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Review

Limits to Paris compatibility of CO_2 capture and utilization

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SUMMARY

The Paris Agreement's temperature goals require global CO_2 emissions to halve by 2030 and reach net zero by 2050. CO_2 capture and utilization (CCU) technologies are considered promising to achieve the temperature goals. This paper investigates which CCU technologies—using atmospheric, biogenic, or fossil CO_2—are Paris compatible, based on life cycle emissions and technological maturity criteria. We systematically gathered and harmonized CCU technology information for both criteria and found that CCU with technology readiness levels (TRLs) of 6 or higher can be Paris compatible in 2030 for construction materials, enhanced oil recovery, horticulture industry, and some chemicals. For 2050, considering all TRLs, we showed that only products storing CO_2 permanently or produced from only zero-emissions energy can be Paris compatible. Our findings imply that research and policy should focus on accelerating development of CCU technologies that may achieve (close to) zero net emissions, avoiding lock-in by CCU technologies with limited net emission reductions.

INTRODUCTION

In the 2015 Paris Agreement, almost all of the world's nations committed to collectively hold "the increase in the global average temperature to well below 2°C above pre-industrial levels and pursuing efforts to limit the temperature increase to 1.5°C above pre-industrial levels."[2] This is also known as the Paris Agreement's long-term temperature goal (LTTG). Climate change mitigation pathways with "no or low overshoot" of 1.5°C[3] are compatible with the Paris Agreement's LTTG[4–7] and characterized by two key numbers: in 2030, global net anthropogenic carbon dioxide (CO_2) emissions are halved compared with 2020 emissions, and in 2050, net CO_2 emissions are zero.[3] This net zero CO_2 target implies that no sector can be excluded from deep emission reductions, that some technologies that still have significant emissions need to be phased out, and that a range of technologies is required to reach these emission reductions.

Carbon (dioxide) capture and utilization (CCU) is among these options for potential emission reduction and is defined here as a process in which CO_2 is technologically captured from CO_2 point sources or ambient air and is subsequently used in or as a product. The reason why CCU could contribute to climate change mitigation is that it replaces fossil feedstocks, avoids upstream emissions, and temporarily keeps CO_2 out of the atmosphere until re-emitted in the use phase of the product.[8–13] CCU is distinguished from (permanent) carbon dioxide removal (CDR) of atmospheric CO_2. The two terms only overlap when CO_2 in a CCU product has recently been removed from the atmosphere and is never re-emitted.[14] CCU appeals to policymakers and the general public because it is seen as part of the circular economy and a form of sustainable waste processing.[15] It also appeals to industry because CCU creates value from waste through CO_2-based products[16,17] while avoiding the storage costs and concerns of geological storage of captured CO_2, known as carbon (dioxide) capture and storage (CCS).[18]

However, the relevance of CCU in climate change mitigation is questioned in the literature, based on several concerns: (1) CCU products may not always substantially reduce emissions compared with their conventional counterparts that do not require the energy-intensive CO_2 capture and conversion steps;[19–22] (2) utilization of captured CO_2, rather than permanent geological storage, may result in a higher global warming effects because utilized CO_2 is typically re-emitted when the CCU product is used or disposed of;[16,19] (3) CCU may not be economically feasible because of the high financial costs associated with the energy-intensive CO_2 capture and conversion steps;[19,23] and (4) CCU may form a political distraction from reducing CO_2 emissions, in particular when replacing CCS, because the scale at which CO_2 could be utilized is limited compared with the scale at which CO_2 could be stored geologically.[21]

The goal of this review is to provide conceptual clarity on what CCU is and what can be expected from different CCU technological routes, in particular in reaching the Paris Agreement's LTTG. We first describe the different process steps and varieties of CCU technologies. Next, we present a framework to assess "Paris compatibility" in the context of CCU, using criteria based on technological maturity and greenhouse gas emissions reductions. We then show the results of a systematic review of the

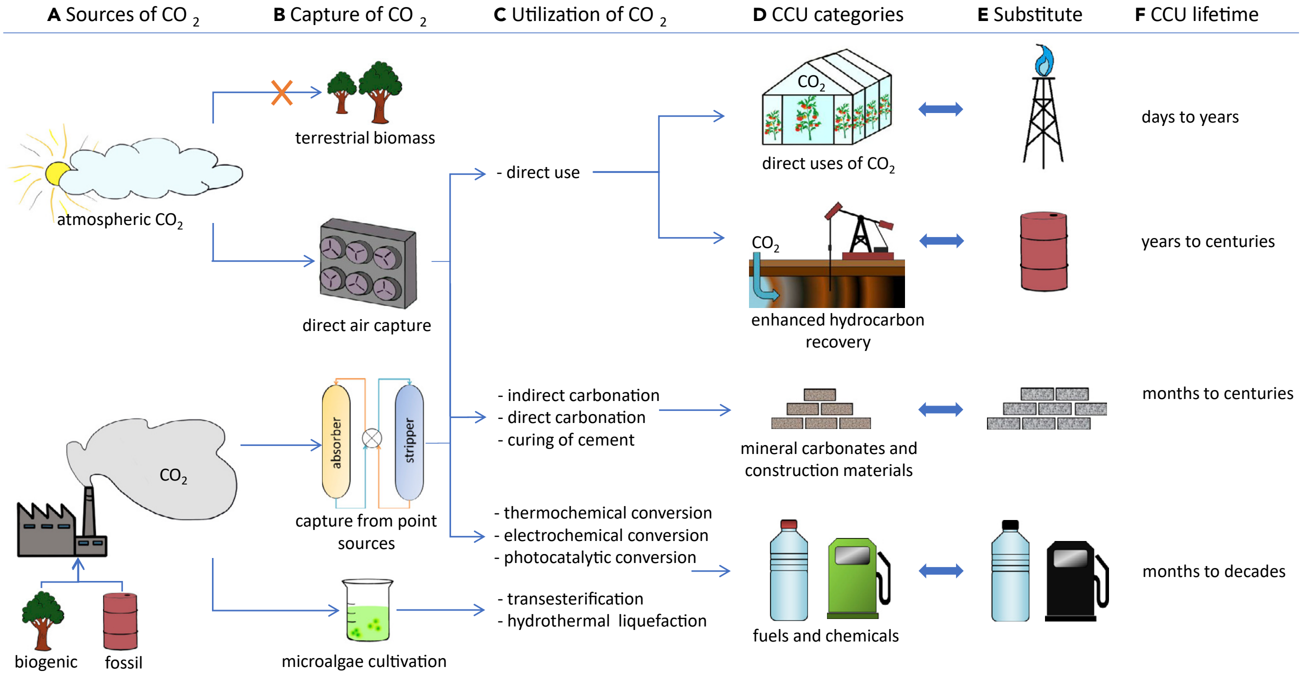


Figure 1. Scope of CCU

This overview of CCU includes sources of CO_2 (A), capture of CO_2 (B), and examples of utilization processes (C) leading to different CCU products in four categories (D), each with its substituted product in the conventional economy (E) and lifetime (F). CO_2 capture via terrestrial biomass (top process under "capture of CO_2") is not included in the this paper's CCU definition.

CCU literature following this framework. Last, we discuss our findings and provide a research and policy outlook for climate change mitigation through CCU. For an overview of acronyms used, see Note S1.

WHAT IS CCU?

Scope of CCU

In line with our definition of CCU, we defined six key characteristics of CCU (Figure 1):

(A) Sources of CO_2. CO_2 can originate from fossil fuel or biomass combustion in power plants or industrial plants, from industrial processes such as the calcination reaction in cement production or biomass fermentation, or from the atmosphere directly using direct air capture (DAC).

(B) Capture of CO_2. CO_2 is captured technologically on an industrial scale by separating CO_2 from a bulk gas stream or the atmosphere using a solvent or sorbent, a membrane, cryogenics, or industrially cultivated organisms, such as microalgae, to photosynthesize CO_2 into biomass.

(C) Utilization of CO_2. CO_2 is used directly or indirectly by converting CO_2 into a range of products, often requiring electricity, heat, and/or catalysts.

(D) CCU categories. The resulting CCU products can be categorized as direct uses, enhanced hydrocarbon recovery (EHR), mineral carbonates and construction materials, and fuels and chemicals.

(E) Substitute. A CCU product is assumed to replace a product in the conventional economy with the same chemical structure, composition, or characteristics, typically produced from fossil fuels and referred to as the substitute.[24]

(F) CCU lifetime. CO_2 is, depending on the CCU product, stored permanently or released into the atmosphere after a certain period of time, called its lifetime, ranging from days to centuries. For example, for fuels, the utilized CO_2 is emitted into the air upon combustion. For the purpose of this paper, we define storage as reaching permanency when it has a duration consistent with geological timescales: centuries or longer.

Because we consider CCU for climate change mitigation, we exclude the use of CO_2 from natural reservoirs because this source of CO_2 does not reduce atmospheric CO_2 concentrations.[25] Our definition of CCU constrains CCU to processes that "technologically capture CO_2," including industrial and engineered biological processes such as CO_2 capture from flue gases by microalgae, and excluding land-based CO_2 sequestration in biomass (in contrast to, for example, Detz and van der Zwaan[13] and Hepburn et al.[11]). Use of biomass for energy and materials is therefore also not in the scope of this review.

CCU is sometimes connected to CDR. CDR is a necessity to limit warming to 1.5°C[3] and has been defined as "anthropogenic activities removing CO_2 from the atmosphere and durably storing it in geological, terrestrial, or ocean reservoirs, or in products."[14] CDR includes methods like bioenergy with CCS (BECCS) and direct air CCS (DACCS).[26] CCU can only be classified as CDR, following the criteria formulated by Tanzer and

Ramírez,[27] when (1) physical CO_2 is removed from atmosphere by capturing CO_2 directly from the air or capturing CO_2 from a biogenic source, (2) the CO_2 is stored permanently and not re-emitted to the atmosphere at a later point, and (3) the net quantity of CO_2 stored permanently through CCU is greater than the quantity of CO_2 emitted over the product's full life cycle, including use of the product and the emissions associated with the energy required for the CO_2 capture and conversion processes. When the criteria for CDR are not met but the life cycle emissions are lower than for the substituted product, CCU is considered a climate change mitigation measure. Avoided emissions compared with the substituted product may be presented as a negative number but should not be confused with physical removal of CO_2 from the atmosphere.[27]

Capture of CO_2

CO_2 capture from a point source

CO_2 is typically captured from a point source, such as flue gas streams at power plants or industrial plants, or from industrial processes where CO_2 is produced as a by-product. Capturing CO_2 from a bulk gas stream entails separating the CO_2 from the rest of the stream. Sometimes CO_2 separation is already required in the primary production process; for example, in ammonia synthesis, natural gas processing, and biogas upgrading to biomethane.[28] This leads to a high-purity stream of CO_2 along with the primary product.

Depending on the concentration and components present in a gas stream, one of three main separation processes may be most suitable[29]: (1) absorption of CO_2 in a liquid solvent, (2) adsorption of CO_2 onto a solid, and (3) using a membrane to separate CO_2 through selective permeability.[30] In CO_2 absorption processes using amine-based solvents like monoethanolamine, CO_2 is chemically bound, and then the CO_2 is stripped to allow the solvent to absorb CO_2 again in the next cycle. This is called regeneration of the solvent and requires heat. Besides single-amine absorption, amine blends are in development where amines are combined for complementary characteristics. An integrated CCU process has also been proposed to reduce the overall energy demand, absorbing CO_2 from raw natural gas in methanol, after which conversion takes place.[31] Other developments include ionic liquids, which are liquids composed entirely of ions with a melting point of less than 100°C.[32] Physical adsorption research has focused on improving the adsorbents as well as the adsorption processes of regeneration.[32] Alternatives, like high-temperature solid adsorption of CO_2, require less energy input for regeneration compared with low-temperature liquid absorption.[33–35] Use of membranes for CO_2 separation is based on creating configurations especially for CO_2 selectivity from polymer or ceramic materials.[33]

Other emerging CO_2 capture technologies are high-temperature solid looping systems: calcium carbonate looping, where calcium oxide is used as a sorbent that binds CO_2 to form limestone, and chemical looping combustion, where a metal oxide is used to separate CO_2 from other components in the flue gas, foregoing the need for gas separation, followed by an exothermic metal oxide regeneration reaction.[32,36] Cryogenic systems are based on the differences in the temperature and pressure at which constituent gases in flue gas become liquid, removing CO_2 from the bulk stream by cooling and condensation.[33] CO_2 from a point source can also be captured photosynthetically or electrosynthetically into biomass by living organisms:[15,37] microalgae are cultivated at CO_2 concentrations of 5%–20%, making flue gas a suitable source, and have a conversion efficiency of solar energy into chemical energy higher than that of terrestrial plants (3%–8% instead of 0.5%).[38] Co-location of the facility at the site of the point source is important to avoid the need for transporting flue gas.[39] Other options for biofixation include microorganisms such as acetogenic bacteria[40] or anaerobic CO_2-sequestering bacteria.[38]

CO_2 capture from ambient air

DAC of CO_2 can be performed using a range of separation processes. Because atmospheric CO_2 concentrations (approximately 400 ppm) are 100–300 times lower than for point sources, energy requirements, and therefore costs, for DAC are substantially larger than for point source capture.[41,42] Expert elicitation leads to expected cost declines to around 200 US dollars (USD) per ton of CO_2 captured by 2050,[43] still higher than the 15–35 USD per ton of CO_2 captured for high-purity point sources and 60–120 USD per ton of CO_2 captured for lower-purity point sources; e.g., steel or cement production.[17,44] An exception would be when lower-purity CO_2 streams are sufficient, such as for microalgae.[45] The two main categories of DAC methods are based on (1) amine-functionalized solid sorbents, which require regeneration at low temperatures or via moisturizing,[46] and (2) alkaline hydroxide capture solutions, which require high-temperature solvent regeneration.[47] Less energy-intensive regeneration processes are under development, including electrochemical regeneration[48] and bipolar membrane electrodialysis.[49]

Utilization of CO_2

Direct uses of CO_2

CO_2 can be used directly, without conversion, in several sectors. In horticultural production, elevating CO_2 concentrations in greenhouses increases crop yields by approximately 50%.[50] This process is called CO_2 enrichment and is traditionally achieved by combustion of fossil fuels such as diesel[50] or natural gas,[51] which has the dual purpose of greenhouse heating. Because more CO_2 is required to reach the desired CO_2 concentration than is produced for heat, captured CO_2 can be used.[50] CO_2 can also be used directly as a refrigerant for supermarket applications, replacing hydrofluorocarbons with higher global warming potentials,[52] reducing risks of leakage and associated global warming effects.[53] CO_2 can also be used as a carbonating agent in sugar production and soft drinks, as a solvent for extraction of flavors, in the decaffeination process, as dry ice, in fire extinguishers, and in the pharmaceutical industry as a respiratory stimulant.[16,21,54]

EHR

Enhanced oil recovery (EOR) is a method where CO_2 is injected into the subsurface to recover oil from almost depleted reservoirs. EOR is a mature technology that has been practiced commercially for decades, starting in the early 1970s in North America.[25] Similarly, CO_2 can be used to recover natural gas from coalbeds (enhanced coalbed methane [ECBM]), although there are currently no active ECBM projects.[55] Although the source of CO_2 for EHR is typically natural CO_2 reservoirs (not part of our definition of CCU), EHR can also be performed with CO_2 captured from point sources or DAC.[45]

Mineral carbonates and construction materials

Mineral carbonation is a process where CO_2 reacts with magnesium or calcium oxides, -hydroxides, or -silicates in an exothermic reaction, forming stable carbonate products;[56] examples of magnesium- or calcium-rich materials are mined minerals[57] or industrial residues such as slag from steel production,[58,59] red gypsum,[60] fly ash,[61,62] or desalination brines.[63] These processes can be categorized into direct or indirect carbonation processes.[59] In the direct carbonation process, a gas-solid reaction takes place between CO_2 and the ground minerals. The produced carbonates can be used in concrete, asphalt, and other construction practices.[64] In the indirect carbonation process, magnesium or calcium ions are first extracted in an alkaline solution, and then they react with CO_2 to form precipitated magnesium carbonate (PMC),[63] precipitated calcium carbonate (PCC)[59] or nano-calcium carbonate (NCC).[65] These carbonates can be used as a pigment or filler material in production of paper, plastics, and pharmaceuticals.[66]

CO_2 can be used to cure concrete, absorbing CO_2 instead of steam in the hardening process. Steam-cured concrete normally re-absorbs roughly 30% of its production's CO_2 emissions during its lifetime,[67,68] so emissions are not reduced by CO_2-accelerated curing in itself. Instead, the reduction is achieved because of the lower steam requirement[69] and the improvements in mechanical properties, reducing the amount of cement required,[70–72] in a similar way as addition of "cementitious" materials like carbonated minerals reduces the need for cement in concrete.[61,73] Other options under development include production of carbon nanomaterials like graphene,[74] nanofibers, or nanotubes from CO_2,[75] which can be used in construction, reducing energy and material demand in the manufacturing process.[76]

Fuels and chemicals

Fuels and chemicals directly based on fossil fuels or fossil feedstocks are energy-dense products. Therefore, producing these chemicals or fuels from CO_2 often requires an energy-intensive conversion process at high pressure and/or increased temperature, supported by catalysts, because CO_2 is an inert and thermodynamically stable molecule. Conversion processes include thermochemical, electrochemical, and photocatalytic conversion.

In thermochemical conversion (sometimes referred to as "hydrogenation"), CO_2 and H_2 are prepared separately and subsequently combined.[77] The hydrogen supplies part of the energy needed for the CO_2 conversion process. This process allows production of chemicals and fuels that would otherwise be of fossil origin, like methane or methanol.[12] Methanol, in turn, can serve as a feedstock for production of other chemicals and fuels, such as ethylene,[78] polyols,[79] and dimethyl ether (DME).[80] Methane production can be a method for long-term storage of renewable energy from intermittent electricity, producing H_2 with water electrolysis, followed by CO_2 hydrogenation to methane (Sabatier reaction) and methane combustion for power generation at a later time (power-methane-power).[81] Another key hydrogenation process is synthesis of liquid hydrocarbon fuels, which consists of synthesis gas (syngas) production via conversion of CO_2 to carbon monoxide (CO) in the reverse water-gas shift (rWGS) reaction and mixing it with H_2 or via steam methane reforming (SMR) and, finally, using the Fischer-Tropsch process to create hydrocarbon chains.[82] The stoichiometric ratio of H_2 to CO in syngas can be adjusted to the targeted end products, as occurs currently via the WGS reaction in conventional syngas-based processes.[83]

In photocatalytic conversion, CO_2 is converted to, for example, methane or methanol using sunlight, water, and dye-sensitized semiconductors.[84] Electrochemical reduction, where CO_2 is reduced at atmospheric temperature using electricity,[85] can be used to produce, for example, ethanol,[12] methanol,[86,87] dimethyl carbonate (DMC),[20] formate,[88] or formic acid.[85] When CO_2 is captured by microalgae, the microalgae can be converted into fuels or chemicals; e.g., via transesterification or hydrogenation of algal oil[89,90] or hydrothermal liquefaction.[91] Microalgae may also be used as fish food[92] or food.[93]

ASSESSING THE PARIS COMPATIBILITY OF CCU TECHNOLOGIES

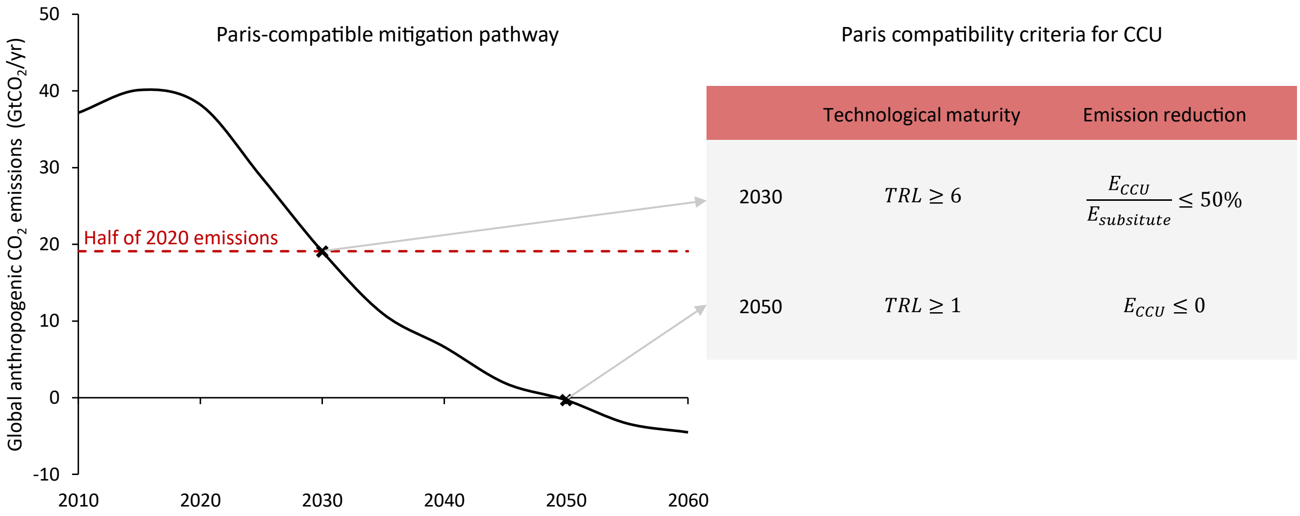
Paris compatibility criteria

In this paper, a CCU technology is classified as Paris compatible when its deployment is expected to be in line with reaching the Paris Agreement' LTTG and corresponding 1.5°C mitigation pathway. This definition results in two questions to assess Paris compatibility: (1) is the CCU technology ready on time and (2) does the CCU technology sufficiently reduce CO_2 emissions? These questions result in the Paris compatibility criteria for 2030 and 2050 outlined in this section (Figure 2). The following two sections present a review of the literature underlying the criteria and explain how these criteria can be applied to CCU technologies.

Maturity criteria

Technological maturing, the process leading from research and development (R&D) to demonstration trials, early market formation, and widespread diffusion, can span several decades and is ridden with financial risks and technological uncertainties.[94] We include technological maturity as a criterion for Paris compatibility because for a technology to be able to contribute to emission reductions in 2030 or 2050, it must be developed sufficiently to be ready for widespread diffusion.

The technological readiness level (TRL) of a technology is an indication of the maturity of a technology on a scale that summarizes detailed information on technological maturity into a single value.[95] The scale has nine levels, spanning from basic concept (TRL 1) to successful, real-life operation (TRL 9). TRL 6 presents a turning point in technological development by requiring an operational system at a relevant scale. The time it takes to progress through the TRL scale differs per technology and is context dependent, but for CCU technologies, 10–15 years is typically assumed to be needed to progress from lab scale to full-scale implementation.[9,96,97] In line with Chauvy et al.,[9] we assume that, for a CCU technology to be ready in 2030, it must be in at least TRL 6 in 2020. Given that it is possible to progress from the R&D phase (TRL 1–3) to real-life operation (TRL 9) in 20–30 years,[9,98] the TRL is no impediment for a CCU technology for 2050. This does not mean that it can be assumed that a technology in the R&D phase in 2020 will be ready in 2050, only that a



	Technological maturity	Emission reduction
2030	$TRL \geq 6$	$\frac{E_{CCU}}{E_{subsitute}} \leq 50\%$
2050	$TRL \geq 1$	$E_{CCU} \leq 0$

Figure 2. Paris compatibility criteria for CCU technologies

Paris-compatible mitigation pathway using scenario data from Huppmann et al.[1] as the mean of no and low overshoot 1.5°C scenarios from which Paris compatibility criteria for CCU technologies for 2030 and 2050 were derived. TRL is the 2020 technology readiness level, E_{CCU} are the emissions associated with the CCU product (in kgCO_2-equiv/kg CO_2 utilized), and $E_{substitute}$ are the emissions associated with the substituted product (in kgCO_2-equiv/kg CO_2 utilized).

technology cannot be excluded from Paris compatibility in 2050 based on its 2020 TRL.

Emissions criteria

1.5°C-pathways are characterized by two key numbers: in 2030, global net CO_2 emissions are halved compared with 2020 emissions, and in 2050, net CO_2 emissions are zero.[3] For CO_2 emission reductions to be in line with 1.5°C pathways, the CCU technology must halve CO_2 emissions associated with production and use of the CCU product by 2030 compared with the emissions associated with the current conventional, substituted product. In 2050, CO_2 emissions associated with the CCU product must be zero to be Paris compatible. Although emission data are often provided for a basket of greenhouse gases (GHGs) instead of CO_2 only, CCU is focused on reducing CO_2 emissions, which is by far the most prominent contributor to total GHG emissions, directly as well as indirectly.[99,100]

Maturity of CCU technologies

The TRL scale has its origin at the American National Aeronautics and Space Administration in the 1970s and became more widely known when the US Department of Defense started using it to improve its technology R&D outcomes.[101] It was first comprehensively described by Mankins,[95] and has since received recognition in policy-making, industry, and academia.[102] For example, in 2010, the European Commission advised projects that received European Union (EU) funding to use TRLs for identification of technological maturity.[103] Its use has also been recommended specifically for CCU techno-economic assessments.[24,104]

The European Commission defines TRLs in a generalized way to allow comparability of technologies in different fields, including energy and climate technologies (Table 1).[105] A downside of a generalized scale is the general description for each level, including criteria that may not be applicable or not specific enough to achieve unambiguous TRL ratings.[102,106] Adaptations to the generalized scale were developed to better rate technologies in a certain field; for example, in the chemical industry[102] and even for CCU technologies[9] (Table 1). Despite the differences between descriptions of TRLs, the commonality is that TRL 6 represents a break from the foregoing TRLs by requiring an operational system at a relevant scale (pilot plant) to have been developed.

To assess whether a CCU technology fulfills the Paris compatibility maturity criteria, its TRL in 2020 must be determined from direct TRL specifications or by applying the milestones as defined by Buchner et al.[102] (Table 1) to descriptions of the technology's state of development. Table 1 includes examples of CCU technologies for each TRL, and Note S2 provides a comprehensive overview of TRL ratings of CCU technologies. Some studies report ranges instead of a single TRL for a CCU technology. This can be done for three reasons: (1) the technology is a "composed" technology made up of multiple processes, each with its individual TRL. Hence, the range of these individual TRLs is reported; (2) more than one technology is in development to produce a CO_2-based product, and the full range is reported for this product instead of for each production process separately; or (3) there is uncertainty because of a lack of data. In this paper, we deal with these ranges in TRL in the following way: in the case of a composed technology (reason 1), the lowest TRL is counted as the overall TRL because it is the weakest link in the chain in the process to commercialization.[102] We specify a TRL for each CCU production process rather than only per CCU product (reason 2). Uncertainty in the TRL is resolved by comparing reported process descriptions (e.g., proof of concept, bench-scale process, pilot plant) with the descriptions of Buchner et al.[102] to find the best match (reason 3), as explained in Note S2.

Emissions of CCU technologies

To assess the Paris compatibility of CCU technologies, we determined the GHG emission intensity of CCU products (kgCO_2-equiv/kg CO_2 utilized) and of substituted conventional products (assuming 1:1 replacement) based on a literature search of life

Table 1. Technology readiness levels for CCU technologies

TRL	Description by European Commission[105]	Description by Chauvy et al.[9] for CCU evaluation	Description by Buchner et al.[102] for the chemical industry	CCU examples
1	basic principles observed	published research that identifies the principles that underlie the technology	idea; ppportunities identified, basic research translated into possible applications	methanol via photocatalytic conversion[9,84]
2	technology concept formulated	publications or other references that outline the application being considered and that provide analysis to support the concept; the step up from TRL 1 to TRL 2 moves the ideas from pure to applied research; amajor part of the work is analytical or paper studies; experimental work is designed to corroborate the basic scientific observations made during TRL 1 work	concept; technology concept and/or application formulated, patent research conducted	ethanol[12] and methanol[107] via electrochemical reduction[108]
3	experimental proof of concept	active R&D has been initiated; at TRL 3, the work has moved beyond the publication phase to experimental work	proof of concept; applied laboratory research started, functional principle/reaction (mechanism) proven, predicted reaction observed (qualitatively)	ethylene via electrochemical reduction,[109] DME via syngas[12,110]
4	technology validated in lab	TRL4–6 represent the bridge from scientific research to engineering; TRL 4 is the first step in determining whether the individual components will work together as a system	preliminary process development; concept validated in laboratory environment, scale-up preparation started, shortcut process models found	PCC and PMC[63,66,97,111]
5	technology validated in relevant environment	basic technological components are integrated so that the system configuration is similar to the final application in almost all respects	detailed process development; process models found, property data analyzed, simulation of process and pilot plant using bench scale information	formic acid via electrochemical reduction in an aqueous environment,[63,66,97,111] sodium bicarbonate using flue gas directly[63,66,97,111]
6	technology demonstrated in relevant environment	represents a major step up in a technology's demonstrated readiness; TRL 6 begins true engineering development of the technology as an operational system	pilot trials; pilot plant constructed and operated with low-rate production, products approved in final application, detailed process models found	Fischer-Tropsch fuels,[97,112] urea from steel gases[113,114]
7	system prototype demonstration in an operational environment	TRL 7 is a significant step beyond TRL 6, requiring an actual system prototype	demonstration and full-scale engineering; parameter and performance of pilot plant optimized, (optional) demo plant constructed and operating, equipment speciation, including components that are type conferrable to full-scale production	CO and syngas via the reverse water gas shift reaction,[12,112] methane and methanol via hydrogenation of CO_2[9,115,116]

(*Continued on next page*)

Table 1. *Continued*

TRL	Description by European Commission[105]	Description by Chauvy et al.[9] for CCU evaluation	Description by Buchner et al.[102] for the chemical industry	CCU examples
8	system complete and qualified	represents the end of true system development; the technology has been proven to work in its final form and under the expected conditions	commissioning; products and processes integrated in organizational structure (hardware and software), full-scale plant constructed	Polyols,[9,117,118] construction materials from carbonated steel slag[51,119,120]
9	actual system proven in an operational environment	technology is in its final form and operates under the full range of operating mission conditions	production; full-scale plant audited (site acceptance test), turn-key plant, production operated over full range of expected conditions in industrial scale and environment, performance guarantee enforceable	CO_2 enrichment in agricultural greenhouses,[121] CO_2 EOR[55,122]

Shown are descriptions of technology readiness levels (TRL) and examples of CCU technologies for each TRL. See Note S2 for an overview of TRLs for all studied CCU technologies.

cycle assessment (LCA) studies on CCU. In this way, 1,041 studies were identified (see experimental procedures). After screening these papers for relevance and excluding reviews and meta-analyses without original data, 106 studies remained (Note S3). We then selected the most recent study with the most complete life cycle inventory for each available combination of CCU technology and type of CO_2 source (fossil, biogenic, or atmospheric). Not all combinations could be found in the literature. This resulted in 30 studies (1 on direct use, 6 on EHR, 8 on mineral carbonates and construction materials, and 15 on fuels and chemicals), together covering 44 unique CCU technologies and resulting in 74 CCU technology-CO_2 source combinations (see Note S3, which also provides an overview of the specific CO_2 sources and CO_2 capture processes covered). These studies were harmonized regarding (1) functional unit, (2) system boundaries, (3) electricity mix, (4) hydrogen production, (5) dealing with multifunctionality, and (6) accounting of temporary carbon storage, as detailed below. For the substituted products, we followed the choices made in the original papers, assuming the CCU product replaces a product with identical molecular structure or, if this does not exist, a product with the same characteristics and function.[24]

Functional unit

The functional unit of CCU products can be end product-based (e.g., kg or MJ product) or input-based (e.g., kg CO_2 utilized). Most CCU LCA studies use an end product-based functional unit, which allows estimating absolute emission reductions when switching to a CCU product. However, to be able to compare different CCU technologies, the functional unit should be the same and, thus, input based.[12,123] We therefore use 1 kg of CO_2 utilized as functional unit in this paper. It is possible to go from an end product-based functional unit to the functional unit of 1 kg of CO_2 utilized by determining the product's GHG intensity in kgCO_2-equiv/kg product and multiplying it by kg product/kg CO_2 utilized.

System boundaries

Most LCA studies quantify "cradle-to-factory-gate" emissions, assuming that emissions beyond the gate are the same for CCU product and substitute. In our analysis, we determine the absolute CO_2 emissions of a CCU product and the percentual CO_2 emission reduction of a CCU product relative to its substitute. This requires a wider "cradle-to-grave" approach that includes end-of-life emissions for both products. In practice, however, data on transport and use-phase emissions often lack in the reviewed LCA studies, except for re-emission of CO_2 upon combustion or dissolution. So where cradle-to-grave data were not available, we used cradle-to-factory-gate emissions and added end-of-life emissions, as proposed by Fernández-Dacosta et al.,[23] assuming that use-phase emissions were negligible. Combustion and dissolution were then included in the end-of-life emissions. We did not harmonize production infrastructure and transport emissions and follow the choices in the respective LCAs because these emissions contribute negligibly to the total GHG intensity.[124–126] For the substituted products, emissions were also determined based on a cradle-to-grave basis.

Electricity mix

To improve inter-comparability of CCU products and take into account the expected decarbonization of the electricity sector,[127] we harmonized the GHG intensity of electricity used in all foreground processes of the studies considered. This includes electricity use in the capture and conversion process and hydrogen production via electrolysis. The harmonized GHG intensity of electricity was set to be in line with emissions pathways limiting global warming to 1.5°C:[3] 0.17 kgCO_2/kWh for 2030 and 0 kgCO_2/kWh for 2050. For zero-emissions electricity, it is assumed that the (limited) emissions of renewable electricity production are compensated by CDR (e.g., from bioenergy with CCS).

Hydrogen

Hydrogen is an important feedstock in the production of fuels and chemicals from CO_2 and is produced using fossil fuels with or without CCS or water electrolysis. Because we assume hydrogen production to decarbonize in Paris-compatible pathways, we harmonize all studies toward electrolysis-based hydrogen following their reported electricity requirements,

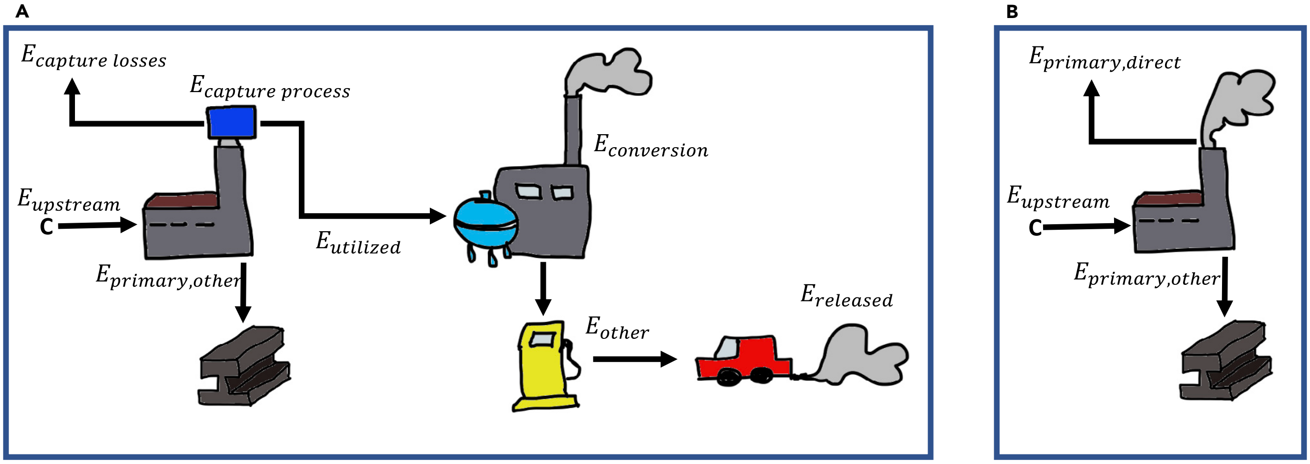


Figure 3. Determining the GHG intensity of CCU products through the substitution approach

(A and B) In the system expansion via substitution approach, the GHG intensity of the CCU product E_{CCU} is determined by the difference between the emissions of (A) the multifunctional system and (B) the marginal production system of the primary product.

unless H_2 is co-produced in the conversion process. For an electricity requirement of 52 kWh/kg H_2 and using the aforementioned electricity carbon intensities for 2030 and 2050, electrolysis-based hydrogen results in 6.85 kgCO_2-equiv/kg H_2 in 2030 and 0 kgCO_2-equiv/kg H_2 in 2050. We use the substitution approach to deal with the co-produced O_2 (experimental procedures).

Multifunctionality

The CCU production chain is typically inherently multifunctional; an industrial facility or power plant produces a primary product (e.g., steel or electricity) as well as CO_2, which is used as feedstock in the secondary CCU process. GHG emissions of this system must be divided between the primary product and the secondary CCU product. DAC-based CCU systems are not multifunctional because there is no primary product. We apply system expansion via a substitution approach to solve this multifunctionality problem following the LCA standard ISO 14044 and LCA guidelines for CCU[104] (Figure 3).

System expansion via substitution assumes that a primary production plant with CCU directly substitutes an identical plant without CCU to ultimately determine the emissions that can be associated with CCU itself. It is formalized as shown in Equation 1 (for a full derivation, see experimental procedures):

$$E_{CCU} = -E_{utilized} + E_{capture\ process} + E_{conversion} + E_{other} + E_{released} \qquad \text{(Equation 1)}$$

where E stands for GHG emissions (in kgCO_2-equiv/kg CO_2 utilized), specifically

E_{CCU} GHG emissions of the CCU product;

$E_{utilized}$ CO_2 utilized in production of the CCU product;

$E_{capture\ process}$ GHG emissions associated with capturing CO_2 and separating CO_2 from the bulk gas stream;

$E_{conversion}$ GHG emissions of the conversion process of CO_2 into the end product;

E_{other} other (remaining) GHG emissions associated with the CCU product, such as GHG emissions at end of life other than release of utilized CO_2 (for example, from fossil feedstock added in the conversion process); and

$E_{released}$ GHG emissions of utilized CO_2 to the atmosphere at the end of life of the CCU product.

In most cases, $E_{utilized}$ and $E_{released}$ cancel each other out. However, $E_{released}$ is less than $E_{utilized}$ when some of the utilized CO_2 is lost in the conversion process (i.e., included in $E_{conversion}$) or when the CO_2 is stored permanently; then $E_{released}$ is zero. A negative value for E_{CCU} can occur when CO_2 is stored permanently and the amount of CO_2 utilized is greater than the combined emissions of the CCU production, use, and end-of-life processes.

The interpretation of a negative value for E_{CCU} depends on the source of CO_2. In the case of CO_2 with a fossil origin, a negative E_{CCU} means that the total GHG emissions of the system with CCU are reduced in comparison with the system without CCU and that this emission reduction can be attributed to CCU. It does not mean that CCU is CDR because CO_2 is not removed from the atmosphere.[27,128] In the case of CO_2 that is removed from the atmosphere, directly (through DAC) or indirectly (through capture of CO_2 with biogenic origin), a negative value for E_{CCU} does mean that CDR takes place. For biogenic CO_2, CDR would also require sustainable sourcing of the biomass used.

The substitution approach is based on the assumption that a point source without capture is 1:1 substituted by the same plant with capture. In the near future (i.e., 2030), this assumption holds because many plants without capture can still be substituted. In the long run, however, the reference system is expected to transform toward a zero-emissions economy. Therefore, in a Paris-compatible 2050 system, we can no longer assume that there are unabated fossil point sources to substitute and associated emissions to avoid. Because emissions are no longer unabated, considering CCU as an option to abate fossil CO_2 emissions means that the responsibility for not emitting shifts to CCU. In terms of calculating the GHG intensity of CCU for a fossil CO_2 source in 2050, this

Table 2. GWP factors ($GWP_{storage}$-100) for temporary storage of CO_2 in CCU products using a 100-year time horizon

Lifetime	0–6 months	0.5–1 year	1 year	5 years	10 years	25 years	50 years	100 + years
$GWP_{storage}$ -100	1	0.99	0.98	0.92	0.85	0.67	0.42	0

means that all emissions from the stack onward are assigned to the CCU product:

$$E_{CCU(fossil\ in\ 2050)} = E_{capture\ losses} + E_{capture\ process} + E_{conversion} + E_{other} + E_{released} \quad \text{(Equation 2)}$$

where E stands for GHG emissions (in $kgCO_2$-equiv/kg CO_2 utilized), specifically

$E_{CCU(fossil\ in\ 2050)}$ GHG emissions of the CCU product (for 2050 in a Paris-compatible scenario where substitution of unabated fossil CO_2 point sources cannot be assumed) and
$E_{capture\ losses}$ CO_2 not captured in the capture process, assuming a capture rate of 95%.[32,81,122]

In the results, we report E_{CCU}, but when this applies to fossil CO_2 sources in 2050, Equation 2 is used for the calculation. For CCU processes based on biogenic or atmospheric CO_2 in 2050, Equation 1 still applies because availability of these sources in 2050 is in line with a Paris-compatible scenario.

Benefits of temporary storage of CO_2

The importance of a product's lifetime for CCU's contribution to climate change mitigation has been stressed in the literature.[26,129] Temporary storage of CO_2 by lowering atmospheric CO_2 concentrations for the duration of the product's lifetime decreases radiative forcing over this period of time.[130] We calculated the GHG intensity of CCU products without (base case) and with the benefit of temporary carbon storage (sensitivity analysis) using a time horizon of 100 years. Global warming potentials are used for this calculation, adjusted for temporary carbon storage (global warming potential $[GWP]_{storage}$). Global warming potentials describe the contribution to radiative forcing of delayed CO_2 emissions compared with direct CO_2 emissions over 100 years. We derived the $GWP_{storage}$-100 factors using the method described by Guest et al.,[131] which is based on Clift and Brandao,[132] combined with the more recent atmospheric CO_2 decay curves described by Joos et al.[133] The factors depend on the lifetime of the CCU product, as shown in Table 2; see the experimental procedures for an overview of CCU lifetimes. To include the storage effect, $E_{released}$ in Equations 1 and 2 is multiplied by $GWP_{storage}$-100. In this way, only captured CO_2 is affected by this temporary storage benefit, not fossil carbon added in some CCU routes as additional feedstock.

FULFILMENT OF PARIS COMPATIBILITY CRITERIA

Paris compatibility in 2030

Figure 4 shows the relative GHG intensity of CCU technologies in 2030 plotted against their 2020 TRL. As explained above, a negative GHG intensity ratio means that an emissions reduction takes place for fossil CO_2 and CDR when the source of CO_2 is biogenic or atmospheric. CCU technologies fulfilling the 2030 emissions criterion can be characterized by one or more of the following attributes: (1) preventing (high) capture emissions, (2) preventing (high) conversion emissions, (3) preventing re-emission of CO_2, and (4) replacing an emission-intensive process.

Paris-compatible CCU technologies in 2030 are CO_2 enrichment in the horticulture industry[50] with CO_2 from a co-located biogas to biomethane upgrading unit, which is considered a by-product (i.e., is characterized by attribute 1) and can be used directly (attribute 2). In the carbonation process of steel slag to produce construction blocks, flue gas is used directly,[58] omitting the capture step (1). CO_2 is stored permanently through an exothermic carbonation reaction[134] (2 and 3). In EOR, CO_2 is used directly (2) and can be Paris compatible for several gas separation methods[135–137] as long as no more than 2 barrels of oil (bbl) are recovered per ton CO_2 injected. This allows a favorable balance of the CO_2 released upon combustion of the recovered oil and the CO_2 stored permanently (3). Urea production from basic oxygen furnace gas (BOFG) uses waste heat from the steel plant for capture and conversion processes (1 and 2) and replaces electricity production from BOFG.[113] Hence, quantification of the GHG intensity of urea includes replacement of electricity produced from BOFG by the 1.5°C-compatible electricity mix (4). Last, close to halving emissions is CO via rWGS with relatively low conversion emissions (2).[12]

Low TRL CCU technologies that only fulfill the emissions criterion can be characterized by the same four attributes. The sodium bicarbonate process uses flue gas directly, avoiding the capture step (attribute 1).[138] Indirect mineral carbonation reactions producing PMC or PPC may have low emissions for high efficiency of the alkaline absorption process, depending on process development. They store CO_2 permanently (3).[63] Formic acid via hydrogenation reduces emissions sufficiently compared with the emission-intensive substitute (4).[12] Electrochemical production of formic acid via supercritical CO_2[139] and methane and methanol via photocatalytic conversion[84] have low conversion emissions (2) because they do not require electricity-intensive hydrogen.

High TRL CCU technologies not fulfilling the emissions criterion can also be characterized by the aforementioned attributes. However, here, these are the reasons for not fulfilling the emissions criterion. CO_2 curing of concrete[72,140] does not sufficiently reduce emissions because of the emissions associated with cement production (2). Fuels and chemicals based on thermochemical conversion typically have high conversion emissions because of electricity-intensive hydrogen production (2). Finally, EOR with a recovery ratio over 2 bbl per ton of CO_2 injected does not store a sufficient volume of CO_2 compared with combustion emissions[141,142] (3).

Benefits of temporary CO_2 storage

Some products store CO_2 for several decades; e.g., polyethylene and polypropylene. Adding the temporary storage benefit based on a lifetime of 50 years leads to a reduced GHG intensity of roughly 25%. This, however, is not enough to lead to a difference in Paris compatibility. For long-lived polyols, the reduction

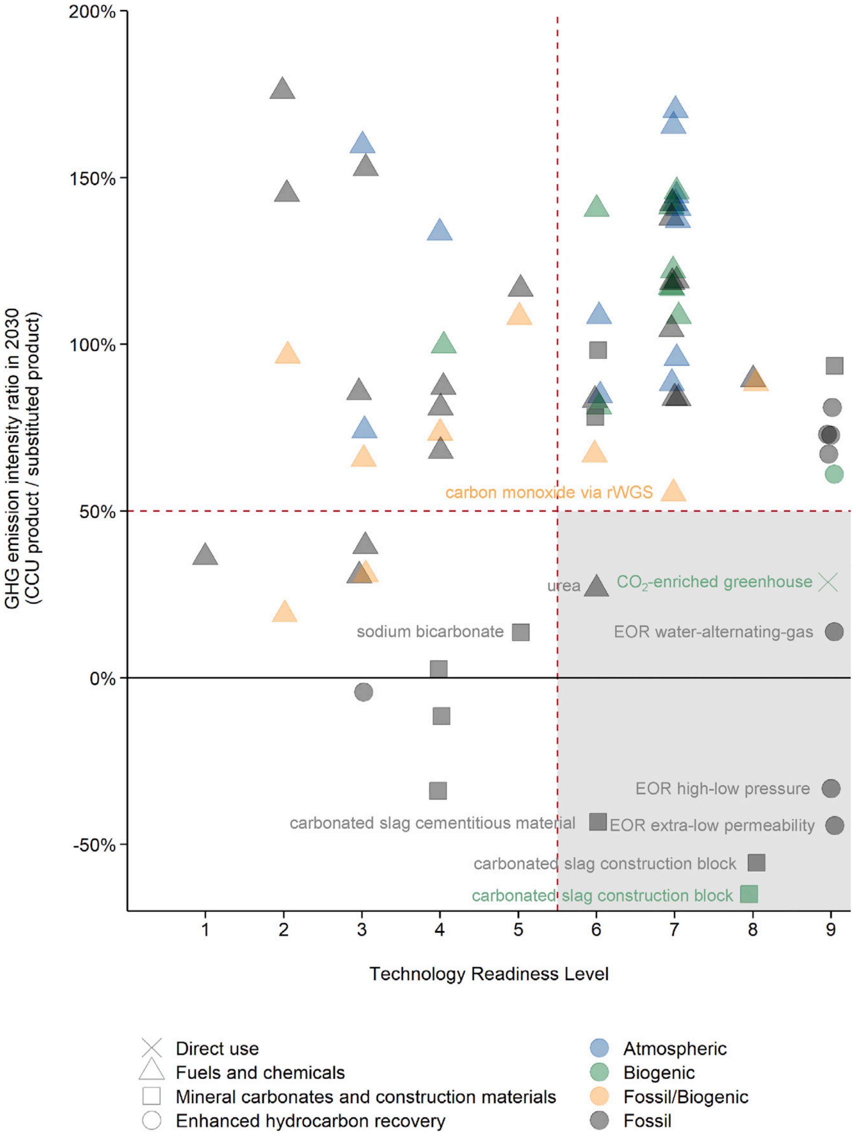


Figure 4. Paris compatibility of CCU technologies in 2030

Technological maturity of CCU technologies in terms of their 2020 technology readiness level and GHG emission intensity ratio of the CCU product in 2030 compared with its substitute. The four shapes represent the four CCU categories, and the colors are used to differentiate between the different types of CO_2 sources. For a CCU technology to be Paris compatible in 2030, it must currently have a TRL of 6 or higher and reduce emissions by at least 50% compared with its substitute. Technologies in the lower right gray area fulfill these Paris compatibility criteria. The full dataset, including outliers not shown in this figure, and source data for this figure are provided in Data S1.

is only a few percent because a large share of the embodied carbon is from fossil feedstock, to which the storage factor does not apply. The lifetime of the other chemicals and fuels is assumed to be 6 months, where the $GWP_{storage}$-100 factor of 0.99 does not affect Paris compatibility. For EHR, construction materials, and most mineral carbonates, permanent storage was already assumed in the base case.

Paris compatibility in 2050

Figure 5 shows the GHG intensity of CCU products in 2050 (E_{CCU}, in $kgCO_2$-equiv/kg CO_2 utilized) (Equations 1 and 2). In 2050, low low-TRL technologies may be Paris compatible. The characteristics of CCU technologies fulfilling the 2050 emissions criterion are simplified to (1) preventing re-emission of CO_2, (2) only using zero-emissions energy, and (3) utilizing CO_2 recently removed from the atmosphere (biogenic/atmospheric).

Construction blocks from carbonation of steel slag using a purified stream of biogenic CO_2[134] are Paris compatible, resulting in CDR because the CO_2 is stored permanently (i.e., criteria 1 and 3 are fulfilled). In addition, steel slag carbonation using fossil flue gas directly to produce cementitious material[58] achieves close to zero emissions because no capture process is required, the carbonation process is exothermic, and the CO_2 is stored permanently (fulfilling criteria 1 and 2).

Because CO_2 is not stored permanently in fuels or chemicals, these products can only be strictly Paris compatible when the CO_2 is of biogenic or atmospheric origin and zero emissions are associated with the capture and conversion processes (fulfilling criteria 2 and 3). This situation is approached when heat integration is applied, electric heat is used, or simply only electricity is required. Processes approaching zero emissions are methane production from H_2 and biogenic CO_2;[143] DMM via condensation with formaldehyde or via direct synthesis, both for a biogenic source;[144] and Fischer-Tropsch fuels from atmospheric CO_2, based on a fully electric process for capture and conversion.[145] The assumption of zero-emission electricity is crucial because the GHG intensity of these options using the 2030 electricity mix is between 1.21 and 1.39 $kgCO_2$-equiv/kg CO_2 utilized (see Figure S1 for the absolute GHG intensity of CCU products in 2030).

Benefits of temporary CO_2 storage

Inclusion of the benefit of temporary storage of 6 months is enough to obtain a negative absolute GHG intensity for DMM via condensation with formaldehyde or via direct synthesis, both for a biogenic source.[144]

DISCUSSION AND RESEARCH OUTLOOK

Maturity

We considered technological maturity in terms of TRLs of individual technologies because data are available for individual technologies. Others have suggested "system readiness levels"[146,147] to reflect the technology's need to embed in a

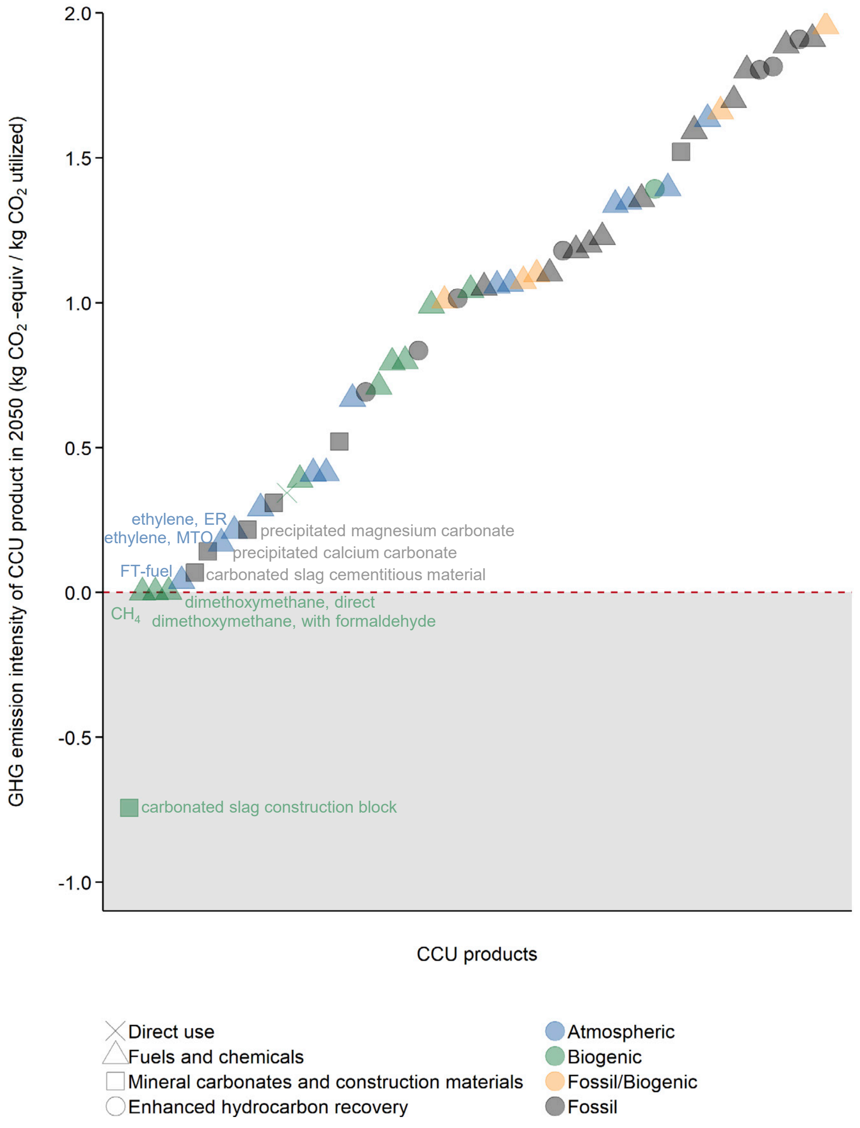


Figure 5. Paris compatibility of CCU technologies in 2050

GHG intensity of CCU products in 2050 (kgCO_2-equiv/kg CO_2 utilized). The four shapes represent the four CCU categories, and the colors are used to differentiate between the different types of CO_2 sources. The TRL is not indicated because 2020 technological maturity does not preclude technologies from fulfilling the Paris compatibility criteria by 2050. Technologies in the lower gray area fulfill these Paris compatibility criteria. The full dataset, including outliers not shown in this figure, and source data for this figure are provided in Data S1.

certain system. Because of this embedding, assessment of technological readiness at a systemic level could lead to lower levels of maturity. This means that Paris compatibility of CCU could be overestimated. On the other hand, concerted, transdisciplinary action on innovation could speed up technological maturing of promising technologies. For example, this could apply to sodium bicarbonate, which fulfills the emission reduction criterion but is in TRL 5. As a next step, "technological innovation system" analyses[148,149] can be performed to provide information on what actors can do to advance such technologies.

Emissions

Our harmonization allowed a comparison of CCU technologies, but some uncertainty in future emissions remains because (1) LCAs of low-TRL technologies may be based on idealized modeling data, underestimating the environmental effects because of an information bias;[108] (2) the environmental effect of technologies is expected to decrease as they mature because of energy efficiency improvements and/or development of catalysts for more efficient conversion;[139,127] (3) some CCU processes could be adapted so that more CO_2 is utilized per product produced (for example, for EOR);[150] and (4) if product lifetimes could be extended, then they could, based on the temporary storage benefit, decrease their contribution to climate change. Future LCA studies could focus on CCU technologies that are developing quickly and for which no LCAs are available or for which only LCAs based on low-TRL data are available or on how CCU processes can be adapted to reduce overall emissions.

Even when the emissions criteria are not fulfilled, it is possible that there are no future alternative technological routes that provide the same product at a lower GHG intensity. Although a CCU technology is not strictly Paris compatible in such cases, CCU could still play a role in the overall energy system transition. In addition, if other environmental effects besides climate are included in the equation, then CCU options may, in certain cases, be viewed more positively than remaining alternatives to our fossil fuel-based production system. For instance, CCU products do not have the land-use effects of crop-based biomaterials (including biodiversity).[151] Further research could include alternative routes for the assessment and define emission reduction criteria per product group. The assessment of emission reduction could be extended beyond the technology level to the sector or even the system level. Determining the global climate change mitigation potential of CCU at scale requires that interactions with other mitigation options as well as broader changes in the economy are considered, which may be achieved using integrated assessment models. Further research could focus on including CCU in these models and determining how much each CCU technology could contribute at scale to CO_2 emission reductions within and across different sectors.

Multifunctionality

The multifunctionality problem is one that needs to be solved carefully and transparently. Otherwise, it is possible to end up with statements that cannot be simultaneously true, such as the following: CCU can make traditionally CO_2-emitting industries (e.g., fossil fuel electricity generation, chemicals, cement) carbon-neutral[10,75] while producing carbon-neutral CCU products.[78,152] The reality is that the eventual emission of CO_2 must be accounted for by one of the processes. And, as shown above, when moving to a future without unabated fossil emissions, this emission has to be allocated to CCU. The right year of this allocation shift is unclear and may depend on the GHG reduction targets of a specific country, sector, or company. Although we consider the year 2050, based on Paris-compatible emission pathways at a global level, other countries or firms may have reasons to select a different point in time where fossil points sources can no longer go unabated.

Decarbonization of electricity

The carbon intensity of electricity of 0.17 $kgCO_2$/kWh that we used for 2030 is higher than the carbon intensity in many of the CCU LCA studies; for example, when based on only renewables. This means that, in these cases, we found a higher 2030 GHG intensity of CCU products compared with the original studies. Still, our 2030 carbon intensity of electricity may be an underestimation because it was based on emission pathways that only include use-phase emissions, not upstream emissions. Our 2030 results, however, are not sensitive to this increase in carbon intensity; doubling the carbon intensity (0.34 $kgCO_2$/kWh) did not affect Paris compatibility of technologies in 2030. However, storing renewable electricity in a fuel or chemical results in a loss of primary energy[153] and has a low energy return on energy invested.[21] Hence, use of low-carbon electricity for mitigation options other than CCU has been shown to achieve higher emission reductions per kWh; e.g., for e-mobility and heat pumps[10] or DACCS.[154] Alternatively, CCU technologies have been proposed to use excess renewable electricity and aid the energy transition by balancing the peaks of renewable power production.[155,156] This suggestion is sobered by its limited economic attractiveness as a result of the low-capacity factors,[81] but further research on the systemic effects of electrification of the CCU process would be of value.

Lock-in

The only CCU technologies that are 2030 and 2050 Paris compatible are construction materials based on carbonation of steel slag, either using fossil flue gas directly or using CO_2 captured from a biogenic source. Arguably, implementing CCU options that only reduce emissions sufficiently for 2030 but not for 2050 could lead to a carbon lock-in, which would occur when large investments are made in 2030, complicating phase-out and discouraging necessary transitions later on.[157,158] Hence, assuming that a CCU production plant built in 2030 is still operational in 2050 would mean that the 2050 emissions criterion should be added to assess 2030 Paris compatibility. In that case, with the exception of some construction materials, none of the 2030-compatible options fulfill the 2050 emissions criterion. A lock-in in these CCU processes could be prevented by implementing an exit strategy for industries relying on CCU processes that are only 2030 compatible. CCU technologies that are currently used or considered and that are not 2030 compatible could lead to a lock-in within years. We recommend that these are critically evaluated regarding their potential for a short-term exit strategy.

Sources of CO_2 and capture processes

We found that energy requirements for CO_2 capture and their associated emissions differ between specific CO_2 sources and capture processes, in line with Müller et al.[128] and von der Assen et al.[159] Whether a CCU process is Paris compatible can thus depend on the capture process itself (e.g., a DAC process based on natural gas[143] rather than renewable electricity[145] may render the entire CCU product non-Paris compatible). Our research has focused on available CO_2 source-CCU technology combinations in the LCA literature, and further research could harmonize CO_2 sources and capture processes to provide additional insights into remaining CO_2 source-CCU technology combinations. This would allow including more innovative CO_2 capture routes and identifying more environmentally optimal combinations of capture processes and conversion processes for different CO_2 sources.

CCU versus CCS

Cuéllar-Franca and Azapagic[16] showed that, per ton of CO_2 captured, CCS results in lower emissions than CCU. Although capture emissions are the same, emissions for compression and injection of CO_2 in geological formations are lower than most CCU technologies' emissions from conversion and ultimate release of CO_2. Based on this, only CCU technologies with low conversion emissions and permanent storage could compete with CCS. Although the avoided emissions of the product that CCU replaces are not taken into account,[16] including these could still lead to the same conclusion, as shown for methanol.[19] To deal with residual flows containing CO_2 from essential industries as long as they exist, further research could focus on systematic comparison of CCS and CCU technologies in light of their product-specific substitutes[123] and TRLs.

CONCLUSIONS AND POLICY RECOMMENDATIONS

By combining the literature on technological maturity of CCU with the global emission reduction requirements consistent with the Paris Agreement, we conclude that only very few CCU options would be Paris compatible. Moreover, some CCU options only meet the criteria in the short run and could lead to a lock-in toward 2050. We find that, for a CCU technology to be Paris compatible in 2030, it has to have low GHG emissions from CO_2 capture and conversion, replace a GHG-intensive substitute, and (in most cases) lead to permanent storage. For 2050, the criteria become more stringent, and Paris compatibility typically depends on a combination of no capture and conversion emissions (e.g., by using zero-emission electricity or waste heat), permanent storage, and use of biogenic or atmospheric CO_2 sources. Achieving CDR via CCU technologies is only possible when biogenic or atmospheric CO_2 is used and stored permanently.

Although our conclusions are robust as a result of the harmonization approach we adopted, we also showed that determining the CO_2 emission reduction associated with a CCU technology depends on a range of assumptions. When designing policies for implementation of CCU technologies, clear guidelines for these assumptions must be in place. We recommend using a future electricity mix and considering the full life cycle, including possible re-emission of CO_2. To clarify the emission reduction potential of CCU, it should also be made very clear against what CCU is compared. We distinguish three benchmarks: (1) emission reduction of CCU compared with emitting CO_2 at the point source, (2) emission reduction of CCU compared with the substituted original product, and (3) emission reduction of CCU compared with other alternative technological routes replacing the original product. This study's assessment of the Paris compatibility of CCU was based on the first two benchmarks, but the third could shed light on the potential trade-offs between CCU and other options for replacing fossil-based production, which may change the verdict on CO_2 source-CCU options close to the Paris compatibility emission frontier.

Our findings have implications for climate and innovation policy. Research funding may currently be allocated to CCU options that are unlikely to be mature in time for the Paris temperature limits or that are not able to sufficiently reduce emissions. To be in line with the objectives of the Paris Agreement, such funding would have to be redirected to mitigation options consistent with the Paris Agreement and unlikely to result in carbon lock-in. CCU encompasses a wide range of technologies with different conversion processes, product lifetimes, and substituted products. Therefore, we recommend that decision-makers recognize this diversity in CCU, base their decisions on the share of emissions an individual CCU technology can reduce, and whether (close to) zero emissions or CDR can be achieved rather than treating CCU as a homogeneous technology. In addition, the technology's current level of maturity and when it is expected to be ready for diffusion should be considered. Such a focus could facilitate creation of strategies that accelerate development of technologies with low TRLs that may lead to (close to) zero emissions or CDR.

EXPERIMENTAL PROCEDURES

Resource availability

Lead contact

For questions related to this article, please contact the lead contact, Kiane de Kleijne (Kiane.deKleijne@ru.nl).

Materials availability

Not applicable to this study.

Data and code availability

For identification of a Paris-compatible mitigation pathway, we used data available from the Integrated Assessment Modeling Consortium (IAMC) 1.5°C Scenario Explorer and Data.[1] The full dataset (Data S1) with harmonized emissions of CCU technologies for 2030 and 2050 and source data for Figures 4, 5, and S1 have been deposited at Data Archiving and Networked Services (DANS) under https://doi.org/10.17026/dans-28h-n6zj.

Identification of a Paris-compatible mitigation pathway

We use all "no-overshoot" and "low-overshoot" 1.5°C pathways as defined in the IAMC 1.5°C Scenario Explorer and Data[1] (version 1.1). We take the mean of the global annual emissions in these pathways from 2010–2060 to arrive at the Paris-compatible mitigation pathway in Figure 2. "Half of 2020 emissions" in Figure 2 is found by multiplying the mean 2020 emissions in these pathways by 0.5.

Selection of LCA studies to determine the GHG intensity of CCU products

To assess Paris compatibility of CCU technologies, we determined the GHG emission intensity of CCU products (kgCO_2-equiv/kg CO_2 utilized) and of substituted conventional products (assuming 1:1 replacement) based on a literature search of life cycle assessment studies on CCU. The Web of Science search string used was as follows:

TOPIC = CCU* OR "carbon capture and utili$ation" OR "carbon capture utili$ation" OR "carbon capture and use" OR "carbon capture and re-use" OR "carbon dioxide capture and utili$ation" OR "carbon dioxide capture utili$ation" OR "carbon dioxide capture and use" OR "carbon dioxide capture and re-use" OR "carbon dioxide utili$ation" OR "carbon dioxide use" OR "CO2 use" OR "CO2 utili$ation" OR "CO2 re-use" OR "CO2-enhanced" OR "CO2-based" OR "CO2-activated" OR "CO2 capture and utili$ation" OR "CO2 capture and use" OR "CO2 capture and re-use" OR "produc* from CO2" OR "produc* from carbon dioxide" OR "carbonat* curing" OR "CO2 curing" OR "carbonat* aggregate*"OR (CO2 calcium carbonat*) OR ("carbon dioxide" "calcium carbonat*") OR (CO2 micro-alga*) OR (CO2 micro-alga*) OR ("carbon dioxide" micro-alga*) OR ("carbon dioxide" micro-alga*) OR (CO2 carbon nanotube*) OR ("carbon dioxide" carbon nanotube*) OR (CO2 carbon nanofib*) OR ("carbon dioxide" carbon nanofib*) OR (CO2 enhanced oil recovery) OR ("carbon dioxide" enhanced oil recovery) OR (CO2 Fischer-Tropsch synthesis) OR ("carbon dioxide" Fischer-Tropsch synthesis)

AND TOPIC = LCA OR life cycle OR life cycle OR "carbon footprint*" OR "climate footprint*" OR "environment* impact*" OR "climat* impact*" OR "GHG balance" OR "greenhouse gas balance" OR "carbon balance" OR "global warming potential" OR "global warming impact*" OR "global warming footprint*" OR "carbon-negative" OR "below zero".

On November 30, 2020, this search string led to 1,041 results. After screening these papers for relevance (1) fulfilling our definition of CCU, (2) determining the GWP and not only LCA endpoints, and (3) including emissions from energy use and excluding conference proceedings and reviews and meta-analyses without original data, 106 studies remained (Note S3). Microalgae biofuel studies were excluded based on the agreement in the most recent reviews, meta-analyses, and harmonized LCA studies that emissions were not reduced consistently compared with fossil fuels[160–162] or, on average, even doubled.[163] For each individual combination of CCU technology and type of CO_2 source (fossil, biogenic, or atmospheric), the most recent study with the most complete life cycle inventory was selected. This resulted in 30 studies (1 on direct use, 15 on fuels and chemicals, 8 on mineral carbonates and construction materials, and 6 on EHR), together covering 44 unique CCU technologies and resulting in 74 combinations based on multiple CO_2 sources. If the same CCU process was presented with small changes in setup or composition, then the process with the lowest overall GHG emissions was used in this study. If several CO_2 sources of the same "type" (fossil, biogenic, or atmospheric) were given in the LCA, the source most likely to be deploying CO_2 capture in a 1.5°C pathway and to still exist in 2030 and 2050 was preferred (i.e., an industrial facility over a fossil fuel-based power plant and a natural gas power plant over a coal-fired power plant). For processes for which a pure biogenic source was not available but a source representing a mix of biogenic and fossil sources was available, the mix was included in addition to the fossil and atmospheric sources. When the emissions for the substitute were not included in the study, the emissions reported in another LCA for the same product were used and are referred to in Note S4.

Derivation of GHG emissions of CCU products in 2030

We apply a system expansion via substitution approach to solve the multifunctionality problem following the LCA standard ISO 14044 and LCA guidelines for CCU.[104] This method assumes that a primary production plant with CCU directly substitutes an identical plant without CCU to ultimately determine the emissions that can be associated with CCU itself. The GHG emissions of these two systems can be determined as shown in Equations 3 and 4:

$$E_{plant\ w/o\ CCU} = E_{upstream} + E_{primary,\ direct} + E_{primary,\ other} \qquad \text{(Equation 3)}$$

$$\begin{aligned} E_{plant\ with\ CCU} = {} & E_{upstream} + E_{primary,\ other} + E_{capture\ losses} + E_{capture\ process} \\ & + E_{conversion} + E_{other} + E_{released} \end{aligned} \qquad \text{(Equation 4)}$$

where E stands for GHG emissions (in kgCO_2-equiv/kg CO_2 utilized). All emissions are scaled to the functional unit of 1 kg of CO_2 utilized. We specifically distinguish the following GHG emissions:

$E_{upstream}$ Upstream GHG emissions associated with extraction of resources or production of the carbon feedstock feeding into the primary production process (as feedstock and/or fuel), including handling and transport to the plant

$E_{primary,\ direct}$ Direct CO_2 emissions emitted at the point source in the primary production process

$E_{primary,\ other}$ Other (remaining) GHG emissions associated with the primary production plant

$E_{capture\ losses}$ Emissions of CO_2 that are not captured in the capture process (capture is not 100% efficient)

$E_{capture\ process}$ GHG emissions associated with capturing CO_2 and separating the CO_2 from the bulk gas stream (e.g., from additional electricity use)

$E_{conversion}$ GHG emissions of the conversion process of the CO_2 into the end product

$E_{released}$ Emissions of utilized CO_2 into the atmosphere at the end of life of the CCU product

E_{other} Other (remaining) GHG emissions associated with the CCU product, such as GHG emissions at end of life other than release of utilized CO_2

The amount of CO_2 that is utilized in CCU can be determined as shown in Equation 5:

$$E_{utilized} = E_{primary,\ direct} - E_{capture\ losses} \qquad \text{(Equation 5)}$$

The GHG emissions that can be attributed to the CCU product via substitution can thus be determined via Equations 6a, 6b, 6c:

$$E_{CCU} = E_{plant\ with\ CCU} - E_{plant\ w/o\ CCU} \qquad \text{(Equation 6a)}$$

$$\begin{aligned} E_{CCU} = {} & E_{upstream} + E_{primary,\ other} + E_{capture\ losses} + E_{capture\ process} \\ & + E_{conversion} + E_{other} + E_{released} - E_{upstream} - E_{primary,\ direct} - E_{primary,\ other} \end{aligned} \qquad \text{(Equation 6b)}$$

Solving and filling in Equation 5 then results in Equation 6c:

$$E_{CCU} = -E_{utilized} + E_{capture\ process} + E_{conversion} + E_{other} + E_{released} \qquad \text{(Equation 6c)}$$

In the case of DAC, there is no multifunctionality problem, but Equation 6c can still be used to determine E_{CCU} because $E_{utilized}$ is simply the amount of CO_2 captured from the air. When the CO_2 must already be separated in the primary process, CO_2 is a by-product, and the capture emissions ($E_{capture\ process}$) are zero.

Equation 6c is used in the main text as Equation 1.

Derivation of GHG emissions of CCU products in 2050

The substitution approach is based on the assumption that a point source without capture is 1:1 substituted by the same plant with capture. In the near future (i.e., 2030) this assumption holds because many plants without capture can still be substituted. In the long run, however, the reference system is expected to transform toward a zero-emissions economy. Therefore, in a Paris-compatible 2050 system, we can no longer assume that there are unabated fossil point sources to substitute and associated emissions to avoid. Because emissions are no longer unabated, considering CCU as an option to abate fossil CO_2 emissions means that the responsibility for not emitting shifts to CCU. In terms of calculating the GHG intensity of CCU for a fossil CO_2 source in 2050, this means that all emissions from the stack onward are assigned to the CCU product (Equation 7):

$$E_{CCU(fossil\ in\ 2050)} = E_{capture\ losses} + E_{capture\ process} + E_{conversion} + E_{other} + E_{released} \qquad \text{(Equation 7)}$$

Equation 7 is used in the main text as Equation 2.

To calculate $E_{capture\ losses}$ in 2050, a high capture rate of 95% is assumed based on the finding that such high capture rates would be needed[32] and are expected to become dominant after 2040[122] under stringent decarbonization targets. Furthermore, in achieving a net-zero CO_2 system, Wevers et al.[81] found that going beyond a 95% capture rate would be energetically unfavorable for deploying DAC for the remaining 5%.

In the results, we report E_{CCU}, but when this applies to fossil CO_2 sources in 2050, Equation 7 is used for the calculation. For CCU processes based on biogenic or atmospheric CO_2 in 2050, Equation 6c still applies because the availability of these sources in 2050 is in line with a Paris-compatible scenario.

Assumptions to arrive at harmonized GHG emissions of CCU product and substitute

To complement the "Emissions of CCU technologies" section and Note S4, which includes detailed assumptions according to the LCA study, this section provides details regarding overarching assumptions in harmonization of GHG emissions of CCU products.

For some CCU products, not all carbon in the end product has its origin in captured CO_2. Instead, some processes use fossil methane or methanol as additional feedstock; for example, in dry reforming of methane, where (captured) CO_2 and methane are used to produce CO and H_2.[164] This results in emission of additional CO_2 besides the $E_{released}$ of 1 kgCO_2/kg CO_2 utilized upon combustion of the chemical/fuel; these additional emissions are included in E_{other}. Conversely, in CO_2 EOR, the CO_2 utilized to produce the hydrocarbon does not end up in the product itself but stays behind in the depleted oil field. The CO_2 is cycled and used several times in the process; the amount of CO_2 utilized per barrel of oil is taken to be the total amount of CO_2 injected in a well over the years of CO_2 EOR operation and divided by the total number of barrels of oil produced. Given that the CO_2 utilized is defined as the CO_2 stored in the well, $E_{released}$ is zero. The combustion of the produced hydrocarbon is included in E_{other}.

Several processes produce co-products with the CCU product or with the feedstock, leading to a multifunctionality problem. In ethylene production, propylene, butene, and hydrogen are co-produced;[78] in the electrochemical reduction process, O_2 and H_2 are co-produced;[107] and for hydrogen production via electrolysis, 7.94 kg O_2/kg H_2 is co-produced. We follow the LCA standard ISO 14044 guidelines and apply the system expansion via substitution approach. In particular, we assume that the production of 1 kg of H_2 via electrolysis avoids the production of 7.94 kg of O_2 via cryogenic air separation. We used the production process of O_2 using air separation in EcoInvent v.3.6 and adapted this process to ensure harmonization of the electricity used in electrolysis and the substituted oxygen production process; we replaced the electricity source for the required 1.42 kWh/kg O_2 in the EcoInvent process by the 2030 and 2050 electricity mix used in the harmonization. In 2030, this results in an emission reduction factor of 22% for H_2 production because of substitution; we multiply the emissions for H_2 with a factor of 0.78; in 2050, with electricity assumed to be emission-free, H_2 production has (close to) zero emissions.

For cement-based substitutes and CCU construction materials, we include the CO_2 uptake during the use phase and end-of-life phase, which is not included in the analyzed LCAs. Xi et al.[67] show that, from 1930 to 2013, 43% of the limestone calcination emissions in cement production were offset because of atmospheric CO_2 absorption in a natural cement carbonation process. Cao et al.[68] project that, from 2015 to 2100, 30% of cement production emissions are absorbed by cement carbonation, largely in line with Xi et al.,[67] considering that calcination of limestone currently accounts for 58.4% of the CO_2 emissions of cement production.[68] To take this uptake of CO_2 into account, we multiply the emissions of cement production by a factor 0.7 to find the full life cycle emissions, including uptake.

In our analysis, we assume that industrial waste as feedstock is impact free. Several processes in the mineral carbonates and construction materials category require input of industrial waste; e.g., steel slag, fly ash, or desalination brines. Although some studies assume that CCU avoids landfilling or processing of these waste streams, assigning avoided emissions to the CCU product, there is no consensus on how much these are (i.e., a factor 4 difference between avoided emissions from slag landfilling of -0.04 kgCO_2-equiv/kg $CaCO_3$[66] and -0.16 kgCO_2-equiv/kg $CaCO_3$[65]). We follow the most regularly used and most conservative approach of assuming impact-free waste streams.

Lifetimes of CCU products

We calculated the GHG intensity of CCU products without (base case) and with the benefit of temporary carbon storage (sensitivity analysis), using a time horizon of 100 years. To include the storage effect, $E_{released}$ in Equations 1 and 2 is multiplied by $GWP_{storage}$-100, whose value depends on the product-specific lifetime (Table 2). For direct use of CO_2 in an agricultural greenhouse, a lifetime of less than 6 months is assumed, based on Mazotti et al.[129] For EHR, permanent storage[129] or at least millennia[11] is assumed. For mineral carbonates and construction materials, a lifetime of centuries to permanent is assumed, based on Hepburn et al.,[11] Pan et al.,[58] and Sanna et al.,[56] except for sodium bicarbonate, which may release CO_2 upon use, assuming a lifetime of 6 months. In the fuels and chemicals category, for more stable chemicals (polyethylene, polypropylene, and polyols), a lifetime of 50 years is assumed, based on the lifetime of months to decades for polymers[11] or decades to centuries for polyurethanes.[129] For the remainder of fuels and chemicals, a lifetime of 6 months is assumed, based on the lifetime of 6 months indicated for methanol and urea by Mazotti et al.,[129] and for methane, Fischer-Tropsch fuels, and DME by Hepburn et al.[11]

SUPPLEMENTAL INFORMATION

Supplemental information can be found online at https://doi.org/10.1016/j.oneear.2022.01.006.

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AUTHOR CONTRIBUTIONS

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