9 Greenhouse gases, ozone-depleting gases

9.1   Introduction

By the end of the 20th century it was widely accepted that carbon dioxide and several other gases are involved in physical and chemical processes in the earth's upper troposphere and stratospere (see Figure 4.2) that may result in global climate change. Already in 1896 Arrhenius forecasted rising global temperatures as a result of fossil fuel combustion (Ponting, 1991, Göttlicher, 1999). So-called greenhouse gases (GHGs), most importantly carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) trap the outgoing solar radiation that is reflected by the earth's surface, which leads to global warming. It is noted, though, that the most abundant greenhouse gas in the atmosphere is water causing $\sim {}^2/_3$ of the greenhouse effect. The result of increasing concentrations of GHGs that cause the other $\sim {}^1/_3$ is referred to as the "enhanced greenhouse effect", or, since it is primarily the result of human activities, the "anthropogenic greenhouse effect" (IPCC 1990, Firor and Jacobsen 1993, Smith and Adams 1997, ABCnews 1998).
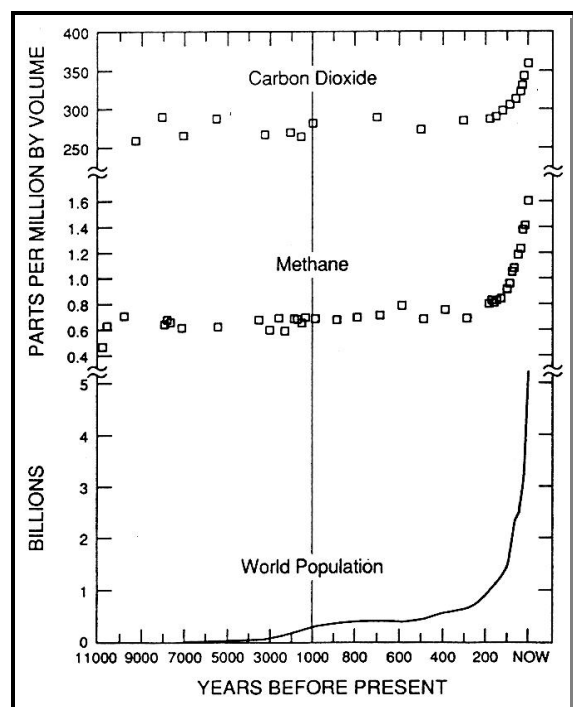
Several other gases such as nitrogen oxides (NOx = $NO+NO_2$), and non-methane VOCs (NMVOCs) such as ethane, CO, tetrachlorocarbon (CCl_4) and chlorofluorocarbons (CFCs) are recognised to be ozone-depleting substances (ODS), *i.e.* substances that do not have a global warming effect but influence the formation and destruction of tropospheric ozone. This may explain the so-called ozone holes over the arctic and antarctic poles. The ozone-layer is crucial for many forms of life on earth by blocking "hard" ultraviolet solar radiation.

Figure 9.1   Increasing world population and carbon dioxide and methane concentrations (picture from Firor and Jacobsen, 1993)

Carbon dioxide is a major product of hydrocarbon combustion and is also found in gasification product gases. Concentrations in flue gases from power plants are ~ 4%-vol CO_2 for natural gas-fired combined cycles (NGCC), ~9%-vol for coal-fired IGCC and ~14%-vol for pulverised coal-fired boilers. It is also present in natural gas, from which is has to be removed down to typically 2.5 %-vol to meet customer specifications (IEA DTI, 2000).

Figure 9.1 gives one of very many illustrations of increasing concentrations of CO_2 and CH_4 in the atmosphere and their correlation with world population, dating back over long periods by using experimental data from *e.g.* air trapped in polar ice. CO_2 concentrations started to rise in ~1800 and are currently increasing at ~1% annually, from about ~355 ppmv in 1990 to ~380 ppmv in 2000. Noticeable increases in CH_4 concentrations are also seen, increasing at ~0.5% annually from 1.7 ppbv in 1990 to ~1.8 ppbv in 2000. Similar trends are seen for the other GHGs and the ODS.

As shown in Figure 9.2, CO_2 is responsible for ~72% of the enhanced greenhouse effect. The combustion of hydrocarbon fossil fuels is the major reason for this.
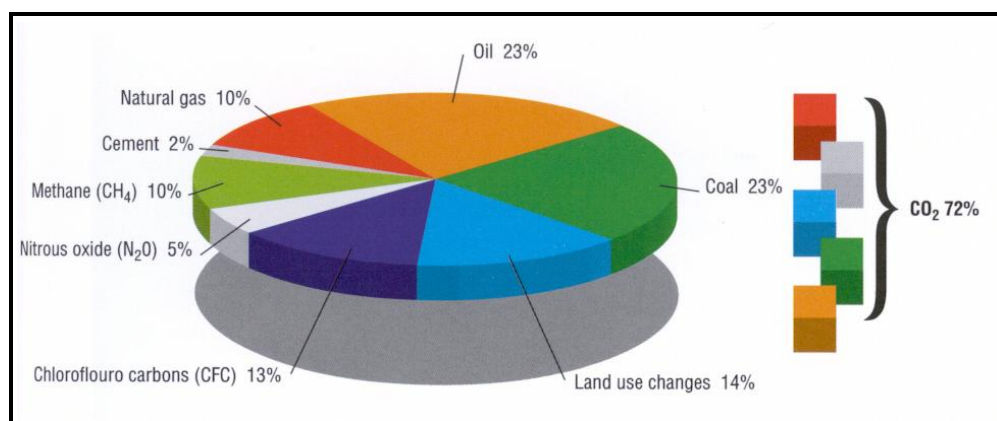

Figure 9.2 Contributions of different gases to the enhanced greenhouse effect (picture from IEA DTI 1993)

Considering other GHG emissions, data from the US from the 1990s given in Table 9.1 shows that fluorocarbon species have minor contributions, appointing 80-85% to CO_2. This table also gives the so-called global warming potentials (GWPs) for the six leadings GHGs that are under discussion in international climate change negotiations. Taking carbon dioxide as a reference with GWP=1 the other GHGs show higher values due to a different radiation absorption behaviour and a different lifetime in the atmosphere. Especially the synthetic GHGs such as the HFCs (which contain H, F and C), PFCs (which contain F and C) and SF_6 are very stable and persistent.

Table 9.1   Six greenhouse gases and their emissions in the US in 1990 and 1998 (data from US EIA, 1997, US EPA 2001a)

GHG compound	Global warming potential (GWP)	% of US GHG emissions (1990)	% of US GHG emissions (1998)
Carbon dioxide, CO_2	1	~85	~ 81
Nitrous oxide, N_2O	310	~2.5	~ 7
Methane, CH_4	21	~12	~ 10
Hydrofluorocarbons, HFC	140 - 11700	< 1	< 1
Perfluorocarbons, PFC	7400	< 1	< 1
Sulphur hexafluoride, SF_6	23900	< 1	< 1

For the different GHG types the three most important sources as reported by the US EPA are listed in Table 9.2.

Table 9.2   US greenhouse gas emissions in 1998: three most important sources per GHG type (data from US EPA 2001a)

CO_2	Fossil fuel combustion industry ~32 %	Fossil fuel combustion transportation ~30 %	Fossil fuel combustion residential ~ 20 %
CH_4	Landfills ~ 33 %	Fermentation ~ 19 %	Natural gas systems ~19 %
N_2O	Agriculture and soil management ~ 70 %	Mobile combustion sources ~ 14 %	Nitric acid production ~ 5 %
HFCs, PFCs, SF_6	Substution of ozone-depleting gases ~ 36 %	HCFC-22 production ~ 27 %	Electrical transmission and production ~ 17 %

It may be concluded that controlling the CO_2 emissions from fossil fuel combustion will have the largest impact on GHG emissions. Large-scale, stationary sources such as power plants and industrial utility boilers, accounting for $\sim^1/_3$ of the fossil fuel-derived CO_2 emissions may be the easiest to control. Transport vehicles may be converted to run on electricity or hydrogen, whilst residential heating may be accomplished with district heating systems or heat pumps (Lindeberg, 1999).

Emissions of methane are very much related to landfills, agricultural activities and land use. Gases emitted from landfills for municipal wastes typically contain ~45-50%-vol CO_2 and 50-55%-vol CH_4. Leachate recycling may increase the gaseous emissions rate. Other methane sources are coal mining (10-100 m³ CH_4 / tonne coal, depending on coal rank and mine depth), rice paddies, and emissions from animals

with a "rumen", *i.e.* pre-stomach, such as cows, sheep and goat. Typical methane emissions are 0.2-0.5 m³/day for a cow, and ten times less for sheep and goat (Japan, 1994). Methane emissions from oil and gas handling and processing on-shore and off-shore may be also significant.

Nitrous oxide emissions are also mainly resulting from agriculture (rice paddies, soils fertilised with *e.g.* $NaNO_3$ and $(NH_4)_2SO_4$) and other types of land-use. Also important are chemical plants such as nitric acid- or nylon processing units. The combustion of fuels containing high concentrations of fuel-nitrogen generally leads to increased NO_x and N_2O emissions, especially when char-N concentrations are high. Fluidised bed combustion (FBC) is most problematic from this point of view: typical N_2O emissions from coal-fired and sewage sludge-fired FBC are 50-150 ppmv and 200-600 ppmv, respectively, against ~1 ppmv for pulverised coal firing. Recent Finnish measurements showed emissions of ~10 mg N_2O/MJ for a wood-fired FBC (say, 20 ppmv N_2O) and 15-40 mg/MJ for a peat-fired FBC (say, 40-100 ppmv) (Leskelä, 2001). Mobile combustion sources, (*i.e.* vehicles) emit more N_2O than stationary sources do: 20 mg/km (plus 50 mg/km CH_4) for a typical gasoline-fired passenger car with three-way catalyst.

Hydrofluorocarbons (HFCs) and perfluorocarbons (PFCs) are synthetic chemicals, produced as alternatives for the ozone-depleting chlorofluorocarbons (CFCs) in response to the "phase out" of CFCs under the Montreal protocol of 1987. HFC-134a is the major substitute for CFCs in refrigerators, the PCFs CF_4 and C_2F_6 are emitted during aluminum 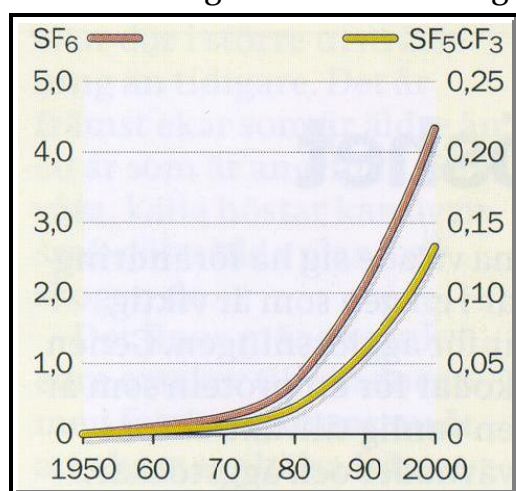smelting. Other PFCs, HFCs and also SF_6 are being used in plasma etching and CVD (chemical vapour deposition) processes. The most important uses and emission sources for SF_6 are electrical transmission systems and circuit brakers, and magnesium production (US EPA, 2001a). It is an extremely stable gas that is inert, non-toxic and non-flammable. Its atmospheric lifetime is estimated at 3200 years, making it the strongest GHG known today. A recent study showed that SF_6 is emitted to the atmosphere ~ 8 months after product sale. Global surface concentrations were found to increase from ~ 0.24 ppt (1 ppt = 10^{-6} ppm) in the early 1970s to ~4 ppt in 1996, as shown in Figure 9.3

Figure 9.3   A t m o s p h e r i c concentrations of SF_6 and SF_5CF_3, **unit : ppt = 10^{-6} ppm** (picture from F&F, 2000)

(Maiss and Brenninkmeier 1998, F&F 2000). SF_6 is responsible for ~0.1 % of the enhanced greenhouse effect. More recently, SF_5CF_3 was detected in the atmosphere. It is not clear whether this compound is made by industry or if it is a breakdown product of SF_6. SF_5CF_3 is an even stronger greenhouse gas than SF_6 and has a long lifetime as well.(Brenninkmeier 2000). Figure 9.3 indicates that current atmospheric concentrations are ~ 0.12 ppt.

Ozone-depleting substances (ODS) influence the formation or destruction of the stratospheric ozone layer. CO, NOx and non-methane VOCs (ethane, propane, butane) are ODS species, stronger though are halogenated compounds such as CFCs and CCl_4. A classification made in the US distinguishes between two types of ODS: class I are the primary ODS comprising the CFCs, CCl_4, and halons (*i.e.* fire extinguishers) such as Halon-1211 (CF_2ClBr) and Halon-1301 (CF_3Br), class II are the partially hydrogenated chlorofluorocompounds HCFCs (US EPA 2001a, b) that pose only a small threat to the ozone layer when compared to CFCs. Based on a reference ozone depleting potential, ODP = 1 for CFC-11, the potential hazard of other ODS can be quantified, *e.g.* ODP = 1 - 10 for halons, 1.2 for CCl_4 and 0.01 - 0.1 for HCFCs. The high ODP for halons is due its bromine content. (US EPA 2001b).

The remainder of this chapter focusses on CO_2 capture and storage and on methods for reducing emissions of other greenhouses and ozone-depleting substances.
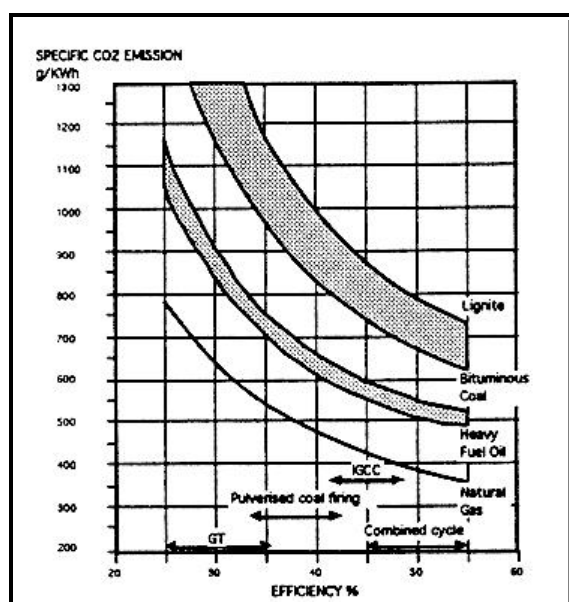

Figure 9.4   Specific CO_2 emissions *versus* combustion process efficiency (GT = gas turbine) (picture from Allam and Spilsbury, 1992)

9.2   Carbon dioxide emissions reduction, capture and storage

Emissions of carbon dioxide are an inevitable result of hydrocarbon fuel combustion for power and electricity generation. This depends partly on the C/H ratio of the fuel: per unit power the CO_2 emissions from CH_4 combustion are less than for coal or lignite. And more efficient processes will produce less CO_2 per unit power generated (*e.g.* g CO_2/kWh electricity): gas turbines are more efficient than condensing steam plants. As illustrated by Figure 9.4 the CO_2 emissions
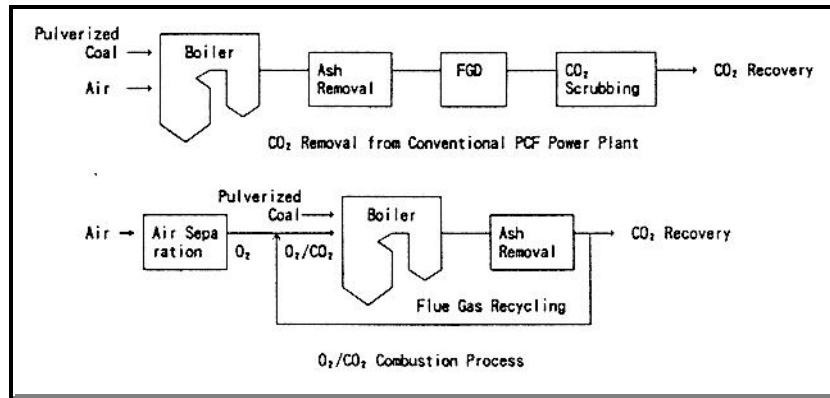
Figure 9.5   Two systems for CO_2 recovery at pulverised coal combustion plants (picture from Nakayama *et al.*, 1992)

per kWh power generated from a low-efficiency coal- or lignite- fired boiler may be 4 times those from a natural gas-fired gas turbine combined cycle plant (NGCC).

Even if a maximum concentration of 500 ppm CO_2 in the atmosphere would be as a limit an emission reduction of ~ 40% has to be achieved by the year 2025, which cannot be achieved by higher increasing energy efficiency alone. Since fossil fuel firing will be the major heat and power source for many years to come, the capture and storage of CO_2 from flue gases cannot be avoided, if such significant emission reductions are enforced. A recent analysis by Göttlicher (1999) lists five approaches to CO_2 retention or "sequestration" from fossil fuel-fired power plants as listed in Table 9.3.

Table 9.3   *Options for CO_2 emissions control from fossil-fuel fired power plants (after Göttlicher, 1999)*

Process type	CO_2 emission control method
Gasification and CO/water-shift	Removal from fuel gas before fuel gas combustion
Conventional combustion	Removal from flue gas
Combustion in O_2/CO_2	Removal of water from flue gas, gives CO_2
Fuel cells	Removal after fuel reforming or from off-gas
"Hydrocarb" *etc.*	Removal of carbon from the fuel before combustion

A conventional process for pulverised coal combustion followed by CO_2 removal from the flue gas and a process where the coal is burnt in a mixture of O_2 and CO_2 are shown in Figure 9.5. A mixture of 70%/30% (vol/vol) CO_2/O_2 gives combustion behaviour very similar to air: due to the higher specific heat (c_p, J/mol.K) of CO_2 compared to N_2 the concentration of O_2 has to be increased from 21 to 30%-vol. CO_2/O_2 combustion was extensively studied during the early 1990s for pulverised coal combustion (Nakayama *et al.*, de Beer *et al.*, 1991) as well as for coal-fired PFBC (Andries and Zevenhoven, 1993). With these processes almost pure CO_2 is obtained

after condensing the water from the flue gas. Costs will be relatively low for CO_2 removal but will be high for the oxygen production (air separation) unit that is needed.

Göttlicher (1999) concludes that for coal-fired systems the most suitable procedure is to gasify the fuel, followed by CO/water shift conversion ($CO + H_2O => CO_2 + H_2$) after water injection, CO_2 removal and combustion of the H_2 gas in a turbine. For gas-fired units the removal of CO_2 from the flue gas is the most suitable method. Similarly, also Lyngfelt and Leckner (1999) conclude that the separation of CO_2 from flue gas or fuel gas will be the least costly option but nonetheless rather expensive.

Removal of CO_2 from flue gases or from gasification product gases has a negative effect on the energy efficiency of the power plant. These losses vary from 7 to 14 percent points and are higher for plants with lower efficiencies. A comparison is given in Table 9.4.

Table 9.4   Effect of CO_2 removal on power plant emissions and efficiency (data from IEA DTI, 2000)

Process	CO_2 capture	Efficiency (% LHV)	CO_2 emissions (g/kWh)
Natural gas CC	-	56	370
	After combustion	47	60
	After CO/shift	48	60
Pulverised coal combustion	-	46	720
	After combustion	33	150
Coal IGCC	-	46	210
	After CO/shift	38	130

The removal of CO_2 from gases is standard procedure for natural gas upgrading, usually in combination with H_2S removal (sour gas stripping), as discussed in chapter 3.13. Chemical or physical sorbents can be used for large gas volumes and/or high concentrations of CO_2. Other methods are adsorption on alumina, activated carbon beds or zeolites, membrane separation or cryogenic methods. For selective removal of large amounts of CO_2, physical and chemical sorbents are the two options.

For a chemisorption process the suitable solvents are MEA (mono-ethanol amine), DEA (di-ethanol amine), ammonia and potassium carbonate, for physisorption the most important processes are the Selexol process which uses DMPEG (di-methyl ether of polyethylene glycol), the Purisol process using NMP (N-methyl -2-
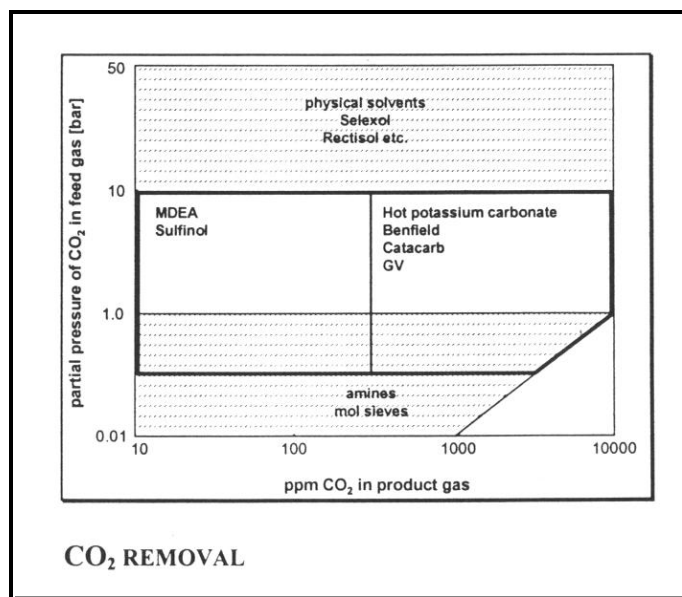
CO₂ REMOVAL

Figure 9.6   Scheme for selecting a CO_2 removal process based on concentration and partial pressure (picture from Bloemendal en Kerkhof, 2000)

pyrrolidone) or the Rectisol process based on cold methanol. Physical sorption is more beneficial for pressurised gases (Bloemendal and Kerkhof, 2000, Göttlicher, 1999, IEA GHG 2000). A selection diagram for CO_2 removal processes is given in Figure 9.6.

For power plant flue gases treatment disadvantages of chemical and physical sorption processes for CO_2 removal are that SO_2 and NO_2, if still present in the gas, react with chemical sorbents which leads to solvent losses. SO_2 is very soluble in physical sorbents such as Selexol and can not easily be recovered (IEA GHG 2001).

After capturing the CO_2 from power plants the more serious problem of long-term disposal or storage arises. Xiaoding and Moulijn (1996) gave an overview of possible chemical products concluding that methanol is an important option. Of course, combustion/oxidation to CO_2 should not be the application for this methanol. Applications of carbon dioxide are very limited when compared to its production in fossil fuel-fired power plants and vehicles, and large-scale sinks for CO_2 must be found that allow for long-term storage without contact with the atmosphere.

Figure 9.7 shows some of five options that are currently considered feasible from an economic as well as an environmental point of view: 1) deep ocean storage, 2) depleted oil/gas fields, 3) enhanced oil recovery (EOR), 4) unmineable coal beds and 5) deep saline reservoirs and aquifiers (see *e.g.* IEA GHG 2001, IEA DTI 2000, Lyngfelt and Leckner 1999, CCT 1999).

Several other options that are considered unattractive from an economical point of view are storage in underground caverns, as solid dry ice and as mineral carbonates. This last option, however is receiving continued attention because of its tremendous potential: enormous natural resources of minerals would be able to fixate CO_2 as
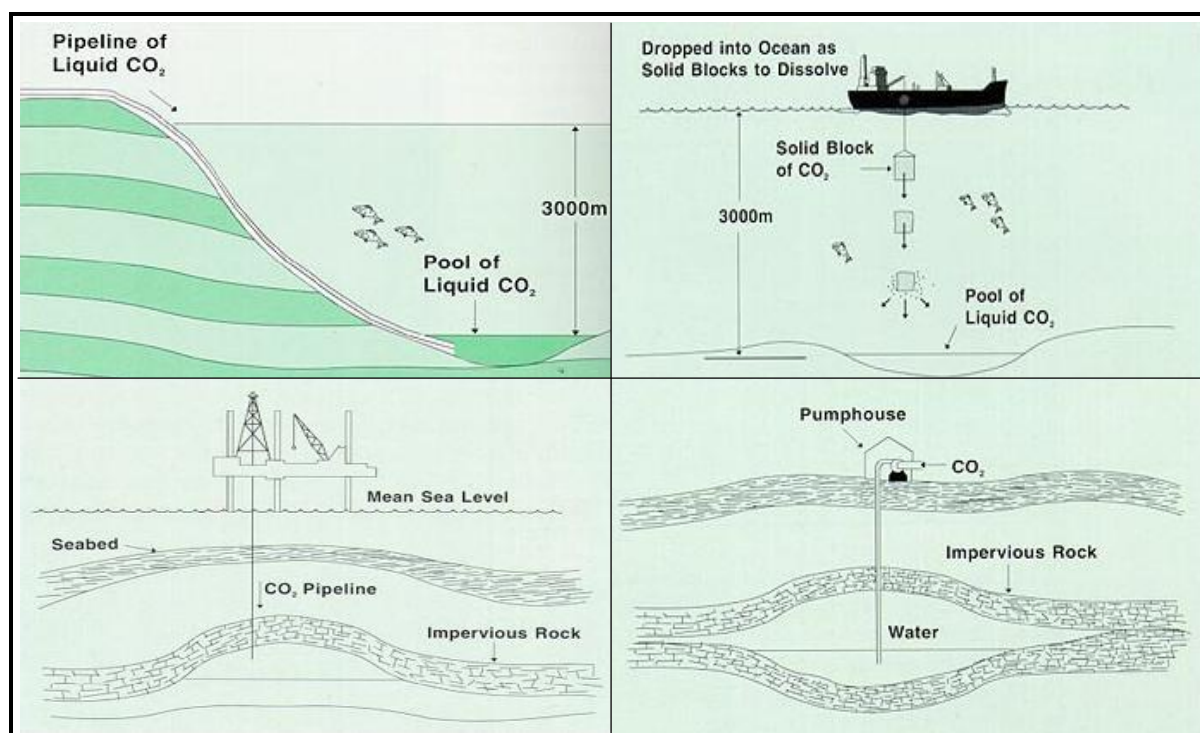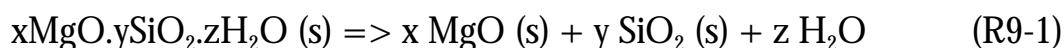
Figure 9.7   Options for large-scale CO_2 storage that are considered feasible: deep ocean storage, deep saline reservoirs or aquifers (pictures from IEA GHG 2001)

mineral carbonates. Especially magnesium silicates (serpentine, olivine, forsterite) have great potential: natural resources (distributed as shown in Figure 9.8) may be capable of binding all fossil fuel -bound carbon (Lackner and Ziock 2000, Ziock 2000).

The chemistry for this CO_2 fixation can be summarised as

$$xMgO.ySiO_2.zH_2O \ (s) => x \ MgO \ (s) + y \ SiO_2 \ (s) + z \ H_2O \qquad \text{(R9-1)}$$
$$MgO \ (s) + CO_2 => MgCO_3 \ (s) \qquad \text{(R9-2)}$$

Large-scale flue gas CO_2 sequestration as mineral carbonates will, however, require enormous amounts of mineral. For a typical power plant the mass flows of fuel and carbonated mineral will be of the same order of magnitude.

Two problems must be solved to make this approach more attractive: 1) extracting the reactive component MgO from the mineral, reducing the mass streams that have to be handled at the power plant, and 2) speeding up the kinetics of reaction (R9-2), which has to take place at temperatures below 300-400°C (and at elevated pressures) (IEA GHG 2000). Resources of magnesium silicate minerals are very large in Finland
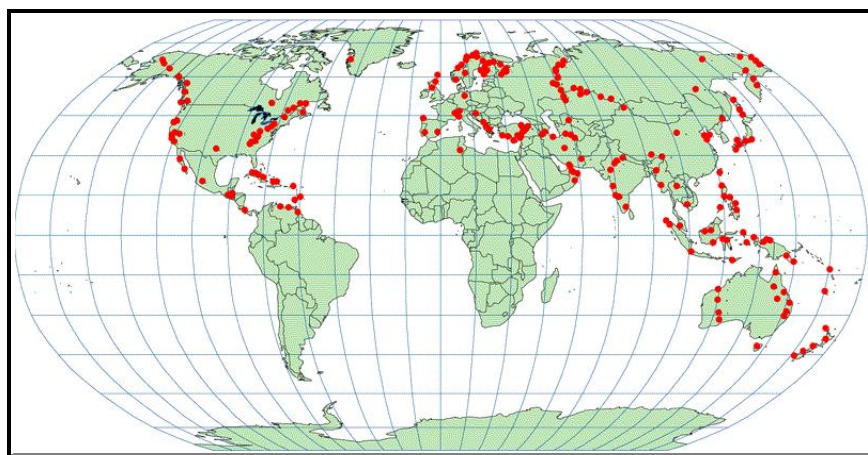
Figure 9.8   World-wide distribution of magnesium silicate minerals (picture from Ziock, 2000)

and since other options that are feasible elsewhere appear to be not applicable to the Finnish situation, research is ongoing on further optimising this option for CO_2 fixation. Mineral carbonation can be done either at the power plant site or at a mine to which CO_2 is then to be transported by pipeline (Kohlmann and Zevenhoven 2001, Kohlmann 2001).

9.3   Emission control for greenhouse gases other than CO_2

Methane emissions are preferably controlled by conversion to CO_2 in a heat or power utility. The GWP of CO_2 is much less than for CH_4. Thermal or catalytic convertors or flares for VOCs (see chapter 6.4) at, *e.g.*, landfills or oil wells, or even fuel cells with a reformer (see chapter 2) may accomplish this.

Nitrous oxide, the most important greenhouse gas after CO_2 when considering combustion, gasification or incineration, was discussed extensively already in Chapter 4. One solution may be to avoid the combustion or incineration of certain high nitrogen-containing fuels at temperatures below 1000°C, such as in FBC.

The other greenhouse gases are organo-halogen compounds that may be adsorbed by activated carbon particles or may be destroyed by thermal or catalytic oxidation (see chapter 6 and 7). High temperatures may be needed for complete oxidation of SF_6, HFCs and PFCs to SO_2, HCl and HF plus CO_2 and H_2O.

9.4   Emission control for ozone-depleting gases

ODS species can be divided into non-methane VOCs and organo-halogen compounds such as CFCs, halons and CCl_4. These can be adsorbed by activated carbon particles or may be destroyed by thermal or catalytic oxidation (see chapters 6 and 7). Oxidation of CFCs, HCFCs and halons gives mainly HCl, HF and HBr, plus CO_2 and H_2O.

Fluidised bed combustion tests burning insulation foams such as extruded poly styrene and poly urethane containing 5-10%-wt CFCs or HCFCs showed > 99.9 % destruction of CFCs at 900EC, whilst HCFCs were destroyed to below detection limits already at 850EC. Some interactions between silicate matter and gaseous fluorine species, mainly HF, were noticed (Mark and Vehlow, 1999).

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