British Columbia Geological Survey
Geological Fieldwork 2002

Geological and Mineral CO_2 Sequestration Options: A Technical Review

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INTRODUCTION

This review results from directed studies at the University of Victoria by the senior author and it covers the main technical aspects of the Greenhouse Gas Sequestration methodology.

The Kyoto Agreement may or may not be ratified, however, the following text highlights those options that British Columbia will have if a need for geological, mineral or deep ocean sequestration arises. Of the six greenhouse gases covered by the Kyoto protocol, carbon dioxide (CO_2) is the greatest contributor to Canada's total GHG emissions (Table 1). Fossil fuel combustion is the main source of anthropogenic CO_2, and it currently supplies over 85% of the global energy demand (Figure 1). The main engineering effort for reduction of CO_2 emissions is therefore aimed at increased efficiency of fossil energy usage, development of energy sources with lower carbon content and increased reliability on alternative energy sources such as wind, solar, geothermal and nuclear. It is not likely that the reduction of CO_2 emissions, in an order of magnitude similar to the Kyoto agreement, could be met using these measures alone. CO_2 sequestration methods that are currently considered, or being evaluated by industrialized countries, are part of the global plan. Each method has its weaknesses and strengths. The methods that we will cover in this review are:

- Storage in Oil and Gas Reservoirs
- Storage in Deep Coal Seams
- Storage in Deep, Saline Aquifers
- Storage in Deep Ocean
- Storage in Salt Caverns
- Mineral Carbonation

Since all geological and mineral CO_2 sequestration methods involve the capture and extraction of CO_2 from flue-gases or industrial streams, transportation of CO_2 and its disposal in an appropriate sink, the next stage of our study will identify the main stationary point sources of CO_2 emissions and the main potential carbon or CO_2 sinks in British Columbia.

TABLE 1
CANADIAN GREENHOUSE GAS EMISSIONS

Carbon Dioxide (CO_2)	**78.90%**
Methane (CH_4)	**12.40%**
Nitrous Oxide (N_2O)	**7.40%**
Other (HFCs*, PFCs$^+$ and $SF_6{}^x$)	**1.30%**

*Hydrofluorocarbons
$^+$Perfluorocarbons
$^\&$ As Carbon Dioxide Equivalent
^{x}Sulphur hexafluorides
(Source: Environment Canada, 2002)

Geographic relationships between the main stationary point CO_2 sources and sinks is an essential piece of the puzzle for CO_2 sequestration planning in British Columbia since transportation is one of the important cost factors.

PHYSICAL PROPERTIES OF CARBON DIOXIDE

It is important to know the main properties of carbon dioxide to understand carbon sequestration methods. Carbon dioxide (CO_2) is an odourless, colourless gas that occurs naturally in the atmosphere. Current ambient atmo-

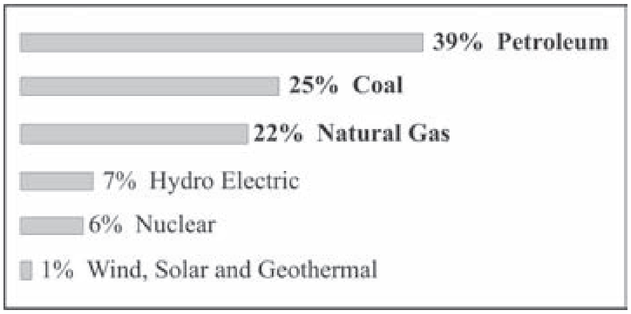

Figure 1. Global energy demand. Fossil fuels supply over 85% of the world's energy. (Source: McKee, 2002)

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spheric concentrations of CO_2 are around 370 ppm (0.037%). Benson *et al* (2002) summarizes the effects of high concentrations of CO_2 on humans and other life forms.

Depending on pressure and temperature, CO_2 can take on three separate phases (Figure 2). CO_2 is in a supercritical phase at temperatures greater than 31.1°C and pressures greater than 7.38 MPa (critical point). Below these temperature and pressure conditions, CO_2 will be either a gas or a liquid. Depending on in situ temperature and pressure, CO_2 can be stored as a compressed gas or liquid, or in a supercritical (dense) phase.

CO2 STORAGE IN OIL AND GAS RESERVOIRS

Both depleted and active fossil fuel reservoirs are potential storage space for CO_2 in underground formations. CO_2 may be injected directly into a depleted or inactive reservoir without expectation of any further oil production, or the CO_2 injection may result in enhanced oil/gas recovery and simultaneous CO_2 sequestration. CO_2 may also be injected into producing oil and gas reservoirs, where CO_2-enhanced oil recovery (EOR) and CO_2-enhanced gas recovery (EGR) will offer an economic benefit. Typically, oil reservoirs have undergone a variety of production and injection processes during primary and secondary recovery (*e.g.* gas, water or steam injection), as described by Jimenez and Chalaturnyk (2002). As a tertiary recovery process, CO_2 can be injected into the reservoir to improve the mobility of the remaining oil (van der Meer, 2002), thereby extending the production life of the reservoir. Injection of CO_2 into producing gas reservoirs for EGR was previously believed to risk contaminating the natural gas reserve (Stevens *et al.*, 2000). However, recent studies by Oldenburg and Benson (2002; 2001) suggest that mixing of the CO_2 and methane (CH_4) in a gas reservoir would be limited due to the high density and viscosity of CO_2 relative to the natural gas. Furthermore, significant quantities of natural gas can be produced by repressurization of the reservoir. According to Davison *et al.* (2001), it is possible that improved oil and gas recovery could more than offset the cost of CO_2 capture and injection.

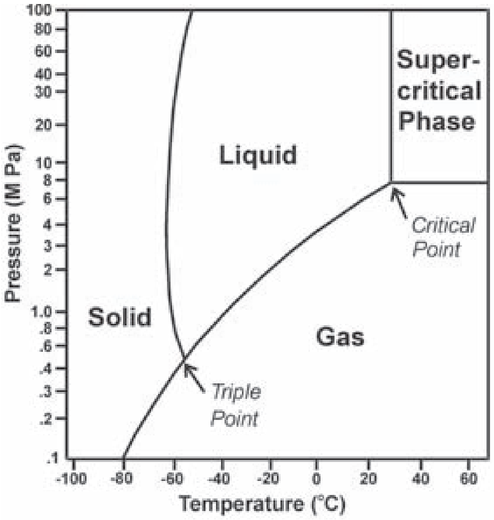

Figure 2. Carbon dioxide phase diagram. The critical point for CO_2, when it reaches supercritical state, is 31.1°C and 7.38 MPa. (Adapted from Koide *et al.*, 1996).

For the purpose of this paper, the term "depleted fossil fuel reservoirs" refers to abandoned oil and gas reservoirs. These reservoirs have undergone primary and secondary recovery and CO_2-enhanced oil recovery is not currently envisaged to generate positive cashflow.

ACTIVE OIL RESERVOIRS

The petroleum industry has been injecting CO_2 into underground formations for several decades (Gentzis, 2000) to improve oil recovery from light and medium oil reservoirs, even before climate change became an issue (Bachu, 2000a). CO_2 injected into suitable oil reservoirs can improve oil recovery by 10-15% of the original oil in place in the reservoir (Davison *et al.*, 2001). When CO_2 is injected into a reservoir above its critical point (typically a reservoir depth greater than 800 m), the gas acts as a powerful solvent. If the pressure is high enough and the oil gravity is greater than 25° API (Bachu, 2001), the CO_2 and oil become completely miscible. According to Aycaguer *et al.* (2001), the miscible flood reduces the oil's viscosity thereby enabling the oil to migrate more readily to the producing wells (Figure 3). At lower pressures CO_2 and oil are not completely miscible, however some fraction of the CO_2 will dissolve in the oil. This is known as immiscible displacement and also enhances oil recovery. CO_2 enhanced oil recovery is now considered as a mature technology (Gentzis, 2000). If EOR is the main objective of CO_2 injection, then the operation is optimized to minimize the cost of CO_2 used and maximize the oil recovery. CO_2 sequestration differs from EOR by CO_2; its main objective is to sequester as much CO_2 in the reservoir as possible for geological time (van der Meer, 2002; Benson, 2000).

A life cycle assessment study on EOR with injection of CO_2 in the Permian Basin of West Texas (Aycaguer *et al.*, 2001) suggests that the amount of CO_2 injected, not includ-

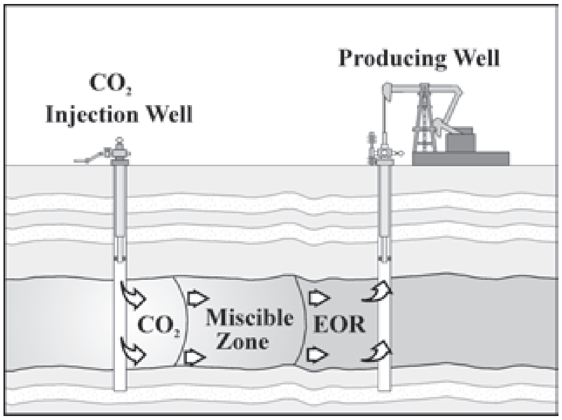

Figure 3. Simplified diagram of a CO_2-enhanced oil recovery (EOR) operation. (Modified from: IEA R&D Programme, 2001).

ing the recycled CO_2, may balance the amount of CO_2 in emissions that ultimately are produced by combustion of the extracted hydrocarbon product. To reduce atmospheric CO_2 and preserve natural CO_2 reservoirs, the source of CO_2 for EOR should come from anthropogenic sources. Most of the existing CO_2-EOR projects in the world use natural CO_2 sources (Whittaker and Rostron, 2002). The CO_2 comes from natural carbon dioxide reservoirs where the infrastructure for distribution is already present, providing delivery without major capital costs (Aycaguer *et al.*, 2001) and without processing (Smith, 1998). Of the 74 CO_2-EOR projects in the USA, only four use anthropogenic CO_2 (Whittaker and Rostron, 2002). A Canadian study done by Tontiwachwuthikul *et al* (1998) on the economics of CO_2 production from coal-fired power plants concluded that flue gas extraction could be an economically viable CO_2 supply source for CO_2-EOR projects in Western Canada, should oil prices increase substantially. Currently, in Weyburn, Canada, a large-scale project for CO_2-EOR has been developed with the aim at implementing a guideline for geological storage of anthropogenic CO_2 (Moberg, 2001; Whittaker and Rostron, 2002; Srivastava and Huang, 1997). The Weyburn project is sponsored by a number of Governments and industries from North America, Europe and Japan.

DEPLETED OIL AND GAS RESERVOIRS

Following more than a century of intensive petroleum exploitation, thousands of oil and gas fields are approaching the ends of their economically productive lives (Davison *et al.*, 2001). Some of these exhausted fields could act as storage sites for CO_2. As in the case of producing fields, the general concept of CO_2 disposal in depleted oil and gas reservoirs is that the hydrogeological conditions that allowed the hydrocarbons to accumulate in the first place will also permit the accumulation and trapping of CO_2 in the space vacated by the produced hydrocarbons (Hitchon *et al.*, 1999; Gentzis, 2000). The caprock that prevented the escape of oil and gas over geological time, should retain the sequestered CO_2 for thousands of years (Bachu, 2001), as long as it is not damaged as a result of overpressuring during the CO_2 injection (van der Meer, 1993), by the presence of unsealed, improperly completed or abandoned wells (Hitchon *et al.*, 1999), tectonic activity or pH change.

About 80% of the world's hydrocarbon fields are at depths greater than 800m (IEA, website), thus meeting the criteria for the pressure and temperature needed to efficiently store CO_2 as a supercritical fluid (van der Meer, 1993). Existing infrastructure and reservoir properties make storage of CO_2 in depleted oil and gas reservoirs a simpler option than other forms of CO_2 sequestration (Bachu, 2000a).

Closed, underpressured **oil reservoirs** that have not been invaded by water should have good sequestration capacity (Bachu, 2001). Oil field primary recovery varies from 5% to 40% (van der Meer, 2002), thus, depending on the extraction technology used and economic conditions that prevailed during the active life of the reservoir (Bachu *et al.*, 2000), significant oil reserves may remain in the reservoir. Therefore, if exhausted oil fields were used for CO_2 storage, substantial amounts of oil could be recovered (van der Meer, 2002). Depleted hydrocarbon reservoirs that are filled with connate water (fully water-saturated reservoirs) offer limited storage capacity. The injected CO_2 would have to displace the connate water of the reservoir. Storage of CO_2 in water-saturated reservoirs would in practice amount to aquifer storage (Bachu, 2000a; van der Meer, 2002) as described later in this paper.

Closed, underpressured, depleted **gas reservoirs** are excellent geological traps for CO_2 storage. Firstly, primary recovery of gas fields usually removes as much as 95% of the original gas in place (Bachu, 2001), creating large storage potential. Secondly, the injected CO_2 can be used to restore the reservoir to its original pressure (Bachu *et al.*, 2000), thereby preventing possible collapse or man-induced subsidence. Thirdly, the trapping mechanism that retained hydrocarbons in the first place should ensure that CO_2 does not reach the surface (Bachu *et al.*, 2000). And lastly, the existing surface and down-hole infrastructure used for production of gas is ideally suited for transportation and injection of supercritical CO_2.

Spatial association between hydrocarbon production and the presence of reservoirs suitable for CO_2 sequestration may result in shared infrastructure and reduction of transportation costs. Furthermore, depleted hydrocarbon fields commonly have an established geological database and as such, reservoir characteristics are well known. Currently, the petroleum industry is reluctant to consider storage of CO_2 in depleted hydrocarbon reservoirs, because abandoned fields will still contain oil and gas resources (US Dept of Energy, 2002), which potentially have economic value if oil prices were to rise enough or new EOR technologies were developed in the future (Davison *et al.*, 2001; Bachu *et al.*, 2000). Today, sequestration of CO_2 in depleted oil reservoirs offers little or no economic benefit for the oil companies, however these reservoirs may become a base of the future CO_2 disposal industry.

CO_2 STORAGE IN COALBEDS

Coalbeds are a potential storage medium for CO_2. British Columbia has abundant coal resources; some of them lie at depths too great to be considered for conventional mining. CO_2 can be injected into suitable coal seams where it will be adsorbed onto the coal, stored in the pore matrix of the coal seams, and locked up permanently. An alternative to CO_2-only storage is injection of flue gas, a mixture of CO_2 and nitrogen (N_2) into coalbeds. According to Reeve (2000), flue gases account for 80% of CO_2 emissions in western Canada. Although in British Columbia flue gases represent much smaller percentage of total emissions, the injection of flue gas may avoid the high cost of CO_2 separation (Law *et al.*, 2002).

CO_2-ENHANCED COALBED METHANE RECOVERY

CO_2 sequestration in coal seams has the potential to generate cashflow through enhanced coalbed methane (CBM) recovery, a process similar to the practice of CO_2-EOR. Recovery of CBM is a relatively well-established technology used in several coalfields around the world (Schraufnagel, 1993; Ivory *et al.*, 2000). A number of companies are looking at producing CBM in British Columbia. Primary CBM recovers about 20-60% of the gas in place (Gentzis , 2000; van Bergen, 2001); some of the remaining CBM may be further recovered by CO_2 enhanced CBM recovery.

The disposal of CO_2 in these methane-rich coalbeds, where applicable, is expected to increase drive pressure and the CBM recovery rate (Hitchon *et al.*, 1999). Thus, injection of CO_2 should enable more CBM to be extracted, while at the same time sequestering CO_2. CO_2-enhanced CBM production could be achieved by drilling wells into the coal deposits, typically a five-spot pattern, with the centre well as the injector and the four corner wells as the producing wells (Wong *et al.*, 2001). After discharging formation waters from the coal, CO_2 is injected into the coal seam. CO_2 has a higher affinity with coal, about twice that of methane (Figure 4), just below the critical point (~7.38 Mpa). Limited data at pressures exceeding the critical point of CO_2 indicate that the extrapolation of the CO_2 adsorption curve above 7.38 Mpa is not justified (Krooss *et al.* 2002). In theory, injected CO_2 molecules displace the adsorbed methane molecules (Wong *et al.*, 2001; Ivory *et al.*, 2000; Hitchon *et al.*, 1999), which desorb from the coal matrix into the cleats (figure 5) and flow to the production wells. CO_2 enhanced CBM can achieve about 72% recovery (Wong *et al*, 2000). A CO_2 enhanced CBM production project terminates at CO_2 breakthrough in one or more of the production wells (Wong *et al.*, 2001).

Flue gas injection may enhance methane production to a greater degree than CO_2 alone (Ivory *et al.*, 2000). However, N_2 has a lower affinity for coal than CO_2 or methane (Figure 5). Therefore, injection of flue gas or CO_2-enriched flue gas results in rapid nitrogen breakthrough at the producing wells (Macdonald *et al.*, 2002; Law *et al.*, 2002). In such cases, N_2 waste could be reinjected into the coal seam (Macdonald *et al.*, 2002; Wong and Gunter, 1999).

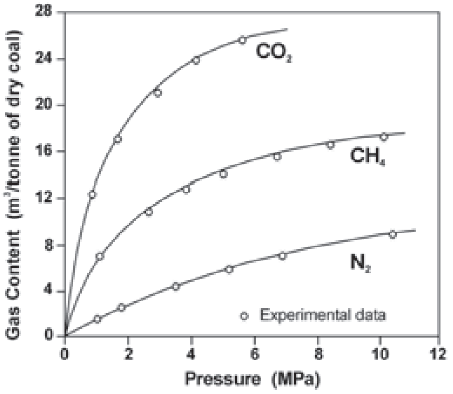

Figure 4. Adsorption isotherms for carbon dioxide (CO_2), methane (CH_4) and nitrogen (N_2) on coal (Adapted from Arri *et al.,* 1992). Limited data is available for CO_2 adsorption at pressures in excess of 7.38 MPa (Krooss *et al.*, 2002) .

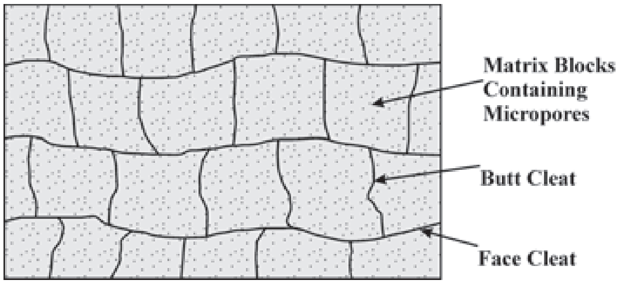

Figure 5. Coalbeds contain both primary and secondary porosity systems. The coal matrix (primary porosity system) contains the vast majority of the gas-in-place volume while the cleats (secondary porosity system) provides the conduit for mass transfer to production wells. (Adapted from Rice *et al.*., 1993).

Sequestration of CO_2 in coal seams, while enhancing CBM recovery, is an attractive option, but the physical characteristics of the coals, for the purpose of CO_2-enhanced coalbed methane recovery (ECBM), are largely unknown. Recent studies (Fokker and van der Meer, 2002; Reeves, 2002) have shown that continued injection of CO_2 in coalbeds induced a decrease in the permeability of the cleat system surrounding the injection well area. In general, desorption of the methane causes shrinkage of the coal matrix, which in turn, causes the cleats to open, thereby allowing the CO_2 injection rate to increase and the methane to flow to the producing well. At the same time, replacement of the methane by the injected CO_2 is believed to cause the coal matrix to swell. This swelling will partially block the cleat system and negatively affect the main flow parameters. The fracturing of the coal and the swelling have opposite effects on the CO_2 injectivity (Fokker and van der Meer, 2002). One possible solution to achieve an acceptable CO_2 injection rate would be to allow the near-well gas pressure in the cleat system to exceed the hydraulic fracturing pressure (Fokker and van der Meer, 2002; Shi *et al.*, 2002). However, if repeated hydraulic fracturing is necessary to maintain connectivity between the well bore and the permeable areas of the coal seam, this in turn may result in over/under burden fracturing (Gale, 2002), and CO_2 leakage.

The Alberta Research Council (ARC) has done extensive applied research in this field and some of the outstanding contributions were published by Wong *et al.* (2000), Law *et al.* (2002), and Mavor *et al.* (2002). There are currently several CO_2-ECBM recovery field projects studying sequestration of CO_2 and flue gas in deep coal seams. These projects range in depth from 760 to 1100 metres.

- Alberta Research Council under an international project, facilitated by the IEA Greenhouse gas R&D Programme, has established a pilot site at Fenn-Big Valley, Alberta, Canada. The project is looking at the enhancement of CBM production rates in low permeability CBM reservoirs using mixtures of CO_2 and N_2 while sequestering CO_2 into coalbeds (Law *et al*., 2002; Reeve, 2000; Ivory *et al*., 2000).

- In October 2000 a three-year government-industry project in the San Juan Basin (USA), known as the Coal-Seq project, was launched. The project studies the feasibility of CO_2-sequestration in deep, unmineable coal seams using enhanced CBM recovery technology (Reeves, 2002).
- In November 2001, the RECOPOL project (Reduction of CO_2 emission by means of CO_2 storage in coal seams in the Silesian Coal Basin of Poland), funded by the European Commission, started with aims to develop the first European field demonstration of CO_2 sequestration in subsurface coal seams (van Bergen *et al*., 2002).

The industry and scientific community will carefully scrutinize the results from these field tests, particularly since they may provide emperical data on CO_2 adsorption behaviour above its critical point (7.38 Mpa). The outcome from these tests will probably determine where new research will be oriented.

CO_2 STORAGE IN DEEP AQUIFERS

Worldwide, deep saline aquifers have larger geological storage capacity than hydrocarbon reservoirs and deep coal seams (Table 2). Deep aquifers are found in most of the sedimentary basins around the world (Bachu, 2001) and typically contain high-salinity connate water that is not fit for industrial and agricultural use, or for human consumption. Deep saline aquifers have been used for injection of hazardous and nonhazardous liquid waste (Bachu *et al*., 2000) and as such provide viable options for CO_2 sequestration. Approximately 2% of the total effective volume in a deep aquifer can be made available for CO_2 storage (van der Meer, 2002; 1993). Thus, from a capacity perspective, deep saline aquifers offer a significant potential for CO_2 storage (Gale, 2002).

Suitable aquifers must be capped by a regional aquitard (*e.g.* shale), which should not contain any fractures or incompleted wells (Bachu *et al*., 1994). The top of the aquifer must be located at a minimum depth of 800 meters (van der Meer, 2002), ensuring that the injected CO_2 will be stored in supercritical state. No single bed or stratigraphic interval is likely to be a potential injection aquifer across an entire basin (Hitchon *et al*., 1999), thus near-well permeability should be high for injection purposes, but regional-scale permeability should be low, to ensure long-term disposal of CO_2 (Bachu *et al*., 1994). When the CO_2 is injected into a suitable aquifer, due to buoyancy effects, it will rise up and gradually spread out forming a layer of CO_2 under the cap rock (Gale, 2002). In the early stages of geochemical reaction, dissolution is expected to be the predominant process (Gunter *et al*., 1997). The surface area of CO_2 in contact with the formation water will control the rate of dissolution. It is believed that during an injection period of 25 years, between 10 and 25% of the CO_2 will be dissolved (Gale, 2002). The undissolved portion of the injected CO_2 will segregate and form a plume at the top of the aquifer as a result of density differences (Bachu, 2001). The CO_2 plume will be driven by both hydrodynamic flow and by its buoyancy (Bachu *et al*., 2000). The greater the density and viscosity differences between CO_2 and the formation fluid, the faster the undissolved CO_2 will separate and flow updip in the aquifer in a process similar to oil and gas migration (Bachu, 2001). Thus, CO_2 should be injected under high pressures to ensure high density of the CO_2 and high CO_2 solubility rate in formation water.

TABLE 2
GLOBAL CAPACITY OF GEOLOGICAL RESERVOIRS

Storage Option	Global Capacity	
	Gt CO_2	% of emissions to 2050
Depleted oil and gas fields	**920**	**45**
Deep saline aquifers	**400-10,000**	**20-500**
Unmineable coal seams	**>15**	**>1**

Source: IEA Greenhouse Gas R&D Programme, 2001

Once outside the radius of influence of the injection well, both the dissolved and immiscible CO_2 will travel with the natural velocity of the formation water (Gunter *et al*., 1997). On the regional scale, the velocity of formation waters in these aquifers is expected to be of the order of 1 to 10cm/year (Bachu *et al*., 1994), suggesting that CO_2 residence time in a deep, low-permeability aquifer could be of the order of tens to hundreds of thousands of years (Gunter *et al*., 1997). The geological time-scale trapping of CO_2 in deep regional aquifers, caused by very low flow velocity, is termed **hydrodynamic trapping**, because it depends on the hydrodynamic regime of formation waters (Bachu *et al*., 1994).

Injection of CO_2 into a siliclastic formation may lead to precipitation of carbonate minerals, in effect storing CO_2 in a stable form. This is referred to as **mineral trapping** (Bachu *et al*., 1994; Gunter, Bachu and Benson, in review) and is based on the same principle as mineral carbonation that will be discussed in the last section. The following chemical reaction is an example of mineral trapping of CO_2 (Bachu *et al*., 1994):

$$CaAl_2Si_2O_8 \text{ [Ca-feldspar]} + CO_2 + 2H_2O \Rightarrow$$
$$Al_2Si_2O_5(OH)_4 \text{ [kaolinite]} + CaCO_3 \text{ [calcite]}$$

Experiments carried out to test the validity of mineral trapping of CO_2, by Gunter *et al.* (1997), concluded that these reactions are expected to take hundreds of years or more to complete. Due to the long residence time of CO_2-charged formation waters within the aquifer, these reactions may eventually trap over 90% of the injected CO_2 (Gunter *et al*., 1997). Mineral trapping will not greatly increase the CO_2 storage capacity of the aquifer; rather its advantage over the hydrodynamic trapping resides in the permanent nature of CO_2 disposal (Bachu *et al*., 1994).

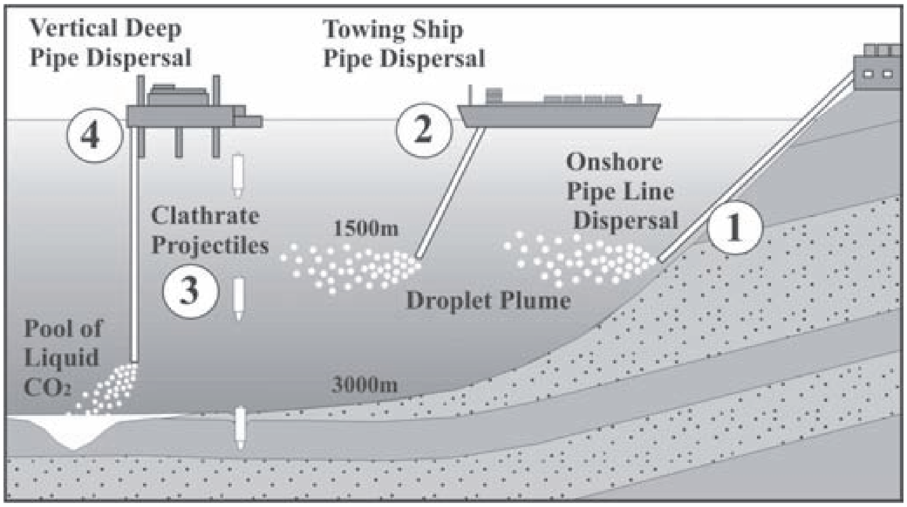

Figure 6. Compilation diagram of proposed methods for ocean disposal of CO_2. Method 1 and 2 dispose of CO_2 by injecting a droplet plume, which dissolves into the ocean water. Method 3 and 4 involve sequestration of CO_2 as clathrate hydrates.

Injection of CO_2 into deep, saline aquifers relies on existing technology. Since 1996, Statoil injects about 1 million tonnes of CO_2 per year into a deep aquifer offshore Norway (Chadwick *et al.*, 2002). Sequestration of the CO_2 waste, a by-product of natural gas production, saves the company from paying a Norwegian CO_2 tax (Gentzis, 2000).

DEEP OCEAN DISPOSAL OF CO2

The ocean is the largest sink available for disposal of CO_2 with a residence time of four to five hundred years (Gentzis, 2000). The oceans contain a stratified thermocline, which is located between the surface layer and the deep ocean. Its waters circulate between surface and deep layers on varying time scales from 250yrs in the Atlantic Ocean to 1000yrs for parts of the Pacific Ocean (Mignone *et al.*, 2002; Ormerod *et al.*, 2002). The atmosphere and the ocean are in contact over 70% of the globe and there is a continuous exchange of inorganic carbon between them. Oceans are, at present time, removing about six gigatonnes CO_2/year from the atmosphere (Ormerod *et al.*, 2002). Disposing anthropogenic CO_2 in the deep ocean would accelerate a natural process. CO_2 could be injected as a liquid below the thermocline at depths greater than 1500m and be sequestered either by dissolution in the water column or by formation of CO_2 hydrates (Figure 6).

STORING CO2 BY DISSOLUTION

One approach involves transporting liquid CO_2 from shore by pipeline and then discharging it from a manifold lying on the ocean bottom, forming a **droplet plume**. Since liquid CO_2 is less dense than seawater, the CO_2 droplets will rise until they are dissolved into the seawater and the CO_2-charged solution spreads laterally into the (stratified) surrounding seawater. The dissolved CO_2 may travel in the thermocline, and eventually (after hundreds of years) circulate back into the atmosphere. The deeper the CO_2 is injected, the more effectively it is sequestered, but injecting deeper requires more advanced technologies (Ormerod *et al.*, 2002). The oil and gas industry have established technology to construct vertical risers in deep water and to lay seabed oil and gas pipelines in depths down to 1600m (Ormerod *et al.*, 2002), suggesting that this method is technically feasible.

Alternatively, liquid CO_2 could be transported by a tanker and discharged from a pipe towed by a moving ship. The Japanese R&D program for ocean sequestration of CO_2 is currently in phase II of a large-scale “moving-ship” scheme in the western North Pacific to assess environmental impact and CO_2-plume behaviour (Murai *et al.*, 2002). Studies by Ozaki *et al.* (2001) have shown that CO_2 injection would be most effective at relatively slower rates (larger droplet size) and at depths greater than 1500m (Ormerod *et al.*, 2002). Such a depth is well within the capability of present day subsea pipeline technology and CO_2 could be transported by a tanker, like those used currently for transportation of liquid petroleum gas (Ormerod *et al.*, 2002).

STORING CO2 AS CLATHRATES

Another method for ocean disposal of CO_2 involves sequestration of CO_2 at depths in excess of 3000 metres. At these depths, due to the high pressure and low temperatures (Ozaki *et al.*, 2001), CO_2 exists in the form of a clathrate hydrate, an ice-like combination of CO_2 and water (Brewer *et al.*, 2000). Pure CO_2-hydrate is denser than seawater and will generate a sinking plume, settling on the bottom of the ocean (Brewer *et al.*, 2000). CO_2 sequestered in this way would form submarine pools in hollows or trenches in the deep sea. Dissolution of CO_2 into the overlying seawater would be reduced significantly due to formation of the CO_2-hydrates. Direct disposal of CO_2 at great depths is currently not technically feasible, however, it may be possible to send cold CO_2 (dry ice) from mid-depth to the ocean floor (Aya *et al.*, 2002). With a density greater than seawater, cold CO_2 will sink to the ocean bottom and be effectively stored. The Monterey Bay Aquarium Research Institute (MBARI) has recently conducted a series of controlled experiments that involve release of cold CO_2 slurry at depths of 350-500m (Aya *et al.*, 2002).

Yet another method proposes disposal of CO_2 as **clathrate blocks**. Studies on this disposal method confirm that streamlined blocks have higher terminal velocity and thus reach the seabed faster than equidimensional blocks (Guever *et al.*, 1996). As large as 1000 tons and shaped like a projectile, these blocks could penetrate into the deep seabed where the solid CO_2 would physically and chemically interact with the sediments before reacting with the ocean water. The retention times could, therefore, be significantly increased as compared to the gaseous or liquid CO_2 disposal methods (Guever *et al.*, 1996). According to the IEA this method is currently not economically feasible (Ormerod *et al.*, 2002).

Further studies on ocean disposal of CO_2 include **fertilising the oceans** with additional nutrients to increase draw-down of CO_2 from the atmosphere (Ormerod *et al.*, 2002). Addition of nutrients such as nitrates and phosphates or iron may increase production of biological material, thereby drawing down additional CO_2 from the atmosphere through photosynthesis of the phytoplankton (Ormerod *et al.*, 2002). Should this method prove to be feasible, the fishing industries may benefit from the resulting increase in the fish population, with atmospheric CO_2 sequestration as a secondary benefit, however the overall impact on the marine ecosystem is not well understood

All the above described ocean disposal methods could potentially cause at least a local change in pH of the ocean water. Marine communities are, in general, intolerant to changes in the pH. Thus, due to environmental impacts on the marine ecosystem and associated public disapproval, ocean sequestration of CO_2 is not currently considered as an attractive option. The situation may change if the development of extensive CH_4 clathrate deposits along the BC coast takes place.

STORAGE IN SALT CAVERNS

Underground caverns, such as mined salt domes, could be created to store CO_2. Salt is generally found as intrusive (domal or ridge) deposits whereby salt from a major underlying source has been forced up into overlying formations. Salt caverns are created by solution mining, a process in which water is injected down a well, to dissolve the salt, and the brine solution is pumped out, creating large cavities. These caverns can be up to 500 000 m^3 in volume (Bachu, 2000a), and since salt is highly impermeable (Murck *et al.*, 1996) these spaces could provide a long-term solution to CO_2 sequestration. Solid CO_2 (dry ice) could also be stored in these repositories, surrounded by thermal insulation to minimise heat transfer and loss of CO_2 gas (Davison *et al.*, 2001). The technology has been developed and applied for salt mining and underground storage of petroleum, compressed air and natural gas (Bachu, 2000a; Crossley, 1998; Istvan, 1983). Although salt and rock caverns theoretically have a large storage capacity, the associated costs are very high and the environmental problems relating to the mined rock and disposal of large amounts of brine are significant (Kolkas-Mossbah and Friedman, 1997). Based on current technology, storage of CO_2 in underground salt caverns is uneconomical for the time being.

MINERAL CARBONATION

Mineral carbonation is a CO_2 sequestration concept where CO_2 is chemically combined in an exothermic reaction with readily available Mg or Ca-silicate minerals to form carbonates and other stable by-products (Seifritz, 1990; Gerdemann *et al.*, 2002; O'Connor *et al.*, 2000). Both Mg and Ca carbonates are stable on geologic time-scale, potentially storing CO_2 for millions of years. Mg-silicates are favoured relative to Ca-silicates because they are more widespread, form larger bodies and contain more reactive material per tonne of rock (Lackner *et al.*, 1997; Kohlmann *et al.*, 2002). Wide variety of Mg-bearing materials, such as enstatite, asbestos tailings (Fauth and Soong, 2001), fly ash and other industrial residues were investigated as potential starting materials for the industrial carbonation process, however, in the light of recent laboratory tests, olivine $[(Mg,Fe)SiO_4]$ and serpentine $[Mg_3Si_2O_5(OH)_4]$ appear as the most promising. The two reactions below illustrate the CO_2 carbonation principle using olivine and serpentine as examples:

$$Mg_2SiO_4[\text{olivine}] + 2CO_2 \Rightarrow 2MgCO_3[\text{magnesite}] + SiO_2 \quad (1)$$

$$Mg_3SiO_3(OH)_4[\text{serpentine}] + 3CO_2 \Rightarrow 3MgCO_3[\text{magnesite}] + 2SiO_2 + H_2O \quad (2)$$

In nature, carbonation reactions involving silicates are slow (Kohlmann and Zevenhoven, 2001). Currently, a sequestration plant can be visualized as a blender operating at high temperature-pressure conditions (Figure 7). For the

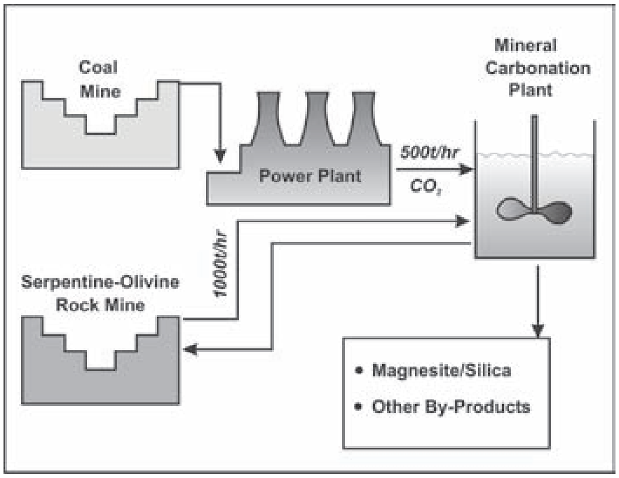

Figure 7. Idealized view of a mineral carbonation plant. (Modified from Bauer, 2001)

industrial CO_2 sequestration applications, carbonation reactions have to be accelerated by increasing surface area of the Mg-silicate, agitating the slurry (O'Connor *et al.*, 1999; Dahlin *et al.*, 2000) and by adding catalysts (for example, NaCl and $NaHCO_3$ and HCl) to the solution/slurry prior to the carbonation process (Dahlin *et al.*, 2000; Goldberg and Walters, 2002; Jia and Anthony, 2002; Fauth and Soong, 2001; Lackner *et al.*, 1998). Optimization of the carbonation process by controlling temperature and partial pressure of CO_2 (P_{CO2}) may be also a major factor (O'Connor *et al.*, 1999; Dahlin *et al.*, 2000).

Subjecting Olivine to supercritical conditions is believed to improve olivine dissolution rates (O'Connor *et al.*, 2000; 1999). Furthermore, in the case of serpentine, an energy-intensive heat pre-treatment (activation-destabilization of the crystal structures) at temperatures above 600°C is required. Such pre-treatment removes chemically bound water and increases overall porosity (Gerdemann *et al.*, 2002; Kohlmann *et al.*, 2002; Goldberg and Walters, 2002).

There is currently no mineral sequestration plant in operation, however members of the Mineral Sequestration Working Group are developing pilot-scale mineral carbonation units and according to their plan a 10 MW demonstration plant will be operational by 2008 (Goldberg and Walters, 2002). The concept is currently incorporated into the design of the coal-fuel electricity generating plant of the ZECA Corporation (New Mexico). However, it may be also applied elsewhere.

ADVANTAGES OF MINERAL CARBONATION

Serpentine and Olivine are the two most likely silicates, which could be used as starting materials in mineral sequestration. Olivine is favoured because it reacts better without the energy-intensive pretreatment that serpentine requires. In contrast to the previously described methods, once the CO_2 is locked into a carbonate (a mineral stable on geological time scale), there is no possibility for an accidental release of CO_2. Furthermore, direct carbonation does not lead to problematic by-products (Lackner *et al.*, 1998). Mineral carbonation may, therefore, benefit from public acceptance (Lackner *et al.*, 1997).

The costs of the CO_2 disposal could be higher than for the injection of CO_2 into oil and gas reservoirs or deep coal seams. These costs may be reduced if the potential for industrial applications of the product (depending on acceptable purity, form, grain size, particle shape and chemical properties), and metal recoveries could be realized. Magnesite has a wide variety of industrial applications (Simandl, 2002) and the same applies for silica. The carbonation process may also become a new source of Fe, Mn, Co, Cr and Ni recovered during the breakdown of Mg silicate's crystal structure (Haywood *et al.*, 2001; O'Connor *et al.*, 2000). Technology breakthroughs and the law of "supply and demand" will determine if, and to what extent the sequestration costs an be offset by these potential byproducts.

Large-scale CO_2 sequestration as mineral carbonates will require enormous amounts of mineral (Kohlmann *et al.*, 2002). For a typical power plant, the mass flows of fuel and carbonated mineral will be of the same order of magnitude. For example, studies suggests that for a single power plant, generating approximately 10 000 tons CO_2 per day, over 23 000 to 30 000 tons per day of Mg-silicate ore would be required (Dahlin *et al.*, 2000; O'Connor *et al.*, 2000). If mineral sequestration becomes a reality and serpentine becomes a workhorse of mineral CO_2 sequestration, no shortage of starting material is likely to occur in BC. However, if forsterite (Mg-end member of olivine) is used as starting material, supplies are limited and geographically constrained. Under ideal conditions, coal and Mg-silicate mines would be located close to each other. In most cases, serpentine is an unwanted by-product of metal and chrysotile mining, but in some locations, this waste may become a sought after commodity when its potential for CO_2 sequestration is realized. Should mineral sequestration of CO_2 become an established technology, then new opportunities will arise for potential producers of magnesium silicates and owners of magnesium silicate-rich tailings.

The British Columbia Geological Survey may not participate in the development of mineral sequestration technology, however, the inventory, characterization and documentation of potential sources of Mg-silicates is in the Survey's interest. It may attract industry to the province, should this technology become accepted.

CONCLUSIONS AND PLANS FOR FUTURE WORK

This review concentrated on the description of the main geological and mineral CO_2 sequestration methods that are currently the focus of intensive research by industrialized nations worldwide. At first glance, the most technologically mature methods are storage in active and depleted oil and gas fields, though most of the emphasis lies on maximizing oil and gas recovery rather than sequestration potential. Research relating to injection of CO_2 into deep coal seams is rapidly advancing, with CO_2-enhanced CBM recovery potentially offsetting sequestration costs. Saline aquifers provide huge storage potential in terms of volume for CO_2 sequestration, but they are much more difficult and expensive to characterize than hydrocarbon reservoirs due to the lack of an existing exploration database. The methods , which currently encounter the most resistance from the public, are storage in salt caverns and ocean sequestration. Mineral sequestration is the only method that truly disposes of CO_2 on geological time scale, with a minimum risk for an accidental CO_2 release.

The next stage of our study will expand and summarize the relative technological maturity of the methods covered in this paper and their potential applicability to British Columbia. Since all geological and mineral CO_2 sequestration methods involve the capture and extraction of CO_2 from flue-gases or industrial streams, transportation of CO_2 and its disposal in an appropriate sink, the next stage of our study will also identify the main stationary point sources of

CO_2 emissions and the main potential carbon or CO_2 sinks in British Columbia. The relative geographic relationships between the main stationary point CO_2 sources and sinks is also an essential piece of the puzzle for conceptual decision-making and a base for rigorous CO_2 sequestration planning in British Columbia if it becomes a necessity.

ACKNOWLEDGEMENTS

We would like to thank Terry, P. Mc Cullough of BC Hydro, Stefan Bachu of Alberta Energy and Utilities Board, Barry Ryan of British Columbia Ministry of Energy and Mines and Alan A. Johnson from ZECA Corporation for reviewing the earlier version of this manuscript. Brian Grant, Derek Brown and Dan Green from British Columbia Ministry of Energy and Mines greatly improved the clarity of the final version.

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