sequestration. In typical ocean temperatures, injected CO_2 will be in liquid form at depths greater than 400–500 m. At high concentrations, the liquid CO_2 may also form a solid hydrate phase. At depths greater than 3000 m, the liquid CO_2 generally becomes compressed to a density greater than that of the surrounding seawater. These properties have led to various suggestions for pumping CO_2 into the oceans at rates sufficient to mitigate rising atmospheric CO_2 levels and to reduce the peak concentrations expected without mitigation in the next century (Caldeira et al. 2005; Adams and Caldeira 2008). For example, it has been proposed that liquid CO_2 could be injected into sediments beneath the deep sea floor (House et al. 2006). However, in addition to permanence, a significant deterrent to oceanic CO_2 sequestration is the growing evidence for negative impacts of acidification caused

by the chemical reactions that occur when CO_2 is dissolved in seawater (Feely et al. 2009). Many marine organisms and ecosystems that depend on the formation of carbonate skeletons and sediments that are vulnerable to dissolution in acidic waters will be impacted. Laboratory and field measurements indicate that CO_2-induced acidification may eventually cause the rate of dissolution of carbonate to exceed its rate of formation in these ecosystems. The impact of ocean acidification on coastal and marine food webs and other resources are poorly understood, however (Guinotte and Fabry 2008; Hoegh-Guldberg and Bruno 2010). In addition, many unanswered environmental uncertainties have deterred proposals to enhance the ocean CO_2 uptake by fertilization of marine ecosystems (Buesseler et al. 2008; Adams and Caldeira 2008). Also, as with other CO_2 sequestration methods, the implementation of oceanic sequestration requires consideration of wide-ranging economic, environmental, social and political constraints. As a result, only minimal research efforts have been implemented in ocean CO_2 sequestration.

13.12   Geochemical Sequestration

Due to the urgency of mitigating the effects of anthropogenic CO_2 emissions and the associated many constraints on implementing CCS at the impactful level or ocean CO_2 sequestration, some scientists have pressed for broad consideration of novel approaches to global carbon cycle management (Cicerone 2006; Broecker 2007). Some of the approaches include geochemical methods that mimic the long-term cycling of CO_2 through the Earth's crust (Lackner 2002). CO_2 forms carbonic acid in water, and reacts on land with carbonate and silicate minerals during chemical weathering to liberate dissolved bicarbonate and carbonate ions. These ions are transported by rivers and streams to the oceans, where they combine with dissolved calcium and magnesium to form carbonate minerals that accumulate in sediments and thus are returned to the Earth's crust. Geochemical sequestration occurs when these reactions lead to net accumulation of carbon in dissolved or mineral form. Their natural geologic effectiveness is conspicuous in the large amounts of carbon dissolved in the oceans and retained in limestones and other carbonate sediments. Deliberate geochemical sequestration involves the acceleration of natural weathering and burial processes. Geochemical methods have been suggested for increasing the rates of weathering of silicate minerals (Lackner et al. 1997; Lackner 2002) and carbonate minerals (Rau and Caldeira 1999), thereby increasing geochemical CO_2 sequestration. Some proposals focus on the accelerated formation of carbonate minerals (i.e., mineral carbonation), which are generally more stable than the combination of silicate and dissolved CO_2 reactants (McGrail et al. 2006; Oelkers et al. 2008). A particularly creative idea is the addition of alkalinity to ocean surface waters, which would enhance natural ocean uptake of atmospheric CO_2 while buffering ocean acidification (Kheshgi 1995). Although high costs and slow rates of reaction make geochemical sequestration less

attractive now than many alternatives, this option certainly merits further investigation and discovery during the current period of nascent carbon cycle management (Stephens and Keith 2008).

13.13 Conclusions

CCS involves the integration of four elements—CO_2 capture, compression, transportation to the storage location, and isolation from the atmosphere by pumping it into appropriate geological formations such as saline aquifers, exhausted oil and gas reservoirs and coalbeds with effective seals to keep it safely and securely trapped underground and isolated from the atmosphere. Storage in other rock types such as basalt, oil and gas shales, and sea bed sediments may also be possible, but much less is known about their potential. Technologies for CCS are at different stages of development, some such as CO_2 compression, or CO_2-EOR is fully mature, while some such as storage in saline formations are in early stages of demonstration. Three approaches are available for capture from power plants and industrial sources that produce CO_2 gas with relatively low concentration of contaminants. These are pre- and post-combustion capture, and oxy combustion capture. Both processes have been routinely used for other applications but their optimization with power plants or most industrial processes have not been fully achieved. Considering full life cycle emissions, CCS technology can reduce up to 65–85% of CO_2 emissions from fossil fuel combustion from stationary source. Small-scale and industrial demonstration projects have provided insights into the feasibility of CCS technologies. Researchers have gained experience in industrial-size demonstration projects that have been running safely for years such as Sleipner in Norway and Weyburn in Canada. Additionally, decades of CO_2-EOR in North America have contributed in added knowledge base. Small-scale geological storage experiments aided by laboratory experiments and modelling has also contributed in development of improved monitoring and performance prediction tools. Nevertheless, costs of CCS implementation have remained prohibitive because of other price factors. Despite the advances in science and technology of CCS, the future of CCS is highly uncertain, and the past decade has shown how dependent the technology is on social, political, and financial context. The financial crisis of 2008, and lack of commitment from Copenhagen climate change conference in 2009, as well as the rise of shale gas in United States, lower cost of renewable energy, a public that has remained skeptical about the role of CCS in mitigating climate change, and rising resources costs have all contributed to diminishing attention to CCS, leading to cancellation of various planned demonstration projects and low investment in the CCS in recent years. Action on climate change at national and/or international either through pricing C, mandating technology on a sectoral basis while also imposing emission standards requires political leaders to make clear choices and commitment against a high C future. There is also a need to assure the public that

geological storage of CO_2 is safe and an effective means to stabilize and in the long-run to reduce atmospheric CO_2 concentration. But credible, accessible, and scientifically sound information sources, appropriate engagement activities are currently lacking, and this has led to lack of community support for CCS. Successful experiences from CO_2-EOR, and ongoing small scale and industrial scale demonstration projects demonstrate that at least on this limited scale, appropriately selected and managed geological reservoirs likely retain nearly all the injected CO_2 for long time and provide the benefits for intended purposes of CCS. However, up to tenfold scale-up in size of current individual industrial scale demonstration would be needed to capture, transport, and store emissions from a large-scale point source such as 500–1000 MW coal-fired power plant, and a thousand-fold scale-up in size of current CCS projects would be needed to reduce CO_2 emissions at the level of Pg C yr^{-1}. This remain a big challenge which need to be addressed. Specifically, is there sufficient capacity to store these quantities of CO_2? At what cost? Does institutional, economic, and technical constraint to implement CCS on this scale be overcome? The answers to these questions, together with social acceptability will determine the future success of CCS.

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