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Environmental Sustainability of Electricity Generation Systems with Carbon Dioxide Capture and Storage

Working Paper

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Introduction

This working paper analyses the environmental sustainability of four electricity production systems that include carbon dioxide capture and storage (CCS):

- Pulverised coal (PC)

- Pulverised coal with oxyfuel combustion (PCOC)

- Natural gas combined cycle (NGCC), and

- Integrated gasification combined cycle (IGCC)

The analysis is based largely on a review of relevant Life Cycle Assessments (LCAs). Thus it considers the environmental sustainability of the entire electricity generation chain from fuel extraction through electricity generation and CO_2 capture to CO_2 storage.

Carbon dioxide capture and geological storage (CCS)

The aim of CCS is to reduce the emissions from industrial point sources of CO_2, which principally result from the combustion of fossil fuels. Figure 1 is a diagram of a coal-based CCS power generation system with storage in an offshore setting. The life cycle starts with the mining of the coal, which is transported to the power station. CO_2 generated in the coal combustion process, which would otherwise be emitted to the atmosphere, is captured at the power plant, compressed and sent to a geological storage site which, in the UK, is most likely to be offshore. Transport to the storage site will probably be by pipeline, although part of the transport route could be by ship. Once at the storage site, the CO_2 will be injected into a deep underground storage reservoir such as a depleted oil or gas field, or a suitable structure in a saline water-bearing reservoir rock (saline aquifer) via a well or wells. The goal of storage is permanent containment of the CO_2. However, the possibility that at some point in the future a portion of the stored CO_2 could leak to domains where it could cause adverse environmental effects also needs to be considered in a full life cycle analysis.
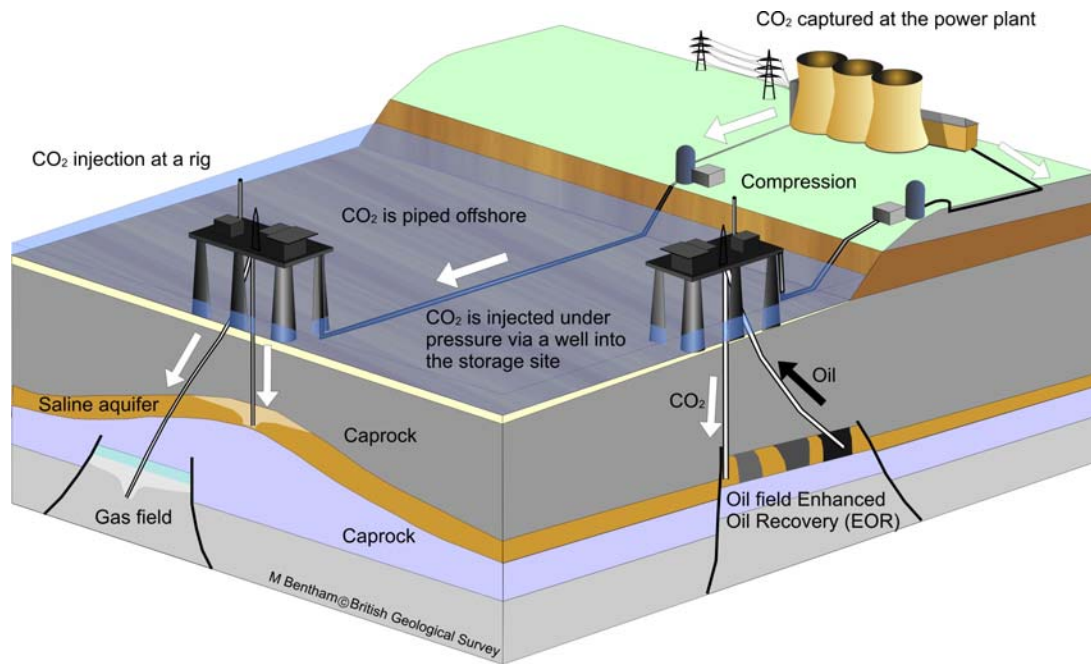
Figure 1. Diagram of the CCS concept

Life Cycle Assessment (LCA)

There can be up to four stages in a typical life cycle assessment: definition of the goal and scope of the study, inventory analysis, impact assessment and interpretation/improvement assessment.

Scope

The scope of the LCA includes definition of the boundaries of the system to be studied. In this review of published LCAs, the boundaries of the systems studied commonly enclose all processes between the fuel extraction (mining of the coal or production of the natural gas) and storage of the captured carbon dioxide in a geological reservoir rock. However, some of the LCAs only consider subsets of this system such as power plants, i.e. they exclude upstream and downstream processes. Some consideration is given to the possibility of leakage from the storage reservoir in one study, but leakage from storage does not lend itself to study by LCA techniques.

Inventory analysis

This step in an LCA involves the compilation of inventories of materials and energy put into the construction, operation and decommissioning of the system, and wastes, materials and energy put out by the system. Diagrams showing the processes involved at each stage of the chain are commonly constructed, indicating the flows of materials and energy through them.

At present, there are no full chain demonstrations of carbon dioxide capture at a power station and geological storage of the captured CO_2. Nevertheless, the flow of energy through power plants fitted for CCS has been analyzed extensively by

industry in both pilot-scale power plants and analogous applications in order to determine the most cost-effective technologies in a range of settings. Consequently, it is well known that CCS reduces the efficiency of power generation systems substantially (Table 1), and will result in higher electricity generation costs. Because the existing literature covers this topic comprehensively, flows of energy through the various systems are not described in detail here.

Table 1. Power plant thermal efficiencies.

Fuel	Power generation technology	CO_2 capture technology	Net efficiency % LHV
Coal	PC (Fluor study)	*None*	*44.0*
		Post-combustion (Fluor)	34.8
	Pulverised fuel (MHI study)	*None*	*43.7*
		Post-combustion (MHI)	35.3
	PCOC (MB study)	*None*	*44.2*
		Oxy-combustion	35.4
	IGCC (Shell)	*None*	*43.1*
		Pre-combustion, Selexol	34.5
	IGCC (GE Energy)	*None*	*43.1*
		Pre-combustion, Selexol	31.5
Gas	NGCC (Fluor/MHI studies)	*None*	*55.6*
		Post-combustion, Fluor	47.4
		Post-combustion, MHI	49.6
	NGCC (MB studies)	*None*	*56.0*
		Oxy-combustion	44.7

Base case plants are in italics, comparable plants fitted for CO_2 capture in regular font. Electrical output is the same for the plants with and without CO_2 capture. PC = Pulverised coal, PCOC = Pulverised coal with oxyfuel combustion, IGCC = Integrated gasification combined cycle, NGCC = Natural gas combined cycle. Fluor = Fluor Daniel, MHI = Mitsubishi Heavy Industries, GE = General Electric. Source: IEAGHG R&D Programme.

Impact assessment

The impacts of obtaining, transporting and using the materials, including the wastes generated along the entire chain from fuel extraction to waste remediation or disposal, need to be taken into account in an analysis of the environmental sustainability of a power cycle with CCS. Such a life cycle assessment, which relates the product or process to its full range of environmental impacts is described as Life Cycle Impact Assessment (LCIA). The results are commonly expressed in a systematic way as an impact table comprising a selection of the commonly used life cycle impact categories shown in Table 2.

Table 2: Life cycle impact categories (from WEC 2004, after US EPA Guidelines and Principles).

Impact category	Scale	Relevant LCI data	Common characterisation factor	Description of characterisation factor
Global Warming	Global	Carbon Dioxide CO_2 Nitrous Oxide N_2O Methane CH_4 Chlorofluorocarbons CFCs Hydrochlorofluorocarbons HCFCs Methyl Bromide CH_3Br	Global Warming Potential (GWP)	Converts LCI data to CO_2 equivalents
Stratospheric ozone depletion	Global	CFCs HCFCs Halons Methyl Bromide	Ozone Depleting Potential	Converts LCI data to trichlorofluoro methane (CFC-11) equivalents
Acidification	Regional Local	Sulphur oxides SOx Nitrogen oxides NOx Hydrochloric acid HCl Hydrofluoric acid HF Ammonia NH_4	Acidification potential	Converts LCI data to hydrogen (H^+) ion equivalents
Eutrophication	Local	Phosphate PO_4^{3-} Nitrogen oxide NO Nitrogen dioxide NO_2 Nitrates NO_3^- Ammonia NH_4	Eutrophication potential	Converts LCI data to phosphate (PO_4) equivalents

Photochemical smog	Local	Non-methane volatile organic compounds NMVOCs	Photochemical oxidant creation	Converts LCI data to ethane (C_2H_6) equivalents.
Terrestrial toxicity	Local	Toxic chemicals with a reported lethal concentration to rodents	LC $_{50}$	Converts LC_{50} data to equivalents.
Aquatic toxicity	Local	Toxic chemicals with a reported lethal concentration to fish	LC $_{50}$	Converts LC_{50} data to equivalents.
Human health	Global Regional Local	Total releases to air, water and soil with a reported lethal concentration to man	LC $_{50}$	Converts LC_{50} data to equivalents.
Resource depletion	Global Regional Local	Quantity of minerals used Quantity of fossil fuels used	Resource Depletion Potential	Converts LCI data to a ratio of quantity of resource used versus quantity of resource left in reserve
Land use	Global	Quantity disposed of in a landfill	Solid Waste	Converts mass of solid waste into volume using an estimated density

In many LCIAs of fossil fuel-fired power generation cycles, only the first four categories are reported. Units used to express these categories are as shown in Table 3. In this study they are expressed per kilowatt hour in order to compare the impacts per unit of electricity generated. One kilowatt hour equals approximately 3.6 megajoules.

Table 3: Commonly used units in power generation cycle LCIAs.

Category	Unit	Unit Abbreviation
GWP	grams of CO_2–equivalent per kilowatt hour	g CO_2-e kWh^{-1}
Stratospheric ozone depletion potential	grams CFC-11 equivalent per kilowatt hour	g CFC-11-e kWh^{-1}
Acidification potential	milligrams SO_2-equivalent per kilowatt hour	mg SO_2-e kWh^{-1}
Eutrophication potential	milligrams PO_4-equivalent per kilowatt hour	mg PO_4-e kWh^{-1}

Improvement assessment and/or interpretation

LCAs have developed as a method to analyse and improve industrial processes. This is not the aim of most of the published LCAs of CCS power generation systems; they commonly interpret the LCA mainly in terms of global warming potential and other facets of environmental sustainability.

Description of the generating plant technologies

The following four generating plant technologies are briefly described below:

1.	Pulverised coal (PC)

2.	Pulverised coal with 'oxyfuel' combustion (PCOC)

3.	Natural gas combined cycle (NGCC)

4.	Integrated gasification combined cycle (IGCC)

In each of the first two systems, a furnace and boiler produce steam which drives a turbine attached to an electricity generator, whereas the latter two employ separate gas and steam turbines to drive generators. All systems use post-combustion flue gas clean-up technologies to meet atmospheric pollution requirements – to reduce emissions of particulates, nitrogen and sulphur oxides – and removal of CO_2 for storage (sequestration) can simply be seen as a further stage in this process.

Pulverised coal

This is the simplest and oldest of the technologies. Coal is milled (pulverised) to a fine grain size and fed into a furnace with an air stream. Combustion in the furnace heats water in a boiler to produce high-pressure steam (Figure 2). The steam is fed through a cascade of turbines, from high-pressure to low-pressure, which drives generators. The exhaust steam is condensed and recirculated as warm water to the boiler to improve heating efficiency. The air stream to the furnace is preheated by heat exchange from the flue gases to improve combustion efficiency. The coal is pulverised to ensure quick and even burning in the furnace.
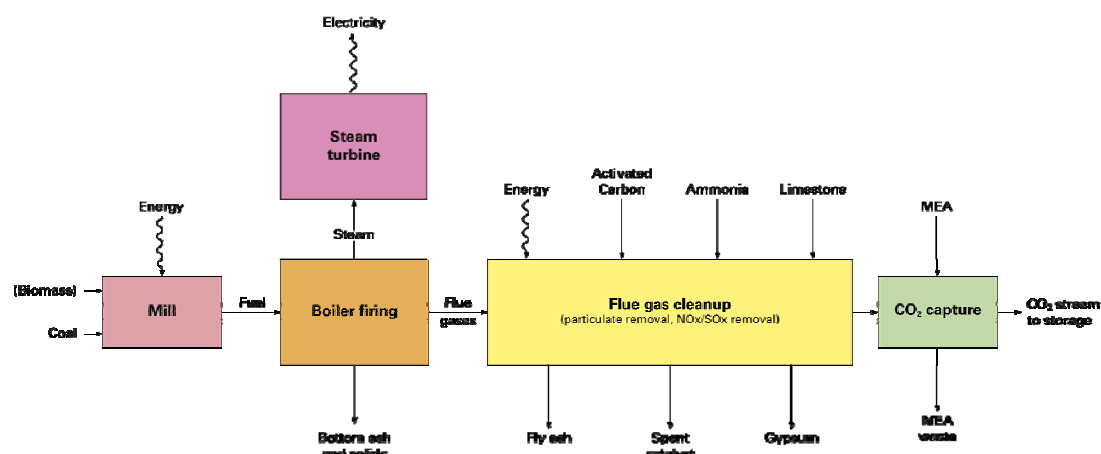

Figure 2. Simplified process flow and material inputs and outputs in the pulverised coal fuel cycle including carbon dioxide capture.

Other fuels may be added to the coal at the milling stage. These include petcoke - an oil refinery waste product, and biomass, such as wood chips or straw. These can be cheaper than coal; and biomass is seen as environmentally acceptable. However, they can reduce burning efficiency and may affect the flue gas clean-up process and the usability of some of its by-products (this is also true of poorer quality coals, e.g. those with high ash or sulphur content). Additives are generally kept below 10% in the fuel feed.

There is a direct solid waste product from the furnace: bottom ash or clinker. All other waste products are entrained in the flue gases. The bottom ash is generally a saleable by-product used in the construction industry.

The furnace flue gas clean-up chain currently includes the following steps to meet stringent air quality criteria in the EU and elsewhere. The order of procedures is important, as they are sequential chemical processes, each having an assumed composition of input gases. For example, the catalyst used in nitrogen oxide removal will be quickly contaminated if the preceding process has not removed the sulphur.

1.   **Particulates removal**. This is usually done by passing the flue gas stream through an electrostatic precipitator. The particulates are attracted to and deposited on electrically charged plates, from which they are mechanically collected as fly ash. This may be a saleable by-product (construction industry) but equally well may be disposed of in landfill, which is environmentally contentious.

2.   **Sulphur removal**. Also known as flue-gas desulphurisation (FGD). In the typical wet FGD process used in major UK power stations, sulphur dioxide in the flue gas is reacted with a slurry of pulverised limestone (calcium carbonate) and water. The sulphur dioxide reacts with the calcium carbonate to produce calcium sulphite and carbon dioxide:

$$SO_2 + CaCO_3 \rightarrow CaSO_3 + CO_2$$
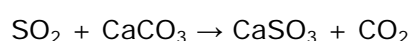
The calcium sulphite then reacts with the oxygen in air introduced into the absorber and water from the slurry and is converted to gypsum (hydrous calcium sulphate):

$$CaSO_3 + 1/2O_2 + 2H_2O \rightarrow CaSO_4.2H_2O$$
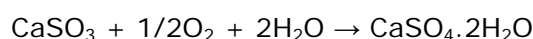
which is collected as a solid by-product and can be sold for use in plaster manufacture (construction industry). Typically the process as currently installed removes about 90% of the SO_2 from the flue gas. The efficiency of FGD would have to be increased to 98% or more in PC systems fitted for CCS because SO_2 reacts with the amine-based absorbents used to capture CO_2 in such systems. Minor amounts of calcium chloride are also produced if there is chlorine in the fuel (Rubin et al. 1991).

3.   **Nitrogen oxide (NOx) removal**. In the selective catalytic reduction (SCR) process, ammonia is injected into and mixed with the flue gases which are then passed over a catalyst at a temperature of about 300—400 °C. The ammonia reduces the nitrogen oxides, yielding nitrogen (N_2; vented to flue) and water (H_2O; vented as steam or condensed out). Various catalysts are used to promote the reaction, depending on the temperature of the exhaust gases. Base metal oxide catalysts have been in use the longest and are useful between approximately 230 °C and 425 °C. Typically these consist mainly of vanadium and titanium oxides and may also contain molybdenum, tungsten and other elements. Zeolite catalysts can be used for high temperature operation (357°C to over 600 °C). Catalyst performance is related to area exposed to contact with the flue gases, so honeycomb structures are used for the metal oxides. The catalysts are not consumed in the reactions but become degraded over time – typically due to residual contamination, e.g. by sulphur, which is not fully removed by FGD. When spent, the catalyst can be at least partially cleaned, regenerated and re-used, or landfilled, which could be environmentally contentious.

The above steps represent a commonly used flue-gas clean-up procedure. There is an alternative method of nitrogen oxide removal: selective non-catalytic reduction (SNCR). This takes place ahead of the other steps, by direct injection of ammonia or urea into the flue gases either within or on leaving the furnace, when they are at a temperature of about 1000 °C. This requires no catalyst, which simplifies the process and reduces costs. However it is less efficient, achieving only a 30—50% reduction in NOx, compared with SCR which typically reduces NOx by >80%, and SNCR uses more of the reducing agent (ammonia/urea) which increases costs. SNCR may also produce N_2O, a greenhouse gas, as a by-product. It is not likely to be used in CCS plants because very low NOx is required to prevent CO_2 capture solvent degradation (see below).

If necessary, trace metals such as mercury can be removed from flue gas e.g. with activated carbon filters, if the fuel contains high levels of such elements. However, it is not expected that mercury removal would need to be implemented specifically because of CO_2 capture, the need depends on the amount of mercury in the (coal) fuel.

Summary of inputs:

Fuel: Coal, optionally co-fired with petcoke, biomass, etc.

Energy: for fuel milling, fuel feed, precipitator, reagent feeds.

Reagents: limestone for FGD, ammonia or urea for SCR/SNCR.

Catalysts: metal oxides and/or activated carbon.

Water for boiler and condensers.

Summary of outputs:

Bottom ash or clinker, from furnace.

Fly ash, from precipitator.

Gypsum, from FGD.

N_2 and water from SCR/SNCR.

N_2O from SNCR.

Unreacted reagents from FGD and SCR/SNCR.

Steam to turbines.

Flue gas to atmosphere or CO_2 capture and storage.

CO$_2$ capture in pulverised coal-fired plants

The CO$_2$ capture process that is most likely to be applied to a conventional supercritical or subcritical pulverised coal-fired power plant is scrubbing using an absorbent such as an amine, which naturally absorbs CO$_2$ at low temperatures and pressures. Amine compounds such as monethanolamine (MEA) are commonly used, in aqueous solution. The process solution also includes additives such as corrosion inhibitors, antifoam agents, oxygen scavengers and salt neutralisers. The capture process and the merits of various amines are described in Thitakamol et al. (2007). In essence, the amines are put in contact with the flue gas in an absorption column, where they absorb most of the CO$_2$ from the flue gas. The CO$_2$-charged amine solution is then taken to a regenerator, where it is heated to drive off the CO$_2$, which is collected and conditioned ready to be sent for storage. The amine is then taken back to the absorption column and re-used. A small proportion of the amine evaporates with the treated flue gas in the absorption column and could go up the stack to atmosphere. However, if a water wash process is applied to the treated flue gas, amine emissions can be reduced to zero or extremely low levels (Thitakamol et al. 2007). Information on the environmental and health impacts of MEA is available at http://www.dow.com/productsafety/finder/mea.htm.

Loss of MEA is also caused by both reversible and irreversible interactions with flue gas components, so some replenishment of the amines used in the process is required. Oxidation of amines results in the production of salts, some of which are heat-stable. The heat-stable salts are mostly formed by reaction of MEA with acids such as carboxylic acid. These acids are generated by reaction of MEA with gaseous components of flue gases such as O$_2$, CO and SO$_2$, or are introduced into the capture process along with water and feed gas streams. The salts typically comprise acetates, formates, glycolates, oxalates, sulphates, thiosulphates and thiocyanates (Thitakamol et al. 2007). They are collected by filters in the amine pipework and also from a slipstream of the amine stream. NaOH can be used to reclaim amines from some of the salts, but the remaining sludge and solid products that result from the irreversible reactions have to be collected and then disposed of by incineration (e.g. in the power plant furnace) and/or landfill. This could be contentious, especially if practised on a very large scale, as this waste is considered to be hazardous (Odeh & Cockerill 2008): it contains heavy-metal corrosion inhibitors (Thitakamol et al. 2007).

A similar amine absorption process is likely to be used to capture CO$_2$ from NGCC power plants.

Pulverised coal with 'Oxyfuel' combustion

So-called 'Oxyfuel' combustion has been used for some time for attaining higher furnace temperatures and more efficient coal burning in industries such as glass manufacture. An oxyfuel system may be retrofitted to an existing furnace or, more effectively, built-in to the design of a new furnace. In either case the mode of operation is the same. The configuration (Figure 3) is that of a PC plant with the addition of an air separator and a flue gas feedback loop.

Air is fed into the separator where the oxygen (O_2) is separated from the remainder of the gases in air. There are several possible and competing separator technologies, including cryogenic, vacuum pressure adsorption and membrane. All these processes consume energy and there is much research and associated debate on their relative efficiencies. The oxygen-deficient air (mainly nitrogen) from the separator is vented to the atmosphere. The O_2 is fed into the furnace with the fuel, in place of the air feed of a conventional PC furnace.
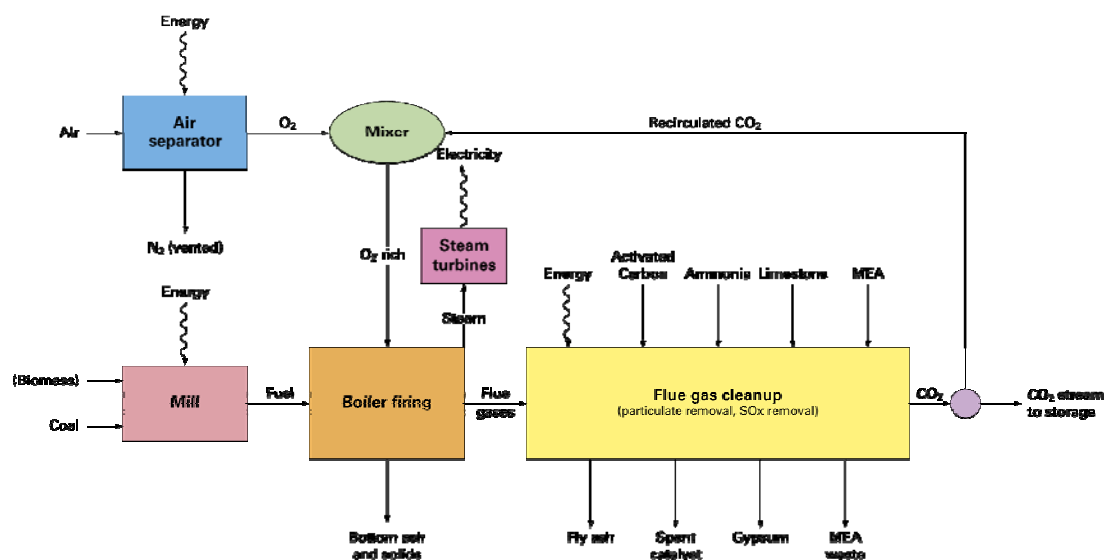

Figure 3. Simplified process flow and material inputs and outputs for oxyfuel-fired plant

Burning the fuel in pure oxygen would produce temperatures too high to contain and would result in oxidation of the furnace components. Therefore, burning is moderated by feeding back some of the oxygen-poor flue gases into the furnace. The flue-gas feedback is typically taken from between the precipitator (so particulates are not recirculated) and the FGD. It runs to a mixer, between the air separator and the furnace. The mixture is adjusted to provide an optimum temperature in the furnace, so that fuel is burned quickly, fully and efficiently but the temperature and oxygen level does not damage the furnace itself.

The flue gas clean-up chain is basically the same as that for a conventional PC plant. However, since the air separator removes the nitrogen, there is negligible NOx in the flue gas stream, just a little from residual nitrogen and any nitrogen compounds in the fuel, so the NOx removal step is unnecessary. Therefore the SCR/SNCR reagent and SCR catalyst are not required. However, flue gas desulphurisation is necessary.

The treated flue gas consists almost entirely of carbon dioxide, so a CO_2 capture step is not required. Note also that the capture rate is extremely high, e.g. 99.5% in the only LCA in which it was modelled (Viebahn et al. 2007). However, there

are significant energy demands from the air separation plant that provides the oxygen for the process.

Although oxyfuel combustion has been in production use in other industries, it is still relatively new in the power generation industry. In early 2009, there are only small-scale technology demonstration and evaluation plants in operation; however, these have produced positive results and both plant manufacturers (e.g. Doosan Babcock) and power generators (e.g. E.ON) hope to have production-scale plants in operation by end 2009. This is likely to be achieved by retrofitting oxyfuel systems to existing PC plant.

Summary of inputs:

Fuel: Coal, optionally co-fired with petcoke, biomass, etc.

Energy: for fuel milling, fuel feed, air separator, precipitator, reagent feed.

Reagent: limestone for FGD.

Oxygen: for fuel combustion.

Water for boiler and condensers.

Summary of outputs:

Bottom ash or clinker, from furnace.

Fly ash, from precipitator.

Gypsum, from FGD.

N_2 from air separator.

Unreacted reagent from FGD.

Steam to turbines.

CO_2 to storage.

Gas combined cycle systems

There are two variants of combined gas cycle systems: natural gas combined cycle (NGCC) and integrated gasification combined cycle (IGCC). Both use the same two-stage generating 'back-end', with a primary gas turbine and a secondary steam turbine generator. The difference is in the fuel gas: NGCC uses natural gas (methane), commonly from the gas grid; IGCC uses 'synthesis gas' which is produced by on-site coal gasification. IGCC (gasification) plants can also produce hydrogen, which can either be sold or used for power generation. Hydrogen can also be produced by steam reforming natural gas.
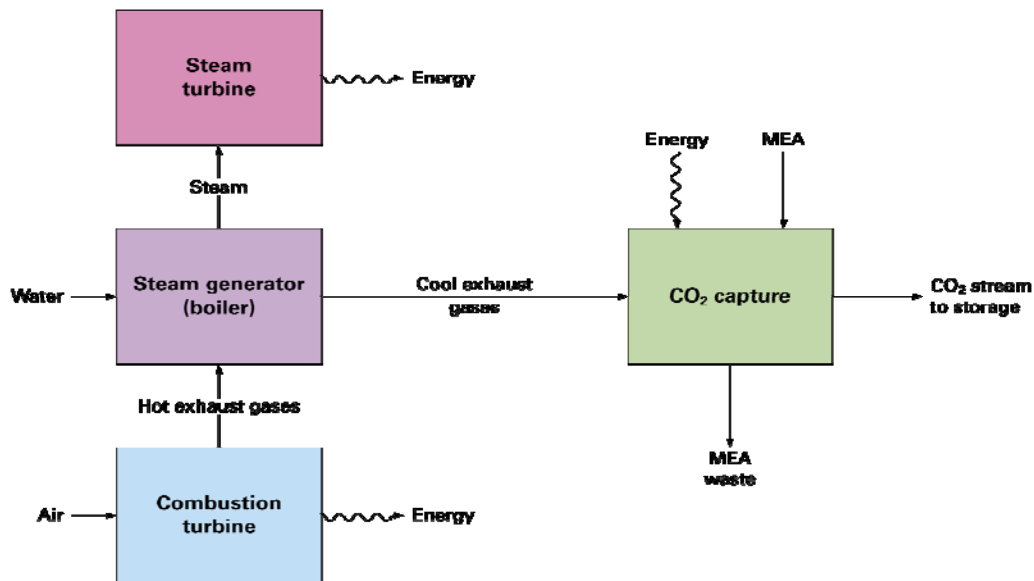
Natural gas combined cycle

Natural gas is the simplest and commonly cited as the cleanest of the GCC systems. It uses natural gas (methane) from the gas grid which is injected into a gas turbine with air to be compressed and burnt (Figure 4). The hot exhaust gases drive the turbine. The turbine shaft drives the gas/air compressor and the generator. The hot exhaust gases from this turbine pass through a boiler, where they are used to produce steam as they are cooled. The cooled exhaust, mostly CO_2 and water vapour, is vented to the atmosphere. The steam from the boiler is fed to a steam turbine generator, and then recycled to the boiler.

There is no requirement for flue gas clean-up at the power station since the natural gas has been processed such that it consists of almost pure methane before being added to the gas grid.

Natural gas processing

Natural gas commonly exists in natural gas fields deep underground as a mixture of methane with variable amounts of higher hydrocarbons. In many fields, other gases are also present, typically water vapour, H_2S, CO_2, helium and nitrogen. This gas mixture has to be purified before it can be added to the national gas grid. Sand scrubbers are commonly installed near the wellheads at the gas field to remove sand that may flow into the well with the gas. Heaters may also be at the gas field, to keep the gas out of the hydrate stability field to avoid methane hydrate plugging any pipelines. Further processing then takes place, either at the production site or at a more distant natural gas treatment plant. Water, any higher hydrocarbons, and any so-called sour gases (H_2S and CO_2) are removed from the gas stream. This usually takes place in four stages: oil and condensate removal, water removal, separation of natural gas liquids and H_2S and CO_2 removal.

Condensate is a term for light hydrocarbons that are gaseous under reservoir conditions underground but liquid at surface temperatures and pressures. Condensates typically comprise a mixture of the lighter straight chain alkanes (the lighter hydrocarbons). Oil and condensate are removed by condensing them

Steam turbine ⟿ Energy

Steam

Energy   MEA

Water → Steam generator (boiler) — Cool exhaust gases → CO_2 capture → CO_2 stream to storage

Hot exhaust gases

MEA waste

Air → Combustion turbine ⟿ Energy

from the raw gas stream. The temperature and pressure of the gas stream is lowered slightly by expanding it, and any oil and condensate condense out. A proportion of any water in the gas may be condensed at this stage as well. Further water is then removed either by dehydrating the gas, e.g. by using glycol or a solid desiccant to absorb water vapour from the gas stream in absorption towers. After this treatment some higher hydrocarbons still remain mixed in with the methane. These are gaseous at surface temperature and pressure but have a higher value when sold separately than when left in the natural gas: when separated they are known as natural gas liquids (NGLs). They comprise a mixture of ethane, propane, butane, isobutane and pentanes. They are separated either by absorption (into oil) or cryogenic separation. Following removal of natural gas liquids, H_2S and CO_2 are removed by amine absorption, as described for CO_2 capture from power plant flue gases above. Thus the amine absorption CO_2 capture techniques that are likely to be used in CCS have been in use in the natural gas processing industry for decades.

Summary of inputs:

Fuel: Natural gas (methane).

Additional energy: For plant start-up only.

Water for boiler.

Summary of outputs:

Steam to secondary turbine.

Treated flue gas to atmosphere

CO_2 to storage.

Figure 4: Simplified process flow and material inputs/outputs for natural gas combined cycle system.

Integrated gasification combined cycle

In IGCC, the fuel gas is produced from an on-site (typically coal) gasification plant (Figure 5). Gasification takes place in the presence of controlled amounts of oxygen, so that reducing conditions are maintained. The gasifier works on the same principle as the old 'town gas' plants common in the UK up until the 1970s.
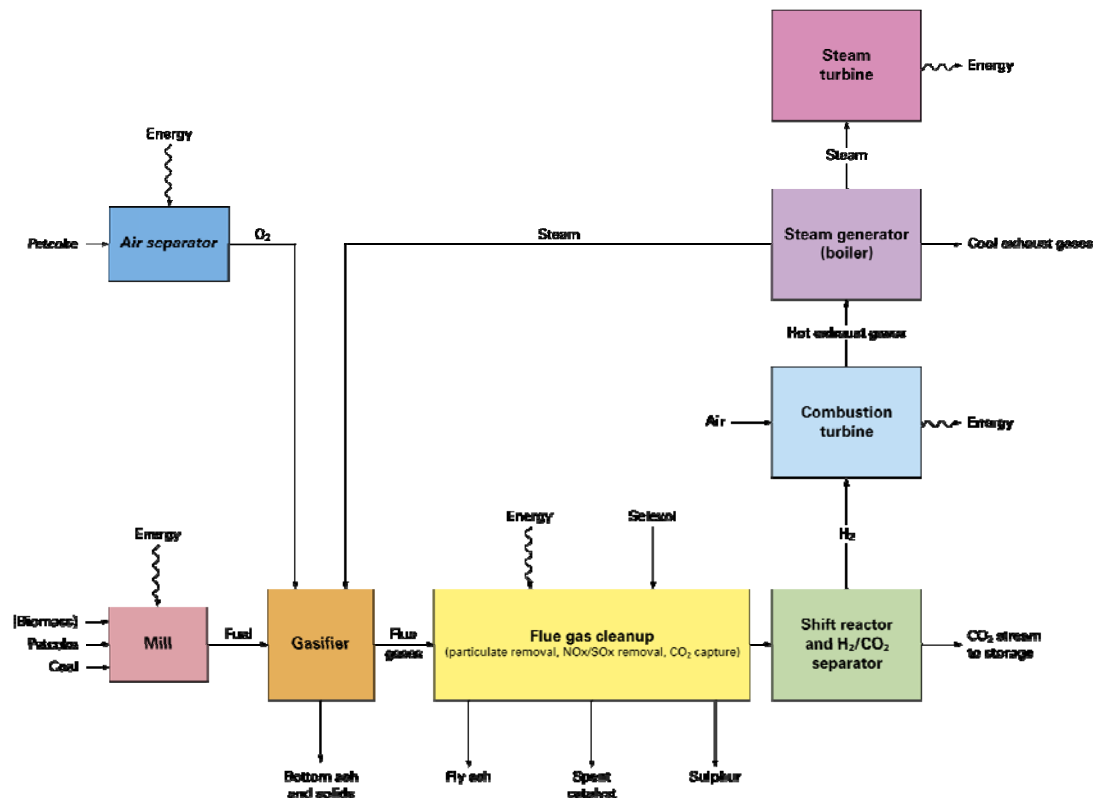

Figure 5. Simplified process flow and material inputs/outputs for integrated gasification combined cycle system.

The gasifier is a pressurised retort which is fed with coal (and optionally an admixture of petcoke, biomass, etc.), oxygen from an air separator and steam siphoned off from the steam turbine component of the system. The reactions in the gasifier produce a mixture of hydrogen and carbon monoxide known as synthesis gas or syngas, and sulphur gases mainly consisting of H_2S but including a little COS (which later in the process is reduced to H_2S), plus impurities such as HCN, NH_3, and volatile trace minerals, along with solid waste products. The solid waste, known as frit, is glassy but otherwise similar to ash and is collected and may be used as a by-product, e.g. for road foundations, or safely landfilled.

Typically, the raw gas stream passes through a system akin to flue-gas clean-up in a PC plant. First, an electrostatic precipitator is used to remove fine particulates, which are disposed of as fly ash. If necessary, chlorine may then be captured. If necessary, activated charcoal is used to capture any mercury derived

from the fuel. The gas stream then passes through a hydrolysis process which reduces the COS to H_2S and then it goes on to a desulphurisation plant. Here, as the gases are at higher pressure than in a PC plant, it is possible to use a glycol-based physical solvent such as Selexol or a refrigerated methanol-based physical solvent such as Rectisol to remove the H_2S. These solvents are cycled through the FGD, where they absorb the sulphur gases under pressure, then through a pressure reduction system that allows the gases to come out of solution and be collected for chemical conversion via the Claus process to sulphur – a useful saleable by-product. Sulphur removal efficiency is typically around 98%. A little solvent can be lost by vapourisation in the absorption process in this cycle – e.g. approximately 0.005 kg Selexol MWh^{-1} in a 500 MWe plant (IPCC 2005). Next the CO and added water are 'shifted' to H_2 and CO_2 via a strongly exothermic shift reaction in the presence of a catalyst. Finally, the CO_2 can be captured. Because the CO_2 is at relatively high pressure it is most economical to use a physical solvent such as Selexol in the capture process. Once captured, the CO_2 can be conditioned and sent to storage, and the syngas, now consisting largely of hydrogen, can be combusted to make electricity or sold. The fact that the IGCC plant produces hydrogen gives it the flexibility to produce either energy vector, which is potentially very useful over a ~40 year lifetime.

Variations on this process have been proposed. In some cases the CO-CO_2 shift reaction may take place prior to desulphurisation, in which case capture of sulphur gases and CO_2 may take place in a single step with a 2-stage pressure let-down to allow separate collection of the H_2S and CO_2 if required.

Summary of inputs:

Fuel: Coal, optionally co-fired with petcoke, biomass, etc.

Energy: for fuel milling, fuel feed, air separator, precipitator, reagent feed.

Reagent: solvent for FGD.

Water for boiler.

Summary of outputs:

Vitrified solids (frit), from gasifier.

Fly ash, from precipitator.

Sulphur, from FGD.

N_2 from air separator.

Spent solvent, from FGD.

CO_2 to storage.

Review of existing LCAs of electricity generation with CCS

Global Warming Potential

Odeh & Cockerill (2008) undertook a life cycle greenhouse gas assessment of power plants with CCS under UK conditions. All their power plant case studies were located in the UK, with CO_2 storage in depleted natural gas reservoirs beneath the Southern North Sea. They selected a subcritical PC plant as their base case and compared it to supercritical PC, NGCC and coal-fired IGCC plants with and without CCS. They used the EcoInvent database (EcoInvent 2006), which contains data applicable to Western Europe in general, to obtain some of the values for process chain analysis.

The results of their life cycle analysis of the global warming potential (GWP) of the various plants with and without CCS are shown below (Table3). Table 3 also illustrates the important point that the percentage reduction in GWP achieved depends on the 'base-case' against which the CCS technology is compared, e.g. typical current UK coal-fired technology without CCS or a plant of the same type with the same electrical output without CCS.

Table 3. GWP of coal-fired power plants with and without CCS (from Odeh & Cockerill 2008).

Plant type	GWP (g CO_2–equivalent kWh^{-1})	% reduction
Subcritical PC*	984	N/A
Supercritical PC	879	0
Supercritical PC with CCS	255	71
NGCC	488	0
NGCC with CCS	200	59
IGCC	861	0
IGCC with CCS	167	81

* Typical UK current coal-fired technology

This study also considers the additional pollutants that are emitted to air as a result of CCS. NOx and ammonia emissions increase in PC and NGCC cycles with CCS. NOx emissions per kWh increase because more fuel is used and there is no upgrading of the SCR plant. The increase in ammonia emissions is due to slippage

of ammonia in the SCR plant and the oxidation of amines used in the capture plant to organic acids and ammonia. These emissions increase the eutrophication potential, which can approximately double in a PC plant with CCS, and increase the acidification potential due to increased NOx. However SO_2 emissions decrease significantly in PC with CCS due to the necessary increase in FGD efficiency. Human toxicology also increases due to the increased emissions of heavy metals to water and the generation of hazardous MEA waste.

This study also considered the sensitivity of GWP to a range of factors. The authors conclude that the length of the CO_2 pipeline has little effect on LC GWP. The greatest sensitivity was to changing the origin of the coal from the UK to Russia (which increased GWP in PC with CCS by 16.9% and IGCC with CCS by 24.4%) and decreasing capture efficiency by 5% (which increased GWP by 11.3 - 25.6%).

These results are broadly similar to those in a range of studies where the cases are in other parts of the world. None of the studies are exactly comparable because they make slightly different assumptions about either fuel transport, fuel supply, CO_2 capture efficiency, CO_2 transport or systems boundaries.

In a very similar study, based on German conditions and confined to IGCC technology, Mayer-Spohn et al. (2007) considered 90% pre-combustion CO_2 capture in a hard coal-fired IGCC with 300 km pipeline transport of the captured CO_2. Life cycle emissions from electricity generation with CCS were calculated to be:

   141 g CO_2 kWh^{-1}

   556 mg SO_2 kWh^{-1}

   615 mg NO_x kWh^{-1}

   2594 mg CH_4 kWh^{-1}

   53 mg NMVOC kWh^{-1}

There is an increase in the last four emissions of approximately 20% compared to the same plant without CCS. The emissions result in:

- Global warming potential of 208g CO_2-equivalent kWh^{-1}

This represents a 77% reduction in GWP, from 904 g CO_2-equivalent kWh^{-1} in the same plant with the same electrical output but without CCS.

- Acidification potential of 1028 mg SO_2-equivalent kWh^{-1}

- Eutrophication potential of 99 mg PO_4-equivalent kWh^{-1}

In an earlier study, Mayer-Spohn et al. (2006) considered the environmental burden linked to carbon capture and storage with 80% CO_2 capture in a hard

coal-fired IGCC using German coal. In this study they used BALANCE software developed for LCA at the University of Stuttgart and the EcoInvent database. They assumed significant transport of the captured CO_2 (80 km by pipeline and 1000 km by ship). They found a reduction in GWP per kWh of 63% compared to the same plant without CCS. Note that in this study 80% capture efficiency was considered and CO_2 transport distances were significantly greater than in the studies cited above. This accounts for the lower GWP reduction.

Viebahn et al. (2007) conducted a life cycle analysis on fossil fuel systems with CCS with 88% capture on PC and NGCC plants and 99.5% on PCOC plants. They used 2020 as a reference year and consequently assumed higher power plant efficiencies than are available at present. They chose Germany as the reference area and used the Umberto software (IFEU and IFU 2006) for the modelling. LCA assessments of conventional power plants, fuel processes and gas pipelines from the Umberto LCA database and the EcoInvent database were used in the analysis. Their results (Viebahn, personal communication) are shown in Table 4 below.

Table 4.   GWP (g CO_2-equivalent kWh^{-1}) from selected fossil fuel-fired power generation cycles with and without CCS from the Viebahn et al. 2007 study, updated 2008 (P. Viebahn, personal communication).

Plant type	PC + hard coal	PC + hard coal + CCS	PCOC + hard coal	PC + lignite	PC + lignite + CCS	NGCC	NGCC + CCS	IGCC + hard coal	IGCC + hard coal + CCS
GWP	791.9	261.5	175.8	897.1	197.9	395.9	132.0	773.5	244.5
% GWP reduction		67	78		78		67		68

Viebahn et al. (2007) also considered the changes in other Life Cycle Impact categories (Table 5).

Table 5. Life Cycle Impacts from pulverised coal-fired power plants across a range of categories from the Viebahn et al. (2007) study, updated 2008 (P. Viebahn, personal communication).

Impact category	PC	PC with CCS	% increase
Photochemical Ozone Creation Potential (in ethylene equivalents (g kWh^{-1})	0.03161	0.06136	94
Eutrophication potential in PO_4 equivalents(g kWh^{-1})	0.06389	0.09206	36
Acidification potential in SO_2 equivalents(g kWh^{-1})	0.07988	0.01110	-10
PM10-equivalents	0.02268	0.02321	2
Cumulative (non-renewable) Energy Demand in kJ*100/kWh,el	0.07739	0.09884	28

Spath & Mann (2004) studied the LC emissions of coal-fired PC, NGCC and biomass-fired power plants. They made slightly different assumptions from the other studies in that for the power generation systems with CCS they kept power production constant by adding capacity generated from an NGCC plant without CO_2 capture rather than calculating the emissions per kWh of the (reduced capacity) plant with capture. Thus their results are not directly comparable to those of the other studies cited here.

They found that their base-case PC power plant without capture had a GWP of 847 g CO_2-equivalent kWh^{-1} and their PC system with CCS emitted 247 g CO_2–equivalent kWh^{-1}, i.e. there was a 71% reduction. With urban-sourced biomass co-firing, the PC system with CCS had a GWP of 45 g CO_2-equivalent kWh^{-1}; a 95% reduction compared to the base-case coal-fired PC plant.

Spath & Mann's (2004) analysis of an NGCC system showed a reduction in GWP from 499 g CO_2-equivalent kWh^{-1} to 245 g CO_2-equivalent kWh^{-1}. The system shows the addition of CCS produces a smaller reduction in GWP than PC (59%), but the systems with CCS are approximately equivalent.

Although outside the scope of this study, Spath & Mann (2004) also consider the LCA of two types of biomass-fired power plants with CCS: a direct-fired biomass power plant using biomass residue, and a biomass-fired integrated gasification combined cycle system using a biomass energy crop. They conclude that biomass-fired power plants with CCS have the potential to produce substantially

negative GWPs because they capture and store CO_2 extracted from the atmosphere by the plants that act as the fuel source.

LCA techniques applied to part of the full CCS chain

Nazarko et al. (2006) considered retrofit and new build PC fired power plants, with 90% post-combustion CO_2 capture using MEA; operations were modelled commencing in 2005, 2010 and 2020. The systems boundary of their study was placed at the plant fence, i.e. upstream and downstream activities in the full CCS power generation chain were not included. GaBi 4.2 software was used for the study.

Their results include figures for the amount of waste and hazardous waste produced by the power plants, and emissions to air and water. They also indicate a strong decrease in CO_2 emissions for the plants with CO_2 capture (from between 769 and 662 g kWh^{-1} to 106-81 g kWh^{-1} depending on when the plant was built and thus its efficiency). This represents a reduction of 86 – 88% for the individual retrofitted or greenfield plants. There was also a significant decrease in the SO_2 emissions. The latter is caused by the necessity to improve flue gas desulphurisation as the presence of SO_2 in the flue gas degrades the MEA solvent.

Life cycle emissions from the 2010 plant retrofitted for capture in 2020 are:

95 g CO_2 kWh^{-1} (an 86.5% reduction compared to the same plant without CO_2 capture)

> 70 mg SO_2 kWh^{-1}
>
> 620 mg NO_x kWh^{-1}
>
> 80 mg NMVOC kWh^{-1}

In a poster presentation, Schreiber et al. (2007) considered the same PC power plants and included upstream but not downstream activities in their analysis. The values for upstream activities were taken from the EcoInvent database. The analysis, as in the previous study by Nazarko et al. (2006), was conducted using the GaBi 4.2 software. They note increases in consumption of hard coal, boiler feed and cooling water, auxiliary power and steam, and increased production of gypsum, sludge, waste, and waste heat, caused by the decreased efficiency of the power plant due to the CO_2 capture facilities. CO_2 and SO_2 emissions decrease (the latter because FGD efficiency has to be increased to make the CO_2 capture process work efficiently). The photochemical oxidation potential is slightly reduced due to the decreased SO_2. Human toxicological potential and acidification potential are increased slightly and eutrophication potential is significantly

increased due to emission of NO_2 and NH_3. Unfortunately exact values are not given.

Livengood et al. (1993) analysed emissions from a 458 MW IGCC plant with and without CCS. Without CCS the net power output was 448 MW with emissions of 872 g CO_2-equivalent kWh^{-1}. With CCS, the net electric power production was 383 MW with emissions of 218 g CO_2-equivalent kWh^{-1} (a 75 % reduction).

Leakage from geological CO_2 storage sites

The goal of CCS is permanent storage. However, analysis of problems at natural gas storage sites (Evans *in press*) indicates by analogy that a proportion of CO_2 storage sites may experience leakage.

The possibility of leakage of CO_2 from the storage site is not commonly included in LCAs, but is briefly considered by Viebahn et al. (2007). They note that leakage could take place at any point or period in 10,000 years or more from the time of storage. They also point out that some LCAs use GWP 100 as a measure of GWP, i.e. the global warming potential over a period of 100 years from the time of the emission and thus implicitly exclude long-term leakage.

The LCA technique is difficult to apply meaningfully to leakage of CCS from storage over the long term because there is no track record on which to base an estimate the likelihood or magnitude of any leakage, and it may never be appropriate to assume an average rate of leakage, because it is clear that any leakage is likely to be highly site-specific. This is because storage sites are highly variable natural systems that, in many cases, have been affected by man's activities in site-specific ways, e.g. by drilling and pressure reduction as a hydrocarbon field is developed (Holloway et al. 2007).

It is possible that in many cases leakage from CO_2 storage sites may be reparable. A high proportion of leaks in natural gas storage systems are associated with wells or above-ground infrastructure. Onshore at least, these commonly can be remediated relatively simply using currently available oilfield techniques. Offshore they may prove much more expensive to remediate as it is difficult if not impossible to re-enter plugged and abandoned offshore wells using currently available techniques.

Leaks that are caused by unidentified migration pathways (such as faults or other fractures) through the natural geological system may prove more difficult or impossible to remediate other than by reducing the pressure within the storage reservoir, effectively curtailing the storage project.

Even if a CO_2 storage project did contain a leak that could not be remediated, it should be borne in mind that a significant proportion (5-30%) of any CO_2 stored is likely to be retained as a residual saturation in the pore spaces of the reservoir rock, even in the event of complete seal failure (Ennis-King & Paterson 2001).

Apart from its GWP impact on the atmosphere, leakage of CO_2 onshore could cause localized damage to the environment (Holloway et al. 2007). Onshore it could also be dangerous to man and other organisms as it is an asphyxiant. In the latter case, it is the concentration rather than flux that is important (Hepple 2005).

Leakage of CO_2 from offshore storage sites could cause damage to the marine environment, both to sediment-dwelling organisms and by acidification of sea water (Turley et al. 2006, Blackford et al. 2008).

Discussion of the LCAs of fossil fuel-fired power generation cycles with CCS

Data issues

Probably because of lack of space, none of the published or otherwise presented LCAs give a full breakdown of all the input/output values for all parts of the CCS chain. These values need to be available if detailed comparisons between LCAs are to be made.

It is not clear in every case where data for process chain analysis comes from. However, it is clear that several of the studies conducted in Europe use the EcoInvent database, which includes data values for Western Europe in general, to provide at least some of the input and output data for process chain analysis.

Major sources of emissions with GWP along the CCS chain

The major sources of emissions with GWP are upstream and at the power plant. Important points of upstream emissions are methane emissions from coal mining, especially deep coal mining, and natural gas production and transport. Natural gas processing and coal mining operations may also produce significant CO_2 emissions.

At the power plant, PC, NGCC and IGCC power plants fitted for CO_2 capture directly emit about 10% of the CO_2 that they generate up the flue gas stack to atmosphere, whereas oxyfuel plants capture almost all the CO_2 that they generate. Significant CO_2 is also generated at the CO_2 compressors prior to transport.

Potential for improvement of environmental performance

In terms of environmental performance, there may be some room for improvement in parts of the chain, particularly the upstream parts. Analysis by Viebahn et al. (2007) points out that if methane emissions from German (underground) hard coal mines were to be reduced by 80%, which is probably technically possible, the LC GWP reduction of the hard coal-fired power plants with CCS studied would be improved by up to ten percentage points relative to the plants without CCS, i.e. from 65% to 75%. CO_2 emissions from natural gas processing plant can be captured and stored (as is taking place at present at the Sleipner facility in the North Sea and the In Salah project in Algeria) and leakage from natural gas production and transportation pipelines could be reduced. A

more efficient power plant, higher CO_2 capture rate or more efficient capture process would also contribute to better environmental performance.

A general point brought out by Spath & Mann (2004) is that the life cycle impacts of the CCS system depend on where the energy to run the capture plant comes from. In a new build plant it is likely that taking energy from the power plant itself would be the most efficient solution. In a retrofit, the energy could be drawn from another source such as wind- or biomass-generated electricity which might reduce the LC GWP further.

Capacity limitations on CCS

Chemicals are used in CCS that would not be used in the four power generation cycles without CCS. These are:

- Solvents for CO_2 capture

    o Amine-based compounds used in low pressure absorption processes

    o Methanol- and glycol-based physical solvents used in higher pressure absorption processes

- Catalysts used in the CO_2 shift reaction in IGCC power generation cycles

- NaOH used to recover amines from the capture process waste

- corrosion inhibitors, antifoam agents, oxygen scavengers and salt neutralisers used in the amine solution in low pressure CO_2 absorption capture processes

Small amounts of solvents can be lost during the capture process, and in the capture process some of the amine-based solvent may react to form solid compounds that are collected and disposed of by incineration or landfill. Further analysis is required to determine whether the widespread use of these solvents could pose environmental issues. Spent catalysts can, at least partially, be regenerated and re-used.

Greater amounts of fuel, flue gas cleanup reagents and catalysts will also be used as a result of the reduction in efficiency of electricity generation when CCS is incorporated into power generation cycles. However, these are unlikely to pose any environmental problems *per se*. Therefore the main capacity limitations are likely to be fuel supply or storage capacity.

CO_2 storage capacity limitations in the UK

In the UK, current studies suggest that CO_2 storage capacity in depleted and partially depleted natural gas and oil fields may be approximately as shown in Table 3 (DTI 2006, Holloway et al. 2006).

Table 3. CO_2 storage capacity in oil and gas fields on the UK Continental Shelf.

Category	Location	Estimated CO_2 storage capacity (million tonnes)
Oil fields	Offshore	1175
Gas fields	Offshore	5140
Gas/condensate fields	Offshore	1200
TOTAL		7515

However, the figures for gas and condensate fields do not include any economic limitations, e.g. small and/or isolated fields may not be economic for CO_2 storage.

Attempts have been made to estimate the theoretical CO_2 storage capacity of various saline water-bearing reservoir formations within the UKCS (DTI 2006, Holloway et al. 2006). These indicate possibly several Gt CO_2 storage capacity in the Southern North Sea Basin and some more limited potential (up to 0.63 Gt) in the East Irish Sea Basin. However, none of these studies take into account all the major factors that should be considered in such an analysis and so the saline aquifer CO_2 storage capacity even of these regions needs to be revisited. In particular, the local and regional pore fluid pressure rise in a potential saline aquifer storage formation needs to be considered as well as the pore volume and the CO_2 saturation that is likely to be achieved in the closed structures. This work is in progress.

A 500 MWe supercritical PC power plant with CCS is calculated to emit about 3.2 million tonnes CO_2 per year operating at a load factor of 75% (Odeh & Cockerill 2008, Table 6). Therefore at 90% capture it will capture about 2.89 Million tonnes CO_2 per year. The amount captured depends on the load factor as well as the plant and capture efficiency. If for simplicity's sake the crude assumption is made that a 2GWe supercritical PC plant with CCS would emit about 10 Mt CO_2 per year (assuming a load factor of about 65%), and such plant have a forty year operating lifetime, the storage capacity in oil and gas fields might be sufficient to store the lifetime emissions from about 18 such plant. Thereafter saline aquifer storage potential would be needed. This probably exists but needs to be quantified as a matter of urgency. For comparative purposes, the UK had 29.3 GW installed subcritical PC-fired capacity in 2008.

Discussion

There are advantages and disadvantages to the adoption of CCS power generation systems. The main advantages are:

- CCS is the only way of significantly reducing the emissions of CO_2 from fossil fuel-fired power generation plant. There is currently serious carbon lock-in to fossil fuel-fired plant because of rapid global expansion of capacity and the need for payback on this investment.

- It can provide baseload power with relatively few interruptions at significantly lower GWP than is available from fossil fuel-fired plant at present.

The disadvantages include:

- Increases in other air and water pollutants, and solid wastes

- Increased cost

- Increased fuel consumption

- Decreased efficiency of power production

- The possibility of leakage of CO_2 from storage

One as yet unanswered question is how well capture and transport systems work if the plant is load following rather than run as base load – which could be the case if there is a large nuclear build programme because nuclear needs to supply base load.

The CCS technology with the lowest Life Cycle GWP varies between studies (Table 6). Part of the background to the study is the likely increase in the use of coal as a fuel in developed and developing countries. Coal has roughly twice the specific carbon emissions of natural gas and unmitigated coal-fired power generation has the potential to increase greatly atmospheric carbon dioxide levels. Therefore it may be more important to develop CCS on coal-fired plant, and technology that can be retrofitted may be the most useful in the short to medium term because of carbon lock-in at the numerous coal-fired power plant recently built and planned in China, India and elsewhere worldwide.

Table 6. GWP (gCO_2-e kWh^{-1}) of five CCS technologies on a life cycle basis from selected studies. N.B. variations in GWP depend principally on study-specific assumptions and system boundaries.

Study	Supercritical PC	Oxyfuel	IGCC	NGCC
Odeh & Cockerill 2008	255		167	200

Viebahn et al. 2007	262	176	245	132
Mayer-Spohn et al. 2007			208	
Livengood et al. 1993			218	
Spath & Mann 2004	247*			245

* Subcritical PC

Conclusions

The results of the LCAs of CCS power generation systems indicate a substantial reduction in GWP compared with fossil fuel fired power plant without CCS. However, the possibility of long-term leakage of CO_2 streams and/or entrained or displaced substances from storage needs to be considered very carefully, particularly as the mass of CO_2 that would have to be stored in order to have an impact on global atmospheric emissions is very high. The risks of leakage should be assessed on a site-by-site basis and it is unlikely that a licence to store CO_2 would be granted by a Regulator unless the risk was considered to be acceptable (OSPAR 2007, London Convention 2007, EU 2008).

With regard to other impact categories, the LCAs indicate that there will be increases in photochemical oxidation potential, eutrophication potential, acidification potential and human toxicology potential for PC plants with CCS.

The main environmental issues other than GWP are centred on the use of large quantities of amines or other solvents in the CO_2 capture process, their potential loss to the atmosphere and the ultimate fate of their degradation products. Thitakamol et al. (2007) state that environmental impact of adding a post combustion absorption-based CO_2 capture unit to a power plant is not severe. Nevertheless, they recommend that an Environmental Management System should be put in place to control pollution, minimize waste production, ensure progress towards environmental goals and provide safety plans for normal plant operation and accidents. They also provide a table of recommendations for leakage reduction from this process.

There may also be issues around the disposal of spent catalysts, some of which may not be reclaimable. This requires further work.

The main capacity constraint for power generation systems with CCS is likely to be CO_2 storage capacity, especially in countries less well endowed with storage space than the UK. A particular capacity issue is that the storage capacity of

saline water-bearing reservoir rocks (saline aquifers) needs to be analyzed as a matter of urgency in many countries, including the UK.

Overall, the above suggests that sustainability issues for the CCS power generation chain are not insuperable. Therefore it is concluded that CCS could be a very useful bridging technology that could mitigate emissions from new and existing fossil fuel-fired power plant, particularly coal-fired plant, which could buy time to move to lower carbon energy systems.

Acknowledgements

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