

Review Article

Carbon capture, storage and utilisation technologies: A critical analysis and comparison of their life cycle environmental impacts



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ABSTRACT

This paper presents a first comprehensive comparison of environmental impacts of carbon capture and storage (CCS) and carbon capture and utilisation (CCU) technologies. Life cycle assessment studies found in the literature have been reviewed for these purposes. In total, 27 studies have been found of which 11 focus on CCS and 16 on CCU. The CCS studies suggest that the global warming potential (GWP) from power plants can be reduced by 63–82%, with the greatest reductions achieved by oxy-fuel combustion in pulverised coal and integrated gasification combined cycle (IGCC) plants and the lowest by post-combustion capture in combined cycle gas turbine (CCGT) plants. However, other environmental impacts such as acidification and human toxicity are higher with than without CCS. For CCU, the GWP varies widely depending on the utilisation option. Mineral carbonation can reduce the GWP by 4–48% compared to no CCU. Utilising CO₂ for production of chemicals, specifically, dimethylcarbonate (DMC) reduces the GWP by 4.3 times and ozone layer depletion by 13 times compared to the conventional DMC process. Enhanced oil recovery has the GWP 2.3 times lower compared to discharging CO₂ to the atmosphere but acidification is three times higher. Capturing CO₂ by microalgae to produce biodiesel has 2.5 times higher GWP than fossil diesel with other environmental impacts also significantly higher. On average, the GWP of CCS is significantly lower than of the CCU options. However, its other environmental impacts are higher compared to CCU except for DMC production which is the worst CCU option overall.

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1. Introduction

Global emissions of carbon dioxide (CO₂) from fossil fuels have been increasing by 2.7% annually over the past decade and are now 60% above 1990 levels, the reference year for the Kyoto Protocol [1]. By contrast, it is estimated that the CO₂ emissions should be reduced by at least 50% to limit the rise of the global average temperature to 2 °C by 2050 [2]. A range of different options that could help towards this target for mitigating climate change are considered worldwide, including carbon capture and storage (CCS) [3,4]. However, CCS faces a number of technical and economic barriers that must be overcome before it can be deployed on a large scale. One of the main economic obstacles is the fact that it is an unprofitable activity that requires large capital investment [5]. In the UK, for example, there are no incentives or subsidies for CCS which is going to make its development and deployment difficult. On the technical side, CO₂ leakage rates are uncertain and in some countries CCS is not a viable option as their geological storage capacity is limited or in some cases only available offshore, thus increasing transportation and injection costs [5,6]. This is the case with the UK, Norway, Singapore, Brazil and India [5,6].

More recently, a related alternative – carbon capture and utilisation (CCU) – has started to attract attention worldwide because it can turn waste CO₂ emissions into valuable products such as chemicals and fuels, while at the same time contributing to climate change mitigation. One of the advantages of CCU over CCS is that utilisation of CO₂ is normally a profitable activity as products can be sold [5]. Furthermore, compared to conventional petrochemicals feedstocks, CO₂ has the advantage of being a ‘renewable’ resource (as long it continues to be emitted by various industrial activities), low in cost and non-toxic [7]. Even though conversion of CO₂ to various products is energy intensive owing to its thermodynamic stability, the potential for providing a secure supply of chemicals and fuels, along with the escalating fossil-fuel prices, could become a powerful driver for CCU [5,7]. Nevertheless, the current global demand for chemicals does not have the capacity to sequester enough CO₂ emissions to contribute significantly to meeting the carbon reduction targets. For example, the annual production of urea and methanol, two of the most commercially important chemicals, would consume only 0.5% of the current 34.5 Gt/yr of the anthropogenic CO₂ emitted globally [8]. Furthermore, using CO₂ for fuel production only delays its emissions rather than removing it over long timescales needed for mitigating climate change. Similarly, the ‘storage’ in some chemicals is also short-lived, depending on their use.

In addition to the above-mentioned aspects, there are other sustainability issues that must be considered before large-scale deployment of either CCS or CCU, notably environmental impacts. This is important to ensure that climate change is not mitigated at the expense of other environmental issues. It is also important that the impacts be assessed on a life cycle basis, to avoid shifting the environmental burdens from one life cycle stage to another. In an attempt to inform the debate in this field, this paper provides a comprehensive state-of-the-art review of different CCS and CCU technologies, analysing their life cycle environmental impacts based on the results of life cycle assessment (LCA) studies found in the literature.

To set the context, the paper starts by an overview of current CCS and CCU technologies in the next section. This is followed in Section 3 by the review of LCA studies, first for different CCS and then for CCU options, discussing and comparing their life cycle environmental impacts. Finally, conclusions and recommendations for future work are given in Section 4.

2. Overview of CCS and CCU technologies

CCS and CCU aim to capture CO₂ emissions from point sources such as power plants and industrial processes, to prevent the release into the atmosphere [9]. The difference between CCS and CCU is in the final destination of the captured CO₂. In CCS, captured CO₂ is transferred to a suitable site for long-term storage [9–15], while in CCU, captured CO₂ is converted into commercial products [5,9]. Different CCS and CCU options are summarised in Fig. 1 and described below. Note that it is not the intention of this paper to provide an in-depth technical review of the CCS and CCU technologies but rather to provide the background and set the context for the main aim of the paper which is a critical review and analysis of the life cycle environmental impacts of these options. For detailed technical (and economic) reviews of CCS, the interested reader may wish to consult Kuramochi et al. [16], Rubin et al. [17], and Markewitz et al. [9]. The latter also provides a short review of CCU options.

2.1. CO₂ capture options

Power plants, oil refineries, biogas sweetening as well as production of ammonia, ethylene oxide, cement and iron and steel are the main industrial sources of CO₂ [5,9]. For example, over 40% of the worldwide CO₂ emissions are caused by electricity generation in fossil-fuel power plants [9]. Therefore, these sources are the main candidates for a potential application of CCS or CCU. As for the CO₂ capture, a one-size-fit-all technology would not be feasible owing to the diversity of the industrial processes generating CO₂ emissions. For that reason, there is a wide variety of CO₂ capturing systems, to ensure compatibility with the specific industry. However, the level of maturity among different capturing systems varies across industries. For example, power plants and oil refineries are getting closer to implementing CO₂ capturing systems at a large-scale, while the cement and the iron and steel industry will still have to overcome the transition from small-scale demonstration plants to industrial deployment [18].

The CO₂ capture options can be classified as post-conversion, pre-conversion and oxy-fuel combustion [18–20]. These are summarised in Fig. 2 and Table 1 and described in turn below. A further option involves biomass fixation of CO₂. Currently, microalgae are used for this purpose because of the drive for biofuels production. Therefore, arguably, this is a CCU rather than CCS option as microalgae would not be cultivated merely to capture CO₂. Thus, CO₂ fixation by microalgae and the related biofuel production are discussed in Section 2.3 which provides an overview of CCU options.

2.1.1. Post-conversion capture

Post-conversion capture involves separation of CO₂ from waste gas streams after the conversion of the carbon source to CO₂ – for example, via combustion of fossil fuels or digestion of wastewater sludge. It can be used to remove CO₂ from various industries, including power plants, production of ethylene oxide, cement, fuels, iron and steel as well as biogas sweetening [10,21]. When used in power plants, post-conversion capture is also known as post-combustion capture [19].

As indicated in Table 1, post-conversion capture methods include absorption in solvents, adsorption by solid sorbents, including porous organic frameworks, membranes and cryogenic separation as well as pressure and vacuum swing adsorption [9,16,22–24]. Among these, absorption by monoethanolamine (MEA) is most commonly used [16,25]. However, this method is not economically viable for all industries as MEA regeneration has high heat consumption. For example, MEA absorption of CO₂ in a cement plant is less well suited than in a combined heat and power

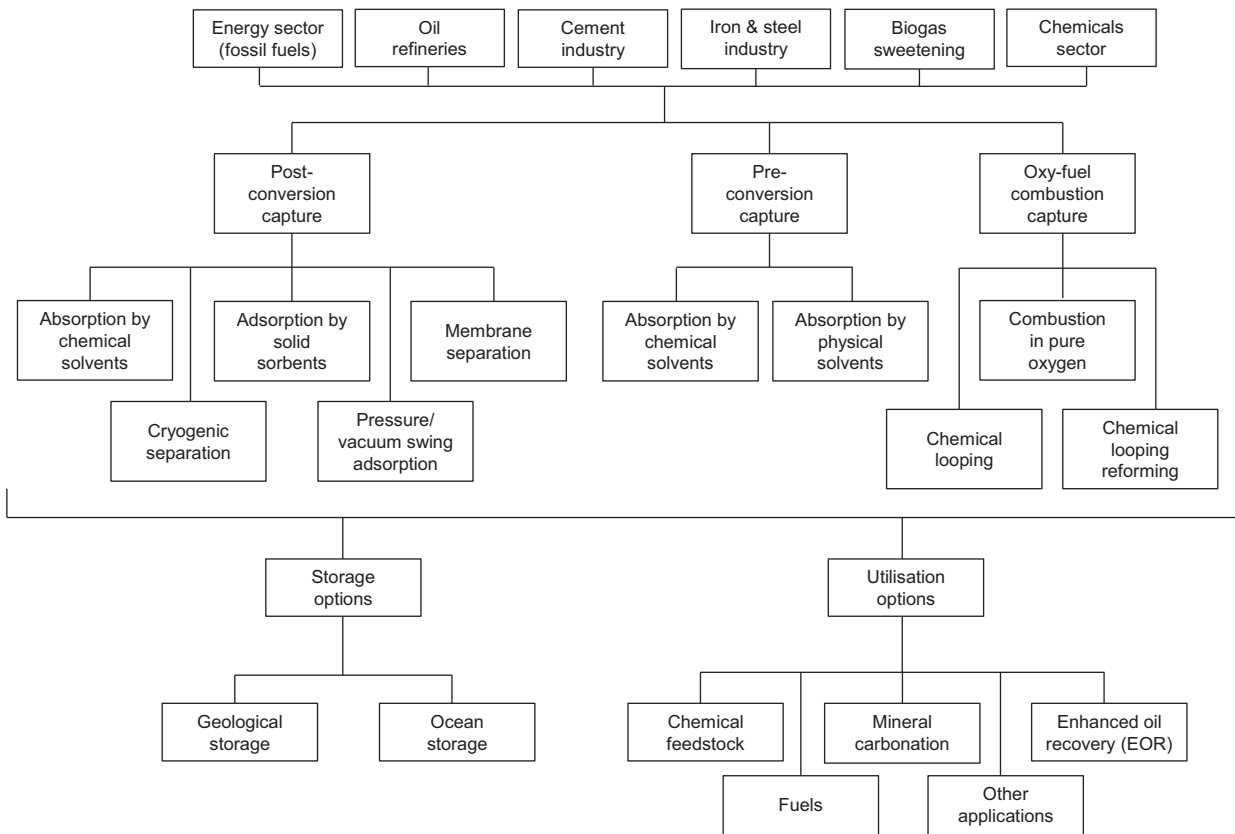


Fig. 1. Different carbon capture, storage and utilisation options.

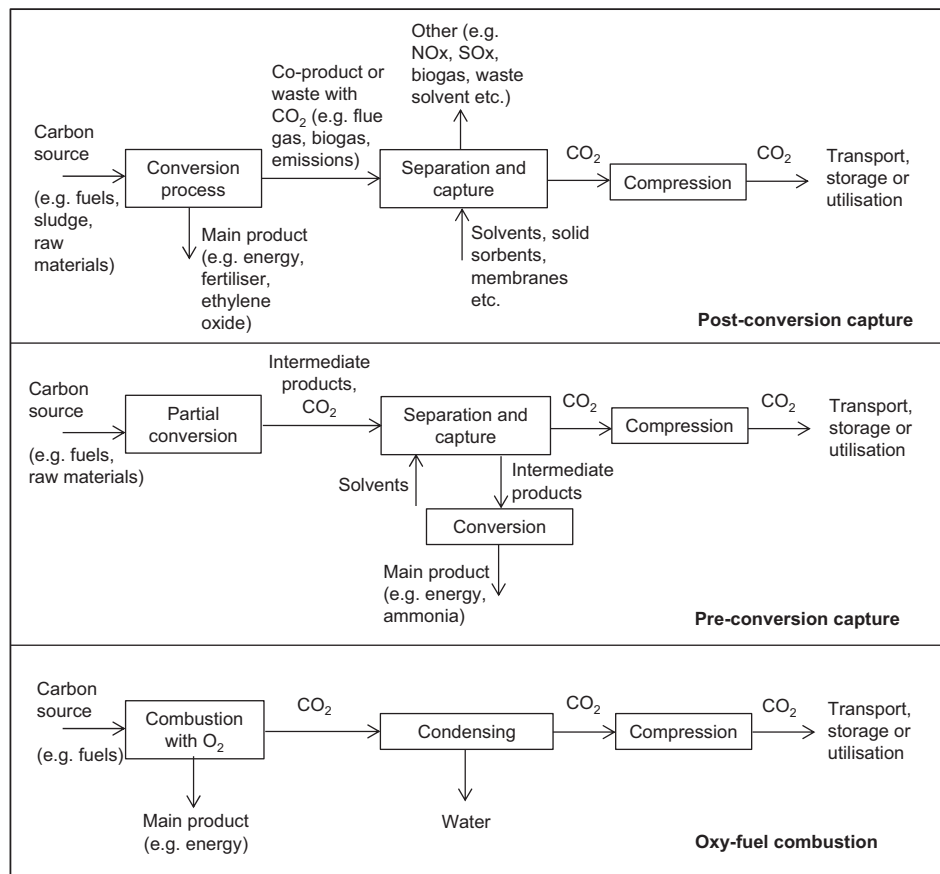


Fig. 2. Carbon capture options. Adapted from UNIDO [18], Singh et al. [19] and Zaimes and Khanna [20].

Table 1
Carbon capture options and applications [5,6,12,16,18,22–25,27,29,30,33,36–40].

Capture option	Separation technology	Method	Applications
Post-conversion	Absorption by chemical solvents ^a	<ul style="list-style-type: none"> • Amine-based solvent, e.g. monoethanolamine (MEA),^b diethanolamine (DEA), and hindered amine (KS-1) • Alkaline solvents, e.g. NaOH and Ca(OH)₂ • Ionic liquids 	Power plants; iron and steel industry; cement industry; oil refineries
	Adsorption by solid sorbents	<ul style="list-style-type: none"> • Amine-based solid sorbents • Alkali earth metal-based solid sorbents, e.g. CaCO₃ • Alkali metal carbonate solid sorbents, e.g. Na₂CO₃ and K₂CO₃ 	No application reported
	Membrane separation	<ul style="list-style-type: none"> • Porous organic frameworks – polymers • Polymeric membranes, e.g. polymeric gas permeation membranes^b • Inorganic membranes, e.g. zeolites • Hybrid membranes 	Power plants Power plants; natural gas sweetening
	Cryogenic separation Pressure/vacuum swing adsorption	<ul style="list-style-type: none"> • Cryogenic separation • Zeolites^b • Activated carbon^b 	Power plants Power plants; iron and steel industry
Pre-conversion	Absorption by physical solvents	<ul style="list-style-type: none"> • Selexol, rectisol 	Power plants (IGCC)
	Absorption by chemical solvents	<ul style="list-style-type: none"> • Amine-based solvent, e.g. monoethanolamine (MEA) 	Ammonia production
	Adsorption by porous organic frameworks	<ul style="list-style-type: none"> • Porous organic frameworks membranes 	Gas separations
Oxy-fuel combustion	Separation of oxygen from air	<ul style="list-style-type: none"> • Oxy-fuel process 	Power plants; iron and steel industry; cement industry ^c
		<ul style="list-style-type: none"> • Chemical looping combustion 	Power plants
		<ul style="list-style-type: none"> • Chemical looping reforming 	Power plants; syngas production and upgrading

^a Mature technology.

^b Commercially available.

^c May become available in the long term (>2030).

plant as the former lacks recoverable heat, incurring additional energy costs [16]. The energy penalty also applies to the other post-conversion technologies, either through the direct energy costs or through a reduced energy efficiency associated with their operation [9,16].

2.1.2. Pre-conversion capture

Pre-conversion capture refers to capturing CO₂ generated as an undesired co-product of an intermediate reaction of a conversion process [18]. Some examples include the production of ammonia and coal gasification in power plants [10,19,26]. In ammonia production, CO₂ that is co-produced with hydrogen during steam reforming must be removed before the ammonia synthesis can take place – absorption in MEA is commonly used for these purposes [10,27]. Similarly, in an integrated gasification combined cycle (IGCC) power plant, CO₂ must be separated from hydrogen. As indicated in Table 1, this is typically achieved using physical solvents such as selexol and rectisol [19,26,28,29]. Porous organic framework membranes can also be used for CO₂ capture owing to their high CO₂ selectivity and uptake; however, no applications have been reported to date [30]. Note that, when applied in power plants, pre-conversion capture is also referred to as pre-combustion capture [19].

Like post-conversion, pre-conversion capture also incurs energy penalties for regeneration of chemical solvents (e.g. MEA); these are lower for the physical solvents as they are regenerated by reducing pressure rather than by heat. Physical solvents are, therefore, more suitable for applications with high operating pressure; they are also more efficient for concentrated CO₂ streams [9].

2.1.3. Oxy-fuel combustion capture

As the name would suggest, oxy-fuel combustion can only be applied to processes involving combustion, such as power generation in fossil-fuelled plants, cement production and the iron and steel industry. Here, fuel is burned with pure oxygen to produce flue gas with high CO₂ concentrations and free from

nitrogen and its compounds such as NO and NO₂. While this avoids the need for chemicals or other means of CO₂ separation from the flue gas, a disadvantage is that oxygen is expensive and the environmental impacts, including CO₂ emissions, associated with its production are high because of the energy intensive air-separation processes [31].

As indicated in Table 1, the alternatives to the oxy-fuel process are chemical looping combustion (CLC) and chemical looping reforming (CLR). Both use a metal oxide to transfer oxygen selectively from an air reactor to a fuel combustor. In CLR, a sub-stoichiometric amount of oxygen is used, leading to the production of syngas, thus making it suitable for syngas generation or upgrading [32]. Some of the advantages of CLR include lower steam demand, higher fuel conversion efficiencies and better sulphur tolerance [32]; it can also handle dilute CO₂ streams [33]. However, a challenge is to operate the system under the high pressure needed to achieve efficiencies equivalent to that of the state-of-the-art oxy-fuel process or post-combustion capture. For CLC, one of the challenges is application to solid fuels and ash handling [32]. Neither of the oxy-fuel technologies is expected to be fully deployed before 2030 [18].

2.2. CO₂ storage options

Once captured, CO₂ is compressed and shipped or pipelined to be stored either in the ground, ocean or as a mineral carbonate [10,13,25]. The first option, known as geological storage, involves injecting CO₂ into geological formations such as depleted oil and gas reservoirs, deep saline aquifers and coal bed formations, at depths between 800 and 1000 m [10,13]. Depending on the characteristics of the site, CO₂ can be stored through different trap mechanisms, including impermeable layers known as “caprock” (e.g. mudstones, clays, and shales) which trap CO₂ underneath as well as in situ fluids and organic matter where CO₂ is dissolved or adsorbed [10]. Subject to the reservoir pressure and temperature, CO₂ can be stored as compressed gas, liquid, or in a supercritical condition [34]. The latter (@31.1 °C and 73.8 bar) makes it denser,

increasing the pore space utilisation and making it more difficult to leak [35].

CO₂ storage in geological formations is at present probably one of the most promising options owing to the previous experience by the oil and gas industry. For example, the industry has good understanding of the structural characteristics and behaviour of depleted oil and gas reservoirs and the existing well-drilling and injection techniques can be adapted for carbon storage applications [10]. Deep saline aquifer formations are also a possibility for storage with a large storage capacity estimated at 700–900 Gt CO₂ [10]. However, very little is known about coal bed formations and further explorations are required before they can be considered a safe storage option [10]. Ocean storage relies on the principle that the ocean bed has a huge capacity to store injected CO₂ at great depths. Yet, ocean storage has never been tested on the large scale even though it has been studied for over 25 years [10,25].

The main concerns with CO₂ storage are its possible leaks and the related damage that a concentrated CO₂ stream would cause if it escaped into the environment. The annual leakage rates reported in the literature range from 0.00001% to 1%, depending on the permeability of the geological structure and its faults or defects [10,19,26].

There are several ongoing CCS projects around the world, ranging from the pilot to commercial scale. The latter include the Sleipner and Snøhvit projects in Norway, the Weyburn-Midale in Canada, the In Salah in Algeria and the Salt Creek project in the USA [10,18]. These projects have been operating in saline aquifer formations (Norway) and depleted oils and gas reservoirs (Canada, Algeria and USA) for more than 10 years.

Finally, mineral carbonation involves reacting CO₂ with metal oxides such as magnesium and calcium oxides, to form carbonates. Carbonation, also known as 'mineral sequestration', can be considered as both storage and utilisation option. The latter applies if the intended application of the carbonates goes beyond storing CO₂ to be used as a material; for example, in the construction industry [41]. Similarly, two other options can also be considered as both storage and utilisation methods: enhanced oil recovery (EOR) and enhanced coal-bed methane recovery (ECBM). For the purposes of this paper, all three of these practices are considered to be utilisation of CO₂ and are discussed further in the next section.

2.3. CO₂ utilisation options

As mentioned earlier, as an alternative to storage, captured CO₂ can be used as a commercial product, either directly or after conversion. Examples of direct utilisation include its use in the food and drink industry and for EOR; CO₂ can also be converted into chemicals or fuels. These and other applications shown in Fig. 1 are described next.

2.3.1. Direct utilisation of CO₂

Several industries utilise CO₂ directly. For example, in the food and drink industry, CO₂ is commonly used as a carbonating agent, preservative, packaging gas and as a solvent for the extraction of flavours and in the decaffeination process [42]. Other applications can be found in the pharmaceutical industry where CO₂ can be used as a respiratory stimulant or as an intermediate in the synthesis of drugs [7,42]. However, these applications are restricted to sources producing CO₂ waste streams of high purity such as ammonia production [7,9,10].

2.3.2. Enhanced oil and coal-bed methane recovery

EOR and ECBM are other examples of direct utilisation of CO₂ where it is used to extract crude oil from an oil field or natural gas from unmineable coal deposits, respectively. While the latter is not

commercially available yet [10], the former has been widely practiced for over 40 years in several oil-producing countries, including Norway, Canada and the USA [10,43].

Also known as tertiary recovery, EOR is used to extract otherwise unrecoverable oil reserves. It involves injection of different agents into the reservoir, including CO₂, natural gas nitrogen, polymers (e.g. polyacrylamides) and surfactants, to remove the oil trapped in the rocks [44]. EOR can extract 30–60% more of the crude originally available in the well, compared to primary and secondary extraction which recover 20–40%. Among the different agents, naturally occurring CO₂ is used most commonly because of its low cost and wide availability [43]. Injected under supercritical conditions, it mixes well with the oil to decrease its viscosity, thus helping to increase the extraction yields [45]. However, most CO₂ returns back to the surface with the pumped oil – although it recycled for economic reasons, some of gas is emitted into the atmosphere.

With the advent of climate change, the possibility of utilising CO₂ from anthropogenic sources in EOR has been considered in recent years [2]. Under special conditions, the injected CO₂ could remain stored underground, similar to geological storage. Nevertheless, the switch from using naturally to anthropogenic sources of CO₂ will depend mostly on the capture costs and incentives for the oil and gas industry.

2.3.3. Conversion of CO₂ into chemicals and fuels

CO₂ can also be utilised by processing and converting it into chemicals and fuels. This can be achieved through carboxylation reactions where the CO₂ molecule is used as a precursor for organic compounds such as carbonates, acrylates and polymers, or reduction reactions where the C=O bonds are broken to produce chemicals such as methane, methanol, syngas, urea and formic acid [5,7,9,41]. Furthermore, CO₂ can be used as a feedstock to produce fuels, for example, in the Fischer–Tropsch process [46].

However, although CO₂ can replace petrochemical feedstocks for production of chemicals and fuels [5], a disadvantage is that its conversion is energy intensive and it requires high-selectivity catalysts since CO₂ is thermodynamically highly stable. Furthermore, chemicals and fuels offer limited storage periods for captured CO₂ because of their short life span (typically less than six months). Consequently, CO₂ is released into the atmosphere before the benefits of the capture can be realised. For that reason, future research efforts should focus on the synthesis of materials and products with longer lifespans. An example is mineral carbonates that can be used in construction [10], as discussed in the next section.

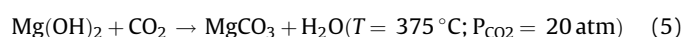
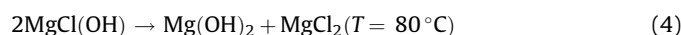
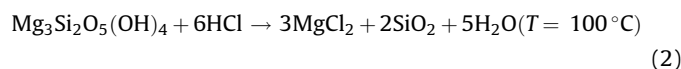
2.3.4. Mineral carbonation

As mentioned earlier, mineral carbonation is a chemical process in which CO₂ reacts with a metal oxide such as magnesium or calcium to form carbonates [10,25]. Magnesium and calcium are normally found in nature in the form of silicate minerals such as serpentine, olivine and wollastonite [10,47]. Large deposits of these minerals exist in Finland, Australia, Portugal and the USA [5].

Mineral carbonation encompasses a series of reactions that can take place in a single or a multi-step process, also known as direct and indirect carbonation, respectively [10,47]. In a single-step process, the extraction of the metal from the mineral matrix and the carbonate precipitation occur simultaneously in the same reactor [10]. Direct carbonation takes place under high pressure conditions in either dry or aqueous media [47]. As an example, the overall carbonation reaction using serpentine is illustrated below [47].



Multi-step or indirect carbonation consists of three main reactions [6,10,47]. The first reaction involves separating the metal from the mineral matrix in the presence of an extracting agent such as hydrochloric acid or molten salts. This is followed by a series of hydration reactions to obtain the metal in the hydroxide form. Finally, the carbonation reaction takes place, where captured CO₂ reacts with the metal in the hydroxide state to form a carbonate. In theory, the carbonation reaction is an exothermic reaction releasing enough heat to make the whole carbonation process self-sufficient [6]. These reactions are illustrated below, using serpentine and hydrochloric acid as an example [47]:



The use of pure CO₂ is not essential for mineral carbonation as the presence of impurities such as NO_x in flue gas will not interfere with the carbonation reaction [10]. Therefore, the separation and capture step that produces a pure stream of CO₂ can be omitted as waste emissions containing CO₂ can be used directly.

The main advantage of mineral carbonation is the formation of stable carbonates capable of storing CO₂ for long periods (decades to centuries) [5], without the risk of CO₂ leakage as in CCS [10,25]. However, this technology is not fully developed for large-scale applications as the energy penalty and costs are still too high [10]. Furthermore, the mining, transportation and preparation of the minerals also have high energy requirements, thus reducing the overall CO₂ removal efficiency [5,6,10,25,38].

2.3.5. Biofuels from microalgae

As mentioned in Section 2.1, CO₂ can be used to cultivate microalgae used for the production of biofuels [5,48,49]. Microalgae have the ability to fix CO₂ directly from waste streams such as flue gas as well as using nitrogen from the gas as a nutrient [5,50]. Cultivation of microalgae can be carried out in open raceway ponds and photo-bioreactors (flat-plate, annular or tubular) [51]. The former require a large land area and process control is difficult, limiting productivity [5]. Photo-bioreactors are better in that respect but are more expensive than open-pond systems. Recently developed vertical flat-panel reactors made from thin polyethylene film have much lower capital costs and energy requirements and it is expected that future systems will be based on these designs [52,53].

As shown in Fig. 3, before microalgae can be converted into fuels, the biomass content has to be harvested and dried [5]. The conversion into fuels can be carried out through thermochemical or biochemical conversion. The former uses heat to produce first syngas and then fuels as well heat and electricity. Some examples of thermochemical conversion processes include gasification, liquefaction and pyrolysis. Biochemical conversion relies on biological and chemical processes, such as anaerobic digestion, fermentation and esterification [48,51].

Unlike crops used in the production of first-generation biofuels (e.g. maize, sugar beet, oilseed rape, etc.), microalgae cultivation does not represent a threat to food markets [5,48]; however, it still requires large land areas, particularly if cultivated in open ponds so that it could ultimately compete for land for food production. Furthermore, large-scale production of biofuels from microalgae is currently not available because of the high production costs, mainly owing to the high energy requirements in the harvesting stage [5,48,49].

3. Life cycle environmental impacts of CCS and CCU

This section reviews and analyses LCA studies for different CCS and CCU options. Overall, 27 studies were found in the literature, of which 11 focused on CCS and 16 on CCU. They are discussed in turn in Sections 3.1 and 3.2, respectively. This is followed by a comparison of environmental impacts of CCS and CCU in Section 3.3. As far as we are aware, this is the first time such a comparison has been carried out.

3.1. Environmental impacts of CCS

Over the past decade, several studies have evaluated the life cycle environmental impacts of CCS technologies for power plants, considering pulverised coal (PC), integrated coal gasification combined cycle (IGCC) and combined cycle gas turbine (CCGT) plants [19,26,28,29,37,40,61–65]. These are listed in Table 2, summarising the assumptions and impacts considered. As can be seen from the table, the goal and scope varied in different studies. For example, Viebahn et al. [29], Pehnt and Henkel [26], Nie et al. [64], and Singh et al. [65] conducted comparative LCA studies of different CCS technologies applied to fossil-fuel based power plants. Viebahn et al. [29] also compared the environmental performance of CCS against those from renewable energy technologies such as wind and solar thermal. The rest of the studies assessed the environmental impacts of fossil-fuel based power plants with and without CCS technologies [26,28,37,40,61–63,65].

The system boundaries, shown in Fig. 4, were consistent across all the studies and considered the extraction and supply of fossil fuels to the power plant, power generation, CO₂ capture, compression, transport, injection and storage. The infrastructure was included in most studies and the functional unit was related to

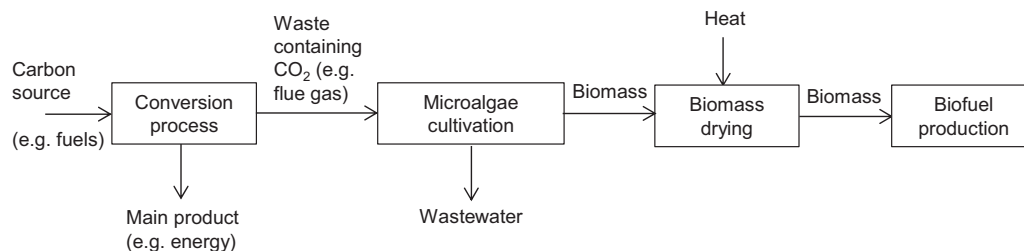


Fig. 3. Utilisation of CO₂ to produce biofuels from microalgae.

Adapted from Passell et al. [54], Lardon et al. [55], Soratana et al. [56], Shirvani et al. [57], Zaimes and Khanna [58], Borkowski et al. [59] and Campbell et al. [60].

Table 2
Summary of LCA studies for CCS technologies [19,26,28,29,37,40,61–65].

Study	Scope	Carbon capture method	Storage option	Functional unit	LCA impacts ^a
Khoo and Tan [37]	Comparative LCA of different CCS technologies applied to a coal-fired power plant in the US, from cradle to grave	Post-combustion via chemical absorption, membrane and cryogenic separation, and pressure swing adsorption	Ocean and geological storage	1 MWh of electricity	GWP, AP
Viebahn et al. [29]	Comparative LCA of CCS and renewable energy technologies applied to PC, CCGT and IGCC power plants in Germany, from cradle to grave	Post-combustion via MEA Pre-combustion via rectisol and oxy-fuel combustion	Geological storage in a depleted gas field	1 kWh of electricity	GWP, AP, POCP, EP
Koornneef et al. [61]	LCA of three different PC power plants in The Netherlands with and without CCS, from cradle to grave	Post-combustion via MEA	Geological storage	1 kWh of electricity	GWP, AP, ADP, ODP, HTP, FAETP, MAETP, TETP, POCP, EP
Odeh and Cockerill [28]	LCA of a PC, CCGT and IGCC power plants in the UK with and without CCS from cradle to grave	Post-combustion via MEA and pre-combustion via selexol	Geological storage in a depleted gas field	1 kWh of electricity	GWP
Pehnt and Henkel [26]	Comparative LCA of different CCS technologies applied to lignite PC and IGCC power plants in Germany, from cradle to grave	Post-combustion via MEA and pre-combustion via selexol Oxy-fuel combustion	Geological storage in a depleted gas field	1 kWh of electricity	GWP, AP, POCP, EP
Korre et al. [40]	LCA of several PC power plants with and without CCS, from cradle to grave	Post-combustion via MEA, PZ, KS-1 ^b	Not specified	1 MWh of electricity	GWP, AP, ADP, HTP, FAETP, POCP, EP
Modahl et al. [62]	LCA of four CCGT power plants in Norway with and without CCS, from cradle to grave	Post-combustion via MEA	Ocean storage below the sea bed	1 TWh of electricity	GWP, EP
Schreiber et al. [63]	LCA of a coal-based power plant in Germany with and without CCS, from cradle to grave	Post-combustion via MEA	Not specified	1 kWh of electricity	GWP, AP, HTP, POCP, EP
Nie et al. [64]	Comparative LCA of different types of CCS in a PC power plant, from cradle to grave	Post-combustion via MEA and oxy-fuel combustion	Geological storage in a saline aquifer	1 MWh of electricity	GWP, AP, ADP, ODP, HTP, MAETP, TETP, POCP, EP
Singh et al. [65]	LCA of a CCGT power plant with and without CCS, from cradle to grave	Post-combustion via MEA	Geological storage in a saline aquifer	1 kWh of electricity	GWP, AP, HTP, FAETP, MAETP, TEPT
Singh et al. [19]	Comparative LCA of different CCS technologies in CCGT and IGCC power plants, from cradle to grave	Post-combustion via MEA, pre-combustion via selexol and oxy-fuel combustion	Ocean storage below the sea bed	1 kWh of electricity	GWP, AP, HTP, FAETP, MAETP, TEPT

^a ADP, abiotic depletion potential; AP, acidification potential; EP, eutrophication potential; FAETP, fresh water aquatic ecotoxicity potential; GWP, global warming potential; HTP, human toxicity potential; MAETP, marine aquatic ecotoxicity potential; ODP, ozone depletion potential; POCP, photochemical ozone creation potential; TETP, terrestrial ecotoxicity potential.

^b PZ, piperazine and potassium carbonate; KS-1, hindered amine.

a unit of electricity generated, expressed either in kWh, MWh or TWh (see Table 2).

Post-conversion capture via chemical absorption using MEA was the most studied method [19,26,28,29,37,40,61–65]. Other

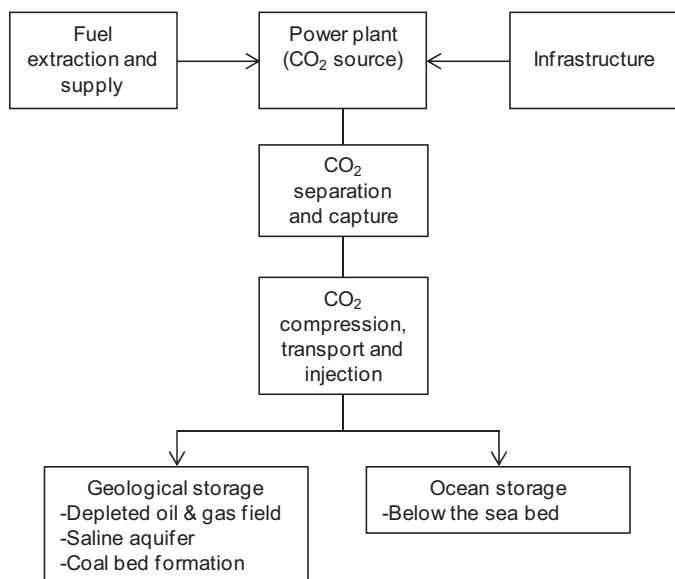


Fig. 4. System boundaries in different LCA studies of CCS technologies [19,26,28,29,37,40,61–65].

solvents included piperazine-promoted (PZ) potassium carbonate and hindered amine (K1-S) [40]. Post-conversion capture via cryogenics and membrane separation as well as pressure swing adsorption (PSA) were considered by Khoo and Tan [37]. The only pre-conversion capture methods studied were physical absorption using selexol and rectisol [19,26,28,29]. The capture rates of CO₂ emissions ranged from 87% to 100%. Most studies considered geological and only two ocean storage [37,62].

The environmental impacts reported in these studies are shown in Figs. 5–15. As can be seen from the figures as well as from Table 2, all the studies estimated the global warming potential (GWP) while the inclusion of other LCA impacts varied widely, from only one impact in addition to the GWP [37], to up to 11 impacts normally considered in LCA [41].

The GWP for the PC, CCGT and IGCC plants are compared in Fig. 5 showing the range of the values reported in different studies; Figs. 6–8 detail the GWP results obtained in each study for the three types of plant, respectively. As can be seen from Fig. 5, the average GWP for pulverised coal (PC) power plants without CCS is 876 kg CO₂ eq./MWh while for the post-conversion capture via MEA the average value is 203 kg CO₂ eq. and for oxy-fuel combustion it is 154 kg CO₂ eq. The equivalent average values for CCS at CCGT power plants are estimated at 120 kg CO₂ eq./MWh for oxy-combustion and 173 kg for post-combustion, compared to 471 kg CO₂ eq./MWh without CCS. The GWP for the pre-combustion capture and oxy-fuel combustion in IGCC plants is similar at 190 and 200 kg CO₂ eq./MWh, respectively, while the average without CCS is 1009 kg CO₂ eq./MWh. Therefore, these results indicate that the greatest GWP reductions (up to 82%) can

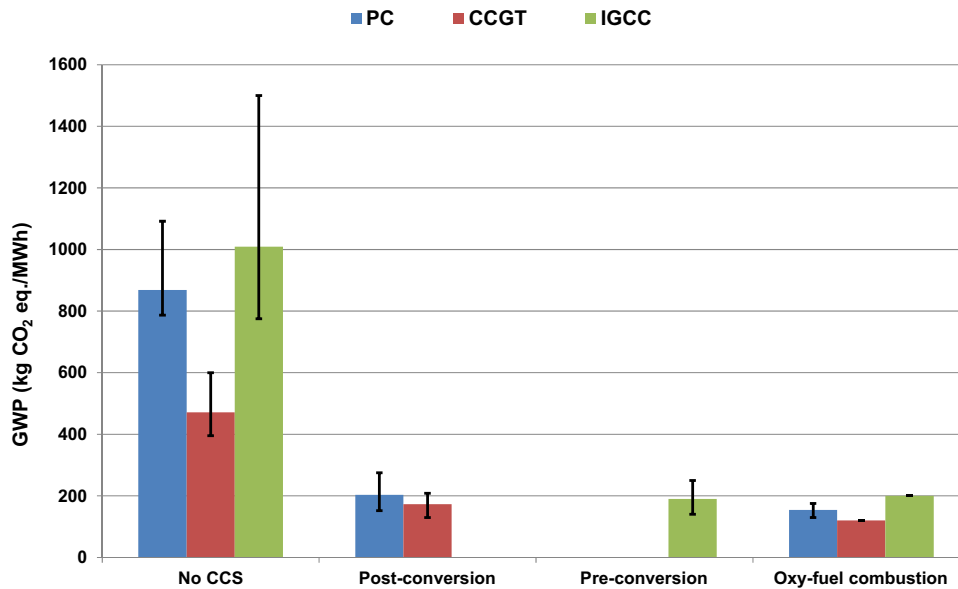


Fig. 5. Global warming potential of CCS options for PC, CCGT and IGCC plants. PC, pulverised coal; CCGT, combined cycle gas turbine; IGCC, integrated coal gasification combined cycle.

be achieved by oxy-fuel combustion in PC and IGCC plants and the lowest by post-combustion capture in CCGT plants (63%).

As can be observed from Fig. 9, fuel supply and CO₂ emissions from power plants are the main contributors to the GWP in the life cycle of CCS, contributing on average 53% and 28%, respectively [28,29,61,62,64]. The GWP from CO₂ capture is only significant for CCGT power plants, adding between 5% and 31%.

The other LCA impacts are shown in Figs. 10–13. As indicated in Fig. 10, the impacts for PC power plants with post-conversion capture vary across the studies, with some reporting higher impacts for the plants with than without CCS. All studies report 2–53% higher abiotic depletion potential (ADP), 17–35% higher ozone depletion potential (ODP), 1–173% higher eutrophication potential (EP) and 9–135% higher freshwater aquatic ecotoxicity potential (FAETP) with than without CCS. ADP and ODP are higher because additional coal is needed to compensate for the loss of energy

efficiency from the use of CCS. Ammonia emissions released during the absorption of CO₂ in MEA increase the EP, and the removal of trace metals by MEA contributes to higher FAETP as the impact from metals is transferred from air (flue gas) and soil (fly ash) to water (liquid effluent potentially reaching freshwater). This is perhaps one of the reasons why all studies find the terrestrial ecotoxicity potential (TETP) to be 36% lower with than without CCS. An increased removal by MEA of HF emissions from flue gases is a further reason for reduced TETP but also for the marine aquatic ecotoxicity potential (MAETP) which is found in all studies to be 89–93% lower for the power plants with CCS. For other impacts, the results vary across the studies. For example, Nie et al. [64], Viebahn et al. [29], Korre et al. [40], and Schreiber et al. [63] report 9–91% higher acidification potential (AP) with than without CCS while Koorneef et al. [61] and Pehnt and Henkel [26] find that the AP is lower (21–24%) with than without CCS. The higher AP is attributed

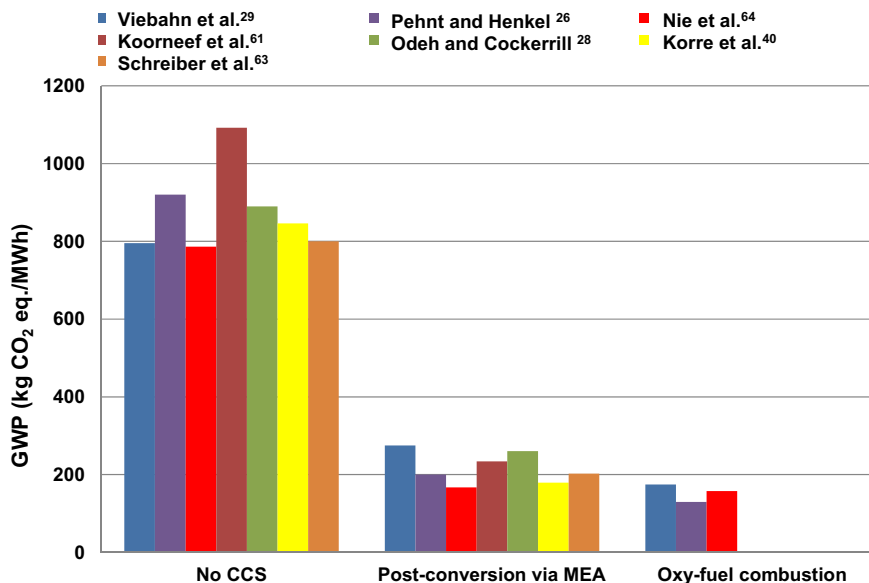


Fig. 6. Global warming potential of pulverised coal power plants with and without CCS [26,28,29,40,61,63,64]. The results reported in Pehnt and Henkel [26], Odeh and Cockerrill [28], Korre et al. [40], Koorneef et al. [61] and Schreiber et al. [63] include energy consumption for the regeneration of MEA.

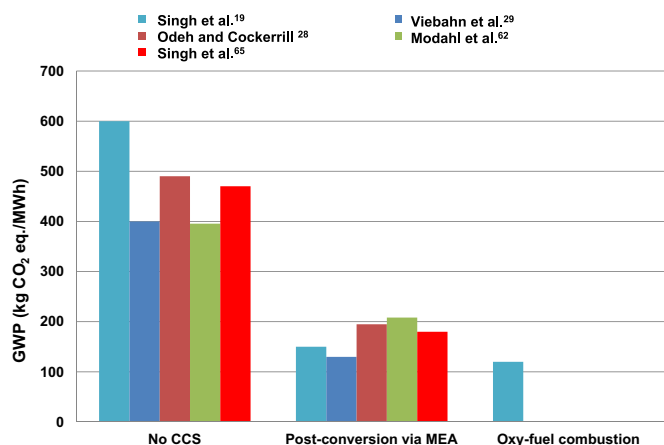


Fig. 7. Global warming potential of combined cycle gas turbine power plants with and without CCS [19,28,29,62,65]. The results reported in Singh et al. [19], Odeh and Cockerrill [28], Modahl et al. [62] and Singh et al. [65] include energy consumption for the regeneration of MEA.

to the additional coal needed, ammonia emissions from the CO₂ capture in MEA as well as NO_x and SO_x emissions from additional coal shipping [29,40,63,64]. However, it has also been reported that higher removal efficiencies of SO₂ and NO_x emissions can lead to lower AP from power plants with CCS [26,61]. Similarly, Nie et al. [64], Koornneef et al. [61], and Schreiber et al. [63] find a 55–183% higher human toxicity potential (HTP) with than without CCS owing to MEA production. However, Korre et al. [40] report a 29% lower HTP because MEA reduces the amount of fly ash and trace metals. Both higher (9–150%) and lower (28–270%) photochemical oxidant creation potential (POCP) for plants with CCS have been reported. This is because volatile MEA emissions to the atmosphere can contribute to higher POCP [26,29,40,63], but also the removal of NO_x and SO₂ emissions by MEA can reduce this impact [61,64].

As indicated in Fig. 11, the impacts also vary across the studies for oxy-fuel combustion plants, although not as much as for the PC plants. For example, Nie et al. [64] report that ADP, ODP, HTP, FAETP and MAETP are all higher for the installations with CCS, ranging from a 26% for the former to 62% for the latter. ADP and ODP are increased because of the additional coal required for power plants with CCS. The conversion of HF air emissions into an effluent discharge to freshwater has a negative effect on the HTP, FAETP and MAETP. Nevertheless, the removal of HF emissions along with other acid gases such as NO_x, SO_x, and HCl, leads to a lower AP, EP and POCP. Moreover, the removal of trace metals

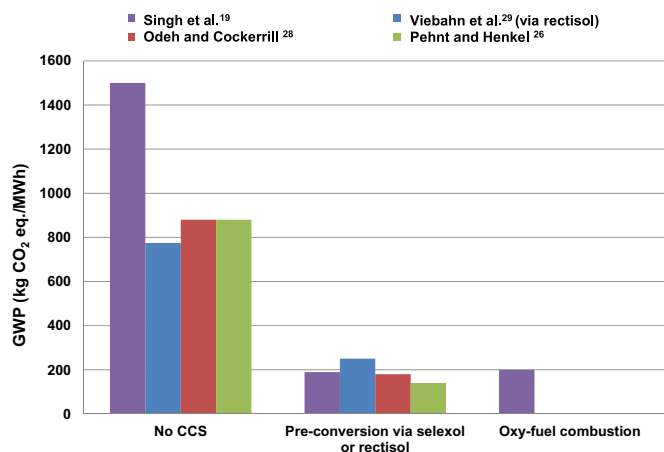


Fig. 8. Global warming potential of integrated coal gasification combined cycle power plants with and without CCS [19,26,28,29].

reduces TETP. Therefore, all these impacts are lower compared to the plants without CCS, from 20% for TETP to 53–120% for POCP.

The impacts are also higher with than without CCS for CCGT and IGCC power plants (Figs. 12 and 13) for both the post- and pre-conversion capture. For example, Singh et al. [65] report that the AP of a CCGT plant is 1.4 times higher with than without CCS with the HTP, MAETP, and TETP being 2.4 times higher. Similarly, Modahl et al. [62] estimate that the EP is also 2.4 times higher with than without CCS. According to Singh et al. [65], FAETP is 28 times higher for a CCGT power plant with than without CCS. All these impacts are increased mainly because of the production and use of MEA, including the emissions of MEA and ammonia to the atmosphere, surface and ground water. For the IGCC power plant, Peht and Henkel find a 17% higher AP, 20% higher EP and 50% higher POCP with than without CCS because of an increase in various emissions to air and water in the CCS supply chain [26].

One study also compared the environmental impacts of post-conversion capture via chemical absorption using three different solvents: MEA, piperazine with potassium carbonate (PZ) and hindered amine (KS-1) [40]. The results shown in Fig. 14 indicate that KS-1 leads to the lowest and MEA highest environmental impacts. This is largely due to the energy used for MEA regeneration [40]. Khoo and Tan also considered MEA and compared it to cryogenic and membrane separations as well as pressure swing adsorption for two impacts only: GWP and AP (see Fig. 15) [37]. Their results suggest that while the absorption in MEA has the lowest GWP, it has the second largest AP (after cryogenic separation). This is due to the release of acid gasses during the CO₂ capture process [37]. Membrane separation has the lowest AP but the highest GWP, which is twice as high as that with MEA. The main reason for this is that membrane separation does not generate air emissions, which is the case in post-conversion capture via chemical absorption. However, membrane separation uses more energy than the MEA process [37]. The GWP for the other two options is almost as high as for the membrane separation, also owing to high energy requirements [37].

3.2. Environmental impacts of CCU

Sixteen LCA studies have been conducted for various CCU options to date globally [6,12,38,51,54–60,66–70]. Similar to the CCS studies, most have considered fossil-fuel power plants as a source of CO₂ with only three studying the use of CO₂ from chemical plants such as ammonia and hydrogen production [51,60,66]. As summarised in Table 3, the CO₂ utilisation options evaluated in these studies are enhanced oil recovery and production of mineral carbonates, chemicals and biodiesel from microalgae. Five out of the six studies presented in Table 3 considered post-combustion capture with absorption in amine-based solvents [6,12,38,66,68], and the remaining one focused on pre-combustion capture using selexol [67]. All studies on microalgae considered direct injection of flue gases from power plants [51,54–60,69]. In addition, five studies included injection of pure CO₂ captured in MEA either in ammonia or power plants [51,58–60,69].

As shown in Fig. 16, the studies considered similar system boundaries comprising CO₂ separation and capture from the source, its compression, transport and utilisation options such as chemical synthesis, carbon mineralisation, EOR or biodiesel production. Specifically, Hertwich et al. [12] and Jaramillo et al. [67] studied the ‘cradle-to-grave’ impacts of separating and capturing CO₂ from flue gases in a CCGT plant in Norway and an IGCC plant in the US, respectively, with the aim of utilising it for EOR. However, unlike the former study, Jaramillo et al. [67] also included the impacts from refining the extracted oil and the combustion of the refined petroleum products. Khoo et al. [6,68]

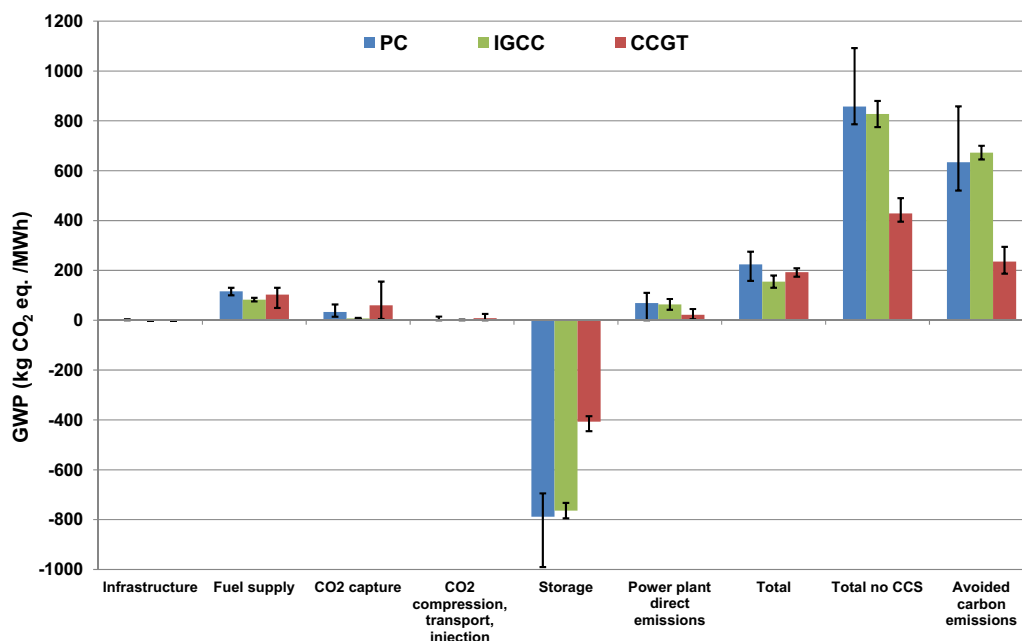


Fig. 9. Contribution to global warming potential of different life cycle stages for PC, IGCC and CCGT power plants with CCS [28,29,61,62,64]. PC, pulverised coal; CCGT, combined cycle gas turbine; IGCC, integrated coal gasification combined cycle.

and Nduagu et al. [38] assessed the environmental impacts of carbon capture and mineralisation of CO₂ from a CCGT in Singapore [6,68], and a coal power plant in Canada [38]. All three studies included the impacts of mining serpentine (Mg₃Si₂O₅(OH)₄), shipping and production of raw materials used in the mineralisation process. The resulting magnesium carbonate (MgCO₃) was assumed to be used for several potential applications, including in the construction industry and as a land reclamation material

(e.g. from landfills). However, the impacts associated with these applications were not considered. For the capture of CO₂ by microalgae and the subsequent production of biodiesel, most authors [54–58,60,69,70] assessed the whole life cycle from ‘cradle to grave’ with only two studies considering the ‘cradle-to-gate’ system boundary [51,59].

As can be seen from Table 3, the functional unit for the studies related to mineral carbonation and EOR varied but five out of six

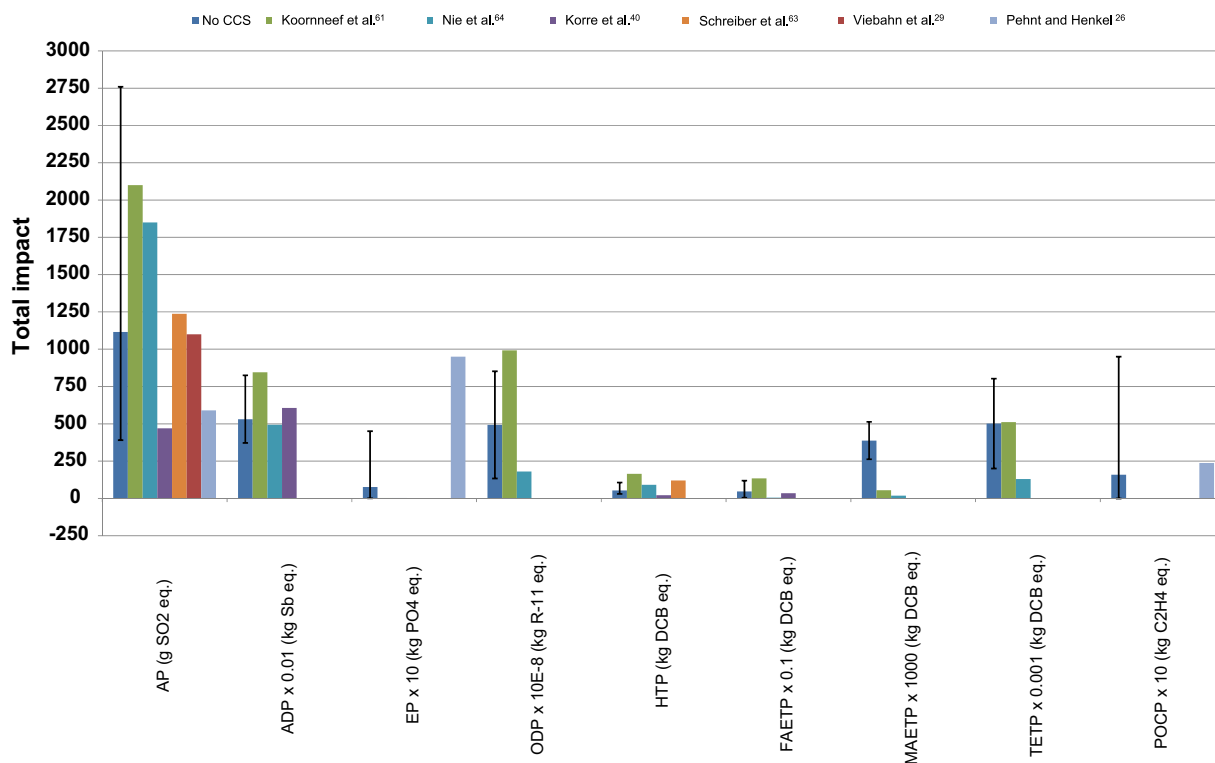


Fig. 10. Environmental impacts (other than global warming potential) of pulverised coal power plants with and without post-conversion CCS [26,29,40,61,63,64]. All impacts expressed per MWh. For impacts nomenclature, see Table 2. Some impacts have been scaled to fit. To obtain the original values, multiply with the factors shown against relevant impacts.

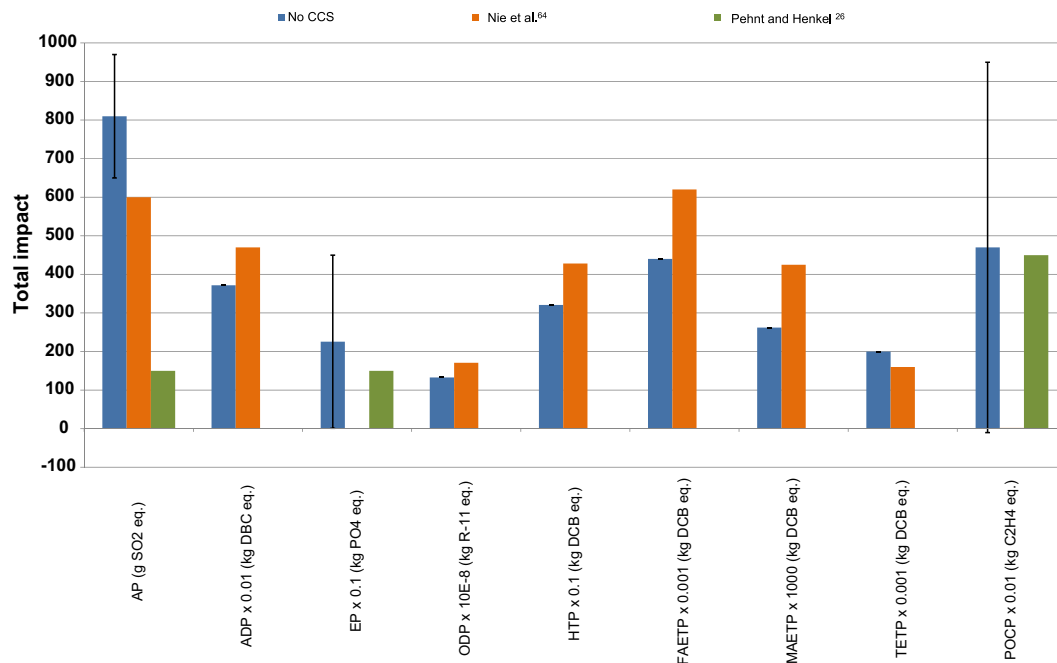


Fig. 11. Environmental impacts (other than global warming potential) of pulverised coal power plants with and without oxy-fuel combustion CCS [26,64]. All impacts expressed per MWh. For impacts nomenclature, see Table 2. Some impacts have been scaled to fit. To obtain the original values, multiply with the factors shown against relevant impacts.

based their analysis on the main product (electricity, chemicals or extracted oil) [6,12,66–68], with only one focusing on the amount of CO₂ removed [38]. On the other hand, most studies on microalgae adopted the functional unit related to the energy content of biodiesel. The only exception to this are two studies which based their analysis on distances travelled [60,69], and another which considered the impacts per tonne of biodiesel produced [70].

The results of the LCA studies for CCU are presented in Figs. 17–23. To be able to compare them, the original results from some studies have been recalculated for the functional unit of “1 tonne of CO₂ removed” [6,12,38,51,56,67], or “1 MJ of fuel produced” [51,56,70], as appropriate. These results are discussed below in turn, first for mineral carbonation, then for the production of chemicals (DMC) and EOR and finally for biodiesel production

from microalgae. This is followed in the next section by a comparison of the impacts between the CCS and CCU options discussed in this paper.

According to Khoo et al. [6], Nduagu et al. [38], and Khoo et al. [68] mineral carbonation to produce MgCO₃ can reduce the GWP from 4% to 48% compared to no CCU (Fig. 17). Although all three studies assumed a multi-step carbonation outlined in Section 2.3.4, they considered different routes, hence the wide-ranging reductions in the GWP: the latter two studies used ammonium sulphate to precipitate Mg(OH)₂ from the mineral matrix while Khoo et al. [6] assumed a hydration reaction. In all three cases this was followed by a carbonation reaction between Mg(OH)₂ and CO₂ to produce MgCO₃. In addition, Khoo et al. [68] considered two different mineral-to-CO₂-carbonation ratios (2.1:1 and 3.1:1), leading to different energy demands. Furthermore, Nduagu et al.

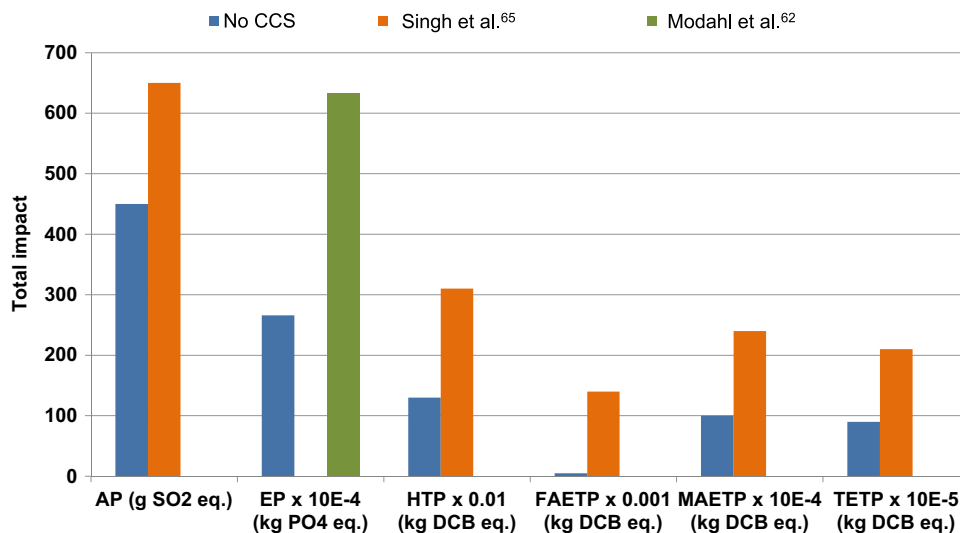


Fig. 12. Environmental impacts (other than global warming potential) of CCGT power plants with and without post-conversion CCS [62,65]. For impacts nomenclature, see Table 2. Some impacts have been scaled to fit. To obtain the original values, multiply with the factors shown against relevant impacts.

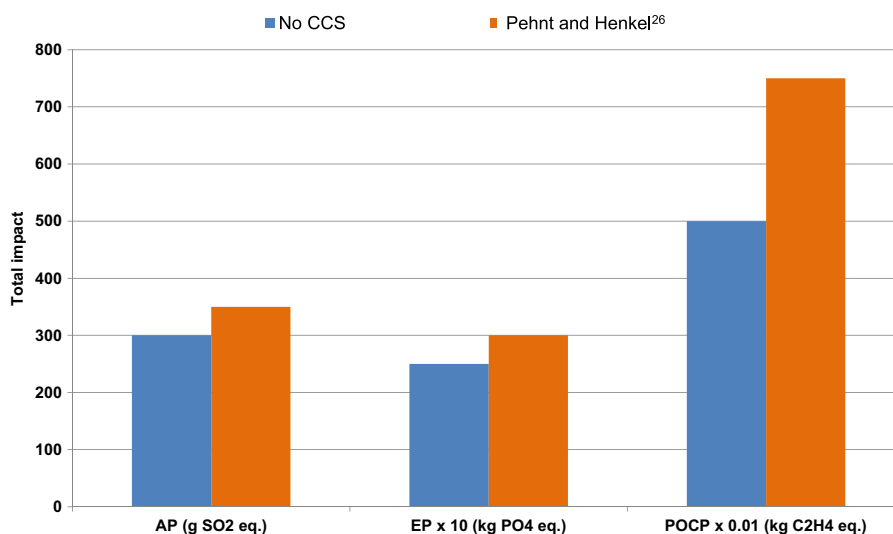


Fig. 13. Environmental impacts (other than global warming potential) of IGCC power plants with and without pre-conversion CCS [26]. For impacts nomenclature, see Table 2. Some impacts have been scaled to fit. To obtain the original values, multiply with the factors shown against relevant impacts.

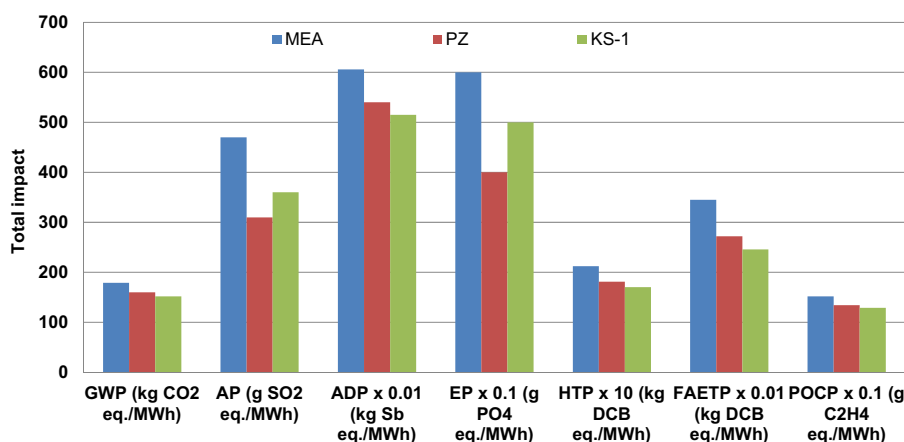


Fig. 14. Environmental impacts of a pulverised-coal power plant with post-conversion capture via MEA, piperazine with potassium carbonate (PZ) and hindered amine (KS-1) [40]. For impacts nomenclature, see Table 2. Some impacts have been scaled to fit. To obtain the original values, multiply with the factors shown against relevant impacts.

[38] and Khoo et al. [6] assumed that the heat produced in the carbonation reaction was recovered and used within the system, while Khoo et al. [68] reported that additional energy had to be supplied as the carbonation process did not produce enough heat

for the whole process. As a result, the estimated GWP ranged from 524 kg CO₂ eq. per tonne of CO₂ removed for the carbonation of CO₂ directly from a power plant to 1073 kg eq./t CO₂ removed when it was first absorbed in MEA and then recovered to be used in

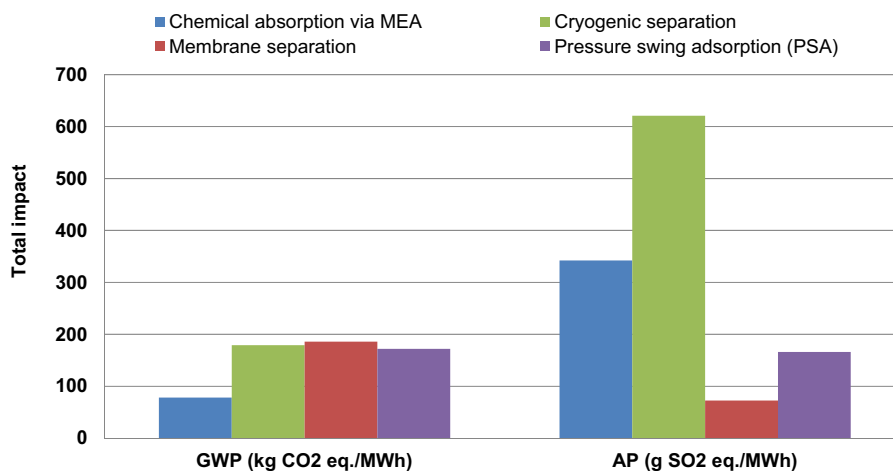


Fig. 15. Global warming potential (GWP) and acidification potentials (AP) of different post-conversion capture methods [37].

Table 3
Summary of LCA studies of CCU.

	Study	Scope	Carbon capture method	Utilisation option	Functional unit	LCA impacts ^a
Mineral carbonation	Khoo et al. [6]	LCA of a CCGT plant in Singapore with carbon capture and mineral carbonation (with and without heat recovery), considering mining and shipment of serpentine from two different locations in Australia	Post-combustion capture via MEA	Mineralisation of CO ₂ into carbonated products used in construction (e.g. as filler material for concrete)	Supply of 1 MWh of electricity from CCGT	GWP
	Khoo et al. [68]	LCA of a CCGT plant in Singapore with and without carbon capture and two mineral carbonation processes, considering mining and shipment of serpentine from Australia	Post-combustion capture via MEA and direct carbonation of CO ₂ from flue gas	Mineralisation of CO ₂ into MgCO ₃ with applications in construction and land reclamation	Production of 1 MWh of electricity from CCGT	GWP
	Nduagu et al. [38]	LCA of coal power plant in Canada including coal and serpentine mining and transport, carbon capture, transport and mineralisation	Post-combustion capture via MEA	Mineralisation of CO ₂ into MgCO ₃ with potential applications in construction and landfilling	Sequestration of 1 tonne of CO ₂ in a mineral silicate	GWP
Production of chemicals	Aresta and Galatola [66]	Comparative LCA of the synthesis of dimethylcarbonate (DMC) via conventional route with phosgene and an alternative route using captured CO ₂ as a feedstock (urea-based synthesis)	Removal from diluted streams via MEA	Production of DMC	Production of 1 kg of DMC	GWP, AP, EP, ODP, POCP
Enhanced oil recovery	Jaramillo et al. [67]	LCA of five IGCC plants in the US with carbon capture, compression, transport and use for EOR, including crude oil refining and combustion of refined products	Pre-combustion capture via selexol	Injection into oil field for EOR	Total production of electricity over the projected lifetime	GWP
	Hertwich et al. [12]	LCA of a 832 MW CCGT plant in Norway with carbon capture, compression, transport and use for EOR	Post-combustion capture via MEA	Injection into oil field for EOR	Production of 1 MWh of electricity; extraction of 1 m ³ of oil	GWP, AP
Diesel production from microalgae	Lardon et al. [55]	Cradle to grave LCA of biodiesel produced from microalgae (<i>Chlorella vulgaris</i>) in an open raceway pond in France	Direct injection from industrial sources	Production of biodiesel	1 MJ of fuel	GWP, AP, EP, ADP, ODP, HTP, MAETP, POCP, land competition and ionising radiation
	Brentner et al. [51]	Comparative cradle to gate LCA of biodiesel produced from microalgae (<i>Scenedesmus dimorphus</i>) through various methods available in the US	Direct injection of flue gas from power plant; pure CO ₂ captured with MEA from ammonia plant	Production of biodiesel	10 GJ of fuels	GWP, EP, energy, land requirement, water use
	Campbell et al. [60]	Comparative LCA of biodiesel production from microalgae in open raceway ponds with canola biodiesel and ultra-low sulphur diesel in Australia	Direct injection of flue gas from power plant; pure CO ₂ captured with MEA from ammonia plant	Production of biodiesel	Tonne kilometre (t km)	GWP
	Clarens et al. [69]	Cradle to grave LCA of biodiesel produced from microalgae (<i>Phaeodactylum</i> sp. and <i>Traselmis</i> sp.) in an open pond in the US	Direct injection of flue gas from power plants; pure CO ₂ captured with MEA from power plant	Production of biodiesel and bioelectricity	Vehicle kilometre travelled (VKT)	GWP, energy, water use
	Shirvani et al. [57]	Cradle to grave LCA of biodiesel produced from microalgae (<i>Chlorella vulgaris</i>) in an open raceway pond in various countries. The results were compared with fossil diesel	Direct injection of exhaust gas from combustion engines	Production of biodiesel	1 MJ of fuel	GWP, energy
	Borkowski et al. [59]	Cradle to gate LCA of biodiesel and renewable diesel produced from microalgae in open ponds located in Phoenix, AZ, USA. The study looked at different allocation scenarios for waste	Direct injection of flue gas from power plants; pure CO ₂ captured with MEA from power plant	Production of biodiesel and renewable (green) diesel	1 MJ of fuel	GWP
	Passell et al. [54]	Cradle to grave LCA of biodiesel produced from microalgae (<i>Nannochloris</i> sp. and <i>Nannochloropsis</i> sp.) in an open pond in Israel. The results were compared with other types of diesel	Direct injection of flue gas from power plants	Production of biodiesel	1 MJ of fuel	GWP, POCP, energy, water use, SO _x and NO _x emissions
	Soratana et al. [56]	Cradle to grave LCA of biodiesel produced from microalgae (<i>Chlorella vulgaris</i>) in a flat-plate photobioreactor in the USA. The results were compared with other types of diesel	Direct injection of flue gas from power plants	Production of biodiesel	8.94 × 1010 MJ of diesel/year	GWP, EP, POCP
	Zaimes and Khanna [58]	Cradle to grave LCA of biodiesel produced from microalgae in open raceway ponds in the US. Multiple production pathways considered	Direct injection of flue gas from power plants; pure CO ₂ from power plants captured in MEA	Production of biodiesel	1 MJ of fuel	GWP, energy
Stephenson et al. [70]	Cradle to grave LCA of biodiesel production from microalgae (<i>Chlorella vulgaris</i>) in open raceway ponds and tubular bioreactor in the UK	Direct injection of flue gases from a CCGT power plant	Production of biodiesel	1 tonne of biodiesel	GWP	

^a For impacts nomenclature, see Table 2.

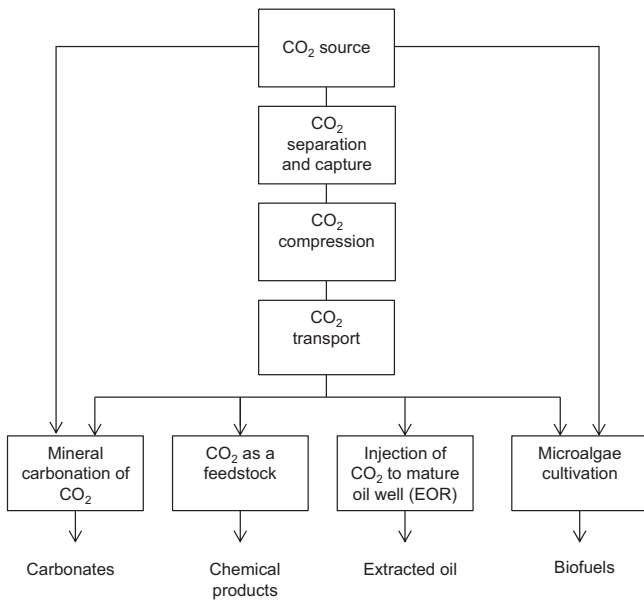


Fig. 16. System boundaries in the LCA studies of CCU technologies [6,12,38,51,54–60,66–70].

the carbonation process [68]. Furthermore, in the scenario with no heat recovery assumed by Khoo et al. [6], the GWP was equal to 937 kg CO₂ eq./t CO₂ removed, compared to 769 kg CO₂ eq. with heat recovery. Finally, applying different allocation methods – system expansion and mass allocation – for iron – and calcium-based (oxy)-hydroxides co-produced with MgCO₃, resulted in the GWP of 738 and 648 kg CO₂ eq./t CO₂ removed, respectively [38].

Therefore, these differing results suggest that the reduction in the GWP is sensitive to CO₂ capture and allocation methods as well as the assumptions for heat recovery from the system.

As indicated in Fig. 18, CO₂ capture to produce dimethylcarbonate (DMC) also leads to significant reductions in the GWP. Using

CO₂ recovered after the capture in MEA to produce DMC in the urea-based process reduces the GWP by 4.3 times compared to the conventional synthesis of DMC from phosgene (31 vs 132 kg CO₂ eq./kg DMC) [66]. The reductions in the other environmental impacts are similar, ranging from 3.6 times for the EP to four times for the AP and POCP to 13 times for the ODP. The main reason for this is a four times lower energy requirement for the production of raw materials used in the urea-based synthesis (hydrogen, ammonia and CO₂ recovered from MEA), than for the production of raw materials used in the phosgene-based process (chlorine and sodium hydroxide).

Utilising CO₂ from power plants for EOR can also reduce the GWP significantly (on average by 2.3 times) compared to discharging CO₂ to the atmosphere (Fig. 19) [12,67]. However, CCU via EOR has a higher GWP than the equivalent CCS option: by 50% in the study by Jaramillo et al. [67] and by 8% in Hertwich et al. [12]. It should be noted that the differences in the results between the two studies are due to different CO₂ capture options and EOR practices assumed (see Table 3).

Hertwich et al. [12] also found that the EOR increases the AP by almost three times compared to the system without CCU (Fig. 20). This is mainly because of the ammonia emissions from the CO₂ capture plant, which contribute 56% to the total AP. This is twice as high as the AP from the acid emissions at the power plant, which contribute 23% to the total; the EOR activities add a further 12%.

Finally, the average GWP of CCU via microalgae capture to produce biodiesel is estimated at 209 kg CO₂ eq./MJ (Fig. 21). This excludes the emissions of biogenic CO₂ captured from the atmosphere by microalgae and then released from the combustion of biodiesel during its use in vehicles. However, as can be observed in Fig. 21, the total GWP varies significantly between the studies, ranging from 19 to 534 g CO₂ eq./MJ. By comparison, the GWP of fossil diesel is 85 g CO₂ eq./MJ [51,54,56,71]. Only two studies found that biodiesel was a better option for the GWP than fossil fuel: Stephenson et al. [70] with 19 g and Brentner et al. [51] with 80.5 g CO₂ eq./MJ (Best case in Fig. 21).

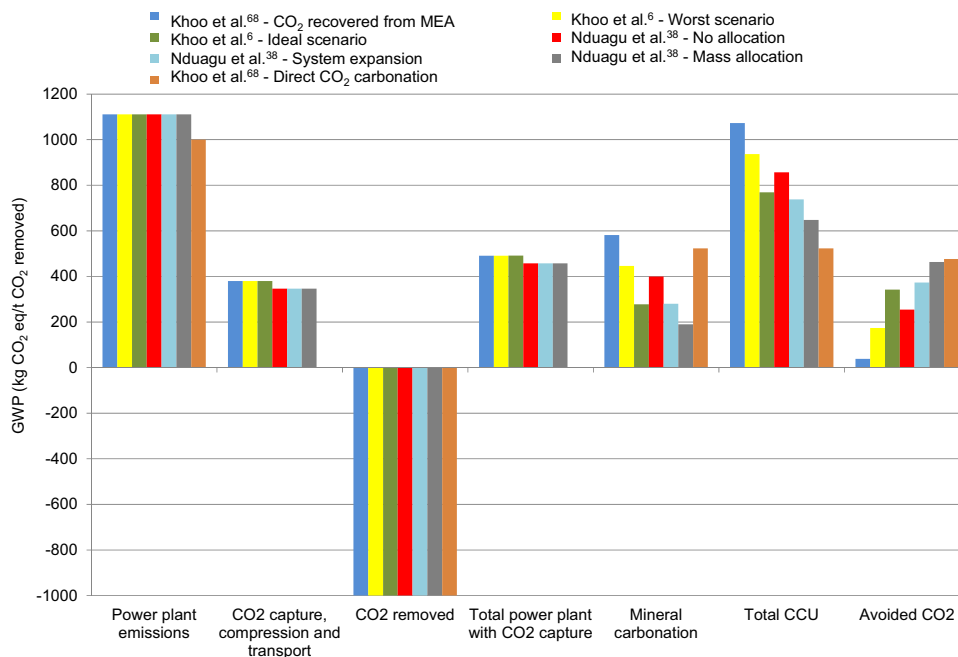


Fig. 17. Global warming potential of mineral carbonation, showing contribution of different life cycle stages [6,38,68]. The results include energy consumption for the regeneration of MEA. Nduagu et al. [38]: system expansion and mass allocation for the production of iron and calcium co-products. Khoo et al. [6]: Ideal scenario – the heat released from the carbonation process is recovered and used within the system. Worst scenario – no heat recovery. Khoo et al. [68]: CO₂ recovered from MEA – CO₂ from flue gas absorbed and then recovered from MEA with the capture efficiency of 90%; Direct CO₂ carbonation – CO₂ captured directly from flue gas with the efficiency of 100%.

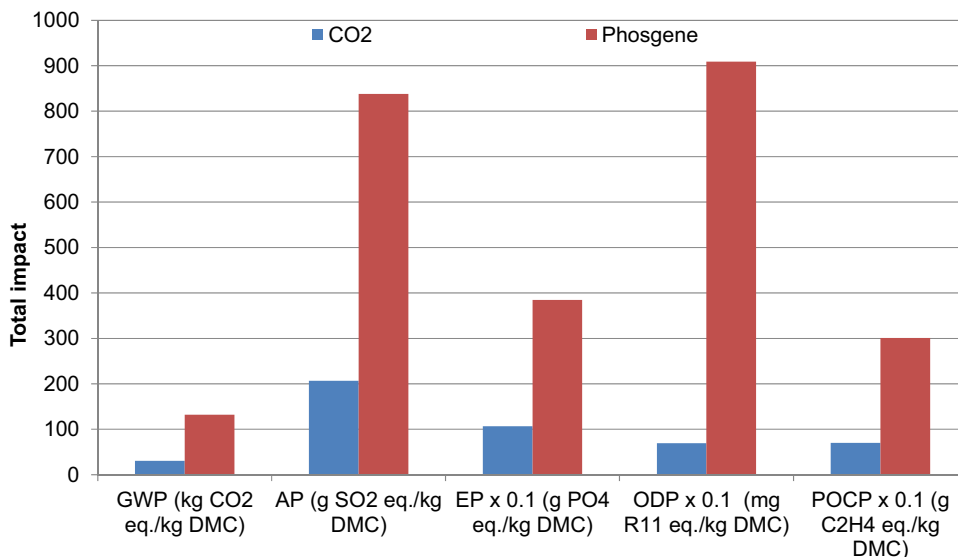


Fig. 18. Environmental impacts of producing dimethylcarbonate (DMC) from captured CO₂ compared to conventional production of DMC from phosgene [66]. Production of DMC from CO₂ in urea-based process. For impacts nomenclature, see Table 2. Some impacts have been scaled to fit. To obtain the original values, multiply with the factors shown against relevant impacts.

The large difference in the GWP results found for the biodiesel in different studies appears to be largely due to the method assumed for disposal of waste biomass generated from microalgae during the production of diesel. For example, despite assuming similar cultivation of microalgae and diesel production processes, Stephenson et al. [70] reported the lowest GWP value and Brentner et al. [51] the highest in the above-mentioned range because the former considered anaerobic digestion of waste to generate electricity and in the latter the waste biomass was landfilled. Landfilling was also found to be the worst waste management option by Shirvani et al. [57] who report the second largest GWP for open raceway ponds of 300 g CO₂ eq./MJ. Processing waste for animal feed appears to be only marginally better at 235 g CO₂ eq./MJ. However, the very low GWP value in Stephenson et al. [70] is out of range compared to the other similar studies. For instance, for the same waste disposal method as well as cultivation and diesel

production, Borkowski et al. [59] report the GWP value of 160 g CO₂ eq./MJ and Zaimis and Khanna [58] 140 g CO₂ eq./MJ (see Scenario 3 and Case 3, respectively, in Fig. 21). Generating electricity by direct combustion of waste appears to be a better option than electricity from biogas produced by anaerobic digestion of waste for the same cultivation and diesel production route as above, with Borkowski et al. [59] estimating the GWP of 130 g CO₂ eq./MJ (Scenario 2 in Fig. 21). This is mainly because of methane emitted to the atmosphere from the anaerobic digestion process. Combusting waste in a combined heat and power plant is a better option still, reducing the GWP to 93–105 g CO₂ eq./MJ [57].

In addition to waste management options, the source of CO₂ also influences the GWP results. As can be seen in Fig. 21, using pure CO₂ recovered after capture in MEA increases the GWP by 30–60%, depending on the waste management option considered. For example, assuming electricity generation from biogas produced by

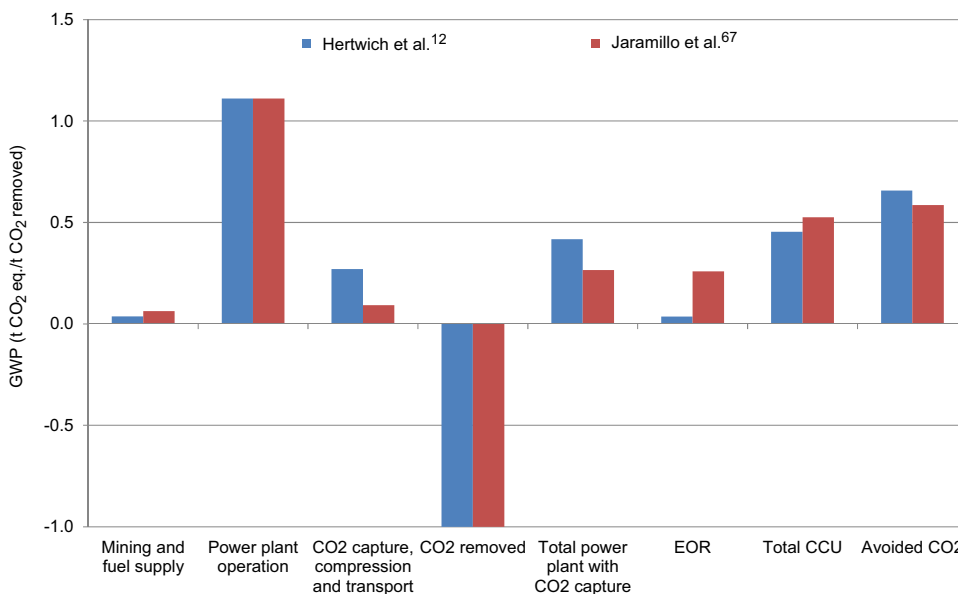


Fig. 19. GWP of enhanced oil recovery, showing contribution of different life cycle stages [12,67]. CO₂ capture efficiency: 90%. The results in Hertwich et al. [12] include energy consumption for the regeneration of MEA.

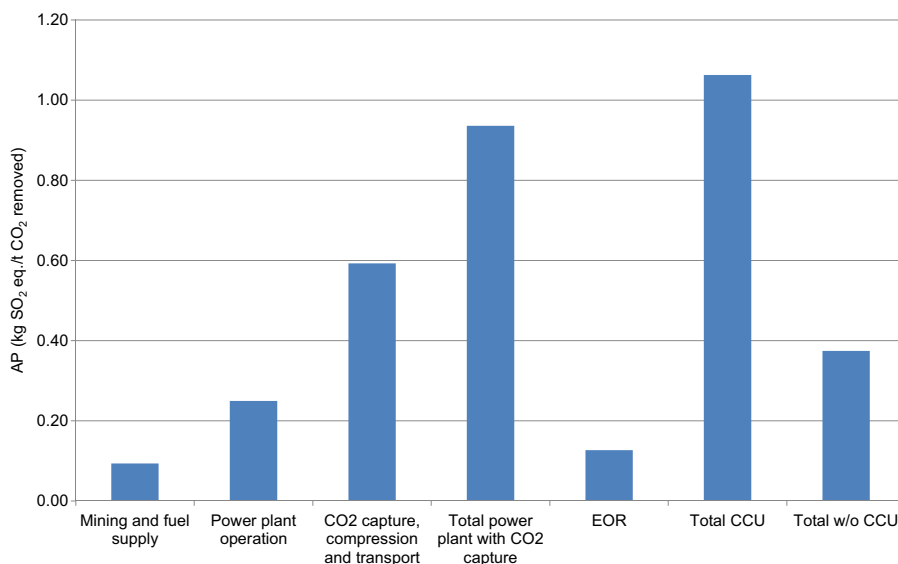


Fig. 20. Acidification potential (AP) of CCGT power plant with and without enhanced oil recovery (EOR), showing contributions of different life cycle stages [12]. CO₂ capture efficiency: 90%; w/o: without.

anaerobic digestion, Borkowski et al. [59] report the GWP value of 230 g CO₂ eq./MJ for CO₂ from MEA (Scenario 1) and 160 g for CO₂ supplied directly with flue gases (Scenario 3). A similar difference in the results is found by Zaimes and Khanna [58] who estimate the GWP for CO₂ from MEA at 188 g CO₂ eq./MJ and 140 g for direct injection of CO₂ (Case 1 and Case 3, respectively in Fig. 21).

Despite landfilling of waste having the highest GWP for cultivation of microalgae in open raceway ponds, Brentner et al. [51] found that the impact can be reduced significantly (from 534 to 81 g CO₂ eq./MJ) if microalgae are cultivated in a flat-plate

photobioreactor instead of using open raceway ponds, regardless of the waste still being landfilled. This is partly due to more efficient cultivation but also due to the use of supercritical methanol instead of hexane for lipid extraction. Soratana et al. [56] also assumed the same reactor and source of CO₂ (direct injection), but estimated a much higher GWP of 455 g of CO₂ eq./MJ; however, disposal of waste biomass was not considered.

Finally, Stephenson et al. [70] considered an air-lift tubular bioreactor assuming the same cultivation and processing conditions as in the studies with open raceway ponds and estimated the

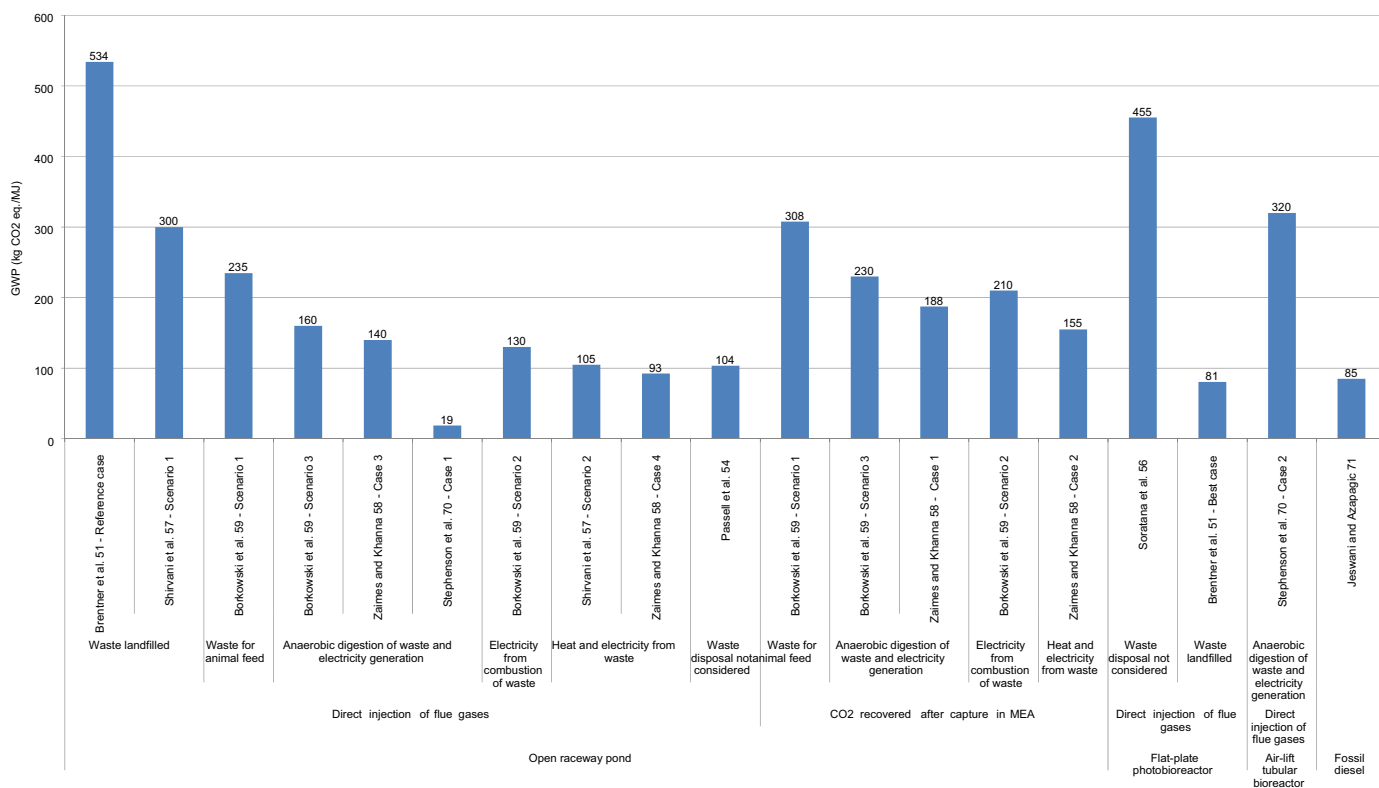


Fig. 21. GWP of biodiesel produced from microalgae in comparison with fossil diesel [51,54,56–59,70]. All studies consider biodiesel production via lipid extraction with hexane and transesterification (LEWH&T) except for Brentner et al. [51] – Best case which assumes lipid extraction with supercritical methanol. All authors assume microalgae harvesting by flocculation except for Brentner et al. [51] – Reference case and Passell et al. [54] who assume centrifugation and Stephenson et al. [70] who consider both. The results in Borkowski et al. [59] and Zaimes and Khanna [58] include energy consumption for the regeneration of MEA.

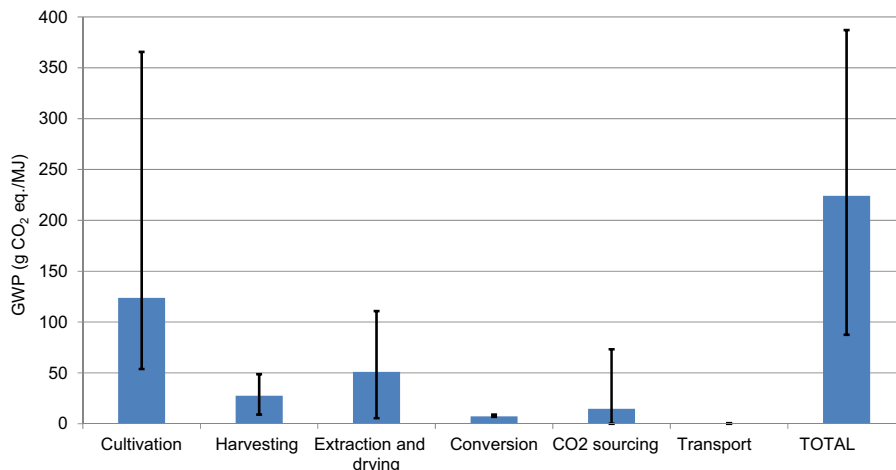


Fig. 22. Global warming potential of biodiesel produced from microalgae, showing contribution of different life cycle stages [54,59,70]. The bars represent an average GWP and the error bars the GWP ranges in different life cycle stages reported in Borkowski et al. [59] (Scenario 1 (MEA) and Scenario 1 (DI)), Passell et al. [54] and Stephenson et al. [70]. For the technologies assumed in each study, see Fig. 21. Note that the total average value of the GWP differs from that in Fig. 21 as only three studies are considered here.

GWP of 320 g of CO₂ eq./MJ (Case 2 in Fig. 21), the third highest value reported for microalgae diesel, despite assuming anaerobic digestion and electricity generation as a waste management option.

Only three studies reported the contribution of different life cycle stages to the total GWP. These results, shown in Fig. 22, suggest that the cultivation stage is the main ‘hot spot’ for biodiesel production from microalgae, contributing on average 55% to the total impact. This is followed by the extraction and drying stage, which adds a further 23%. Harvesting and CO₂ sourcing contribute 12% and 7%, respectively and the conversion the remaining 3%. As also indicated in Fig. 22, the contributions from the life cycle stages vary depending on the technologies and process conditions considered. For example, the (maximum) GWP of microalgae cultivation using an air-lift tubular bioreactor is estimated at 366 g CO₂ eq./MJ [70], while the impact from the cultivation in open raceways ponds ranges between 50 and 70 g CO₂ eq./MJ [54,59,70]. Furthermore, the GWP of harvesting by flocculation is found to be 5.4 times higher than by centrifugation (49 vs 9 g CO₂ eq./MJ, respectively) [54,59].

Three out of the nine studies for microalgae biodiesel estimated other environmental impacts in addition to the GWP, specifically, the EP and POCP. As shown in Fig. 23, the EP for biodiesel is found to

be much higher than for fossil diesel (0.14–0.67 g PO₄ eq./MJ vs 0.05 g PO₄ eq./MJ) because of the energy-intensive harvesting, with the results varying depending on the harvesting technology used. For example, flocculation has three times and centrifugation 13 times higher EP compared to fossil diesel [51,56]. The POCP for biodiesel, on the other hand, varies from being 2.5 times lower to being six times higher than for fossil diesel [54,56]. The higher value is due to the low aerial productivity (25 g/m²/day), which is seven times lower than that reported in Soratana et al. [56] (180 g/m²/day), thus requiring higher energy for cultivation and resulting in a higher POCP.

3.3. Comparison of CCS and CCU

This section compares the environmental impacts of different CCS and CCU technologies discussed in Sections 3.1 and 3.2, respectively. To enable the comparison, the functional unit is defined as “1 tonne of CO₂ removed” and the original results have been recalculated accordingly as detailed in Supplementary material. The results are shown in Figs. 24–26.

As indicated in Fig. 24, the average GWP in the CCS studies is estimated at 276 kg CO₂ eq./t CO₂ removed, which is significantly lower than for the CCU options. The worst CCU option appears to be

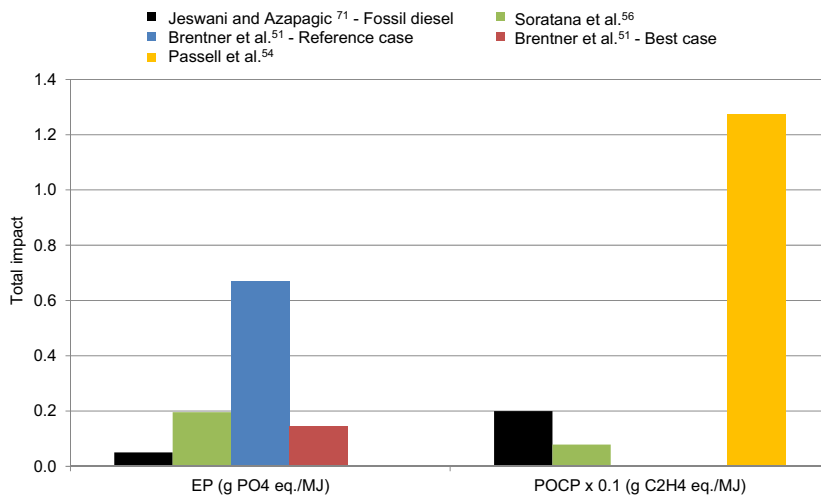


Fig. 23. Eutrophication potential (EP) and photochemical oxidants creation potential (POCP) of biodiesel from microalgae in comparison with fossil diesel [51,54,56,71]. The POCP in Passell et al. [54] and Soratana et al. [56] was expressed in different units (g NO_x and g NMVOC eq., respectively). To enable comparisons with the other studies, these units have been converted in this paper into g C₂H₄ eq. The POCP has been scaled to fit. To obtain the original value, multiply by 0.1.

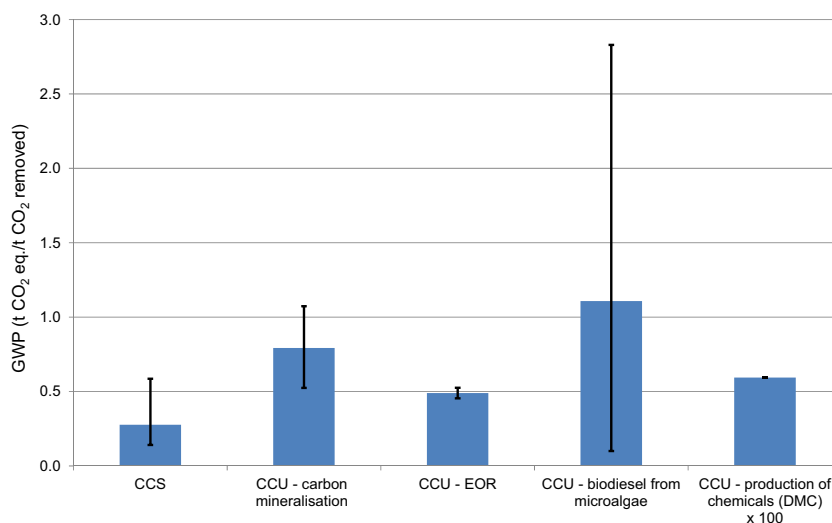


Fig. 24. Comparison of the global warming potential for different CCS and CCU options. CCS [19,26,28,29,40,61–65]; CCU – carbon mineralisation [6,38,68]; CCU – EOR [12,67]; CCU – biodiesel from microalgae [51,54,56–59,70], CCU – production of chemicals (DMC) [66]. The latter has been scaled to fit. To obtain the original value, multiply by 100.

the production of chemicals, specifically DMC from waste CO₂, which has the average GWP 216 times higher than CCS (59.4 vs 0.276 t CO₂ eq./t CO₂ removed). Almost two tonnes of DMC need to be produced in order to remove one tonne of CO₂, requiring large quantities of reactants such as ammonia and naphtha, which increases the impact relative to CCS [66]. However, as there is only one rather dated study of the production of chemicals from CO₂ and is specific to DMC only, in the absence of other data these findings cannot be corroborated or extrapolated to other types of chemical. This indicates a research gap in terms of LCA studies related to CCU via production of chemicals.

The second worst option is biodiesel production with the average GWP four times higher than for CCS. However, the GWP for this option varies significantly depending on the technologies and process conditions assumed (for details, see Section 3.2) so

that at the lower range biodiesel has a lower GWP than the lowest value for CCS: 0.1 vs 0.14 t CO₂ eq./t CO₂ removed. The former refers to biodiesel from microalgae cultivated in open raceway ponds and the latter to CCS using pre-combustion via selexol coupled with IGCC and ocean storage below the sea bed [19,70].

Carbon mineralisation and EOR have the average GWP 2.9 and 1.8 times higher than CCS, respectively. This is further explored in Fig. 25 which shows the contribution of different life cycle stages to the total GWP from these options, including the avoided carbon emissions. As indicated, PC and IGCC power plants with CCS have the highest CO₂ avoided, removing between 0.80 (PC) and 0.88 (IGCC) tonnes of CO₂ eq. per tonne of CO₂ treated. By comparison, CCU via EOR removes 0.59–0.66 tonnes and CCU via carbon mineralisation 0.26–0.36 t/t CO₂ removed.

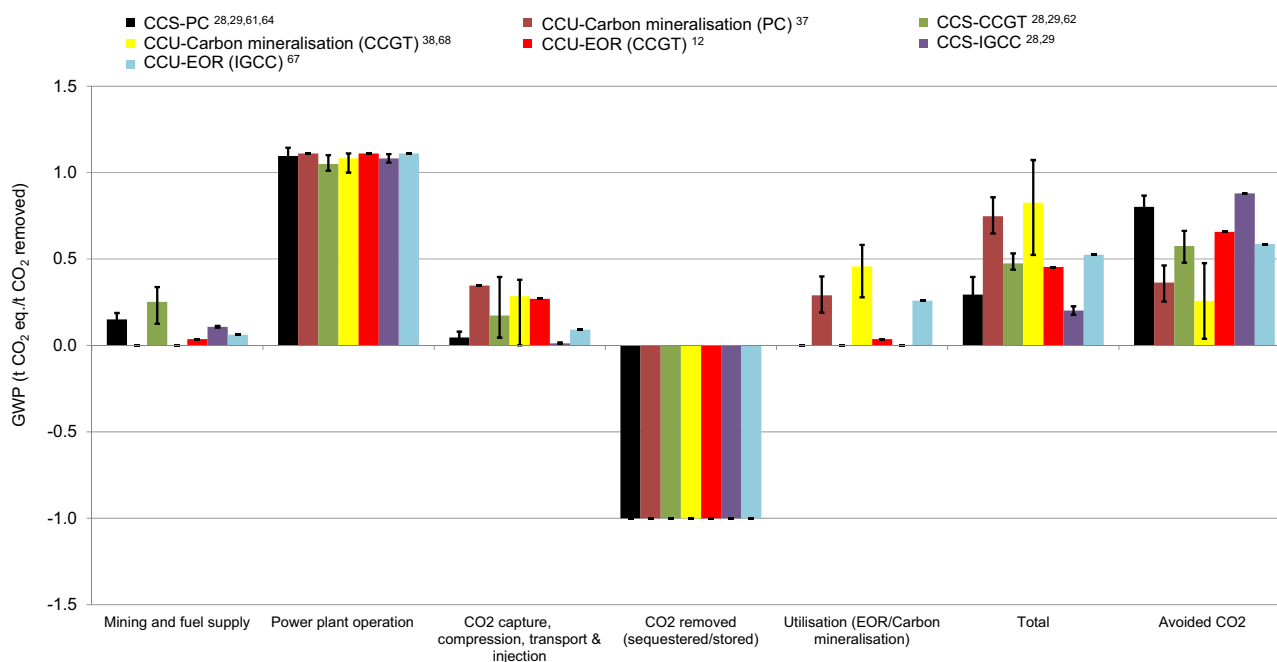


Fig. 25. Comparison of the global warming potential for CCS and CCU for different power plants, showing the contribution of life cycle stages. CM – carbon mineralisation. All studies for PC and CCGT power plants considered post-conversion capture in MEA. Studies for IGCC power plants considered pre-conversion adsorption on rectisol [29] and selexol [28,67].

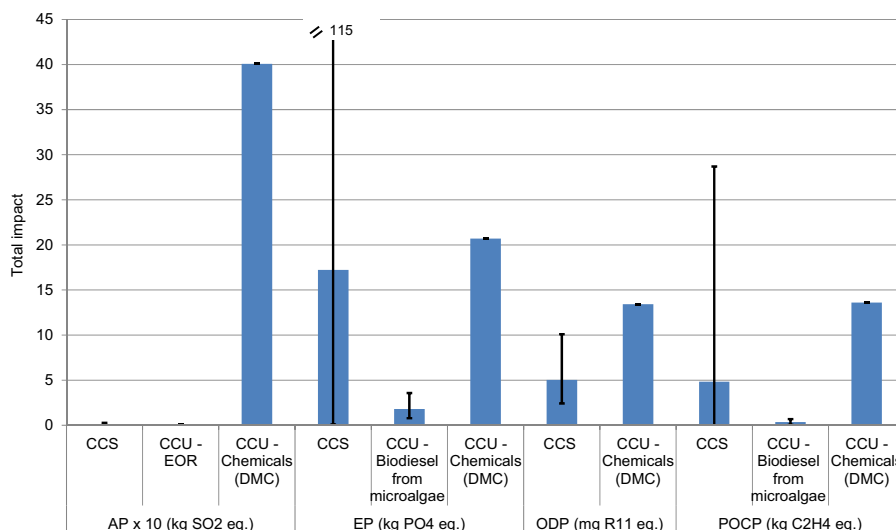


Fig. 26. Environmental impacts (other than the GWP) of CCS and CCU for different utilisation options. CCS: acidification potential (AP) from [26,29,40,61,63–65], eutrophication potential (EP) from [26,29,40,61–64], ozone layer depletion potential (ODP) from [61,64], photochemical oxidants creation potential (POCP) from [26,29,40,61,63,64]. CCU-EOR: AP from [12]. CCU-Chemicals (DMC): All impacts from [66]. CCU-Biodiesel from microalgae: EP from [51,56] and POCP from [54,56].

Fig. 25 also shows that the GWP of PC power plants with CCU via carbon mineralisation (0.75 t CO₂ eq./t CO₂ removed) is 2.6 times higher than for the same plant using CCS instead (0.29 t CO₂ eq./t CO₂ removed). The same difference is found for the GWP of IGCC plants with CCU via EOR (0.53 t CO₂ eq./t) in comparison with CCS (0.2 t CO₂ eq./t CO₂ removed).

For CCGT power plants, the GWP depends on the CCU option used. For example, the GWP of CCGT power plants with CCU via carbon mineralisation (0.83 t CO₂ eq./t CO₂ removed) is 1.7 times higher than for the plants using CCS (0.48 kg CO₂ eq./t CO₂ removed) but it is 4% higher than for CCU via EOR.

As also indicated in Fig. 25, the main hot spots for power plants with CCS are upstream processes such as mining and transport of fuels, which on average contribute 52% of the total GWP, followed by power plant emissions (after capture) with 28% and CO₂ capture activities with 19%. Conversely, the main hot spots for power plants with CCU are shared equally between the CO₂ capture and utilisation activities, each contributing on average around 39% to the total but with quite a wide spread of results, depending on the option assumed (see Fig. 25). Net power plant emissions and upstream processes contribute on average 18% and 5%, respectively.

Apart from the GWP, only four other environmental impacts of CCS and CCU have been considered in the literature, notably the AP, EP, ODP and POCP. These are compared in Fig. 26. As can be seen, similar to GWP, all four impacts for the production of chemicals (DMC) are higher than for CCS with the AP being 320 times higher, ODP and POCP around 2.8 times and EP 20% higher. This is because production of DMC requires large quantities of reactants including ammonia and naphtha which increase the impacts relative to CCS.

Only the AP has been estimated for EOR which is found to be 14% lower than for CCS. The EOR study assumed a CCGT power plant as the source of CO₂ while the AP for CCS comprises various power plants including PC and IGCC, which have higher acid emissions than CCGT power plants [12].

The results in Fig. 26 also suggest that CCU via biodiesel from microalgae has respectively 9.6 and 13 times lower EP and POCP than CCS. The main reason for this is that the flue gases from power plants are injected directly into microalgae ponds thus avoiding the impacts of CO₂ capture in MEA and the subsequent recovery needed in CCS.

As mentioned at the beginning of this section, the results and comparisons of the studies considered here should be regarded as a guide only because of the methodological inconsistencies in the

LCA studies for CCS and CCU. This also highlights the need for standardised LCA guidelines for CCS and CCU technologies (such as, for example, those for the construction industry [72]) to enable consistent and fair comparisons between different options.

4. Conclusions and recommendations for future work

This paper has analysed the life cycle environmental impacts of various CCS and CCU options for the capture, storage and/or utilisation of CO₂ emitted by power plants and other industrial sources. The main CO₂ capture options are post- and pre-conversion capture and oxy-fuel combustion. Post-conversion capture via chemical absorption using monoethanolamine (MEA) is the most mature and widely used technique, especially in the power generation sector. However, the use and regeneration of MEA is a significant contributor to the emissions of CO₂ and related global warming potential (GWP), so that the development of more environmentally sustainable sorbents is one of the challenges for both CCS and CCU.

The captured CO₂ can be stored in geological formations, also known as geological storage, or in the oceans. The former represents a more viable option as the properties of depleted oil and gas reservoirs and deep saline aquifers are better understood. In particular, it is not clear how disposal in the oceans would affect the acidity and marine species. Besides storage, CO₂ can be used directly in different industrial sectors, including the food and beverage as well as pharmaceutical industry. It can also be converted into high-demand products such as urea, methanol and biofuels.

The results of the LCA studies of different CCS options found in the literature indicate that the GWP from power plants can be reduced by 63–82% per unit of electricity generated, depending on the CO₂ capture option. The average GWP for pulverised coal (PC) power plants without CCS is 876 kg CO₂ eq./MWh while for the post-conversion capture via MEA the average value is 203 kg CO₂ eq. and for oxy-fuel combustion it is 154 kg CO₂ eq. The equivalent average values for CCS at CCGT power plants are estimated at 120 kg CO₂ eq./MWh for oxy-combustion and 173 kg for post-combustion, compared to 471 kg CO₂ eq./MWh without CCS. The GWP for pre-combustion capture and oxy-fuel combustion in IGCC plants is similar: 190 and 200 kg CO₂ eq./MWh, respectively, while the average without CCS is 1009 kg CO₂ eq./MWh. Therefore, the greatest GWP reductions (up to 82%) can be

achieved by oxy-fuel combustion in PC and IGCC plants and the lowest by post-combustion capture in CCGT plants (63%).

The results for the other environmental impacts vary across the studies. However, the large majority reported higher impacts for the plants with than without CCS. This is mainly attributed to the additional coal mining and shipping needed to compensate for the energy efficiency losses from the use of CCS, MEA production and ammonia emissions released during the absorption of CO₂ in MEA. Therefore, the impacts are transferred from power plants, further up or downstream from the power plants.

For CCU systems, the CO₂ savings depend largely on the utilisation option. For example, mineral carbonation to produce MgCO₃ can reduce the GWP from 4% to 48% compared to no CCU. The estimated GWP ranged from 524 kg CO₂ eq. per tonne of CO₂ removed for the carbonation of CO₂ directly from a power plant to 1073 kg eq./t removed when CO₂ is first absorbed in MEA and then recovered to be used in the carbonation process. Using CO₂ recovered after the capture in MEA to produce DMC in the urea-based process reduces the GWP by 4.3 times compared to the conventional synthesis of DMC from phosgene (31 vs 132 kg CO₂ eq./kg DMC). Utilising CO₂ from power plants for EOR can also reduce the GWP significantly (on average, by 2.3 times) compared to discharging CO₂ to the atmosphere. As for the CCU via microalgae capture to produce biodiesel, the average GWP is estimated at 209 kg CO₂ eq./MJ, which is 2.5 times higher than for conventional fossil diesel.

The results for the other environmental impacts of CCU also vary widely across the utilisation options. For example, production of DMC from CO₂ can reduce eutrophication by 3.6 times, acidification and photochemical oxidants by four times and ozone layer depletion by 13 times compared to the conventional phosgene-based process. This is because of four times lower energy requirements for the former than the latter. On the other hand, for EOR, acidification is three times higher with than without utilisation. This is mainly due to the ammonia emissions from the CO₂ capture plant. In the case of CCU via microalgae capture to produce biodiesel, eutrophication is found to be much higher than for conventional diesel because of the energy intensive harvesting. The photochemical oxidants creation potential is reported to be both higher and lower than for conventional diesel as a result of the different assumptions made for aerial productivity.

This study has also compared the environmental impacts of CCS and CCU. The findings indicate that the average GWP of all CCS options is estimated at 276 kg CO₂ eq./t CO₂ removed, which is significantly lower than the GWP of any of the CCU options considered. For example, the average GWP of CCU via chemicals production (DMC) is estimated at 59.4 t CO₂ eq./t CO₂ removed, which is 216 times higher than for CCS. These results indicate that, compared to CCS, this particular option is not an efficient method for removing CO₂ emissions. This is because almost two tonnes of DMC need to be produced to remove one tonne of CO₂, which requires large quantities of reactants, including ammonia and naphtha, generating additional CO₂ eq. emissions. Biodiesel production also has higher GWP than CCS, by four times on average. However, this impact varies significantly among studies depending on the technologies and process conditions assumed. For CCU via EOR and carbon mineralisation, the average GWP is 1.8 and 2.9 times higher than for CCS, respectively.

Similar to the GWP, the other environmental impacts of CCU to produce DMC are higher, with acidification being 320 times higher, ozone layer depletion and photochemical oxidants around 2.8 times and eutrophication 20% higher than for CCS. CCU via biodiesel from microalgae has respectively 9.6 and 13 times lower eutrophication and photochemical oxidants than CCS. For EOR, only acidification has been estimated in the literature and it is found to be 14% lower than for CCS.

However, these comparisons between CCS and CCU should be used as a guide only as the inconsistencies in the system boundaries and functional units in the studies make it difficult to compare them on an equivalent basis. For that reason, specific guidelines or 'product category rules' should be developed for the application of the LCA methodology to CCS and CCU technologies. Specifically, the guidelines on the definition of the system boundaries and functional unit should be established for different systems. Further recommendations for future research related to LCA of CCS and CCU include:

- consideration of a wider range of LCA impacts from CCS and CCU, rather than focusing on the GWP only – as shown in this paper, while the GWP may be reduced, other impacts can increase;
- consideration of the influence on results of different allocation methods;
- assessment of the uncertainty in the data and results, particularly as most systems are still at the development stage and real operational data are not available;
- assessment of the effect on the results of different assumptions, particularly CCS and power plant efficiencies and energy penalties, both today and in the future, fuel composition and related emissions, quality of CO₂ captured, potential CO₂ leakage rates from storage and the actual lifetime storage of CO₂ in products made from it;
- further studies of various CCU options, particularly production of chemicals;
- studies of different sources of CO₂, including biogenic CO₂, for example from anaerobic digestion; and
- comparisons of environmental impacts of CCS and CCU with other options, such as renewable energy;

In conclusion, **even though both CCS and CCU technologies seek to mitigate climate change, they can only be regarded as temporary solutions, particularly those options which merely delay the emissions of CO₂ rather than eliminate them permanently. Although from an economic perspective, CCU would appear to be a better option than CCS as the latter is an unprofitable activity, the cost-effectiveness as well as the environmental impacts of CCU have to be evaluated carefully on a life cycle basis to ensure a positive economic and environmental balance.** As demonstrated in this analysis, the latter in particular may not always be the case. Moreover, the potential of CCU is still limited as the current global demand of chemicals and other products does not have the capacity to sink enough CO₂ emissions to contribute significantly to meeting the carbon reduction targets. A further significant issue for CCU is that the 'storage' time of CO₂ is limited by the short lifespans of the chemicals and fuels produced. Therefore, future research should focus on the development of materials and products with longer lifetimes to enable long-term storage of CO₂. While CCS overcomes this problem through long-term storage, there is a risk of CO₂ leakage which could potentially cause more damage than if dilute emissions were to continue unabated. Equally significant is the fact that deployment of large-scale CCS is not expected until well into the 2020s by which time it may be too late to reverse the impacts of climate change. Nevertheless, **if the above concerns can be addressed, both CCS and CCU could play a role in mitigating climate change, together with other options such as energy demand reduction, renewables and other low-carbon technologies.**

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:10.1016/j.jcou.2014.12.001.

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