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Negative Emissions in the Industrial Sector

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NEGATIVE EMISSIONS IN THE INDUSTRIAL SECTOR

Dissertation

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by

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for Chris, who loves the world

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"Forty-two!" yelled Loonquawl. "Is that all you've got to show for seven and a half million years' work?"
"I checked it very thoroughly," said the computer, "and that quite definitely is the answer. I think the problem, to be quite honest with you, is that you've never actually known what the question is."
"But it was the Great Question! The Ultimate Question of Life, the Universe and Everything!" howled Loonquawl.
"Yes," said Deep Thought with the air of one who suffers fools gladly, "but what actually is it?"

from *The Hitchhiker's Guide to the Galaxy*, Chapter 28, by Douglas Adams

Summary

The ongoing climate crisis has led to record heatwaves, increased the severity of droughts and heavy rainfall events, resulting in floods, fires, and death. Preventing even greater catastrophic impacts requires the rapid and dramatic reduction of anthropogenic greenhouse gas emissions. However, we have already emitted so many greenhouse gases that the reduction of future emissions alone is unlikely to limit global warming to “well below 2°C”, the ambition set by the 2015 Paris Climate Agreement and reaffirmed in the 2021 Glasgow Climate Pact.

Therefore, it is highly likely that we will need to remove large quantities of greenhouse gases from the atmosphere to compensate for greenhouse gases that we have already emitted or will continue to emit in the future. In particular, the industrial sector is expected to be a source of residual carbon dioxide emissions due to production technologies that are difficult to electrify (e.g., kilns requiring high-temperature heat), that produce carbon dioxide as part of non-energy chemical conversion processes (e.g., the calcination of limestone), or that produce products that emit carbon dioxide during use or end-of-life (e.g., liquid fuels and fertilisers).

Technologies that deliberately—and permanently—remove carbon dioxide or other greenhouse gases from the atmosphere are referred to as “carbon dioxide removal” or “negative emission technologies” (NETs). These include:

- **afforestation**, the large-scale increase of the land area and carbon stocks of forests;
- **enhanced weathering**, the anthropogenic stimulation of the carbonation of minerals to increase their rate of atmospheric CO_2 removal;
- **biomass with carbon capture and storage** (bioCCS), the use of biomass as a fuel or feedstock where the biogenic CO_2 produced by its combustion or conversion is captured and permanently stored; and
- **direct air capture and storage**, the use of chemicals to directly remove CO_2 from the atmosphere, which is then permanently stored.

One option for compensating the residual emissions of industry is the direct integration of negative emission technologies, such as biomass with carbon capture and storage, into carbon-intensive industrial process such as steelmaking, cement production, and chemical synthesis. However, negative emission technologies do not yet exist on large scales and research on how they could be effectively deployed is still limited, particularly for the industrial sector.

This dissertation explores under what conditions the integration of these so called “negative emission technologies” could allow for industries to achieve or exceed carbon neutrality within the system of production, rather than needing compensation elsewhere in society. Specifically, this dissertation sought to answer the following research question:

To what extent and under what conditions could negative emission technologies (NETs) play a role in the decarbonisation of the industrial sector?

To answer, this dissertation both analysed existing research on negative emission technologies and modelled possible configurations of negative emission technologies in industry. Two systematic literature reviews were conducted: one to map the usage of the term “negative emissions” and propose operational criteria for their attainment, and another to review the current available literature on industrial bioCCS. A series of case studies was used to estimate the decarbonisation potential of bioCCS in the carbon-intensive industries.

The case studies in this dissertation explored the impact of key technological, systemic, and modelling choices on the estimated decarbonisation potential for the use of bioCCS in the industries of fuels, fertilisers, steel, and cement. All case studies included the use of biomass from sustainable agriculture or forestry, the use of biofuels to satisfy the thermal energy demand of CO_2 capture, and the permanent storage of captured CO_2 in geologic formation.

The foundation of the case studies were process models of industrial production, biofuel production, and CO_2 capture, transport, and storage. For this, a software library to facilitate comparative black box modelling, blackblox.py was built, and has been released open-source with this dissertation. These process models were then used as the basis of life cycle accounting of CO_2 emissions and removals and other greenhouse gas emissions, as well as a technoeconomic assessment of the cost of bioCCS.

In the end, the main research question was answered by subdividing it into four subquestions:

1. ***What criteria must negative emissions technologies meet to result in a net decrease of atmospheric greenhouse gases?***

   A literature review of over 400 studies published between 2014 and 2018 determined that the term “negative emissions” was being used to refer to a variety of concepts. Most commonly, the stated or implied goal of negative emissions in these studies was the net decrease of the atmospheric greenhouse gases for the purpose of decreasing global warming. However, some studies used “negative emissions” to refer to the relative reduction of greenhouse gases from one system to another (so called “avoided emissions”), the storage of fossil CO_2, or the use of captured atmospheric CO_2 in a short-lived fuel or chemical product (that will result in re-emission of the removed CO_2). As none of these results in the permanent removal of greenhouse gases, they cannot achieve the goal of decreasing atmospheric greenhouse gases. To align the use of the term "negative emissions" with its oft-stated goal, we proposed four minimum criteria that a negative emission technology should satisfy:

1. *Physical greenhouse gases are removed from the atmosphere.*
2. *The removed gases are stored out of the atmosphere in a manner intended to be permanent.*
3. *Upstream and downstream greenhouse gas emissions associated with the removal and storage process, such as biomass origin, energy use, gas fate, and co-product fate, are comprehensively estimated and included in the emission balance.*
4. *The total quantity of atmospheric greenhouse gases removed and permanently stored is greater than the total quantity of greenhouse gases emitted to the atmosphere.*

2. ***To what extent could NETs reduce the CO_2 footprint of carbon-intensive industrial processes?***

BioCCS was determined to be the most suitable negative emission technology for integration into carbon-intensive industrial processes, and was explored in a series of case studies. In particular, this dissertation investigated if bioCCS could allow for "CO_2-neutral" or "CO_2-negative" products; this requires not only that the system results in negative emissions, as described above, but that the quantity of negative emissions is greater than the quantity of CO_2 emitted in the upstream supply chains and downstream use and disposal of the product itself.

Five technologies of steel production were considered in this dissertation. Blast furnace ironmaking, which accounts for 70% of global steel production, relies on the structural properties of coke, so the replacement of fossil fuels with charcoal in existing blast furnaces is limited to approximately 40%. Because of this limited replacement rate, the initial model of bioCCS in blast furnaces did not result in estimated negative emissions, though net emissions fell from 2.4 to 0.1 t CO_2/t steel. In another case, where "top gas recycling"—a novel technology that improves energy efficiency and reduces coke demand—was modelled to be retrofitted into the blast furnace along with the use of bioCCS, estimated net emissions reached -0.05 t CO_2/t steel. Replacing the blast furnace with a HIsarna furnace, a novel design that eliminates coke use yet also only allows for partial charcoal replacement, could further reduce net emissions by 0.1 t CO_2/t steel. The potential for negative emissions in steelmaking was greater when gas-fired direct reduction ironmaking is used, as, theoretically, a completely biogenic reducing gas could be used, without the need for any fossil fuels in the ironmaking process. Due to this, the estimated net CO_2 for direct reduction steelmaking with bioCCS reached as low as -0.5 t CO_2/t steel.

The production of cement is inherently fossil-carbon intensive due to the calcination of limestone into lime ($CaCO_3$ -> CaO + CO_2), which accounts for roughly 60% of cement kiln CO_2 emissions. In chapters 6 and 7, the use of biofuel, such as charcoal, to fire the kiln combined with the capture of both calcination and combustion emissions resulted in an estimated net CO_2 of -0.3 to -0.1 t CO_2/t cement. When the use of cement in concrete was also considered, these increased to -0.2 to 0.1 t CO_2/t cement, which accounts for both CO_2 emissions during concrete production and the natural uptake of CO_2 by concrete during its lifetime and after demolition.

For maize-based ethanol, CO_2-negative production required both the capture of CO_2 produced by the fermentation process and by the combustion of biofuel for heat provision. The capture of fermentation CO_2 alone was not sufficient to compensate for the CO_2 emitted during the combustion of ethanol, the decomposition of the distiller grains (a by-product of ethanol fermentation), and in the upstream supply chains.

Hydrogen does not contain carbon and, when it is produced using steam reforming of biomethane, allows for a large quantity of biogenic CO_2 to be captured and stored, and thus to be CO_2-negative, even when accounting for the use of hydrogen in the production of ammonia. However, if that ammonia is used for urea production, which typically uses CO_2 captured from steam reforming, the bioCCS system was no longer able to compensate for all emissions, though net emissions reduced from 5.7 t CO_2/t urea to 0.1 t CO_2/t urea. Other system optimisations, such as improving conversion efficiency or the increased use of renewable electricity, which were not considered in this dissertation, may further decrease net emissions.

3. ***What aspects of integrating NETs into industrial processes have the greatest influence on their potential decarbonisation performance and costs?***

In this dissertation, the most influential aspects of the bioCCS system on its ability to reduce net CO_2 emissions were:

- the CO_2 emitted in the supply chains of biomass production and processing.
- the CO_2 emitted in the supply chains of other energy sources. As the modelled technology for CO_2 compression had a high electricity demand, this led to a high sensitivity to the CO_2 intensity of electricity generation.

- the amount of carbon that could be captured during industrial production versus the amount embodied in the product or the amount emitted in upstream supply chains.

The most influential aspects of the bioCCS system on its costs seen in this dissertation were:

- the price of biomass, which varied in literature studies from 0-15€$_{2020}$/GJ, and the price of other energy sources such as electricity.
- the transport distance of CO_2 relative to the volume transported. Transport costs in industrial bioCCS literature ranged from 5-380€$_{2020}$/t CO_2, with the highest estimates seen for small quantities of CO_2 transported long distances, such as from ethanol distilleries in central Brazil to offshore storage sites.
- the capital expenses of the CO_2 capture system and pipeline, particularly when cost estimates account for the extra expenses expected of a near-term "first of a kind" installation.
- assumptions made about taxes on emissions of fossil CO_2 or available credits for the storage of (biogenic) CO_2.

4. ***How do modelling choices affect the perceived environmental and economic performance of selected industrial NETs?***

System boundaries used for estimates of abatement potential and cost of bioCCS systems vary widely across literature. In the bioCCS-in-cement technoeconomic case study, the apparent net CO_2eq of a single bioCCS-in-cement system ranged from -660 to 16 kg CO_2eq/t cement solely by varying the system boundaries to reflect boundaries used across bioCCS literature. Notably, while many bioCCS studies focus on the CO_2 emitted at the industrial production site, most emissions in a bioCCS system occur elsewhere, particularly in the upstream supply chains of energy and biomass. Furthermore, accounting for non-CO_2 greenhouse gases is also necessary to assess whether the use of bioCCS can result in a decrease of global warming. In particular, methane emissions from biofuel upgrading, such as charcoal production, were significant in the steel and cement case studies.

A common assumption in bioCCS studies is that biomass is "carbon neutral". However, biogenic CO_2 resides in the atmosphere as biomass is regrown, and therefore has a temporary warming effect, which can be significant for long rotation biomass. In this dissertation, this issue was explored both by using a global warming potential factor for biogenic CO_2 and, in one model, by by explicitly mapping emissions and removals of CO_2 over time.

This dissertation also explored the limitations of the "net CO_2eq" metric, which can obscure the carbon intensity of a bioCCS system, both due to the inclusion of "positive"

> emissions and "negative" removals. The metric also excludes CO_2 that is not emitted or removed within the system boundaries of the study, such as stored or reused CO_2, which further obscures how much carbon the system contains. Furthermore, if the final fate of the removed CO_2 is not permanent storage but short-term reuse or otherwise emitted, a bioCCS system can emit more CO_2 than an unabated system.
>
> Cost estimates are also influenced by system boundary choices. While CO_2 capture and compression were the largest part of the costs estimated in the bioCCS-in-cement technoeconomic case study, the wide range of estimates seen in literature highlights that the cost of transport and storage cannot be neglected. Furthermore, most available cost estimates for bioCCS are affected by both the assumptions embodied in estimates of both cost and abatement potential. While the wide variety of avoidance cost estimates in literature (13-388€$_{2020}$/t CO_2 abated) is partially attributable technological configuration and parametrical assumptions of input prices, production efficiencies, modelling choices also play a critical role. The estimated CO_2(eq) avoidance cost for a near-term retrofit of bioCCS at a cement plant ranged from 68-321€$_{2017}$/t CO_2 avoided by varying the system boundaries to those seen in bioCCS literature.

This dissertation contains some of the initial charting of the research field of industrial negative emissions. It provides first-order estimates of the decarbonisation potential of bioCCS in industry, key contributors to those potentials, and highlights the importance of system boundary choices in negative emission technology systems. There, of course, remains substantial work to be done if bioCCS is to be used as an effective tool in reaching "net zero" global greenhouse gas emissions. For example, this dissertation did not intend to provide the level of detail or optimisation necessary to assess the net emissions and costs of specific potential bioCCS installations. Furthermore, this dissertation focused on the global warming potential of the emission of CO_2 and other greenhouse gases, but there are many other environmental, as well as social and political, issues that must be addressed for bioCCS to be implemented safely and justly.

Future research on net greenhouse gas emissions of industrial NETs must be inclusive and transparent and always account for emissions in the systems of biomass production and use and CO_2 capture, transport, and storage, and related supply chains. As NETs depend on the permanent storage of CO_2, the fate of all CO_2 in the system must be clearly accounted for. Furthermore, if an industrial system is to be labelled "carbon neutral" or "carbon negative", negative emissions must be sufficient to compensate for all emissions from the system of industrial production and product use. Studies on negative emissions should also clearly decompose the carbon flows in the system, as carbon intensity can be easily obscured by single-point metrics such as "net CO_2" or "relative abatement".

The judicious use of negative emission technologies has the potential to be a valuable tool for balancing residual emissions from difficult-to-decarbonise sectors, or for removing a limited amount of historical CO_2 from the atmosphere. However, it will always substantially less resource-intensive to not emit CO_2 than to remove it later. Negative emissions are not a panacea and will be most effective if they are used only as a supplement to rapid and comprehensive reduction of greenhouse gas emissions.

Samenvatting

De aanhoudende klimaatcrisis heeft veel negatieve gevolgen. Op sommige plekken veroorzaakt dit hittegolven, ernstige droogtes en bosbranden, terwijl andere plekken kampen met overmatige regenval en overstromingen. Om nog grotere catastrofen te voorkomen, is een snelle en drastische vermindering van de antropogene uitstoot van broeikasgassen vereist. Helaas is alleen de vermindering van toekomstige emissies niet genoeg om de opwarming van de aarde te beperken tot de doelstelling “ruim onder de 2°C”, zoals vastgelegd in het Klimaatakkoord van Parijs van 2015 en opnieuw bevestigd in het Glasgow Climate Pact 2021.

Het is zeer waarschijnlijk dat we grote hoeveelheden broeikasgassen uit de atmosfeer moeten verwijderen om de broeikasgassen te compenseren die we al hebben uitgestoten of in de toekomst zullen blijven uitstoten. Van de industriële sector wordt verwacht dat het een bron van resterende CO_2-emissies zal blijven. Dit is bijvoorbeeld het geval bij productietechnologieën die moeilijk te elektrificeren zijn (bijvoorbeeld ovens die warmte op hoge temperatuur vereisen), die CO_2 produceren als onderdeel van niet-energetische chemische omzettingsprocessen (bijvoorbeeld het calcineren van kalksteen), of die tijdens gebruik of aan het einde van hun levensduur CO_2 uitstoten (bijvoorbeeld vloeibare brandstoffen en meststoffen).

Technologieën die opzettelijk - en permanent - CO_2 of andere broeikasgassen uit de atmosfeer verwijderen, worden "carbon dioxide removal" of "negatieve emissietechnologieën" (NET's) genoemd. Waaronder:

- **bebossing**, grootschalige vergroting van het landoppervlak en de koolstofvoorraden van bossen;
- **enhanced weathering** (verwering van gesteenten), de antropogene stimulering van de carbonatatie van mineralen om hun snelheid van atmosferische CO_2-verwijdering te verhogen;
- **bio-energie met CO2-verwijdering en opslag** (verder aangeduid als bioCCS), het gebruik van biomassa als brandstof of grondstof waarbij de biogene CO_2 die door de verbranding of omzetting wordt geproduceerd, wordt opgevangen en permanent opgeslagen; en
- **direct air capture** ("directe opname van CO_2 uit de lucht"), het gebruik van chemicaliën om CO_2 direct uit de atmosfeer te halen, dat vervolgens permanent wordt opgeslagen.

Eén optie om de restemissies van de industrie te compenseren, is de directe integratie van negatieve emissietechnologieën, zoals bioCCS, in koolstof-intensieve industriële processen zoals staalproductie, cementproductie en chemische synthese. Deze negatieve emissietechnologieën bestaan echter nog niet op grote schaal en het onderzoek naar hoe ze effectief kunnen worden ingezet is nog beperkt, met name voor de industriële sector.

Deze dissertatie onderzoekt onder welke omstandigheden de integratie van deze zogenaamde negatieve-emissietechnologieën industrieën in staat zou kunnen stellen om minimaal koolstofneutraliteit binnen het productiesysteem te bereiken, in plaats van compensatie elders in de samenleving nodig te hebben. Dit proefschrift beoogt de volgende onderzoeksvraag te beantwoorden:

In welke mate en onder welke voorwaarden zouden negatieve-emissietechnologieën (NET's) een rol kunnen spelen bij het koolstofvrij maken van de industriële sector?

Om deze vraag te beantwoorden worden in dit proefschrift enerzijds bestaand onderzoek naar negatieve emissietechnologieën geanalyseerd en anderzijds nieuwe mogelijke configuraties van negatieve emissietechnologieën in de industrie bekeken. Er zijn twee systematische literatuuronderzoeken uitgevoerd: in de eerste wordt het gebruik van de term "negatieve emissies" te geanalyseerd en worden operationele criteria voorgesteld voor het bereiken ervan; in de tweede wordt de huidige beschikbare literatuur over industriële bioCCS beoordeeld. Vervolgens is in een reeks casestudies het decarbonisatiepotentieel van bioCCS in de koolstof-intensieve industrieën bepaald.

In de casestudies in dit proefschrift is onderzocht wat de invloed is van belangrijke technologische, systemische en modelkeuzes op het decarbonisatiepotentieel van bioCCS in de productie van brandstoffen, meststoffen, staal en cement. Alle casestudies omvatten het gebruik van biomassa uit duurzame land- of bosbouw, het gebruik van biobrandstoffen om te voldoen aan de thermische energiebehoefte van CO_2-afvang en de permanente opslag van opgevangen CO_2 in geologische formaties.

De basis van de casestudies zijn modellen van industriële productieprocessen, productie van biobrandstoffen en CO_2-afvang, -transport en -opslag. Hiervoor is een softwarebibliotheek gemaakt om vergelijkende "black box"-modellering te vergemakkelijken, blackblox.py, en deze wordt tegelijk met dit proefschrift als open-source vrijgegeven. Deze procesmodellen werden vervolgens gebruikt als basis voor de levenscyclusboekhouding van CO_2- en andere broeikasgasemissies en CO_2-verwijdering, en voor een techno-economische beoordeling van de kosten van bioCCS.

De hoofdvraag is beantwoord door deze op te delen in vier deelvragen:

1. *Aan welke criteria moeten technologieën voor negatieve emissies voldoen om te leiden tot een netto afname van broeikasgassen in de atmosfeer?*

Een literatuuronderzoek van meer dan 400 studies die tussen 2014 en 2018 zijn gepubliceerd laat zien dat de term 'negatieve emissies' naar heel verschillende concepten kan verwijzen. Meestal was het gestelde of impliciete doel van negatieve emissies in deze onderzoeken de netto afname van de atmosferische broeikasgassen met als doel de opwarming van de aarde te verminderen. Sommige studies gebruikten echter "negatieve emissies" om te verwijzen naar de relatieve vermindering van broeikasgassen van het ene systeem naar het andere (de zogenaamde "vermeden emissies"), de opslag van fossiele CO_2, of het gebruik van opgevangen CO_2 uit de atmosfeer in een brandstof of chemisch product, waarbij na gebruik de koolstof opnieuw als CO_2 wordt uitgestoten. Aangezien geen van deze resulteert in de permanente verwijdering van broeikasgassen, kunnen ze aan het doel van het verminderen van atmosferische broeikasgassen niet bijdragen. Om het gebruik van de term 'negatieve emissies' aan te laten sluiten op het eerst genoemde doel, hebben we vier minimumcriteria voorgesteld waaraan een technologie voor negatieve emissies moet voldoen:

1. *Fysieke broeikasgassen worden uit de atmosfeer verwijderd.*
2. *De verwijderde gassen worden permanent buiten de atmosfeer opgeslagen.*
3. *Upstream en downstream broeikasgasemissies van het verwijderings- en opslagproces, zoals de emissies die ontstaan bij het telen van biomassa, het energieverbruik in de keten, en de behandeling van nevenproducten, worden volledig meegenomen in de emissiebalans.*
4. *De totale hoeveelheid atmosferische broeikasgassen verwijderd en permanent opgeslagen is groter dan de totale hoeveelheid broeikasgassen die in de atmosfeer wordt uitgestoten.*

2. *Hoeveel kunnen NET's de* CO_2*-voetafdruk van koolstof-intensieve industriële processen verkleinen?*

BioCCS bleek de meest geschikte negatieve-emissietechnologie voor integratie in koolstof-intensieve industriële processen, en werd onderzocht in een reeks casestudies. In dit proefschrift is met name onderzocht of bioCCS “CO_2-neutrale” of “CO_2-negatieve” producten mogelijk zou kunnen maken; dit vereist niet alleen dat het systeem leidt tot negatieve emissies, zoals hierboven beschreven, maar ook dat de hoeveelheid negatieve emissies groter is dan de CO_2-emissies in de toeleveringsketens en downstream gebruik en verwijdering van het product zelf.

Vijf technologieën voor staalproductie worden behandeld in hoofdstuk 5. De ijzerproductie in hoogovens, die 70% van de wereldwijde staalproductie uitmaakt, is afhankelijk van de structurele eigenschappen van cokes, dus de vervanging van fossiele brandstoffen door houtskool in bestaande hoogovens is beperkt tot ongeveer 40%. Vanwege deze beperkte vervangingsratio resulteerde het initiële model van bioCCS in hoogovens niet in negatieve emissies, hoewel de netto-emissies daalden van 2,4 naar 0,1 t CO_2/t staal. In een ander geval, waar "top gas recycling" - een nieuwe technologie die de energie-efficiëntie verbetert en de vraag naar cokes vermindert - werd gemodelleerd om achteraf in de hoogoven te worden ingebouwd, samen met het gebruik van bioCCS, bereikte de netto-uitstoot -0,05 t CO_2/t staal. Het vervangen van de hoogoven door een HIsarna-oven, een nieuw ontwerp dat het gebruik van cokes elimineert maar ook slechts gedeeltelijke vervanging van houtskool mogelijk maakt, zou de netto-uitstoot met 0,1 t CO_2/t staal verder kunnen verminderen. Het potentieel voor negatieve emissies bij de staalproductie is groter wanneer ijzer wordt geproduceerd middels directe reductie, omdat theoretisch een volledig biogeen gas als reductiemiddel kan worden gebruikt, zonder dat er fossiele brandstoffen nodig zijn in het ijzerproductieproces. Hierdoor bereiken de netto CO_2-emissies voor deze staalproductietechnologie met bioCCS -0,5 t CO_2/t staal.

De productie van cement is inherent CO_2-intensief door de calcinering van kalksteen tot kalk ($CaCO_3 \rightarrow CaO + CO_2$), die goed is voor ongeveer 60% van de CO_2-uitstoot van cementovens. Het gebruik van biobrandstof, zoals houtskool, om de oven te stoken in combinatie met het opvangen van zowel calcinatie- als verbrandingsemissies in een netto CO_2-emissiesvan -0,3 tot -0,1 t CO_2/t cement. Wanneer ook de toepassing van cement in beton wordt meegenomen, neemt de netto CO_2-emissie tot -0,2 tot 0,1 t CO_2/t cement, door het netto-effect van de CO_2-uitstoot tijdens de betonproductie en de natuurlijke opname van CO_2 door beton tijdens de levensduur en na de sloop.

Voor ethanol op basis van maïs vereist CO_2-negatieve productie het afvangen van zowel de CO_2 geproduceerd in het vergistingsproces als de CO2-uitsoot door verbranding van biobrandstof voor warmtevoorziening. Het afvangen van alleen de fermentatie- CO_2 was niet voldoende om de CO_2-uitstoot te compenseren die vrijkomt bij de verbranding van ethanol, en elders in de productieketen.

Waterstof bevat geen koolstof. Wanneer het wordt geproduceerd met stoomreforming uit biomethaan kan een grote hoeveelheid biogeen CO_2 worden opgevangen en opgeslagen. Dus de productie van waterstof kan CO_2-negatief zijn. Dit geldt ook als het gebruik van waterstof bij de productie van ammoniak wordt meegerekend. Als die ammoniak echter wordt gebruikt voor de productie van ureum, waarbij doorgaans gebruik wordt gemaakt van CO_2 die wordt afgevangen bij de stoomreforming, is het bioCCS-systeem niet langer in staat om alle emissies te compenseren, hoewel de netto-emissies werden verlaagd van 5,7 t CO2/t ureum tot 0,1 t CO_2/t ureum. Andere systeemoptimalisaties, zoals het verbeteren van de conversie-efficiëntie of het toegenomen gebruik van hernieuwbare elektriciteit, die in dit proefschrift niet zijn meegenomen, kunnen de netto-emissies verder verlagen.

3. *Welke aspecten van de integratie van NET's in industriële processen hebben de grootste invloed op hun* CO_2*-reductiepotentieel en -kosten?*

In dit proefschrift werden de factoren geïdentificeerd die het meest van invloed zijn op het CO_2-emissiereductiepotentieel. Op basis van literatuuronderzoek en casestudies blijkt dat de belangrijkste zijn:

- de CO_2 die wordt uitgestoten in de toeleveringsketens van de productie en verwerking van biomassa;
- de CO_2 die wordt uitgestoten in de toeleveringsketens van andere energiebronnen. Omdat de gemodelleerde technologie voor CO_2-compressie een hoge elektriciteitsvraag heeft, leidt dit tot een hoge gevoeligheid voor de CO_2-intensiteit van elektriciteitsopwekking;
- de hoeveelheid koolstof die kan worden afgevangen tijdens industriële productie versus de hoeveelheid die in het product zit of de hoeveelheid die wordt uitgestoten in de toeleveringsketens .

De meest invloedrijke aspecten van het bioCCS-systeem op de kosten van vermeden CO2 zijn:

- de prijs van biomassa, die in literatuurstudies varieerde van 0-15 $€_{2020}$/GJ, en de prijs van andere energiebronnen zoals elektriciteit;
- de transportafstand van CO_2 ten opzichte van het vervoerde volume. Transportkosten in industriële bioCCS-literatuur varieerden van 5-380 $€_{2020}$/t CO_2, waarbij de hoogste schattingen werden gezien voor kleine hoeveelheden CO_2 die over lange afstanden werden vervoerd, zoals van ethanoldistilleerderijen in centraal Brazilië naar offshore-opslaglocaties;

- de kapitaalkosten van het CO_2-afvangsysteem en de pijpleiding, met name wanneer rekening wordt gehouden met de extra kosten die verwacht worden bij een "first of a kind" installatie op korte termijn;
- aannames gemaakt over belastingen op uitstoot van fossiele CO_2 of credits voor opslag van (biogene) CO_2.

4. *Hoe beïnvloeden modelleringskeuzes de waargenomen ecologische en economische prestaties van geselecteerde industriële NET's?*

   De systeemgrenzen die worden gebruikt voor schattingen van het reductiepotentieel en de kosten van bioCCS-systemen variëren sterk in de literatuur. De netto emissies van een cement-met-bioCCS-systeem varieerde van -660 tot 16 kg CO_2eq/t cement, alleen door de variatie van systeemgrenzen zoals die in de bioCCS-literatuur worden gebruikt. Terwijl veel bioCCS-onderzoeken zich richten op de CO_2die wordt uitgestoten op de industriële productielocatie, vinden de meeste emissies in een bioCCS-systeem elders plaats, met name in de toeleveringsketens van energie en biomassa. Verder is het ook nodig om rekening te houden met niet- CO_2-broeikasgassen om te beoordelen of het gebruik van bioCCS kan leiden tot een afname van de opwarming van de aarde. Met name de methaanemissies die vrijkomen bij de opwaardering van biobrandstoffen, zoals de productie van houtskool, waren significant in de staal- en cement-casestudies.

   Een veel voorkomende aanname in bioCCS-onderzoeken is dat biomassa CO2-neutraal is. Biogeen CO_2 bevindt zich echter in de atmosfeer wanneer biomassa opnieuw wordt gekweekt en heeft daarom een tijdelijk opwarmend effect, wat significant kan zijn voor biomassa met een lange groeiperiode. In deze dissertatie werd dit probleem onderzocht door òf een global-warming-potential te gebruiken voor biogene CO_2 òf door emissies en verwijderingen van CO_2 in de tijd expliciet in kaart te brengen.

Kosteninschattingen worden ook beïnvloed door de keuze van systeemgrenzen. Alhoewel het afvangen en comprimeren van CO_2 het grootste deel uitmaakten van de kosten in de casestudie voor cement, blijkt ook uit het brede scala aan schattingen in het literatuuronderzoek dat de kosten van transport en opslag niet kunnen worden verwaarloosd. Bovendien worden de meeste beschikbare kostenramingen voor bioCCS beïnvloed door de veronderstellingen die zijn gemaakt voor enerzijds de bepaling van de kosten en anderzijds de bepaling van het reductiepotentieel. Hoewel de grote verscheidenheid aan schattingen van vermijdingskosten in de literatuur (13-388 €$_{2020}$/t CO_2 verminderd) gedeeltelijk toe te schrijven is aan de technologische configuratie en aannames over prijzen van procesinputs en productie-efficiëntie, spelen ook modelkeuzes een cruciale rol. De geschatte kosten voor het vermijden van CO_2(eq) voor een retrofit van bioCCS op korte termijn in een cementfabriek kunnen variëren van 68 tot 321 $€_{2017}$ per t vermeden CO_2 door alleen al de variatie van de systeemgrenzen in de bioCCS-literatuur toe te passen.

Dit proefschrift geeft een eerste schets van het onderzoeksgebied van industriële negatieve emissies. Het biedt eerste-orde schattingen van het decarbonisatiepotentieel van bioCCS in de industrie, de belangrijkste sectoren die een bijdrage leveren aan dat potentieel en benadrukt het belang van systeemgrenskeuzes in negatieve-emissietechnologiesystemen. Vanzelfsprekend moet er nog veel werk worden verzet om bioCCS in te zetten als een effectief instrument om de wereldwijde uitstoot van broeikasgassen naar 'net zero' te brengen. Dit proefschrift was bijvoorbeeld niet bedoeld om de details of optimalisaties te bepalen voor specifieke potentiële bioCCS-installaties. Verder concentreerde dit proefschrift zich op het opwarmingspotentieel van de uitstoot van CO_2 en andere broeikasgassen, maar er zijn veel andere ecologische, evenals sociale en beleidsmatige zaken die moeten worden aangepakt voordat bioCCS veilig en rechtvaardig kan worden geïmplementeerd.

Toekomstig onderzoek naar de netto broeikasgasemissies van industriële NET's moet inclusief en transparant zijn. Ook moet er altijd rekening worden gehouden met emissies bij biomassaproductie en -gebruik en CO_2-afvang, -transport en –opslag, en in andere toeleveringsketens. Aangezien NET's afhankelijk zijn van de permanente opslag van CO_2, moet de bestemming van alle CO_2 in het systeem duidelijk worden verantwoord. Bovendien, om een industrieel systeem als " CO_2-neutraal" of " CO_2-negatief" te bestempelen, moeten de negatieve emissies voldoende zijn om alle emissies van het systeem van industriële productie en productgebruik te compenseren. Studies over negatieve emissies zouden daarom ook duidelijk de koolstofstromen in het systeem moeten ontleden, aangezien de koolstofintensiteit gemakkelijk kan worden verdoezeld door enkelvoudige meetwaarden zoals "netto CO_2" of "relatieve vermindering".

Het weloverwogen gebruik van negatieve emissietechnologieën kan een waardevol instrument zijn om de restemissies van moeilijk te decarboniseren sectoren in evenwicht te brengen of om een beperkte hoeveelheid historische CO_2 uit de atmosfeer te verwijderen. Het zal echter altijd aanzienlijk minder grondstofintensief zijn om geen CO_2 uit te stoten dan om het later te verwijderen. Negatieve emissies zijn geen wondermiddel en zullen het meest effectief zijn als ze alleen worden gebruikt als aanvulling op een snelle en alomvattende vermindering van de uitstoot van broeikasgassen.

Units and Abbreviations

Units

Mass	
g	gram
Gt	gigatonne (1 000 Mt)
kg	kilograms
kt	kilotonne (1 000 t)
Mt	megatonne (1 000 000 t)
t	metric tonne
tkm	tonne-kilometer (1 t transported 1 km)
Energy	
EJ	exajoule (1 000 000 000 GJ)
GJ	gigajoule (1000 MJ)
kWh	kilowatt hour (3.6 MJ)
MJ	megajoule
TJ	terajoule (1000 GJ)
Dimensions	
km	kilometer (1000 meters)
m	meter (length)
m^2	square meter (area)
m^3	cubic meter (volume)
Nm^3	Normal cubic meter (volume of gas at 0°C and 1.013 bar)
Pressure	
bar	100 000 pascals
Mpa	megapascal (1 000 000 pascals)

Abbreviations

AC	Accelerated carbonation
BECCS	Bioenergy with carbon capture, transport and storage
BF	Blast furnace
BioCCS	Biomass with carbon capture and storage
BOF	Basic oxygen furnace
CCS	Carbon capture, transport and storage
CEM I	Portland cement type (no more than 5% other materials)
CEM II	Portland hybrid cement type (no more than 35% other materials)
CO_2eq	"CO_2 equivalent" 100-year global warming potential
DACCS	Direct air carbon capture, transport and storage
DRI	Direct reduction of iron
EAF	Electric arc furnace
EPCCI	European Power Plant Construction Cost Index
EOR	Enhanced oil recovery
EU	European Union
EUR	Euro (€)
FOAK	First of a kind
GHG	Greenhouse gases
HRC	Hot rolled coil (of carbon steel)
IAM	Integrated assessment model
LCA	Life cycle assessment
MEA	Monoethanolamine
M EUR	Million Euro (1 000 000€)
NL	Netherlands
NET	Negative emission technology
NOAK	Nth of a kind
OPC	Ordinary Portland concrete
TEG	Triethylene glycol
TGR	Top gas recycling
UK	United Kingdom
US	United States of America
USD	United States Dollar ($)
VPSA	Vacuum pressure swing absorption

Chapter 1:
Introduction

1 Background

The ongoing climate crisis has resulted in record heatwaves and increased the severity of droughts and heavy rainfall events [1], resulting in floods, fires, and death [2]. In the four short years over which this dissertation was written, anthropogenic emissions of greenhouse gases—a scientifically indisputable cause of global warming [1]—exceeded 220 Gt CO_2eq [3], including a 3% increase in total atmospheric carbon dioxide [4]. Those same four years saw the EU, China, Japan, the US, the UK and 50 other countries commit to combat climate change by reaching either "carbon neutrality" or "net zero" greenhouse gas emissions gases between 2050-2070 [5]. However, the ambitions pledged in the updated nationally determined contributions announced for COP26 in Glasgow imply emissions of approximately 50 Gt CO_2eq in 2030 [6], not even 10 Gt less than was emitted in 2019 [3], and still over 20 Gt higher than what is needed to keep global warming to 1.5°C

Limiting global warming by massively and rapidly reducing our greenhouse gas emissions will require an unprecedented scale and speed of technological change and international cooperation. Furthermore, so many greenhouse gases have already been emitted that the reduction of future emissions alone is unlikely to limit global warming to "well below 2°C", the ambition set by the 2015 Paris Climate Agreement and reaffirmed in the 2021 Glasgow Climate Pact [7]. In the 2018 IPCC special report on limiting global warming to 1.5°C, all but the most ambitious emission reduction scenarios models also required the deliberate removal of carbon dioxide from the atmosphere, up to 20 Gt/year in the second half of the century [8].

Even if our efforts to deeply reduce greenhouse gas emissions are successful, there will remain emissions of that must be compensated with removals if a steady state of "net zero" is to be maintained. Residual greenhouse gas emissions are expected from agriculture, unintentional combustion, and any remaining use of fossil carbon in fuels and other short-term products. Industry is expected to be a major source of residual CO_2 emissions, as many industrial processes not only produce CO_2 from the use of fossil-based energy sources but also from other chemical reactions, such as those in **table 1**. A review of models that project limiting global warming to 1.5°C [9] estimates that process emissions such as these, along with difficult-to-decarbonise energy use in heavy industry, will be responsible for 300 Gt of CO_2 through 2100.

The removal of atmospheric carbon dioxide may also be needed, or desired, to compensate for prior emissions of greenhouse gases from the atmosphere. This may be to offset a limited amount of overshoot from insufficiently rapid emission reduction [8] or via the efforts of individual parties to compensate for their own historic emissions, such as Microsoft's 2030 ambition to remove sufficient CO_2 to compensate for all its direct and electricity-related emissions since its founding [10].

Table 1. Examples of non-energy CO_2 emissions from the production and use of industrial products [11]

Industry	Process	Conversion Chemistry
Cement, paper, glass, and others	Calcination of limestone	$CaCO_3 + \text{heat} \rightarrow CaO + \mathbf{CO_2}$
Ethanol fuel, food and beverages	Fermentation of sugar	$C_6H_{12}O_6 + \text{yeast} \rightarrow 2C_2H_5OH + 2\mathbf{CO_2} + \text{heat}$
Steel	Iron ore reduction	$3CO + Fe_2O_3 \rightarrow 2Fe + 3\mathbf{CO_2}$
Magnesium	Magnesium ore reduction	$2MgO + C \rightarrow 2Mg + \mathbf{CO_2}$
Aluminium	Aluminium ore reduction	$2Al_3O_2 + 3C \rightarrow 4Al + 3\mathbf{CO_2}$
Fertiliser, refining	Methane reforming	$CH_4 + H_2O \rightarrow CO + 3\ H_2$; $CO + H_2O \rightarrow \mathbf{CO_2} + H_2$
Fertiliser	Urea hydrolysis	$NH_2CONH_2 + H_2O \rightarrow \mathbf{CO_2} + 2NH_3$
Soda ash	Calcination of sodium bicarbonate	$2NaHCO_3 \rightarrow Na_2CO_3 + H_2O + \mathbf{CO_2}$

2 Negative emission technologies

Technologies that deliberately remove carbon dioxide from the atmosphere are referred to as "carbon dioxide removal" (CDR) technologies or "negative emission technologies" (NETs). Beyond afforestation, the first NET mentioned in the reports of the Intergovernmental Panel on Climate Change was bioenergy with carbon capture, transport, and storage (bioCCS, also known as BECCS), which was first mentioned in the 4th IPCC Assessment Report [12] in 2007. In the 5th Assessment Report in 2014, a variety of NETs were discussed [13], not only afforestation and bioCCS, but also enhanced weathering and direct air carbon capture with transport and storage, all of which are described briefly below.

In the 2014 report, bioCCS was included in many scenarios, particularly those assessed to hold global warming at or below 1.5°C or allowed for temporary "overshoot" of emission reduction targets [13]. Four years later, in the 2018 IPCC special report on limiting global warming to 1.5°C, the models included 100-1000 Gt of CO_2 removal through 2100. In the models that allowed for limited or no overshoot in achieving a 1.5°C target, the interquartile range of removals included a cumulative 360-660 Gt of bioCCS, potentially exceeding 20Gt/year of removals by 2100. However, the envisioned rapid deployment of negative emission technologies is complicated by the fact that they do not currently exist on the gigatonne scale and there are many uncertainties about what scale on they may be feasible.

2.1 Afforestation

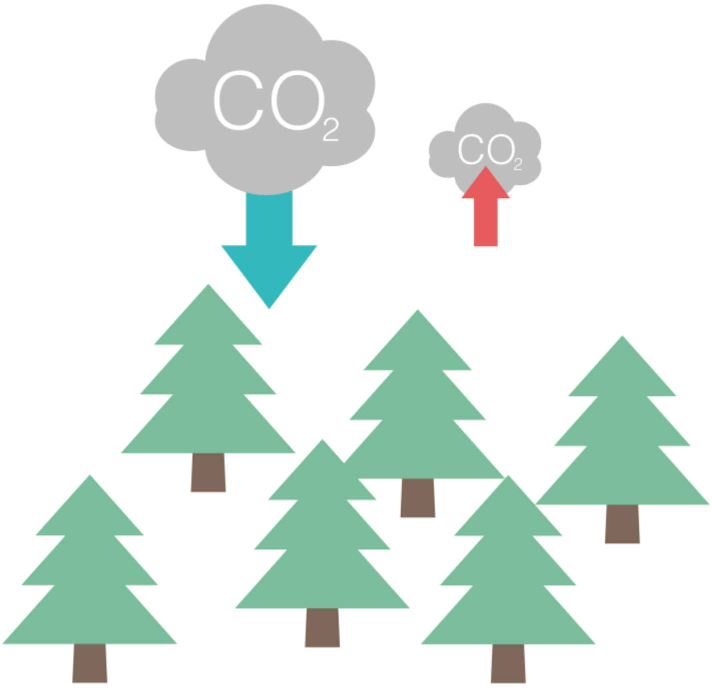

Figure 1. Biomass removes atmospheric carbon via photosynthesis, after which some of the carbon remains stored in the biomass for as long as the biomass is alive, such as in the trunks of trees. Forests also release carbon to the atmosphere as biomass—such as leaves, fruits, grasses, and dead trees—decay.

Afforestation (**figure 1**), the deliberate increase of the land area and carbon stocks of forests, is the most common form of large-scale carbon dioxide removal proposed. Strategically implemented, afforestation can also increase biodiversity, reduce local warming impacts, and improve ecosystem resilience to climate change.

Afforestation requires long-term management and monitoring to be successful. Careful selection of species is necessary to ensure compatibility with the local ecosystems and prevent depletion of water and nutrients in the local environment. Forestry is also land-intensive and the carbon stock of a forest plateaus as the forest matures. Even after this, the forests must be monitored and maintained indefinitely to mitigate incidences of disease, forest fires, drought, neglect, and opportunistic logging, all of which can re-release stored carbon into the atmosphere.

2.2 Enhanced weathering and accelerated carbonation

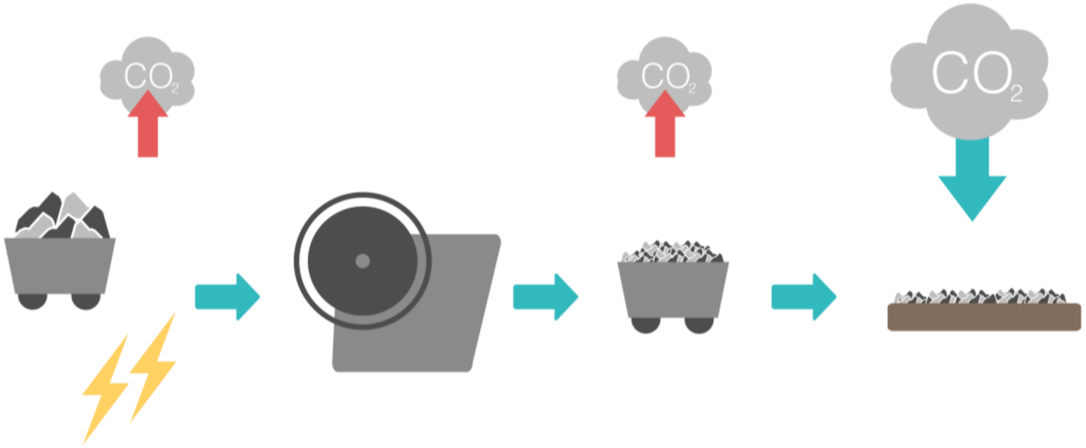

Figure 2. Minerals remove atmospheric CO_2 via carbonation, binding the carbon into the mineral, though it can later be released by processes such as calcination, e.g., as in cement production. Increasing the surface area of the minerals, e.g., via crushing, increases the rate of CO_2 uptake.

In the natural carbon cycle, carbon dioxide is removed from the atmosphere not only in biomass but also, much more slowly, by minerals. This process of carbonation happens in carbonate minerals, such as limestone and dolomite, and silicate minerals, such as olivine and basalt. Accelerated carbonation (**figure 2**) is the anthropogenic stimulation of the mineralization process to increase its rate of atmospheric CO_2 removal.

One proposed form of accelerated carbonation, called "enhanced weathering", involves grinding silicates to increase their surface area and then spreading them over a large surface area, such as beaches, so they take up CO_2 from the atmosphere [14, 15]. The rate of CO_2 uptake is sensitive to rock surface area and ambient conditions, such as temperature [16]. Enhanced weathering is also thought to have potential co-benefits, such as improving agricultural soils, though field research is needed to confirm this, as well as determine real-world removal rates [17]. The potential impact of enhanced weathering on ecosystems and human and animal health is also still unknown [14].

Silicates can also be carbonated by exposing them to concentrated CO_2 streams, such as industrial process emissions [18]. If that CO_2 is of atmospheric is origin (e.g., from biofuel use), this could potentially result in negative emissions. In both cases, the negative emission potential of enhanced silicate weathering is particularly affected by the energy intensity of the mineral grinding and distribution process [14, 17].

Accelerated carbonation can also involve the carbonation of manmade mineral materials, such as lime, fly ash, and steel slag, all of which are created in processes that separate CO_2 from their source minerals (e.g., via the calcination of limestone or combustion of coal). If the

CO_2 released by these processes is captured and stored, and atmospheric CO_2 is then carbonated into these materials, this could result in a net removal of CO_2 from the atmosphere.

2.3 Biomass with carbon capture, transport, and storage (BioCCS)

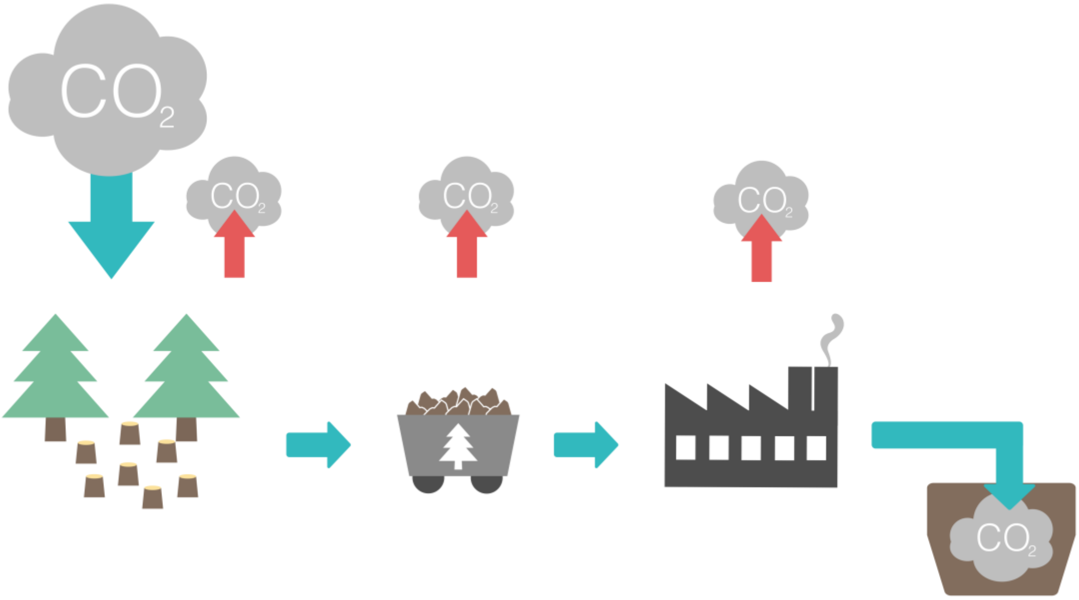

Figure 3. Biomass removes atmospheric carbon via photosynthesis, after which the biomass is harvested to be used as a fuel or feedstock. The CO_2 resulting from the combustion or processing of the biomass is then captured before it reaches the atmosphere and stored permanently, such as in a geologic formation.

Biomass with carbon capture and storage (**figure 3**), also called bioCCS, or BECCS when referring to bioenergy use specifically, is the capture and storage of biogenic CO_2. CO_2 is removed from the atmosphere via the photosynthesis of biomass, which is then used as fuel or feedstock. The CO_2 resulting from the combustion or other chemical conversion of that biomass is then captured and sent to long term storage, such as injected into a geologic formation, such as a depleted gas or oil field or a saline aquifer.

Besides afforestation, bioCCS is the most common negative emission technology assumed to be available in integrated assessment models, typically as a replacement for fossil fuel use in power or industry. In the IPCC Special Report, the interquartile range of models limiting global warming to 1.5°C assumed the deployment of large scale bioCCS as early as 2030, with 7-16 Gt/year of biogenic CO_2 captured and stored by 2100 [8].

BioCCS combines the challenges of increased biomass use, which can be land, water, and nutrient intensive [19], and CCS, which has seen particular difficulty in securing funding for full-scale implementation, with numerous proposed projects cancelled in recent years due to high projected costs [20]. In the UK, the power company Drax has pilot scale bioenergy installations with carbon capture, but they do not yet have access to geologic storage, and therefore the CO_2 is currently re-released after capture [21].

2.4 Direct air carbon capture, transport, and storage (DACCS)

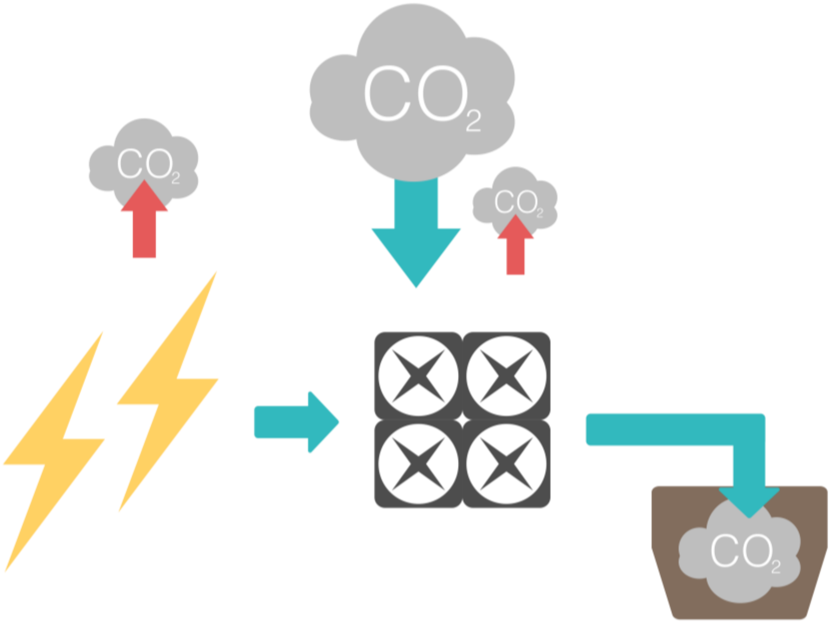

Figure 4. Chemicals or minerals are used to remove CO_2 from the atmosphere, after which the CO_2 is removed from the chemical or mineral and stored permanently, such as in a geologic formation.

Direct air capture (**figure 4**) is the use of chemicals (or minerals) to directly remove CO_2 from the atmosphere. The CO_2 is then separated from those chemicals and sent to long-term storage. As atmospheric CO_2 is very dilute—around 0.04%—, direct air capture is energy intensive, especially in comparison to capturing CO_2 from industrial flue gases, which are typically 5-30% CO_2. DACCS has the potential to be independent from any other industrial or ecosystem process, and thus could be sited in locations where there is both available storage and low-carbon energy sources.

DACCS is currently seeing a surge of commercial interest, with a 4 kt/year installation that began operation in Iceland in 2021 [22], supported in large part by corporations seeking to offset their own emissions [23]. In the US, atmospheric CO_2 removed and stored via DACCS is also covered by the 45Q tax credit for carbon oxide sequestration, but only for installations that are at least 10 kt/year in size [24]. A 1 Mt/year DACCS plant that can take advantage of this scheme is currently under construction with the intent to store its capture CO_2 in the Permian Basin [21].

3 Industrial applications of negative emission technologies

As substantial residual CO_2 emissions are expected from industrial processes even in a "net zero" society, negative emission technologies are likely to be needed to compensate for them. One option, explored in this dissertation, is the direct integration of negative emission technologies into carbon-intensive industrial processes such as steelmaking, cement production, and chemical synthesis.

BioCCS has been identified as a promising candidate for industrial integration, with the IEA's most recent Energy Technology Perspectives report [25] envisioning 4.5 Gt of industrial bioCCS as part of its "below 2 degrees" pathway, and an unspecified amount of industrial bioCCS is found in similar scenarios of other climate change mitigation models [26–30]. Industry is a promising candidate for near-term bioCCS deployment, as both biomass and CCS use are more developed in industry than in the power sector. Industry used twice the amount of biomass—as fuel and feedstock—than the power sector [25] in 2019, and in 2020, 95% of the 40 Mt of CO_2 that was stored from large scale CCS projects was from industrial sources [31]. However, while biomass use, and CO_2 capture, transport, and storage are independently commercialised in various industries and technologies, their combined use as bioCCS is, to date, is limited to the capture and storage of 1 Mt/year of fermentation CO_2 at a US ethanol plant. Still, industries such as cement and chemicals already expect that a multi-pronged decarbonisation strategy, involving both biomass and CCS, will be necessary to reach carbon neutrality [32–34].

4 Motivation for this dissertation

Despite the increasing discussion about negative emission technologies, they do not yet exist on large scales and research on potential technologies and how they could be effectively deployed, while growing rapidly (**figure 5**), is still limited. Notably, while large scale integrated assessment models increasingly include NETs as first-tier options for mitigating climate change, they do not always consider where NETs can be feasibly or effectively implemented within industry [8, 26–30]. In particular, while industry is a potentially promising near-term option for deployment of NETs such as bioCCS, there are few studies on its integration into industries with only four studies on industrial bioCCS use published in 2017, when this research project began [35–38]. This dissertation is part of initial charting of the territory for the potential use of NETs in industry and aims to understand on what scale NETS could contribute to reducing the net CO_2 of industrial processes.

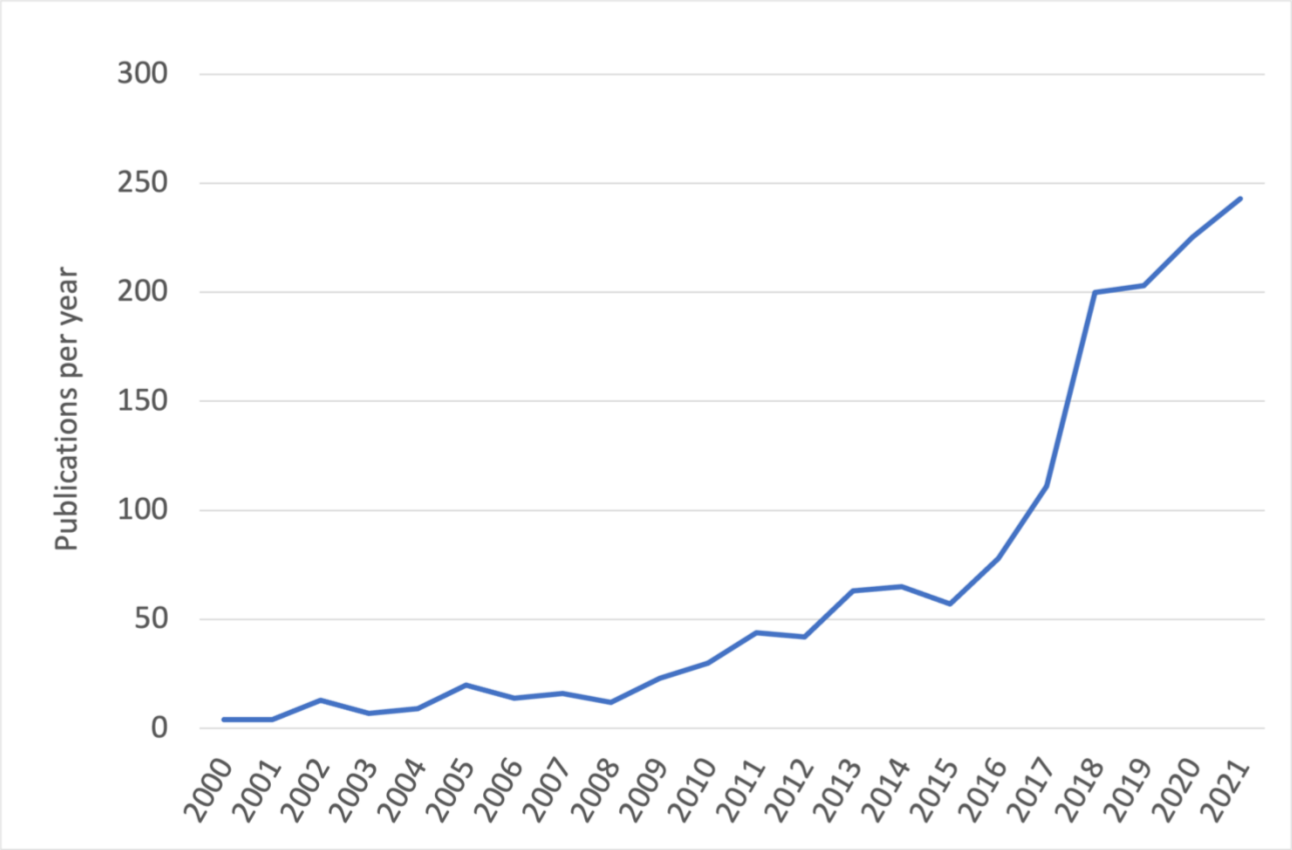

Figure 5. Scholarly Publications on Negative Emissions (Entries in Scopus of articles, conference papers, reviews, and book chapters whose title, keyword, or abstract include "negative emissions", "negative CO_2", or "carbon negative".)

5 Research questions

To direct the research of this dissertation, the overarching research question was formulated as:

To what extent and under what conditions could negative emission technologies (NETs) play a role in the decarbonisation of the industrial sector?

Which has been subdivided into the following subquestions:

1. *What criteria must negative emissions technologies meet to result in a net decrease of atmospheric greenhouse gases?*
2. *To what extent could NETs reduce the CO_2 footprint of carbon-intensive industrial processes?*
3. *What aspects of integrating NETs into industrial processes have the greatest influence on their potential decarbonisation performance and costs?*
4. *How do modelling choices affect the perceived environmental and economic performance of selected industrial NETs?*

6 Overview of main methods used

To answer these questions, this dissertation involved both the analysis of existing research on negative emission technologies as well as explorative modelling of possible NETs-in-industry configurations.

Two systematic literature reviews were conducted. The first was motivated by the heterogeneity of systems that were all labelled as producing “negative emissions”, despite variance in the origin, fate, and accounting of greenhouse gases in the studies. By mapping how the term “negative emissions” and related phrases have been used in literature over the past several years, this dissertation sought to ascertain whether a consensus existed that the intended purpose of negative emissions is to result in a net decrease of atmospheric greenhouse gases. Once the purpose was confirmed, the literature review was also used to propose the minimum criteria NETs needed to meet to fulfil that purpose and to categorise and to explain situations where the term “negative emissions” was used even though the criteria were not met.

The second literature review analysed peer-reviewed literature that estimated the cost or decarbonisation potential of specific industrial bioCCS options. No previous meta-analysis of industrial bioCCS literature was available, so this review provided a first identification of sectors under research, trends in cost and decarbonisation estimation, and existing knowledge gaps.

The remainder of the dissertation involved the construction of exploratory models to generate first estimates of the decarbonisation potential of possible industrial NETs. The modelling in this dissertation is primarily *ex-ante*, that is, the models mostly consider technology systems that are not currently deployable on a commercial scale, with the goal of guiding further research and development by providing comparison with current technology systems [38]. These models are not designed to optimise for cost or efficiency but rather to explore which configurations of NETs-in-industry could lead to negative emissions and to understand the impact of key technological, systemic, and modelling design choices on the estimated decarbonisation potential.

The basic structure of these models is illustrated in **Figure 6**. Each model began with a process model of the technology under investigation that was then used as the basis for a life cycle accounting of CO_2 emissions and removals, as well as for other greenhouse gases in some models. This process model was also the input into the economic assessment used to estimate the potential costs of the proposed technologies. A summary of how these three main modelling components—process modelling, life cycle assessment, and technoeconomic assessment—were used in this research is provided below.

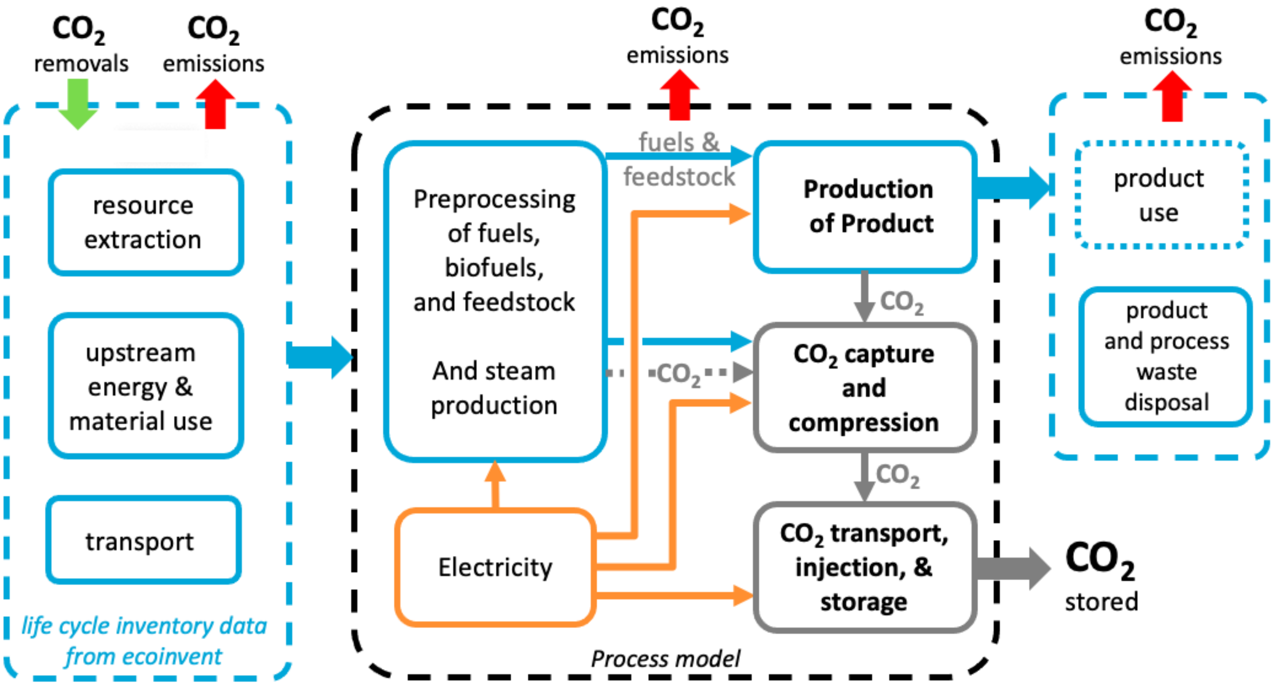

Figure 6. Overview of modelling structure used to evaluate configurations of negative emissions in industry in this dissertation

6.1 Process modelling

The primary modelling focus of this dissertation was the use of bioCCS in different industrial production pathways, with comparisons to the use of biomass or CCS alone and to the unabated production of ethanol, ammonia, urea, hydrogen, steel, and cement. For each technological configuration explored, a process model was created to estimate the use of energy and materials, as well as emissions and removals of CO_2. The models primarily explored the combination of technological elements (e.g., biofuels or CO_2 capture options) that are commercially or near-commercially available, though not used in current commercial practice or used in combination, and therefore also primarily considered present-day efficiencies of industrial production. As exceptions, **chapter 5** includes the use of bioCCS in future steelmaking technologies, and **chapter 6** explores future scenarios of cement production with higher production efficiencies.

Each model consisted of a connected set of unit processes, fixed-ratio black box models representing a single industrial subprocess (e.g., a clinker kiln, ethanol fermenter, or iron furnace) that focused on the flows of major material, and energy inputs and outputs, with a particular emphasis on carbon flows. This level of modelling does not consider individual reactions or the optimisation of kinetics, but instead allows for the comparison of a large set of possible configurations to determine first-order potentials that can be used to understand promising configurations or limiting factors.

For each industrial production pathway, a reference model was first constructed from literature references. The reference models were then adapted for each alternate configuration, e.g., via the replacement of fossil fuels by biofuels or the routing of CO_2 flows to a CO_2 capture subprocess. The model outcome for each case is the overall inflows and outflows of the technical system under consideration. As much as feasible, the models were kept consistent to increase comparability, including the use of standardised emission factors for fuel combustion and the use of standard efficiencies for auxiliary processes such as steam boilers, oxygen separation, and biofuel production.

The construction of these models was facilitated by the development of a python library, blackblox.py, that allows for rapid constriction and comparison of linear fixed-ratio process models. This library, along with many of the models constructed for this dissertation, is available open source. The software, and how to install and use it, is described briefly in **appendix A** at the end of this dissertation.

6.2 Life cycle assessment

Life cycle assessment (LCA) expands the modelling system from the industrial production process itself to include upstream supply chains of material and energy production and resource extraction and the downstream supply chains of (by-)product use and disposal and disposal of wastes. The goal of this “cradle to grave” modelling system is to understand the total resource use and environmental impacts attributable to a product or service, rather than only those impacts that are visible at the point of production or use, and explore which pieces of the supply chain were the source of most variability in the net greenhouse gas balance.

In this dissertation, life cycle assessment was used to estimate the emissions of CO_2 and other greenhouse gases, removals of atmospheric CO_2 (e.g., via the photosynthesis of biomass), and the permanent storage of CO_2 (e.g., injection into geologic reservoirs) that make up the “NETs-in-industry” systems under consideration. The primary metric was the “net CO_2(eq)” balance of emissions and removals over the lifetime of the NET-in-industry system. When other greenhouse gases were included, they were characterised by their “100-year CO_2 equivalent” (CO_2eq) warning potential, that is, the amount of CO_2 that is estimated to result in an equal average amount of global warming during the 100 years after the greenhouse gas has been emitted to the atmosphere.

The process models provided the estimates of direct emissions of greenhouse gases from industrial production, as well as biofuel processing, CO_2 transport, and CO_2 storage. The process models were also the source of the inventory of material and energy inflows and outflows. This inventory was connected to the life cycle inventory database ecoinvent 3 [39], which provided estimates of greenhouse gas emissions in upstream and downstream supply chains, assuming present-day efficiencies. Finally, CO_2 emissions from product use or disposal, as well as CO_2 uptake by biomass were considered stoichiometrically.

6.3 Techno-economic assessment

The process models also served as the basis for the economic assessment conducted in **chapter 7**. The cost model used in this dissertation followed recently released guidelines for techno-economic assessment of CCS systems [41] that take into account the specific complexities of estimating costs for CO_2 capture, transport, and storage.

For capital expenses, the modelled inflows and outflows of each subprocess are used to estimate the scale of equipment needed if the technology were to be implemented in real life. Using these expected equipment sizes and literature values for equipment costs, a scaling methodology was used to estimate the potential total capital expenses of retrofitting the negative emission technology into the industrial process. The equipment costs were scaled to the expected equipment sizes, and additional factors were then added to estimate the cost of installation, ancillary construction such as buildings and access ways, labour, legal fees, financing, and unexpected costs (contingencies).

The operational expenses were the sum of the variable costs of material and energy inputs and waste disposal as well as the fixed costs of labour, maintenance, taxes, and financing. These operating expenses, which include the annualised cost of the capital expenses, were considered in terms of both the change in cost of producing the industrial product and in terms of the cost of CO_2 avoidance, that is, the increase in cost per unit reduction of CO_2 emitted. For the cost of CO_2 avoidance, the cost model was linked to the net CO_2eq metric of the life cycle assessment.

7 Dissertation outline

This dissertation is presented as follows:

Chapter 2: Defining negative emissions

Negative emission technologies are complex systems with many uncertainties. Of paramount concern is understanding the conditions in which they result in a net decrease of atmospheric CO_2. This chapter provides a literature review on the usage of the term “negative emissions”, clarify the critical role of system boundary choices in modelling negative emission technologies, and provide operationalised criteria for evaluating when “negative emissions technologies” can result in actual negative emissions.

Chapter 3: Overview of existing research on industrial applications of bioCCS

This chapter provides an overview of recent research on the use of bioCCS in the production of iron and steel, cement and concrete, ethanol and transport fuels, paper and pulp, and platform chemicals. It describes how bioCCS can be used in each of these sectors and discusses existing estimates of their decarbonisation potential and costs.

Chapter 4: Near-term implementation options for bioCCS in the chemical industry

This chapter models the deployment of bioCCS into the production of ethanol, hydrogen, ammonia, and urea as potential "low hanging fruit" options for industrial NETs. Each of these industries produce a concentrated stream of CO_2, facilitating CO_2 capture, and existing production is either already biogenic (in the case of ethanol) or has a feedstock, which can be potentially substituted with a "drop-in" biomass replacement.

Chapter 5: Comparing the potential for bioCCS in different steelmaking technologies

Steel, the largest industrial emitter of CO_2 is considered a "difficult to decarbonise" industry, as the dominant form of iron production, in blast furnaces, relies on coal not only for fuel, but to control the reactions of iron reduction. This chapter models the use of bioCCS in five steelmaking technologies, including the commercialised pathways of blast furnace steelmaking and the MIDREX-process direct reduction of iron, as well as the three novel steelmaking technologies of blast furnace iron making with top gas recycling, HIsarna smelt reduction, and ULCORED direct reduction.

Chapter 6: Timing of removals and emissions for bioCCS and accelerated carbonation in concrete

This chapter models the deployment of both bioCCS and accelerated carbonation in the production of Portland concrete. As both biomass and concrete absorb CO_2 over time, this chapter focuses on the timing of CO_2 emissions and removals over the life cycle of both bioCCS and concrete. Besides describing the decarbonisation potential of the technologies, this chapter highlights the trade-offs of increased near-term emissions from biomass use versus lower longer-term net CO_2 emissions after reabsorption by biomass and concrete.

Chapter 7: Scoping cost and abatement metrics for biomass with carbon capture and storage

Estimates of abatement potential, technological cost, and CO_2 avoidance cost depend not only on the assumptions of configuration, technology, and prices, but also on what the modeller chooses to account for. This chapter illustrates the variation in cost and abatement metrics possible from using different system boundaries seen in bioCCS literature and proposes guidelines for clearer and more comparable metric use for bioCCS studies.

Chapter 8: Conclusions

This final chapter summarises the findings of this dissertation by explicitly answering each of the research (sub)question. It also reflects on the process of comparative black box modelling, outlines the limitations of the research, and provides recommendations for future research as well as considerations for real world implementation of industrial NETs.

Enjoy!

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Chapter 2:
Defining negative emissions

Negative emission technologies (NETs) have seen a recent surge of interest in both academic and popular media and have been hailed as both a saviour and false idol of global warming mitigation. Proponents hope NETs can prevent or reverse catastrophic climate change by permanently removing greenhouse gases from the atmosphere. But there is currently limited agreement on what "negative emissions" are. The chapter highlights inconsistencies in negative emission accounting in recent NET literature, focusing on the influence of system boundary selection. A quantified step-by-step example provides a clear picture of the impact of system boundary choices on the estimated emissions of a NET system Finally, the chapter proposes a checklist of minimum qualifications that a NET system and its emission accounting should be able to satisfy to determine if it could result in negative emissions.

This chapter was originally published as Tanzer, S. E., & Ramírez, A. (2019). *When are negative emissions negative emissions?* Energy and Environmental Science, 12 (4). doi: 10.1039/c8ee03338b. The publication's electronic supplementary information includes the complete literature review in tabularised form.

Note: Spellings, formatting, and abbreviations have been standardised throughout the dissertation.

1 Introduction

Without immediate and comprehensive mitigation of anthropogenic greenhouse gas emissions, the prevention of catastrophic impacts from global warming may come to depend on the deliberate removal of massive quantities of greenhouse gases from the atmosphere. This concept of "negative emissions" gained increasing attention after its initial inclusion in the 4th IPCC assessment report in 2009 and then in the vast majority of integrated assessment models in the 5th report in 2014. The ambitious "well below 2°C" target of the 2015 COP21 Paris climate agreement may already be unachievable without negative emissions [1–4]. Indeed, all modelling scenarios in the 2018 IPCC special report on limiting global warming to 1.5°C rely on the removal of carbon dioxide from the atmosphere [5]. In a 2017 review [6], all included 1.5°C scenarios depended on permanently removing an annual 3 to 30 gigatonnes of CO_2 from the atmosphere—up to 80% of current global emissions—before the end of this century.

Some of the technologies designed to achieve negative emissions are based the encouragement of natural processes that uptake and store atmospheric carbon, such as afforestation (AF) [7, 8] and soil carbon sequestration (SCS) [7, 9]. Other negative emission technologies (NETs) rely on human engineering, such as capture and storage of CO_2 from the combustion of biomass for energy (bioenergy with carbon capture and storage, bioCCS) [7, 10], or the chemical removal of CO_2 directly from air [7, 11] and subsequent storage (direct air capture with storage, DACCS).

Achieving massive-scale negative emissions requires an unprecedented fast-tracking of technological development and an unprecedented level of cooperation between political, industrial, and consumer stakeholders [12, 13]. For while negative emission strategies are based on proven technological components, such as biomass cultivation, energy use, logistics, and gas storage, each of these components have financial costs, greenhouse gas emissions, and other environmental and social impacts. NETs rely on connecting these components into complex systems, further increasing risk and uncertainty [13]. An overarching necessity is to ensure that the total effect of all components within the complex system of a NET is the permanent removal of atmospheric greenhouse gases, and thereby a net decrease in the greenhouse gas concentration in the atmosphere.

If massive-scale negative emissions are to be achieved, a clear, comprehensive, and consistent definition of when negative emissions occur is a necessary prerequisite for the effective implementation of incentives, regulations, and accounting. However, this is not currently the case. The 2018 IPCC special report [5] defines "negative emissions" explicitly only as the "removal of *atmospheric* greenhouse gases," though long-term storage is a feature of all greenhouse gas removal technologies discussed. A recent report by the European chemical industry [14] argues that CO_2 use—including in fuels and other short-lived chemicals—can be counted as "negative emissions", regardless of the origin of the CO_2 or fate of the product. A proposed EU policy [15] for the emission accounting of manure-based biogas allows methane diverted from traditional waste treatment to be labelled "negative emissions". That is, even if the biogas is later combusted and the resulting CO_2 is released to the atmosphere, since the emissions were prevented from happening during the waste treatment process itself, they are considered "negative." The above examples each come from a document relevant to policy and industry decision makers and each example uses the term "negative emissions" to refer to a different concept, including the removal (and implicit storage of) atmospheric greenhouse gases, the utilisation of greenhouse gases in products, and the prevention or delay of greenhouse gas emissions.

The chapter shows that this lack of clear consensus is due to the use of different system boundaries when considering what to count as "negative emissions." The chapter reviews the variations in the explicit and implicit usage of the term "negative emissions" and related terminology in studies from 2014 to 2018. To clarify the impact of system boundary selection on the perceived emission balance of a NET, a simplified example is used to illustrate the differences in emission accounting for a hypothetical NET when different system boundaries are used. Finally, we propose an operational set of minimum criteria for evaluating whether a system could result in negative emissions.

2 Literature review methods

Recent peer-reviewed academic literature on negative emissions was collected via a Web of Science topic search on the terms "negative emission," "negative CO_2", "negative greenhouse gas", "CO_2 negative", and "carbon negative" from 2014 through June 2018. This search resulted in 433 citations, of which 147 were neglected; 31 for lacking peer-review, 14 for being inaccessible, and 102 for being on unrelated topics, such as carbon electrode design or short-term natural carbon fluxes.

In the remaining 286 studies, the use of the term "negative emissions" was evaluated on whether the usage encompassed:

- the physical removal of greenhouse gases from the atmosphere,
- the storage of atmospheric greenhouse gases, and whether the storage was specified to be permanent,
- whether the emissions associated with both the upstream and downstream supply chains of the negative emission technology (life cycle emissions) were considered, and

- whether other concepts were encompassed by the term, including the storage of non-atmospheric greenhouse gases, the re-emission of captured gases to the atmosphere, or the inclusion of avoided emissions.

Usage was evaluated first by any explicit definition provided and also by any clear implicit criteria. For example, if negative emissions were only referred to as resulting from technologies that store atmospheric greenhouse gases in geologic formations (e.g. bioCCS, DACCS), removal and permanent storage were assumed to be implicit criteria of that study's definition of negative emissions. Usage features for each paper were collected in a tally spreadsheet, which is provided in the supplemental information to the chapter.

3 Overview of the usage of negative emissions terminology in recent literature

Half of the 286 papers reviewed provided an explicit definition of the term "negative emissions" (or "negative CO_2", "negative greenhouse gas", "CO_2 negative", and /or "carbon negative," if those were used additionally or instead). **Table 1** shows that these explicit definitions were not always consistent. 143 (50%) of studies specified the removal of atmospheric greenhouse gas, but only 82 (29%) specified any sort of storage of the greenhouse gas. 23 papers (9%) considered negative emissions to be generated from processes that explicitly re-release the gas into the atmosphere in the short term, such as via conversion to fuel. A further 33 studies (12%) also explicitly considered negative emissions to come from processes that do not remove greenhouse gases from the atmosphere, such as carbon capture and storage (CCS) of fossil fuel emissions or emission reduction technologies. The full list of papers reviewed, tagged with usage features is available in the supplemental information as a sortable spreadsheet.

If implicit usage is also considered, a further 34% (84% of total) of the studies likely consider negative emissions to involve the removal of atmospheric greenhouse gases, and a further 44% (65% of total) likely include the permanent storage of greenhouse gases. However, there is high variance in how clearly these terms are used and, without an explicit definition, it is ambiguous whether these are intended as necessary or optional criteria of negative emissions.

The most consistent usage feature was that 70% (199) of papers state that purpose of negative emissions is to reduce global warming or, more specifically, to reduce atmospheric concentrations of greenhouse gases. Therefore, logically, the quantity of greenhouse gas in the atmosphere must be lower after NET use than before it. This requires not only that greenhouse gases are removed from and stored outside the atmosphere, but also ensuring that any greenhouse gases emissions that result from this process are not greater than the amount of greenhouse gases removed. Of the papers reviewed, only five [10, 16–19] (2%)

explicitly acknowledge that all emissions associated with the use of NETs, including those upstream and downstream of the removal process, are needed determine whether a technology actually results in in an overall decrease of atmospheric greenhouse gases. The system boundary selection example below illustrates the potential importance of these upstream and downstream emissions on the overall GHG balance of an NET system.

Table 1. Summary of results from the literature review on the usage of the term "negative emissions"[1]

Features of Usage	**Number of reviewed papers with feature**	**(% of total)**
States that the goal of negative emissions1 is to reduce global warming or the atmospheric concentration of greenhouse gases	199	(70%)
Provides an explicit definition of negative emissions that includes:		
the removal of greenhouse gas from the atmosphere	143	(50%)
the storage of the removed gases	82	(29%)
and specifying permanent storage	58	(20%)
an accounting of greenhouse gas emissions to the atmosphere that result from the use of negative emission technology	5	(2%)
Uses the term negative emissions[1] to include:		
the capture and/or storage of non-atmospheric greenhouse gases (e.g., from the combustion of fossil fuels)	17	(6%)
greenhouse gases that are explicitly re-emitted to the atmosphere	23	(9%)
greenhouse gases that would be prevented from being emitted to the atmosphere when compared to a reference scenario (avoided emissions)[2]	16	(6%)

For the full article list with usage features marked per article, please refer to the supplemental information of the published article.

1: including the alternate terms: "negative CO_2", "negative greenhouse gas", "CO_2 negative", and "carbon negative".

2: Including 11 of the 27 (41%) life cycle assessments papers that are in the literature review. For the full article list with usage features marked per article, please refer to the electronic supplemental information of the published article.

4 Avoided emissions and enhanced oil recovery

In 11 of the 27 (41%) life cycle assessment (LCA) studies included in the literature review, avoided emissions are labelled as negative emissions. However, while calculations for avoided emissions can result in negative numbers, they are distinct from the physical removal of greenhouse gases from the atmosphere, and a brief clarification of the distinction is warranted.

Avoided emissions are an estimation of emissions that are assumed to be potentially prevented by switching from a system of reference to the system studied in the LCA, based on specific assumptions of future system behaviour. They are a feature of a method to account for the emission-reduction potential of co-products that are produced in a system analysed by an LCA, known as "displacement" or "system expansion" [20]. As an example, in [21], a palm oil biorefinery is assumed to produce —among other products— ethanol and electricity. The study assumes that this ethanol and electricity directly replace gasoline and coal-based electricity, and therefore, if the biorefinery is in operation, these fossil fuels will not be used. It then follows that the greenhouse gas emissions attributable to the production and use of the gasoline and electricity from coal will also not be produced; these emissions are said to be "avoided". The study then subtracts these "avoided emissions" from the emissions of the biorefinery. As the resulting difference is a negative number, the biorefinery is said to result in negative emissions.

In short, the negative greenhouse gas emission numbers in these LCAs are not physical emissions. They are the potential reduction of emissions in a hypothetical scenario where a specific technology replaces another specific technology, and will change depending on the reference scenario selected. Avoided emissions refer to the potential of adding a smaller, but still positive, amount of greenhouse gas to the atmosphere. This is in contrast to how the term negative emissions is used in the context of pathways to reach 1.5°C mitigation targets, which refers to greenhouse gases that are physically removed from the atmosphere. Some LCAs [22, 23] further conflate these terms by lumping together physical removal and assumed avoidance of greenhouse gases while other LCAs simply use the term negative emissions to refer to avoided emissions without any removal of atmospheric greenhouse gases at all [22, 24, 25]. The full list of LCAs in the review that conflate the term negative emissions with avoided emissions is available in the supplemental information.

The term negative emissions is also sometimes used to refer to CCS applied to fossil fuels, particularly in papers within the field of enhanced oil recovery (EOR) [26–28]. In EOR, CO_2 is used to extract otherwise unrecoverable oil from otherwise depleted oil fields. Some EOR studies label the balance of CO_2 (CO_2 trapped in the geological formation minus CO_2 released when oil is combusted) negative emissions, regardless of the origin of the CO_2, which, in most cases, is either extracted from natural formations or from the flue gas from the combustion of fossil fuels. Storage of fossil CO_2 does not involve any removal of CO_2 from the atmosphere, and therefore cannot result in any decrease in atmospheric greenhouse gases. Furthermore, even when removed atmospheric CO_2 is used and permanently stored in the process of EOR, the CO_2 emissions from the use of the recovered oil can be greater than the atmospheric CO_2 removed and stored, thus leading to a net increase in atmospheric CO_2. In at least one study [29], the emissions from the combustion of the recovered oil —which otherwise would have remained in the ground— are excluded from the CO_2 balance, and the whole quantity of stored CO_2 is considered negative emissions.

5 How system boundaries selection matters for negative emissions

To illustrate the impact of system boundary selection on the estimated greenhouse gas emissions of a NET system, the following example looks at the way the emission estimate changes for a steel mill implementing bioCCS based on different boundary selection. The system itself, an overview of which is shown in **figure 1**, is the same in every case; it is only our perspective of it that changes, as indicated by the different system boundary lines.

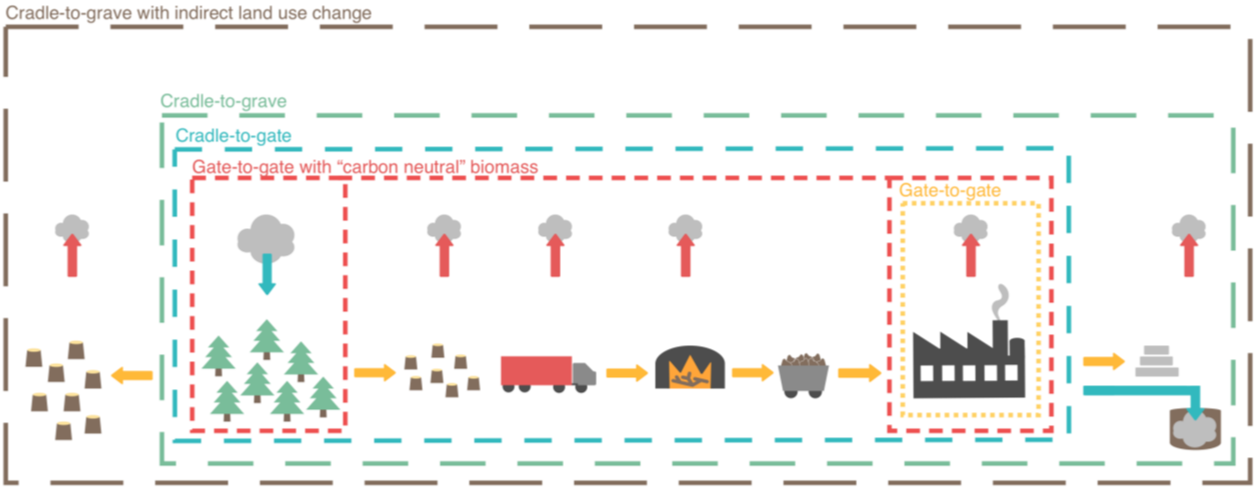

Figure 1. Different technology assessments boundaries applied to a bioCCS steel plant. A "gate-to-gate" system only considers the emission within the steel plant itself. Bioenergy assessment also often includes the uptake of atmospheric carbon by the biomass without also including the biomass processing and transport in a "cradle-to-gate" or "cradle-to-grave" system, the latter also including the impacts of product use and waste processing after they leave the steel plant. In bioenergy systems, unintended (or "indirect") land use change may also need to be included to achieve a full picture of the system impacts.

Figure 1 provides an overview of system boundaries common in technology assessment. A "**gate-to-gate**" system considers only the processes and emissions that occur within the steel plant itself. Studies on bioenergy often use a modified gate-to-gate boundary, that additionally includes an amount CO_2 removed by biomass from the atmosphere that is assumed to be exactly equal to the CO_2 emitted from its combustion, and thus the bioenergy is considered to be "carbon neutral." A "**cradle-to-gate**" system includes upstream emissions and resource use, such as land use, cultivation, harvest, transportation of biomass, and the production of other inputs, but nothing downstream of the factory gate, such as product use or waste treatment. The inclusion of both upstream and downstream emissions is a "**cradle-to-grave**" system. Since bioenergy systems often involve changes in land use that many not be temporally or geographically immediate to the cultivation or harvest of biomass, a further expansion of the boundaries to encompass indirect land use change (ILUC) is also used. The below example illustrates that without a "cradle-to-grave" perspective, it is not possible to determine whether the use of a NET will result in an overall decrease in atmospheric greenhouse gas concentration and thereby achieve negative emissions.

This example, illustrated in **figure 2**, considers a steel mill that first implements capture and geologic storage of its CO_2 emissions (CCS), and later also switches its energy source from coal to wood charcoal (bioCCS). For clarity, the example assumes a heavily simplified steel mill that produces one type of steel and derives all its energy and emissions from the combustion of one type of fuel. Since the focus of this example is CO_2 emissions, the mining of iron ore and use of the steel product are excluded. The quantities used in this example are heavily simplified and intended only for illustrative purposes. This example illustrates only a single possible configuration, and many other choices of technology, production methods, and transport are available. Furthermore, a full inventory of greenhouse gas emissions from the supply chain of steel production, charcoal, and CCS would be much more extensive, but is neglected for clarity.

Figure 2(a) and **(b)** show the steel mill as viewed from gate-to-gate perspective. In **(a)**, the steel mill produces one metric tonne (t) of steel using the energy from the combustion of 0.4 t of coal, which emits 1.0 t of CO_2 to the atmosphere. In **(b)**, the steel mill has installed CCS technology that captures 90% of the CO_2 produced at the mill. However, the energy required for carbon capture increases the mill's coal consumption to 0.5 t, thus increasing the total amount of CO_2 produced by combustion to 1.3 t. The CCS technology captures 1.2 t of this CO_2, which is then sent to for storage in a geologic formation. The uncaptured 0.1 t of CO_2 is still emitted to the atmosphere. Therefore, from a gate-to-gate perspective, the addition of CCS reduces the steel mill's atmospheric CO_2 emissions from 1.0 t to 0.1 t.

Figure 2(c)-(g) assume that the steel mill with CCS that has also switched its energy source from coal, a fossil fuel, to charcoal, a biogenic fuel. Fossil fuels contain carbon that has been removed from the carbon cycle for geologic time periods, and CO_2 emissions from fossil fuels increase the level of CO_2 into the atmosphere. In contrast, CO_2 emitted via the combustion of biogenic fuels contains carbon that was recently removed from the atmosphere via photosynthesis of growing biomass. Theoretically, if the biomass harvested for combustion is replaced by an equivalent amount of new planting, the replacement biomass will eventually absorb an equivalent amount of CO_2 from the atmosphere, resulting in a net zero addition of CO_2 to the atmosphere. In a system emitting fossil CO_2, the maximum impact of CCS is that emissions can be reduced to near-zero. If a system emits biogenic CO_2, it is possible to generate a flow of CO_2 from the atmosphere to some form of permanent storage, thus potentially generating negative emissions.

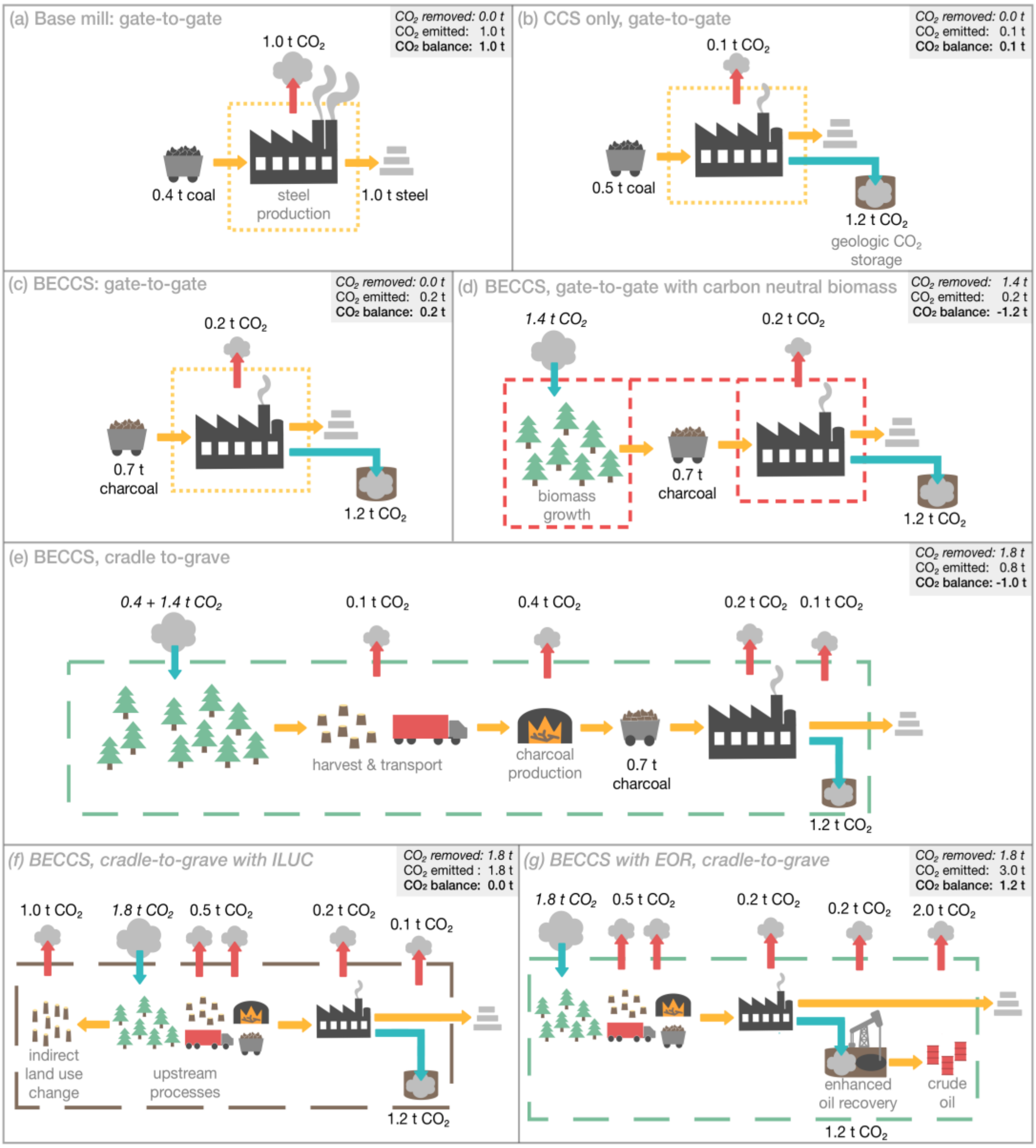

Figure 2. Perceived CO_2 emissions of a simplified steel production system when viewed from different system boundaries. The dashed line in each subfigure represents the system boundaries used to estimate the overall CO_2 emissions in the upper right corner of each figure. The system design and numbers used are heavily simplified for illustrative purposes. (a), (b), and (c) show the gate-to-gate CO_2 emissions of a steel mill, considering only the CO_2 produced at the mill itself for normal production (a), with the use of carbon capture and storage (b), and the use of bioenergy with carbon capture and storage (c). (d) expands the system boundaries to include the photosynthetic absorption of the exact amount of CO_2 released by the combustion; the assumption that the charcoal is “carbon

neutral." (e) shows a simplified "cradle-to-grave" system, including in its boundaries the CO_2 absorbed by the wood that is lost in the charcoal production process, the CO_2 emissions from biomass harvest and transport, the CO_2 emissions of charcoal production, and the CO_2 emissions CO_2 storage. (f) is a variant where the production of biomass has significant emissions from indirect land use change (ILUC). (g) is a variant where the geologic storage of CO_2 to the production and combustion of fossil fuels whose CO_2 emissions outweigh the CO_2 stored.

In this example, the charcoal has a lower energy content than coal, therefore 0.7 t is necessary to provide the same amount of power as the 0.5 t of coal in **(b)**. In Figure **2 (c)-(g),** the combustion of charcoal generates 1.4 t of CO_2, of which 1.2 t are captured and stored in a geological formation, and 0.2 t are uncaptured and emitted to the atmosphere.

Figure 2(c) looks at this bioCCS steel mill from a gate-to-gate perspective, which only considers the emissions at the mill itself. The biogenic origin of the charcoal is outside the system boundaries. From this perspective, the estimated emissions from the bioCCS mill are the 0.2 t of uncaptured CO_2, still 0.8 t less than the original mill, but 0.1 t more than the mill using coal and CCS.

In **figure 2(d)**, the system is extended to include the assumption that the charcoal used is "carbon neutral." That is, since the combustion of the charcoal resulted in generation of 1.4 t of CO_2 emissions, the charcoal is assumed to have been produced from biomass that removed exactly 1.4 t of CO_2 from the atmosphere. Therefore, from the perspective of a "gate-to-gate with carbon neutral biomass" system, a net 1.2 t of CO_2 is estimated to be permanently removed from the atmosphere via bioCCS.

Figure 2(e) takes a cradle-to-grave view of the bioCCS steel mill, including the upstream emissions of biomass harvesting, charcoal production, and transport, and the downstream emissions of CO_2 transport and storage. In **(d)**, it was assumed that biomass absorption of CO_2 was equal to the CO_2 it produces when it is combusted, neglecting any losses between photosynthesis and combustion. The emission accounting for the cradle-to-grave system includes these losses, which encompass an additional 0.4 t of CO_2 absorbed from the atmosphere that is re-emitted during charcoal production. Furthermore, biomass harvest and transport here use energy from fossil fuels, emitting 0.1 t of CO_2. For CO_2 transport and storage, 0.1 t of fossil CO_2 is emitted while providing the energy needed to transport, inject, store, and monitor the CO_2. Leakage of CO_2 from storage is assumed to be negligible. In total, the cradle-to-grave boundaries encompass 1.8 t of CO_2 removed from the atmosphere via photosynthesis, of which 1.2 t is captured after combustion for energy and stored in a geologic formation, and 0.6 t is emitted to the atmosphere during charcoal production and from CO_2 capture losses. Additionally, 0.2 t of fossil CO_2 is emitted to the atmosphere during the upstream processing of biomass and the downstream processing of CO_2. Overall, the cradle-to-grave perspective accounts for an additional 0.4 t of CO_2 removal and 0.6 t of CO_2

emissions than is estimated by using the gate-to-gate system boundaries of **(d)**. Here, a net 1.0 t CO_2 is estimated to be permanently removed from the atmosphere via bioCCS. Nothing in the system has changed, but more of the supply chain is now included in the boundaries used to estimate the emission balance.

Figure 2(f) is an example of the possible impact of indirect land use change (ILUC). ILUC is when a change in land use triggers unintentional changes in land use elsewhere [30, 31]. In this specific example, the charcoal is assumed to come from a forestry plantation that replaced a sheep pasture. The pasture owner then clears woodland elsewhere to replace the grazing space lost to timber production. The clearing releases the CO_2 stored by the woodland into the atmosphere, as well as removes the CO_2 storage capacity provided by the woodland. If this results in CO_2 emissions equivalent to 1.0 t CO_2 per tonne of steel, as in this example, the negative emissions seen in **figure 2(e)** are completely negated.

Figure 2(g) presents a variation where the CO_2 is permanently stored into a geologic formation after being used for enhanced oil recovery. Here, 1.2 t of CO_2 allows for the recovery of 0.6 t of crude oil, a co-product of the CO_2 storage [32]. The oil extraction and associated processes emit about 0.2 t of fossil CO_2 and the combustion of the 0.6 t oil emit about 2.0 t of fossil CO_2 [32]. Therefore, the total emission balance of the bioCCS+EOR system is 1.2 t of CO_2 *added* to the atmosphere.

Figure 2(c)-(g) all describe the same system of steel production with bioCCS, using the same amount of bioenergy, and permanently storing the same quantity of atmospheric CO_2. However, the estimated balance of emissions varies from 1.2 t of CO_2 removed to 1.2 t of CO_2 emitted, depending on which system boundaries are used and whether the upstream or downstream system generates indirect emissions. This dramatic variation for the exact same bioCCS installation underlines the importance of selecting inclusive system boundaries when estimating whether a technology or intervention will result in negative emissions. Quantified estimates of negative emissions should take into account, as fully as possible, all greenhouse gas removals and emissions in the cradle-to-grave system, including indirect emissions when pertinent (e.g., from indirect land use change or the combustion of system coproducts such as EOR oil). While any emissions estimate is limited by the available data, the use of as broad a system boundary as possible minimised the possibility of inconsistent or short-sighted system boundary selection leading to emission estimates that are misleading, contradictory, and possibly very wrong.

6 Further consideration for biomass-based NETs

As several NETs rely on the large-scale cultivation of biomass, it is relevant to briefly highlight the limitations of the above example with regard to biomass production and use, particularly as it only describes a single possible system configuration. In the above example, the bioenergy system of cultivation, harvest, processing, and combustion, by itself (excluding CCS) resulted in a positive balance of CO_2 emitted to the atmosphere. However, depending on the method of cultivation and processing, bioenergy can be carbon positive, carbon negative, or carbon neutral [33, 34]. Factors that influence the emission balance of bioenergy systems include the growth rate and harvest frequency of the biomass, the preparation of the land for biomass cultivation (direct land use change), the energy intensity and energy source for biomass harvest, transport, and processing, and the management of soil and biomass residues, among others [34]. Furthermore, while significant emissions from ILUC were included in the example for illustrative purposes, whether and how much land use change occurs, direct or indirect, is highly specific to the geographic considerations, such as existing available land and land use patterns, of each bioenergy system [35].

Besides the physical considerations of the biomass system, the accounting method can significantly influence the estimated emissions of a bioenergy system, particularly for slow-growth biomass such as forestry. In particular, as highlighted in [33], the geographic and temporal scale of the bioenergy system, whether CO_2 removals and emissions are assumed to be instantaneous or occur over time, and whether the time boundary begins at biomass planting or biomass harvest, can all substantially influence the emission balance. The development of emission accounting methods for bioenergy and biomass systems is an active area of research [33, 36–38].

7 Conclusions

The use of “negative emissions” terminology is not consistent in recent literature. Misinterpreting or miscounting negative emissions could have unintended, and possibly dangerous, consequences, such as policy incentives that reward increasing atmospheric greenhouse gas concentrations under the guise of negative emissions. While cradle-to-grave system analysis is not within the scope of all research on NETs, it is vital for researchers and decision-makers to be aware of the system boundaries they explicitly or implicitly use, and the limitations of those boundaries, particularly when estimating quantities of negative emissions. As shown in the simplified example above, emission negativity cannot be determined without accounting as fully as possible for all emissions and removals of greenhouse gases in the cradle-to-grave system. Based on the most common defining elements seen in explicit and implicit usage of the term “negative emissions,” and keeping in mind the goal of negative emissions—reducing atmospheric level of greenhouse gases—four key criteria can be considered “minimum qualifications” for determining whether a technology results in negative emissions:

1. Physical greenhouse gases are removed from the atmosphere.

2. The removed gases are stored out of the atmosphere in a manner intended to be permanent.
3. Upstream and downstream greenhouse gas emissions associated with the removal and storage process, such as biomass origin, energy use, gas fate, and co-product fate, are comprehensively estimated and included in the emission balance.
4. The total quantity of atmospheric greenhouse gases removed and permanently stored is greater than the total quantity of greenhouse gases emitted to the atmosphere.

While the above criteria require a cradle-to-grave system perspective for emissions accounting, they do not endorse a specific methodology for emission accounting, as evaluating the merits and limitations of the different accounting practices is outside the scope of the chapter. However, a clear distinction should always be made between physical negative emissions, as defined above, and the emission reduction potential of one technology in comparison to another (avoided emissions), which can appear as negative numbers in LCAs. The use of the term “negative emissions” for both physical removals and assumed avoidance has a particular risk for counterproductive misunderstanding in decision-making and incentive design.

Furthermore, the impact on atmospheric greenhouse gas concentrations is just one of several impacts that a negative emission technology could have that may affect global warming. Others include changes in albedo [39], the response of natural carbon sinks [40], or a rebound effect of increased consumption [41]. Additionally, other environmental impacts, such as biodiversity loss, acidification, and water use, also require consideration when evaluating the utility of a specific NET [39, 42]. It is also important to leave space for impacts that are currently beyond our knowledge—the unknown unknowns—and to adapt analysis as understanding of the impacts of negative emissions increases.

Finally, it should be emphasised that negative emission technologies are nascent and the scale on which they could be effectively implemented is uncertain. Preventing catastrophic climate change is a race against the clock requiring unprecedented levels of global cooperation and technological development. While it is imperative to develop long-term technological options such as negative emission technologies, they do not reduce the necessity of immediate and drastic reductions in global greenhouse gas emissions.

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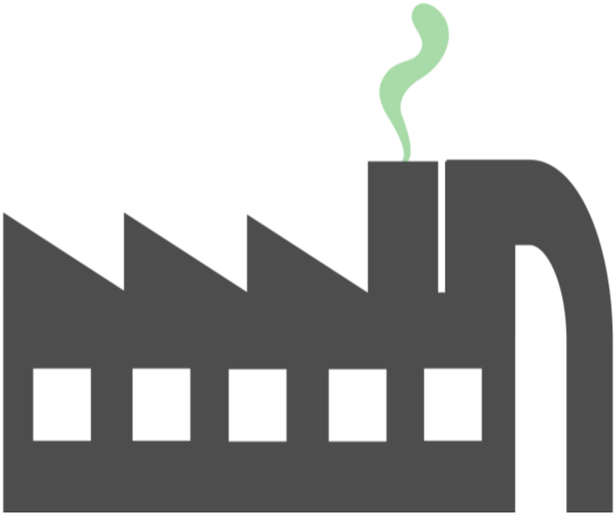
Chapter 3:
Overview of existing research on industrial applications of bioCCS



This chapter reviews recent literature on the combined use of bioenergy with carbon capture and storage (BECCS, or bioCCS) in the industries of steel, cement, paper, ethanol, and chemicals, focusing on estimates of potential costs and the possibility of achieving "negative emissions."

Bioethanol is seen as a potential near-term source of negative emissions, with CO_2 *transport as the main cost limitation. The paper industry is a current source of biogenic* CO_2*, but complex* CO_2 *capture configurations raise costs and limit bioCCS potential. Remuneration for stored biogenic* CO_2 *is needed to incentivise bioCCS in these sectors. BioCCS could also be used for carbon-neutral production of steel, cement, and chemicals, but these will likely require substantial incentives to become cost competitive. While negative emissions may be possible from all industries considered, the overall* CO_2 *balance is highly sensitive to biomass supply chains. Furthermore, the resource intensity of biomass cultivation and energy production for* CO_2 *capture risks burden-shifting to other environmental impacts.*

Research on bioCCS-in-Industry is limited but growing, and estimates of costs and environmental impacts vary widely. While negative emissions are possible, transparent presentation of assumptions, system boundaries, and results is needed to increase comparability. In particular, the mixing of avoided emissions and physical storage of atmospheric CO_2 *creates confusion of whether physical negative emissions occur. More attention is needed to the geographic context of bioCCS-in-Industry outside of Europe, the USA, and Brazil, taking into account local biomass supply chains and* CO_2 *storage siting, and minimise burden-shifting.*

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Note: Spellings, formatting, and abbreviations have been standardised throughout the dissertation.

1 Introduction

We emitted 59 billion tonnes (CO_2eq) of greenhouse gases in 2019 [1], yet limiting catastrophic climate change requires global emissions to be net-zero within the next few decades. Results from integrated assessment models (IAMs) indicate that, beyond rapidly reducing emissions, this transition will require permanently removing greenhouse gases (GHGs) from the atmosphere, or "negative emissions," to compensate for residual or historic emissions [2]. One of the most studied potential negative emission technologies is bioenergy with carbon capture and storage (bioCCS). In a bioCCS system, CO_2 is removed from the atmosphere via biomass, which is then combusted for energy. The resulting biogenic CO_2 is captured and permanently stored, such as in a geologic formation, and the biomass is regrown. BioCCS can result in negative emissions—that is, a decrease in atmospheric CO_2—if, and only if, more biogenic CO_2 is permanently stored than CO_2 is emitted throughout the supply chains of biomass cultivation and use and of CO_2 capture and storage.[1] [3 (**chapter 2**)]

IAMs typically assume bioCCS deployment in the power sector and/or for biofuel production [2, 4]. However, the industrial sector, responsible for 20% of global GHG emissions [1], including 8.5 Gt of CO_2/year [5], is a stronger candidate for near-term deployment. Currently, industry's bioenergy use is more than double that of power [6], and, so far, 95% of the CO_2 stored from large-scale CCS operations has been from industry [7]. Furthermore, industry is the expected source of many residual emissions in a net-zero society, as industry uses carbon as a feedstock, reducing agent, or other stoichiometric necessity, and while the use of bioenergy or CCS alone can significantly decrease CO_2 emissions, only in combination can they result in negative emissions.

In the past five years, 50 peer-reviewed papers[2] considered the combined use of biomass and CO_2 capture in the five largest CO_2-emitting industries: iron steel, cement, paper, platform chemicals, and transport fuels, whose CO_2 emissions and current status of biomass and CO_2 capture are summarised in **table 1**[3]. The papers reviewed broadly fall into three categories:

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[1] BioCCS systems do not *necessarily* result in negative emissions. Thorough cradle-to-grave accounting of GHG emissions and removals is a critical step to determine whether net removal actually occurs.

[2] See supplementary information for search queries used and descriptions of all literature reviewed.

[3] While transport fuels are often assigned the energy sector, they are included here as manufactured energy storage *products* is distinct from the on-demand provision of energy. Due to space constraints, smaller industries, such as beverage manufacturing and ceramic and glass production were excluded, as were industry-generic bioCCS options, such as bioCCS-hydrogen, bioCCS-boilers, and CHP.

1. Retrofitting CCS into existing biomass-based industries as an early opportunity for negative emissions, compensating for CO_2 emitted *elsewhere* in society.
2. Retrofitting bioCCS into carbon-intensive heavy industry, compensating for CO_2 emitted *during production*.
3. Integrating CCS into novel biobased production pathways for carbon-based chemicals (e.g., fuels, olefins), compensating for CO_2 emitted *during product use or disposal*.

In this work, we review the proposed configurations and challenges for bioCCS-in-Industry reported in these papers. We then discuss estimated costs and environmental impacts, focusing on the potential of negative emissions via bioCCS-in-Industry.

Table 1. Overview of major CO_2 emitting industries and their current use of bioenergy and CO_2 capture. [5, 7–9]

Industry	**Direct CO_2 emissions (2019, global)**	**Status of biomass use**	**Status of CO_2 capture**
Cement	2300 Mt, fossil 30-80 Mt, biogenic[1]	Commercial, with individual kilns firing up to 35-40% biomass, typically wastes [10]	Demonstration, up to 75 kt/year
Steel	2100 Mt, fossil	Commercial partial replacement of coal with charcoal. Primarily used in small-scale production in Brazil	Demonstration for blast furnace steelmaking. Commercial for direct reduced iron steelmaking.
Petrochemical refining	1400 Mt, fossil	Early commercialisation for methanol (1 facility) and biomass-to-liquids from biowastes (multiple facilities under construction)	Commercial for methanol and coal-to-liquids, up to 100 kt/year
Paper	200 Mt fossil 700-800 Mt biogenic[2]	Commercial. Process is inherently biobased. Residues used for cogeneration of heat and electricity	Demonstration, 11 kt/year [11]
Ethanol	82 Mt, biogenic[3]	Commercial. Process is inherently biobased, with maize and sugarcane as primary feedstocks. Sugarcane bagasse is used for cogeneration of heat and electricity. Early commercialisation of fermentation of cellulosic biomass.	Commercial for capture of high purity fermentation CO_2, including 1 Mt/year to dedicated storage

1: 3-6% biogenic fuel mix [10, 11], assuming 0.8-1.1 t CO_2/GJ [12].

2: For an approximate global production of 400 Mt pulp [13], assuming an average biogenic CO_2 intensity of 1.9 t CO_2/t pulp [14, 15].

3: Stoichiometrically, 82 Mt CO_2 from the fermentation of 86 Mt of ethanol in 2019 [14].

2 CCS for existing biogenic industries

Some industries already use biomass as a feedstock and emit biogenic CO_2 during production. Notably, the production of bioethanol and paper emit over 800Mt of biogenic CO_2 per year, not including CO_2 embodied in products. As such, the addition of CCS to these industries may by itself be sufficient to result in negative emissions.

The most discussed industry in the recent literature is bioethanol, often highlighted as a "low-hanging fruit" for bioCCS [15–27]. As the CO_2 released from ethanol fermentation is nearly pure (98%vol [24]), it could be prepared for transport and storage via compression alone. Currently, 1 Mt/year of ethanol fermentation CO_2 is injected into dedicated geologic storage in Illinois and three more CCS projects are under development [8].

Bioethanol is typically produced from maize, sugarcane, or other starchy food crops. Alternatively, cellulosic biomass, such as grasses and coppice wood grown on less-arable land or agricultural wastes, can also be fermented. Currently, only a few ethanol distilleries produce cellulosic bioethanol, primarily from maize and sugarcane residues [14, 28]. However, several recent bioCCS-in-ethanol studies envision dedicated facilities fermenting corn stover [29, 30], switchgrass [29–31], miscanthus [29, 32], and wood [29, 33], with captured CO_2 sent to dedicated geologic storage.

Not all CO_2 from bioethanol production is as easy to capture as the high purity CO_2 from fermentation. In Brazilian distilleries, sugarcane residues are combusted to cogenerate heat and electricity, producing up to 90% of total distillery CO_2 in dilute flue gas streams, the capture of which was explored by [16–18, 23], all assuming post-combustion amine-based capture, whose energy demand was estimated to reduce distillery electricity exports by 50-75% [16, 17, 23].

Pulp and paper mills also cogenerate heat and electricity, and the biogenic CO_2 from the combustion of process wastes typically accounts for over 75% of on-site emissions [34, 35]. Flue gases are typically less than 20% CO_2 and distributed between several point sources [34, 36–40]. Some studies estimated that energy demand of full CO_2 capture can switch paper mills from being net energy exporters to energy importers [39] or require supplemental fuel [38, 41]. If only on-site energy is used, estimates of capturable CO_2 ranged from less than 30% in [38, 40], to 90% (with an 80% reduction in electricity exports) in [41], for post-combustion amine-based capture. Two studies [35, 39] considered the integration of a calcium looping

CO_2 capture unit[4] into the lime kiln[5] of a pulp mill, which could lower the net energy intensity of CO_2 capture.

Despite these challenges, bioCCS-in-paper could be particularly significant in the USA, whose mills produces a quarter of the world's paper [13], with biogenic CO_2 accounting for over 115 Mt CO_2/year [38] and in countries like Sweden, where pulp and paper mills account for over 60% (ca. 20 Mt CO_2/year) of large-scale CO_2 emitters [36, 42].

3 Retrofitting bioCCS into carbon-intensive industries

BioCCS could also be used in industries that are large CO_2 emitters but are not currently major biomass consumers, such as steel and cement, which together emitted 5.0 Gt CO_2 in 2018 [6]. While low-carbon production technologies are under development, they will not be available on a large scale for a few decades [6]. Retrofitting bioCCS could allow existing steel mills and cement plants to continue operating at or near carbon neutrality.

Globally, over 70% of steel is produced in blast furnace mills [43] that use high-grade coal as a fuel and reducing agent, emitting around 2-3 t CO_2/t steel [44–46] from the blast furnace and associated energy production. CO_2 capture in steel has been considered by a number of studies [24, 47–49] and demonstration facilities [8], and the use of charcoal as a partial coal replacement is common in Brazil [6, 50]. However, as blast furnaces rely on the mechanical properties of coal as a process control mechanism, biomass replacement is likely limited to around 30% of coal use in current large blast furnaces [45, 51.]

Only five studies of the fifty studies reviewed considered bioCCS for blast furnace steelmaking [44, 46, 52–54]. They estimated that partial charcoal use with full CCS could reduce steel mill emissions over 80% but was unlikely to compensate for emissions from charcoal production or CO_2 transport and storage to allow for negative emissions. Still, bioCCS deployment at 30 EU steel mills could mitigate up to 200 Mt CO_2 per year [52]. However, this requires capturing CO_2 from most point sources within the mill. If capture is limited to the largest CO_2 source, the blast furnace itself, bioCCS has the potential to reduce direct CO_2 emissions by approximately 50% [46, 52].

Other steelmaking methods are more amenable to bioCCS. Direct reduction of iron (DRI), which accounts for 7% of global steelmaking [43], typically uses natural gas or gasified coal to reduce iron, and CO_2 capture can be integrated into reducing gas preparation. This is already

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[4] Calcium looping CO_2 capture works by cycling carbonation ($CaO + CO_2 – CaCO_3 + heat$) and calcination ($CaCO_3 + heat – CaO + CO_2$) to first remove CO_2 from a gas stream, and then, in an oxygen-environment, release a pure stream of CO_2 for capture.

[5] used to regenerate paper-making process chemicals.

the case at Emirates Steel in Abu Dhabi, where 0.8Mt CO_2/year is captured for use in enhanced oil recovery (EOR) [8]. Combined with CCS, a biogenic reducing gas [55, 56] could theoretically allow for "carbon negative" DRI steel [44, 46]. Similarly, bioCCS in smelt reduction steelmaking routes, such as Corex and the under-development HIsarna process, which are also more fuel-flexible than blast furnace steelmaking, could also allow for carbon-neutral or -negative steel [54].

Like steel, cement production is also CO_2 intensive. At a cement plant, roughly 60% of the CO_2 emitted results from the calcination of limestone. This fossil CO_2 is stoichiometrically unavoidable and bioCCS may be the only path to CO_2-neutral cement production [57, 58].

CO_2 capture at cement plants currently operate on scales of 50-75 kt CO_2/year [6], and demonstrations plants capturing 400-600 kt CO_2/year are under development [8]. Furthermore, cement kilns already partially cofire biomass or biogenic wastes. An estimated 3-6% of global kiln fuel is biogenic, with individual kilns co-firing up to 37% biomass [10, 59].

Despite this, only four studies in the past five years explicitly consider bioCCS-in-cement [54, 57, 58, 60]. [58] concluded that CO_2-negative cement and concrete are plausible via fully charcoal-fired cement kiln with post-combustion CCS. Two other studies concluded that partial biomass use with CCS can reduce emissions over 70% [54, 57].

4 BioCCS-integrated biochemical production

The chemical sector emitted 1.4 Gt CO_2 in 2018 from direct energy use and process emissions [6], but half of its carbon inputs leaves as products, such as fuels, fertilisers, and olefins, which then release CO_2 during use or disposal. Both CCS integration and biobased production are under development to reduce the net CO_2 of chemical production [6], and some biobased production pathways also integrate CCS into their designs, aiming for carbon-neutral [54, 61] or carbon-negative [62–68] production.

The majority of these studies focus on biomass gasification technologies [54, 61, 63–65, 69–71]. Biomass gasification breaks the biomass into its component parts (H_2, H_2O, CO, CO_2), followed by catalytic processes to reassemble these components into the desired hydrocarbons, such as diesel and kerosene [70] or methanol and olefins [64–66, 70]. As CO_2 removal is typically a necessary step before catalytic reassembly, capturing the CO_2 for storage represents a relatively minor addition to the proposed process. Two studies did not consider gasification, but used hydrogen separated from biogenic processes gases, requiring CO_2 removal [62, 67]. Most of these technologies are generally at an early stage of development, though currently two plants gasify biomass into methanol, and fossil-based CO_2 capture is commercialised in methanol production [6].

5 Costs of bioCCS-in-industry

Cost estimates from bioCCS literature are difficult to compare, as they embody widely varying assumptions regarding technical performance, technology maturity, system boundaries, financing, commodity pricing, coproduct sales, and carbon taxation[6]. **Table 2** summarises the abatement costs of bioCCS-in-Industry from the reviewed studies, in comparison with literature on CCS alone. When possible, costs of CO_2 capture were separated, but cost estimates were often not broken down into their components. Only one study [54] estimated costs across multiple industries. Their estimates for bioCCS integration into steel, cement, transport fuels, and pulp, ranged between 50-90€$_{2020}$/t CO_2 avoided. However, underlining the difficulty of direct comparison, their CO_2 abated includes emissions from upstream fossil and bioenergy supply chains, unlike most other studies but did not include distance-specific transport costs.

Table 2. CO_2 abatement cost estimates of bioCCS-in-industry, compared to cost estimates for CCS-in-industry, €$_{2020}$/t CO_2[7]. Values in parentheses refer to cost of CO_2 capture only

	BioCCS, this review (*CO_2 capture only*)		**BioCCS, [72]**	**CCS only, [73]**	**CCS only, [54]**	**CCS only, [47]**	**CCS only, [24]**	**CCS only, [74]**
Ethanol, fermentation CO_2 only	22-388 *(11-31)*	[17–21, 26, 75] *[15, 18, 24, 25]*	20-180	-	-	-	13	-
Ethanol, fermentation and cogeneration CO_2	47-120 *(13-115)*	[17, 18, 30, 75] *[18, 31, 75]*	-	-	-	-	-	-
Paper	82-95 *(41-110)*	[36, 54] *[34, 36–39, 42]*	20-70	55-87	26-91	56-58	-	-
Steel	61-200	[44, 52, 54]	-	62-69	26-193	10-118	30-34	35-60
Cement	55-88	[76]	-	55-110	10-132	17-163	25-40	30-65
Drop-in transport fuels	68	[62]	20-40	-	-	-	-	-
Olefins and mixed chemicals	13-102	[54, 71]	-	153-200	23-230	28-247	96	35

The wide uncertainty in costs estimates is also a function of sparsity of bioCCS-in-Industry studies as well as the need to incorporate multiple system changes—bioenergy use, CO_2 capture, and CO_2 transport and storage—whose individual uncertainty is compounded by

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[6] A breakdown of what each study's cost estimates include is available in the Supplementary Information of the published version of this chapter (DOI: 10.1007/s40518-021-00195-3.)

[7] Costs have been standardised to €$_{2020}$ by first adjusting for inflation in the source currency and then converting to Euros. If no basis-year was provided, the annual average for the year preceding the publication year was assumed.

their interaction. Nevertheless, we can discuss the influential cost components seen in the recent literature.

Biomass price: Wood-based biomass was used in 30 of the 41 studies that were not about sugarcane or maize ethanol. Prices ranged from 0-8.6€$_{2020}$/GJ for forestry and mill residues [27, 38, 52, 62, 63, 69], to 1.9-7.5€$_{2020}$/GJ for wood chips and stem wood [27, 34, 39, 55, 61, 62, 64, 67, 70], to 7.2-15.4€$_{2020}$/GJ for charcoal and torrefied wood [44, 54, 71, 77]. Currently, global export prices of wood chips are 4-8€$_{2020}$/GJ [78], and biomass pellet prices in the USA and EU are 10-22€$_{2020}$/GJ [79, 80]. As biomass demand increases, however, prices of sustainably produced biomass are likely to increase.

CO_2 capture: Capture costs typically include the cost of equipment, labour, chemicals, and energy to capture and compress CO_2 so that it is transport-ready. Capture costs ranged from 3-30€$_{2020}$/t CO_2 [15, 19–21, 24–26, 30] for near-pure fermentation CO_2, to 42-110€$_{2020}$/t CO_2 for complex configurations that use amine-based solvents to capture CO_2 from multiple dilute streams, such as in paper mills [36].

CO_2 transport: In papers that assumed fixed CO_2 transport costs, those values ranged from 5-17€$_{2020}$/t CO_2 [34, 36, 39, 54, 54, 61, 71]. In studies that calculated transport costs on volume and distance, the range was much wider: 5-380€$_{2020}$/t CO_2 [17, 19–21, 26, 30, 38, 42, 52, 69], varyingly accounting for topography, existing land use, compression boosting, seasonality of biomass, shared pipelines, or multi-modal transport. However, in only four of these studies, all on Brazilian bioethanol production, was it possible to decompose costs by distance, with average costs typically between 0.2-0.4€$_{2020}$/tkm CO_2, with higher costs typically the result of low volumes transported over long distances [17, 19, 20, 69]. The use of intermediate pipeline hubs [19, 20, 69], short-distance truck transport for low-volume distilleries [19], and shared capacity with CO_2 captured from fossil sources [18] all led to lower transport costs estimates.

Tax on fossil carbon: Beyond absolute costs, an important factor is the cost of bioCCS relative to the cost of fossil-based production. In several studies [42, 57, 70, 81, 82], an estimated 70€$_{2020}$/t CO_2 tax on fossil emissions was necessary for bioCCS processes to be considered cost-competitive with fossil ones. Alternatively, several bioCCS studies on drop-in biofuels [61, 63, 67, 69] estimated the crude oil price necessary for the biofuels to break even, typically between 120-180€$_{2020}$/bbl.

Credits for stored (biogenic) CO_2: Existing biobased industries may not emit enough fossil CO_2 to be financially impacted by a fossil carbon tax. Therefore, several studies considered compensation for stored CO_2. One proposal is tradable “negative emission credits” [30, 34, 39] for stored biogenic CO_2, which can be sold to CO_2 emitters as offsets on emission trading networks. Another option is subsidies for stored CO_2, such as the 45Q scheme in the USA, which provides up to $50/t CO_2 stored, regardless of CO_2 origin. Sanchez et al [25] estimated that a $50/t CO_2 credit would be sufficient to incentivise the storage of 20-25 Mt/year of CO_2

from bioethanol distilleries, but for most distilleries an additional \$20-40/t CO_2 credit would be necessary to cover transport costs [26, 38]. Higher credits would be needed to incentivise may US paper mills as \$50/t CO_2 may be insufficient to cover even the costs of CO_2 capture alone [38].

6 Achieving negative emissions via bioCCS-in-industry

38 of the bioCCS-in-Industry studies claimed their system could result in negative emissions, but few provided sufficient detail to estimate if negative emissions occur. As negative emissions are intended to physically decrease GHGs in the atmosphere [2], they require that, as stated in [3]:

> 1. *Physical greenhouse gases are removed from the atmosphere.*
> 2. *The removed gases are stored out of the atmosphere in a manner intended to be permanent.*
> 3. *Upstream and downstream greenhouse gas emissions associated with the removal and storage process, such as biomass origin, energy use, gas fate, and co-product fate, are comprehensively estimated and included in the emission balance.*
> 4. *The total quantity of atmospheric greenhouse gases removed and permanently stored is greater than the total quantity of greenhouse gases emitted to the atmosphere.*

Estimating negative emissions requires scrutinising the complete systems of biomass production and use and carbon capture and storage. Only 9 of the bioCCS-in-Industry papers performed cradle-to-grave life cycle assessment [30, 32, 33, 46, 58, 60, 61, 63, 67], though a further 9 considered a “cradle-to-gate” system, including impacts of upstream biomass and energy production, but not end of product life or CO_2 transport and storage [29, 31, 44, 54, 62, 64, 65, 71, 75].

Many of the bioCCS-in-Industry studies that claimed to result in negative emissions added together estimates of net permanent storage of atmospheric CO_2 with estimates of avoided emissions from bioCCS products replacing fossil-based production [29–31, 54, 60, 61, 63, 67, 68, 83]. However, avoided emissions refer to an assumed *relative* change in emissions from one system to another, while negative emissions are an *absolute* reduction in CO_2 in the atmosphere via the *physical* removal and permanent storage of atmospheric CO_2. Caution is needed when interpreting such negative numbers to determine whether they actually represent net physical removal of atmospheric CO_2.

Estimates of GHG emissions from biomass supply chains ranged from 32-173 kg CO_2eq/t biomass and varied with biomass type, cultivation technique, transport method and distance, and greenhouse gases considered [16, 33, 44, 46, 54, 58, 61, 63, 64, 71, 84], with the lowest emissions for residual biomass and the highest for charcoal or torrefied pellets. Biomass system emissions are also challenging to estimate due to the variability of land use change and change in soil carbon stocks, which few studies included. In Field et al [31], converting forest to switchgrass production for cellulosic bioethanol released CO_2 both from the destruction of forest and loss of soil carbon, resulting in higher CO_2 emissions than uninterrupted forest growth. However, in the bioCCS system, the estimated biogenic CO_2 stored via CCS was more than double the total carbon storage of continued forest growth, even when considering indirect land use change. In [29] replanting marginal land with native grasses for use in bioCCS ethanol or electricity production was estimated to result in net carbon storage from both CCS and from increased soil carbon stocks. In contrast, in Fan & Friedmann [44], the inclusion of land use CO_2 emissions nearly negated the original estimated decarbonisation of bioCCS-in-steel.

With regard to downstream impacts, in the studies that separated emissions from CO_2 transport and storage [46, 58, 63, 67], estimates ranged from 5-20 kg/t CO_2 for pipeline transport to dedicated geologic storage and were not a major contributor to total emissions. However, not all studies assumed that the CO_2 was sent to dedicated geologic storage. Several studies assumed that the CO_2 would be used in enhanced oil recovery [16, 18, 19, 23, 27, 61]. While EOR does lead to geologic storage of injected CO_2, it also leads to CO_2 emissions from the extracted oil, which was not considered in any of the studies. While it is possible for EOR systems to store more CO_2 than is emitted by the recovered oil, if the system is designed to maximise permanent CO_2 injection [85], that is not typically the case [86–88], and CO_2 emitted by recovered CO_2 would mute the potential “negative emissions” from bioCCS systems.

Geologic storage of CO_2 is likely to store CO_2 for millennia [89] and can be considered effectively permanent. Carbon storage in concrete [58] or buried biochar [77] may also result in long-term storage, though biochar carbon may be partially re-released over time, and carbon storage in concrete is dependent on how the concrete is disposed. In contrast, carbon in short-lived products such as urea, paper products, or olefins, as considered in [40, 55, 64–66] will re-release CO_2 during use or disposal, and thus carbon in these products should not be counted towards negative emissions.

Timing of CO_2 storage and emissions is also relevant to upstream biomass cultivation. Biomass for bioenergy is typically combusted shortly after harvest, and CO_2 is then reabsorbed by replacement biomass, allowing CO_2 from biomass combustion to be part of the short-term carbon cycle. However, while biomass regrowth can be 1-2 years for grasses or 5-10 years for coppiced or fast-growing tree species such as eucalyptus or poplar, common boreal species such as Scots pine or Norwegian spruce take 50-100 years to mature, and CO_2 emitted from their combustion contributes to global warming for decades [90, 91]. In [58]'s models of bioCCS-in-concrete, the bioCCS systems resulted in higher atmospheric CO_2 than a fossil-based CCS system for up to a third of the biomass's rotation period and carbon-negativity was not reached until the after the biomass had been regrown (and CO_2 was reabsorbed by concrete), 50 years after the concrete was produced

Beyond global warming, in the four studies that look at other environmental impacts [22, 32, 33, 60], the bioCCS system resulted in higher acidification, human toxicity, ecosystem toxicity, water depletion, eutrophication, and ozone depletion compared to fossil-based production. These higher impacts resulted from the land and water use of bioenergy production, particulate matter and NO_X formation of biomass combustion, and the energy use of CO_2 capture. However, these studies only considered variations in the industrial production system; options for decreasing burden shifting in the bioenergy or CCS systems were not considered.

7 Conclusions

As both bioenergy and CCS are more developed in industry, industry is a likely candidate for near-term bioCCS implementation. In particular, bioethanol is a potential early source of negative emissions, as fermentation CO_2 can be cheaply captured. However, when bioethanol plants are far from geologic storage, transport network design is of crucial concern to costs. Pulp and paper mills represent the other major existing biogenic industry, but CO_2 capture is likely to be costly due to the complex configuration to capture multiple point-sources of dilute CO_2. BioCCS could also be retrofitted into the carbon-intensive production of steel or cement while low-carbon production technologies are developed. CCS integration into novel biobased chemical production pathways also allow for carbon neutral production of short-lived carbon-based products, such as olefins or transport fuels.

Many uncertainties remain about bioCCS-in-Industry, which is predominantly a prospective technology. However, interest is growing, with 16 studies published in 2020 alone, the same as in 2016-2018. From the studies available, it is clear that bioCCS-based production will require fossil carbon taxes as well as incentives for biogenic stored CO_2 to be cost-competitive on the global market. Furthermore, while bioCCS can reduce GHG emissions, achieving negative emissions is sensitive to specific system configurations and assumptions, and requires thorough and accurate assessment of emissions across the biomass and CCS supply chains.

While ongoing research on separate CCS and bioenergy use in industry and on bioCCS-in-Power will benefit bioCCS-in-Industry, we emphasise the following research needs for bioCCS-in-Industry:

- Life cycle assessment of bioCCS-in-Industry configurations outside of Europe and the Americas, and particularly in centres of industrial production in China and India, that take into account local availability of biomass and CO_2 storage.
- Evaluation of the logistical impacts of retrofitting both combined biomass and CO_2 capture at industrial facilities, particularly on space demand, heat recovery, and siting relative to both biomass and CO_2 storage.
- System designs that incorporate optimisation of both biomass production and CCS supply chains to minimise environmental burden shifting.
- Interactions and optimisation between bioCCS and other decarbonisation options available to industry, taking into account the timing of investment decisions, technological change, and received benefit.
- The incorporation of bioCCS-in-Industry into IAMs, using industry and geography-specific parameters and limitations.

As estimates of costs and environmental impacts of bioCCS systems are highly sensitive to studies' assumptions, it is crucial that these assumptions as well as system boundaries are clearly documented. bioCCS-in-Industry studies should ensure that they account for all carbon in their system and refrain from estimating negative emissions without a cradle-to-grave life cycle assessment. Avoided CO_2 should be accounted for separately from CO_2 that is physically and permanently removed the atmosphere. Furthermore, CO_2 avoidance cost estimates explicitly state both what costs and CO_2 emissions are included, and provide clearly decomposed costs of CO_2 capture, transport, and storage to facilitate comparisons between studies. Finally, environmental impacts beyond GHG emissions need more attention, taking into account the local context of biomass cultivation and CO_2 fate.

BioCCS is not a substitute for immediate and rapid decarbonisation of industry via increased efficiency, novel production methods, and, above all, reduced consumption and waste. Rather, the judicious use of bioCCS can allow for limited continued use of fossil carbon or limited removal of historical CO_2 from the atmosphere. With or without bioCCS, the transition to a "net-zero" society requires confronting the hard limits of our resource-constrained world.

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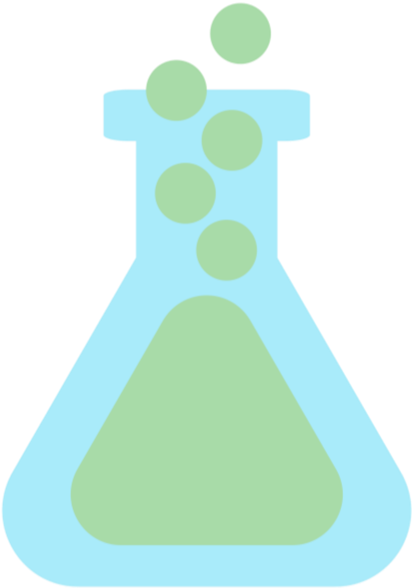
Chapter 4:
Near-term implementation options for bioCCS in the chemical industry



The chemical sector is hydrocarbon intensive, using primarily fossil fuels as both fuel and feedstock. To achieve carbon-neutrality, it is likely that negative CO_2 emissions will be needed to offset the carbon embodied in chemical products. This chapter presents first-order estimates of the decarbonisation potential of combining bioenergy and biofeedstock use with carbon capture and storage (bioCCS) for ethanol, ammonia, urea, and hydrogen. For each, net CO_2 of emissions minus atmospheric removals was estimated over the whole life cycle including chemical synthesis, upstream supply chains, product use, and waste disposal.

With aggressive bioCCS using technologies that are currently commercially available, CO_2 negative production was estimated to be possible for all chemicals modelled, except urea. With the use of biomass for both feedstock and fuel and capture of both high-purity and dilute CO_2 streams, the estimated net CO_2 was -30 g/MJ for maize bioethanol; -50 g/MJ for stover bioethanol; -50 g/MJ for merchant hydrogen; -1.2 t/t N for ammonia and 0.2 t/t N for urea. The potential for negative CO_2 emissions is higher in cases where more CO_2 can be captured during chemical production. However, all cases were sensitive to assumptions regarding the specific configuration and upstream supply chains.

This chapter has been expanded from a paper presented at 15th International Conference on Greenhouse Gas Control Technologies, which was published in the conference proceedings as: Tanzer SE, Blok K, Ramirez Ramirez A. *Negative Emissions in the Chemical Sector: Lifecycle* CO_2 *Accounting for Biomass and CCS Integration into Ethanol, Ammonia, Urea, and Hydrogen Production*. 15th International Conference on Greenhouse Gas Control Technologies, GHGT-15. 2021. doi: 10.2139/ssrn.3819778. The raw dataset for this chapter is available at doi: 10.5281/zenodo.6369409.

1 Introduction

The chemical sector is responsible for 30% of global industrial energy use, 85% of which is hydrocarbon based [1]. As half of these hydrocarbons are used as feedstock, for the chemical sector to reach CO_2 neutrality, it must address not only CO_2 emissions from fuel combustion, but also from the synthesis, use and disposal of chemical products.

One pathway to a CO_2 neutral chemical sector is the integration of bioenergy and biofeedstock along with carbon capture and storage (bioCCS). Theoretically, the permanent storage of biogenic CO_2 can result in "negative CO_2 emissions"—the net removal of CO_2 from the atmosphere—that could help compensate for the carbon embodied in the chemical products. Negative CO_2 emissions require that the quantity of atmospheric CO_2 removed and stored is greater than the CO_2 emissions in the life cycle of the bio-CCCS technology system [2 (**chapter 2**)].

There is substantial research and early commercialisation of the integration of biomass and/or CCS in various chemical production pathways, e.g. [3–7]. However, there are no studies that compare the near-term decarbonisation potential of bioCCS into different production pathways from a life cycle perspective. This chapter therefore provides comparative first-order estimates of the decarbonisation potential of bioCCS for "low hanging fruit" chemicals, including first- and second-generation bioethanol, ammonia and urea, and hydrogen, and only considers biomass and CO_2 capture options that are currently commercially available and could plausibly be retrofitted into existing chemical production installations. These chemicals were selected as near-term possibilities for bioCCS integration as all generate a high-purity CO_2 sidestream, and CCS has been studied or applied [6–8] for all.

2 Methods

For each chemical production pathway, a process model was create using a custom Python3 library (described in **appendix A**) to generate mass balances for the selected configurations with and without the integration of (additional) biomass use and/or CCS, following the modelling procedure described in **chapter 1**.

For each chemical pathway, the following cases were considered:

- A **base** case, representing the existing chemical production process without any intervention.
- A **biomass** case, where biomass replaces fossil fuels, both as a feedstock and as fuel to generate process heat.
- A **high purity CCS** case, where high-purity streams of CO_2 produced as a by-product of the chemical process are captured, compressed, and sent to geologic storage. In all

cases except hydrogen, this CO_2 by-product stream is sent directly to compression, without the need for further purification. In the hydrogen case, the high-purity stream requires additional purification before compression.

- **A full CCS** case, which captures the dilute streams of CO_2 (e.g., flue gases from steam generation), and along with the high-purity CO_2 streams, are compressed and sent to geologic storage.
- A **biomass with high purity CCS,** combining the features of the bioenergy and high purity CCS cases.
- A **biomass with full CCS** (full bioCCS), combining the features of the bioenergy and full CCS cases, with biomass also used to provide heat for CO_2 capture reboiler duty.

Table 1 summarises key process model parameters for each chemical pathway. As urea is produced from ammonia and CO_2, typically from ammonia production, it is not included separately in this table. Full model parameters are provided in the **appendix** to this chapter.

The process models for each chemical pathway and the assumptions regarding biomass and CCS use are briefly elaborated below. The choice of biomass and CCS technologies were selected for their current commercial availability and retrofitability. In particular, CO_2 capture by chemical absorption via MEA was included as it remains the most studied solvent, though the development of more advanced solvent is underway [9]. It allows for a conservative estimate of energy consumption (as reducing energy demand of solvent regeneration is a major focus of solvent development), which allows for a more conservative estimate of whether CO_2-neutral or -negative production is possible.

Determining whether negative CO_2 emissions can be achieved, and thus whether CO_2-neutral or -negative chemical production is possible, requires scrutiny of the CO_2 flows throughout the entire cradle-to-grave system to ensure that atmospheric CO_2 is permanently stored, and that total CO_2 emissions do not exceed the permanent removal of atmospheric CO_2. In this chapter, the life cycle CO_2 accounting includes upstream CO_2 emissions from supply chains of process inflows and downstream CO_2 emissions of product use and disposal. We assumed that the carbon contained in the product, by-products (e.g., fermentation digestate), and wastes was emitted to the atmosphere as CO_2 during use or disposal. Product transport was excluded, due to its dependence on the specific product use case but should be included when evaluation a real-world implementation of bioCCS.

For CO_2 emissions outside our process model, data from the life cycle inventory database ecoinvent 3.6 [10] was used to represent the average CO_2 emissions of supply chains of system inputs, including the production of material inputs, energy use, transportation, and the extraction of natural resources, including emissions of fossil, biogenic, and direct land transformation CO_2. Estimating impacts of indirect land use change was considered outside the scope of this chapter. The selection of data for upstream emissions follows the assumption that production occurs in northwest Europe, but otherwise this chapter was geographically generic. The life cycle inventory values used in this chapter are provided in the appendix.

Our target metric is the net CO_2 balance of each system, which is the CO_2 removed from the atmosphere by biomass minus CO_2 emitted to the atmosphere during the chemical production process, biomass preparation, CCS, and in the upstream and downstream supply chains. For clarity, our model only considered physical emissions and removals of CO_2 and did not include avoided emissions or displaced CO_2.

Table 1. Feedstocks and energy demand for each chemical pathway

Product and main data sources	**Feedstock Type, reference case**	**Feedstock Demand,** ***t per t product***	**Thermal Energy Demand,** ***GJ per t product***	**Electricity Demand,** ***kWh per t product***
Ethanol, starch [11]	Maize (45%C, dry basis)	2.7	6.2	228
Ethanol, lignocellulosic [12, 13]	Maize stover (49% C, dry basis	4.0	8.6	768[1]
Ammonia[2] [8]	Natural gas (93% CH_4)	0.5	9.0	139
Hydrogen [6]	Natural gas (93% CH_4)	2.9	23.1	135[1]
Biomethane [16]	Mixed agricultural and food wastes (50% H_2O, 27% C, wet basis)	8.6	1.4	812

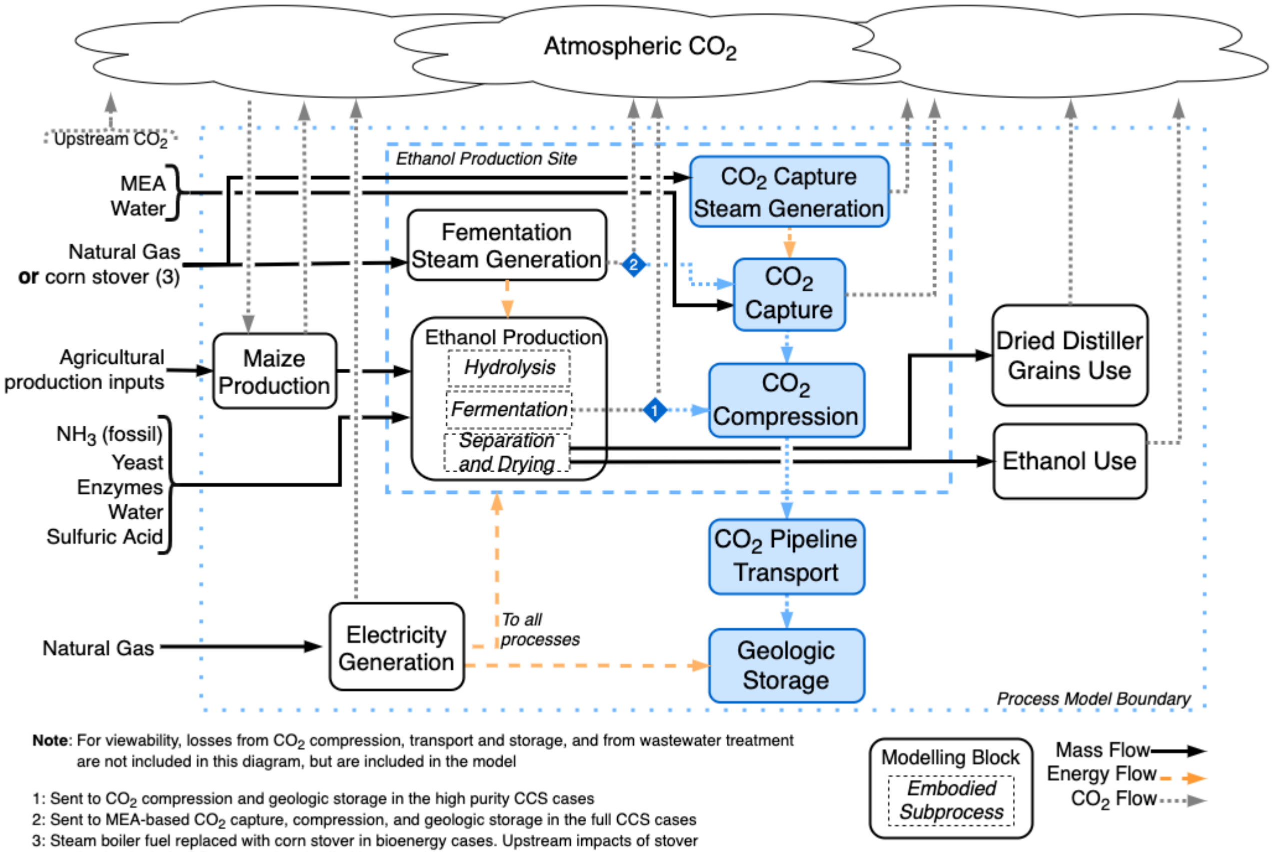
1: Cogenerated.

2: This model is also used for the ammonia and co-produced CO_2 inputs for urea production.

2.1 Bioethanol

As 60% of global bioethanol is produced via the fermentation of maize [17], dry-milled production of maize bioethanol was selected for the reference case, summarised in **figure 1**, using production parameters from [11]. A pure CO_2 stream results from fermentation of starch into ethanol. Unfermented carbon in the maize is assumed to be embodied in the dried distiller grains. The carbon in the ethanol and dried distiller grain products is assumed to be fully combusted or decomposed into CO_2 and emitted to the atmosphere.

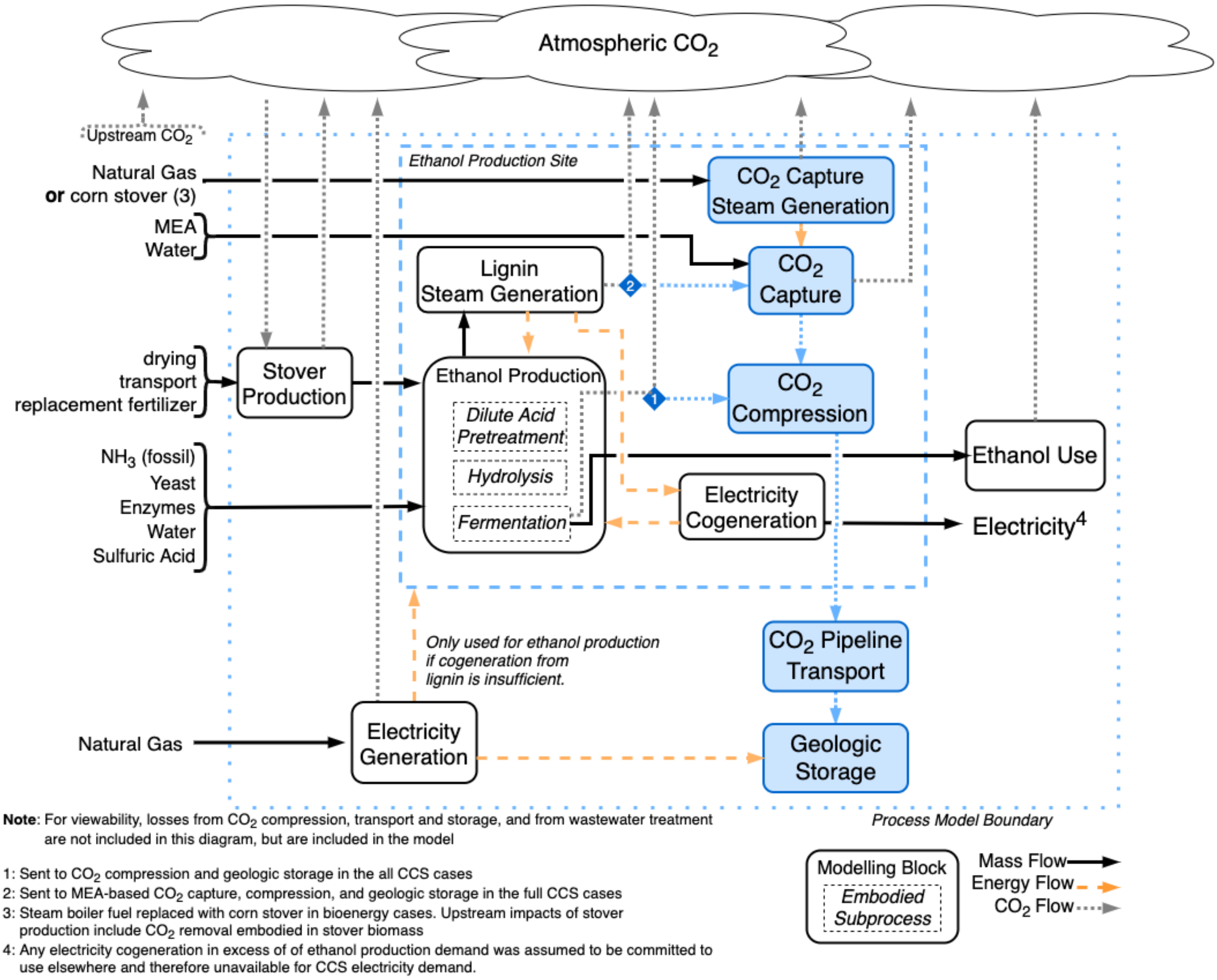
Additionally, lignocellulosic biomass is a potentially valuable feedstock for bioethanol production, as it is less likely to compete with food production, though it requires additional pre-processing to separate its fermentable cellulose from its lignin. In our model, summarised in **figure 2**, corn stover was chosen as the lignocellulosic feedstock, with dilute acid pre-treatment prior to hydrolysis and fermentation, and used the production parameters from the underlying stoichiometric model from [12]. The separated lignin was assumed to be recovered and combusted to provide heat and excess steam used to generate electricity.



Note: For viewability, losses from CO_2 compression, transport and storage, and from wastewater treatment are not included in this diagram, but are included in the model

1: Sent to CO_2 compression and geologic storage in the high purity CCS cases
2: Sent to MEA-based CO_2 capture, compression, and geologic storage in the full CCS cases
3: Steam boiler fuel replaced with corn stover in bioenergy cases. Upstream impacts of stover production include CO_2 removal embodied in stover biomass

Figure 1. Process model system for corn starch ethanol production



Note: For viewability, losses from CO_2 compression, transport and storage, and from wastewater treatment are not included in this diagram, but are included in the model

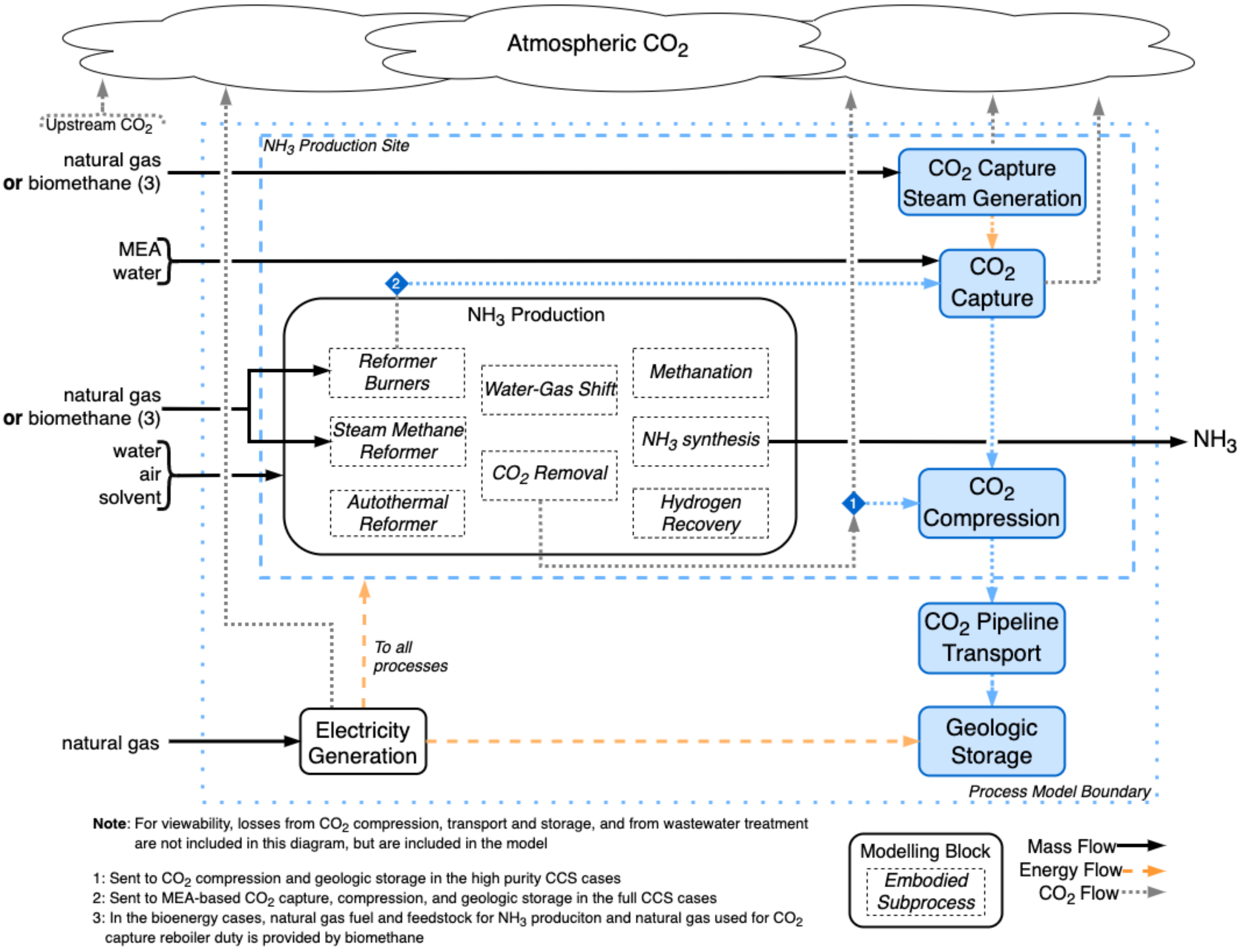
1: Sent to CO_2 compression and geologic storage in the all CCS cases
2: Sent to MEA-based CO_2 capture, compression, and geologic storage in the full CCS cases
3: Steam boiler fuel replaced with corn stover in bioenergy cases. Upstream impacts of stover production include CO_2 removal embodied in stover biomass
4: Any electricity cogeneration in excess of of ethanol production demand was assumed to be committed to use elsewhere and therefore unavailable for CCS electricity demand.

Figure 2. Process model system for corn stover lignocellulosic ethanol production

2.2 Ammonia and Urea

The modern ammonia synthesis process combines nitrogen from air with hydrogen, typically reformed from natural gas. The model in this chapter is based on [8] and is summarised in **figure 3**. Nitrogen enters the syngas during secondary reforming in the autothermal reformer. Syngas separation includes CO_2 removal, resulting in a CO_2 stream of 96% purity. For the cases where biomass replaced fossil fuel, biomethane was substituted for natural gas on a methane-content basis, which would allow continued use of existing infrastructure in the near-term.

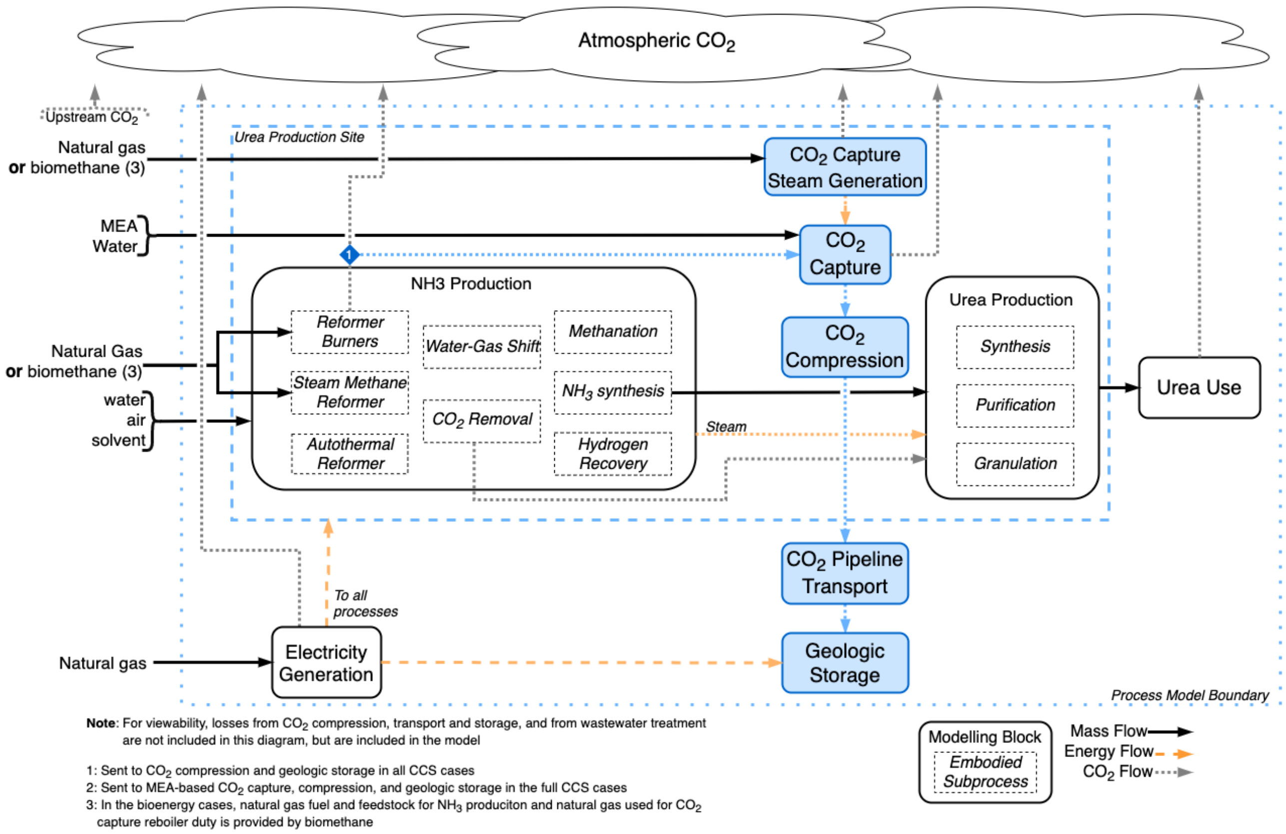
Approximately 20% of commercially produced ammonia is used for the production of urea [18], which typically uses the CO_2 separated from syngas in the ammonia production process. Therefore, the production of urea via an integrated ammonia plant has been considered, as in **figure 4**, using additional data from [8]. Heat demand is integrated with the ammonia plant, and the additional electricity demand is 19 kWh/t urea [8]. Since the high-purity CO_2 stream is used for urea production, the high-purity CCS cases were not considered for urea. All carbon in the urea product is assumed to be released as CO_2 during use.



Note: For viewability, losses from CO_2 compression, transport and storage, and from wastewater treatment are not included in this diagram, but are included in the model

1: Sent to CO_2 compression and geologic storage in the high purity CCS cases
2: Sent to MEA-based CO_2 capture, compression, and geologic storage in the full CCS cases
3: In the bioenergy cases, natural gas fuel and feedstock for NH_3 produciton and natural gas used for CO_2 capture reboiler duty is provided by biomethane

Figure 3. Process model system for ammonia production



Note: For viewability, losses from CO_2 compression, transport and storage, and from wastewater treatment are not included in this diagram, but are included in the model

1: Sent to CO_2 compression and geologic storage in all CCS cases

2: Sent to MEA-based CO_2 capture, compression, and geologic storage in the full CCS cases

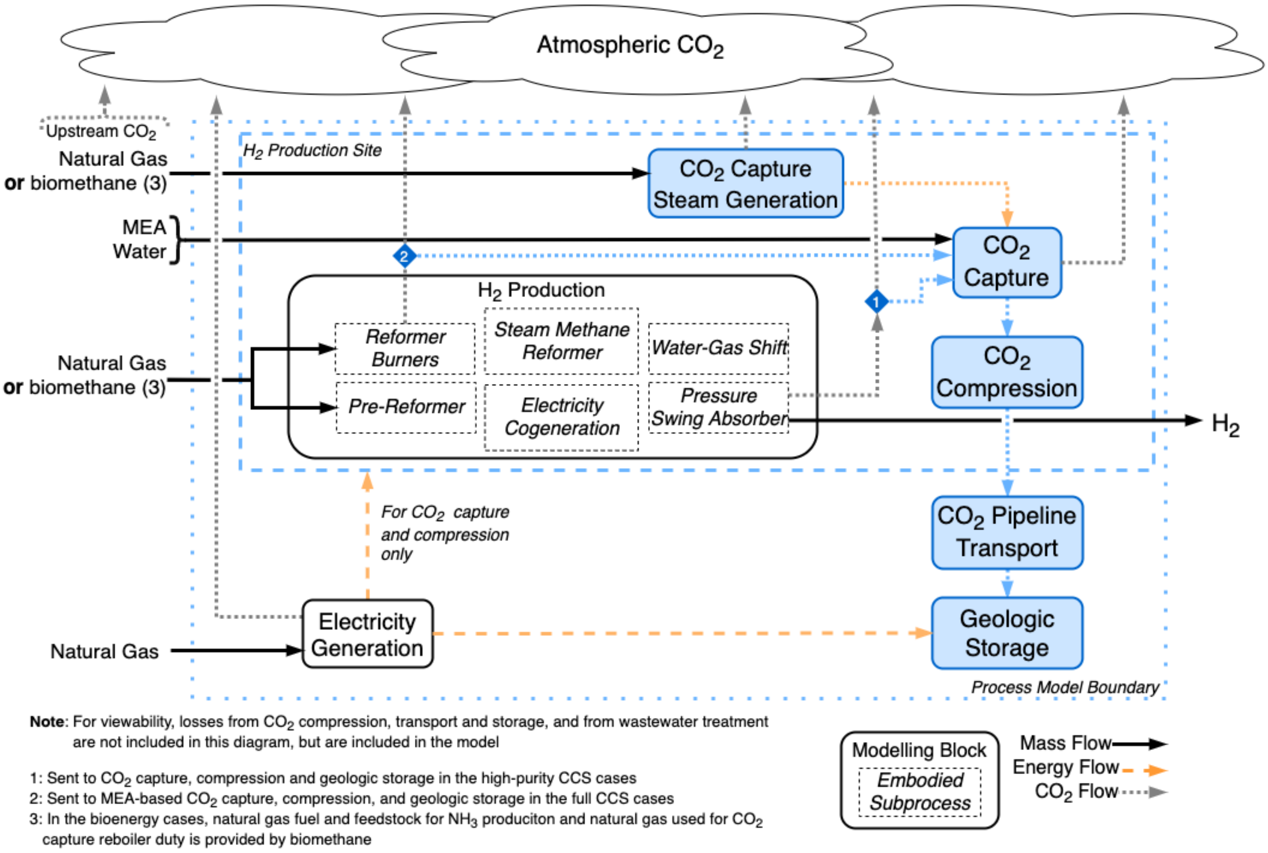
3: In the bioenergy cases, natural gas fuel and feedstock for NH_3 produciton and natural gas used for CO_2 capture reboiler duty is provided by biomethane

Figure 4. Process model system for urea production

2.3 Hydrogen

Commercially, 90% of hydrogen is produced for use in the ammonia, methanol, and oil refining industries [6], typically as an integrated process. However, we have also considered the production of merchant hydrogen, based on the reference model of [6], as in **figure 5**.

In the reference model, the tail gas from pressure swing absorption has a CO_2 content of 51%. Based on [19], this was therefore assumed to still require additional CO_2 purification, with an energy requirement of 3.0 GJ/t CO_2.



Note: For viewability, losses from CO_2 compression, transport and storage, and from wastewater treatment are not included in this diagram, but are included in the model

1: Sent to CO_2 capture, compression and geologic storage in the high-purity CCS cases
2: Sent to MEA-based CO_2 capture, compression, and geologic storage in the full CCS cases
3: In the bioenergy cases, natural gas fuel and feedstock for NH_3 produciton and natural gas used for CO_2 capture reboiler duty is provided by biomethane

Figure 5. Process model system for hydrogen production

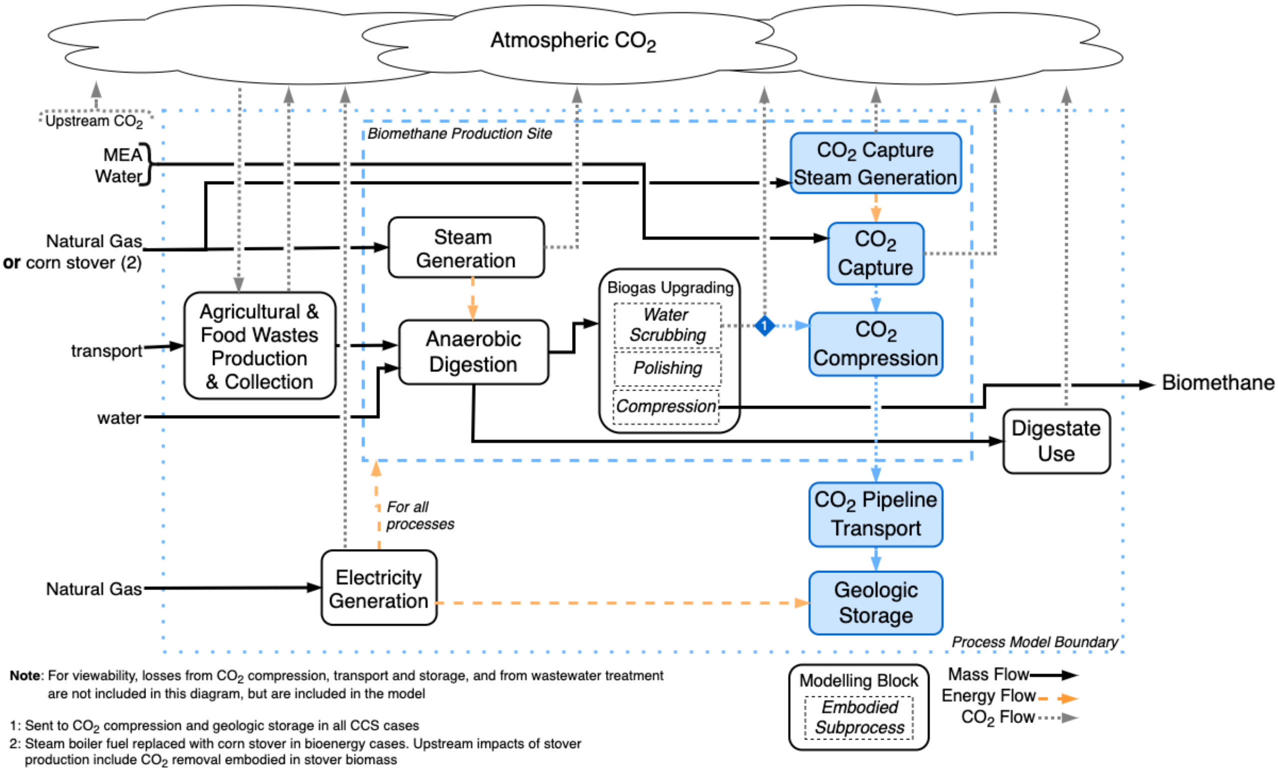
2.4 Carbon capture and storage

Except in the case of merchant hydrogen production, the high purity CO_2 streams were assumed to only require compression, transport, and storage. For CO_2 from dilute flue gas streams, a solvent-based absorption capture process, using monoethanolamine is assumed, requiring 22 kWh/t CO_2 [8] and 3.2 GJ of low-pressure steam, provided by a dedicated boiler. All captured CO_2 was compressed to 110 bar, requiring 80 kWh/t CO_2 [6] and transported 200 km by pipeline to geologic storage, with a conservative estimate of 1.5% of CO_2 lost to transport and injection [20], with injection requiring 8 kWh/t CO_2 [21].

2.5 Biomass and bioenergy production

In the biomass and bioCCS cases for bioethanol, corn stover replaces natural gas for process heat provision for both ethanol production and CO_2 capture. The estimated CO_2 emissions from maize cultivation, harvesting, and drying are from ecoinvent 3.6. The production of corn stover uses data from the US LCI database [22], and includes CO_2 emissions related to harvesting, baling, drying, cutting, transport, and storage of the stover, as well as additional fertiliser and fodder use to replace the stover that now cannot be used for that purpose.

For ammonia, urea, and hydrogen, biomethane replaced natural gas for both feedstock and fuel in the biomass and bioCCS cases. The biomethane is assumed to be produced from food and agricultural wastes, anaerobically digested, followed by regenerative water scrubbing, as in **figure 6**, to a purity of 95% CH_4 [14–16]. The high-purity stream of CO_2 produced in biomethane production is assumed to be emitted in cases without CCS and sent to compression and storage in the cases with CCS. Upstream emissions attributed to the wastes are the transport needed for the collection of waste, assumed to average 100km. Biomethane production was assumed to be co-located with the chemical plant.



Note: For viewability, losses from CO_2 compression, transport and storage, and from wastewater treatment are not included in this diagram, but are included in the model

1: Sent to CO_2 compression and geologic storage in all CCS cases

2: Steam boiler fuel replaced with corn stover in bioenergy cases. Upstream impacts of stover production include CO_2 removal embodied in stover biomass

Figure 6. Process model system for biomethane production

2.6 Utilities

Except in the case of lignocellulosic bioethanol and merchant hydrogen, electricity was assumed to be produced at an off-site natural gas combined cycle natural gas (NGCC) power plant with a net efficiency of 56.6% with a CO_2 intensity of approximately 360 g CO_2/kWh, in line with the reported OECD average grid intensity (385 g CO2/kWh in 2017) [22]. In the case of lignocellulosic bioethanol, excess steam from lignin combustion was used to cogenerate electricity with a net efficiency of 30%. Cogenerated electricity was considered available to the chemical production processes but not to any CCS processes, to maintain an equal basis of comparison with the other models.

For steam provision, a boiler with a 90% efficiency is assumed. In cases with CO_2 capture, it was assumed that no waste heat was available for use in the CO_2 capture system. CO_2 process heat was assumed to be provided by a separate boiler, and the CO_2 there produced was not captured. These assumptions were made to provide a clearer and more generic assessment of the energy demand of CCS in each system.

3 Results

Table 2 presents the estimated net CO_2 for each case modelled in this chapter. The net CO_2 includes CO_2 emitted during chemical production; bioenergy production; CO_2 capture compression, transport, and storage; upstream impacts from production inputs and infrastructure use; and product and by-product end use.

Table 2. Net life cycle CO_2 for chemical products under different cases of (additional) biomass and/or CCS integration.

Feedstock	Unit	No (Additional) Biomass			Biomass		
		No CCS	CCS of pure CO_2	CCS of all CO_2	No CCS	CCS of pure CO_2	CCS of all CO_2
Bioethanol (maize)	t CO_2/t ethanol	1.2	0.2	-0.1	0.8	-0.2	-0.8
	g CO_2/MJ	*45*	*5*	*-5*	*30*	*-10*	*-30*
Bioethanol (stover)	t CO_2/t ethanol	1.8	0.8	-0.9	1.8	0.8	-1.4
	g CO_2/MJ	*70*	*30*	*-35*	*70*	*30*	*-50*
Ammonia	t CO_2/t NH_3	2.0	0.9	0.6	1.0	-0.6	-1.0
	t CO_2/t N	*2.4*	*1.1*	*0.7*	*1.2*	*-0.7*	*-1.2*
Urea	t CO_2/t Urea	1.3	*n.a.*	1.1	0.6	*n.a.*	0.1
	t CO_2/t N	*2.7*	*n.a.*	*2.3*	*1.3*	*n.a.*	*0.2*
Hydrogen	t CO_2/t H_2	9.7	4.5	2.9	4.5	-4.1	-5.8
	g CO_2/MJ	*80*	*40*	*25*	*40*	*-35*	*-50*

Negative CO_2 emissions require both that CO_2 is removed from the atmosphere and that CO_2 is permanently stored. Therefore, neither CCS alone nor biomass use alone results in CO_2-negative systems. The decarbonisation potential for bioCCS is highest for chemicals where the majority of carbon in the feedstock is converted to CO_2 during the chemical production process, such as in merchant hydrogen and merchant ammonia production, where it is capable of being captured and sent to permanent storage. The contributing factors are discussed below, along with a case-by-case breakdown of CO_2 emissions and removal (**figures 5-7**).

3.1 Bioethanol, maize

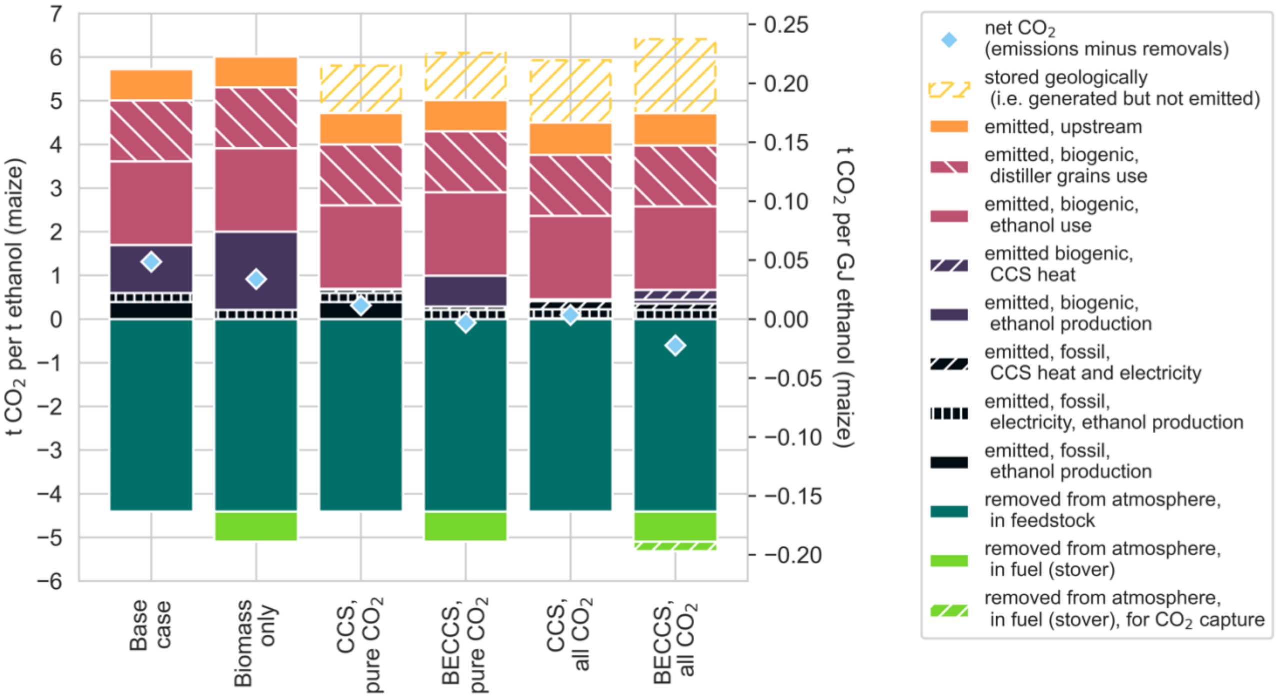


Figure 7. Life cycle net CO_2 emissions and removals for bioethanol produced from maize in different cases of biomass and CCS use

The base maize bioethanol system was estimated to emit 5.6 t CO_2/t bioethanol, of which 4.4 t are biogenic (**figure 7**). As visualised in **figure 8**, approximately 30% of the total CO_2 is emitted during bioethanol production (including 0.1 t CO_2/t bioethanol for electricity generation), slightly less than during the combustion of bioethanol, with an additional 25% resulting from the decomposition of the distiller's grains. The remaining is from upstream supply chains, predominantly from fertiliser production and grain drying. The result is a reference case net CO_2 of 1.2 t CO_2/t bioethanol (45 g/MJ).

As a single intervention, the replacement of natural gas used for heat provision with corn stover only reduced net CO_2 by a third (from 1.2 to 0.8 t CO_2/t), as **figure 7** shows. Due to the large fraction of unabatable emissions occurring both upstream and downstream of bioethanol production, storage of only fermentation CO_2 was insufficient to result in substantive negative net CO_2, though both CCS cases with natural gas or stover providing process heat, reached near carbon neutrality, at 0.2 t CO_2/t (5 g CO_2/MJ) and -0.1 t CO_2/t (-5 g CO_2/MJ) bioethanol, respectively. Note that the inclusion of transport emissions, which were excluded from this model, will likely raise the balance to above CO_2 neutrality.

In the bioCCS cases, the impact of the additional capture of the dilute CO_2 from steam generation for fermentation was more pronounced than in the CCS only cases, with the full bioCCS case modelled to have a net CO_2 of -0.8 t CO_2/t (-30 g CO_2/MJ). Combusting stover rather than natural gas creates a biogenic CO_2 stream larger than the stream of high-purity fermentation CO_2, which allows the capture CO_2 to be sufficient to compensate for the CO_2 emitted by the bioethanol combustion and distiller grain decomposition (**figure 9**), as well as CO_2 emissions from upstream supply chains. The full bioCCS case was estimated to require 0.5 dry tonnes of stover per tonne of bioethanol, which is approximately a fifth of the stover coproduced by the 2.7 dry tonnes of maize required [23].

Approximately 96 Mt of bioethanol were produced in 2018, of which 60% was from maize [17]. If all maize bioethanol was produced according to our "full bioCCS" case, approximately 45 Mt of negative CO_2 could be possible. However, the CO_2 balance of bioethanol is highly dependent on the specific system configuration, particularly with regard to the origin of the maize. This chapter assumed efficient and local European production of maize (225 kg CO_2/dry t maize). If the maize production emissions were instead in line with ecoinvent estimate for the global market average of maize production was used (488 kg CO_2/dry t maize), CO_2 emissions would increase by 0.7 t CO_2/t ethanol, all but negating the negative emission potential of the full bioCCS case.

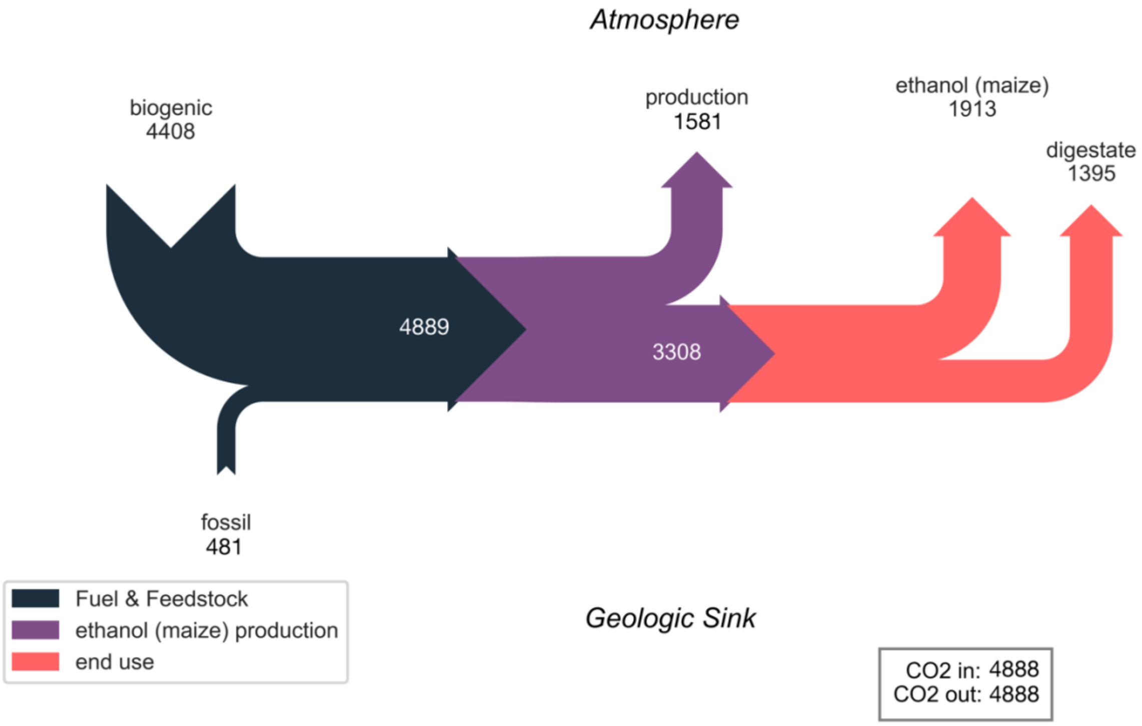


Figure 8. Carbon flow of maize bioethanol production and use (including electricity generation, excluding supply chain carbon), reference case, in kg CO_2eq/t bioethanol

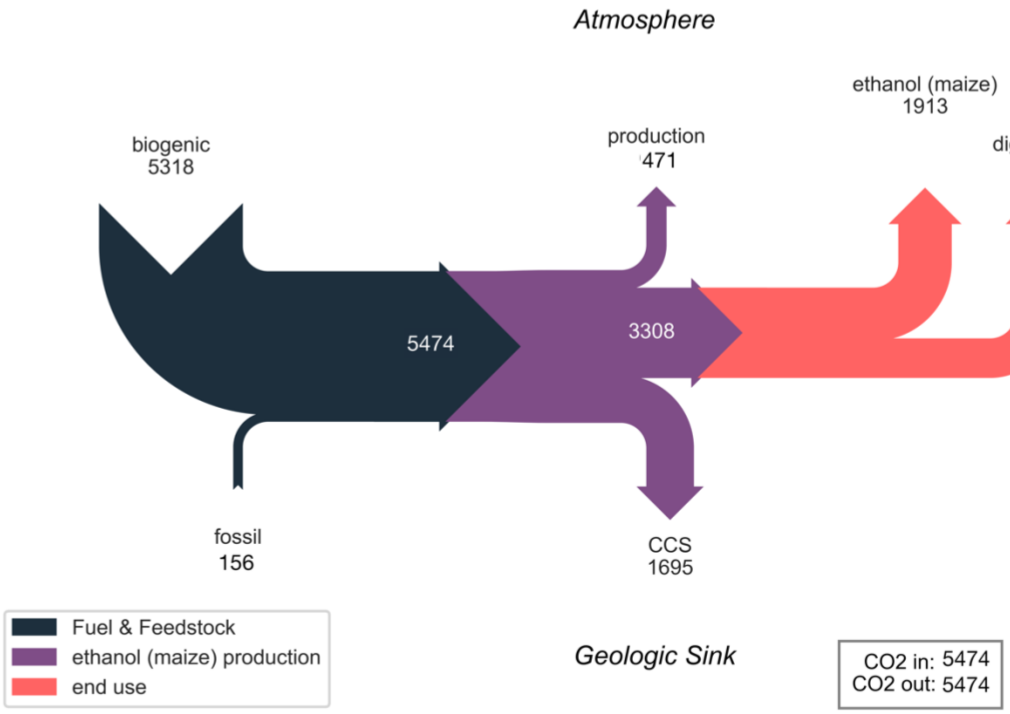


Figure 9. Carbon flow of maize bioethanol production and use (including electricity generation, excluding supply chain carbon), full bioCCS case, in kg CO_2eq/t bioethanol

3.2 Bioethanol, stover

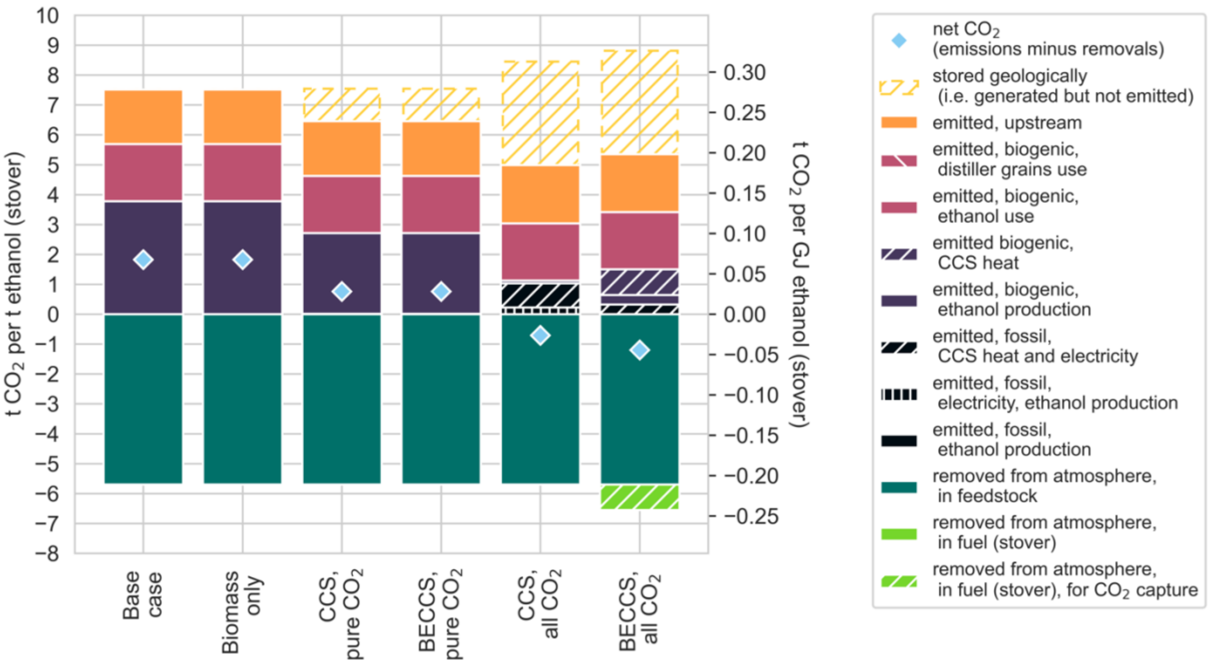


Figure 10. Life cycle net CO_2 emissions and removals for bioethanol produced from corn stover in different cases of biomass and CCS use

In this chapter, we also considered the fermentation of stover for bioethanol production, with co-generation of heat and electricity from the separated lignin. As shown in **figure 10**, the reference case resulted in a net CO_2 of 1.8 t CO_2/t bioethanol (70 g CO_2/MJ), equivalent to the emissions from upstream supply chains, as the CO_2 emitted during production and use are all biogenic, and thus compensated by the uptake of CO_2 by regrown biomass. As modelled, the co-generation from the lignin wastes was sufficient for the energy demands of bioethanol production, and no supplemental natural gas or electricity was required. The cogeneration results in CO_2 emissions at the bioethanol refinery that are more than double that of maize ethanol production and are also completely biogenic (**figure 11**) since no supplementary natural gas is needed.

Upstream CO_2 accounts for 1.8 t of the 6.9 t CO_2/t bioethanol emitted in the reference case, of which 1.3 t were attributed the enzymes needed to process the stover into fermentable starch. The enzymatic loading in the model was 33 g/kg stover, in line with [24], which also showed a significant lifecycle CO_2 impact from enzyme production. The upstream data represented generic enzyme production, including enzymes used for ethanol production, but when assessing the CO_2 balance for lignocellulosic ethanol, particular attention should be paid to the CO_2 emissions of the specific situation of enzyme production and use.

As the reference case has no fossil sources of process CO_2 to abate, the "additional biomass only" is equivalent to the reference case. Likewise, the high purity CCS and bioCCS cases are equivalent to each other, only requiring additional electricity for CO_2 compression, and reduce reference case net CO_2 by 60% to 0.8 t CO_2/ t (30 g CO_2/MJ) bioethanol, but insufficient to compensate for the upstream emissions. The cases with full CCS, with and without the use of supplemental bioenergy, result in negative CO_2 balances, with full bioCCS having a net CO_2 roughly 500 kg CO_2/t bioethanol lower than its CCS-only counterpart, at -1.4 t CO_2/t bioethanol (-50 g CO_2/MJ). In the full bioCCS case, the cogenerated electricity is insufficient for CO_2 capture and compression, and thus requires a small import of electricity from natural gas, as seen in **figure 12**.

Stover is a by-product of maize production, and bioethanol production can also be considered in terms of land use, as in **table 3**. In our model's reference case, the production of 1 tonne of bioethanol required 4 tonnes of stover, requiring 50% more land than maize bioethanol. However, per hectare of maize, stover bioethanol with full bioCCS integration generates a third more negative CO_2 than maize bioethanol. Additionally, if 1 tonne of maize bioethanol was produced as in the full bioCCS case, with the approximately 2 tonnes remaining stover from maize cultivation used for additional ethanol production with full bioCCS integration, an additional 0.4 tonnes of bioethanol and 0.8 tonnes of negative CO_2 emissions could be delivered. Theoretically, in this manner, the current maize-ethanol production could be met requiring 30% less land, and generating a third more negative emissions, totalling -62Mt/year, subject to the same caveats as discussed for maize ethanol, above.

Table 3. Bioethanol production potential from one hectare of land

Case	Stover Input	Maize Input	Ethanol Output	Net CO_2
	t/ha	*t/ha*	*t/ha*	*t CO_2/ha*
Maize bioethanol only	0.5	4.0	1.5	-1.2
Stover bioethanol only	4.0	0.0	1.0	-1.4
Maize and stover bioethanol	4.0	4.0	2.4	-2.4

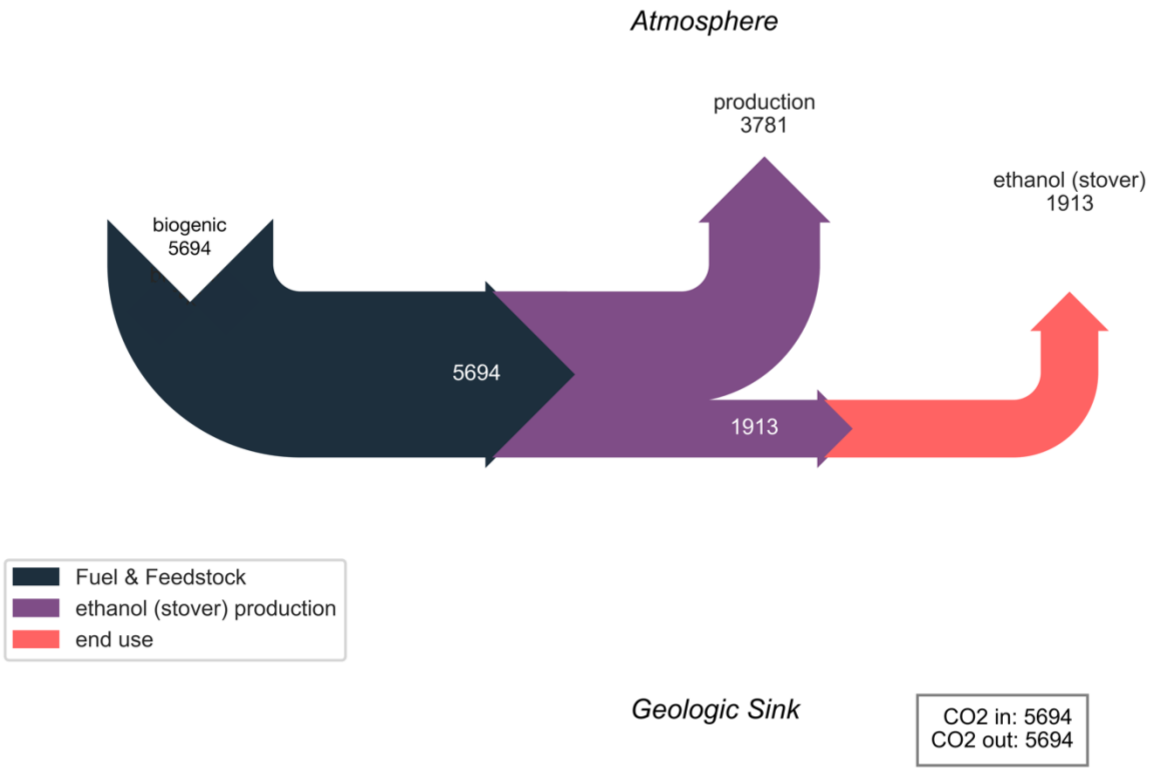


Figure 11. Carbon flow of corn stover bioethanol production and use (including electricity generation, excluding supply chain carbon), reference case, in kg CO_2eq/t bioethanol

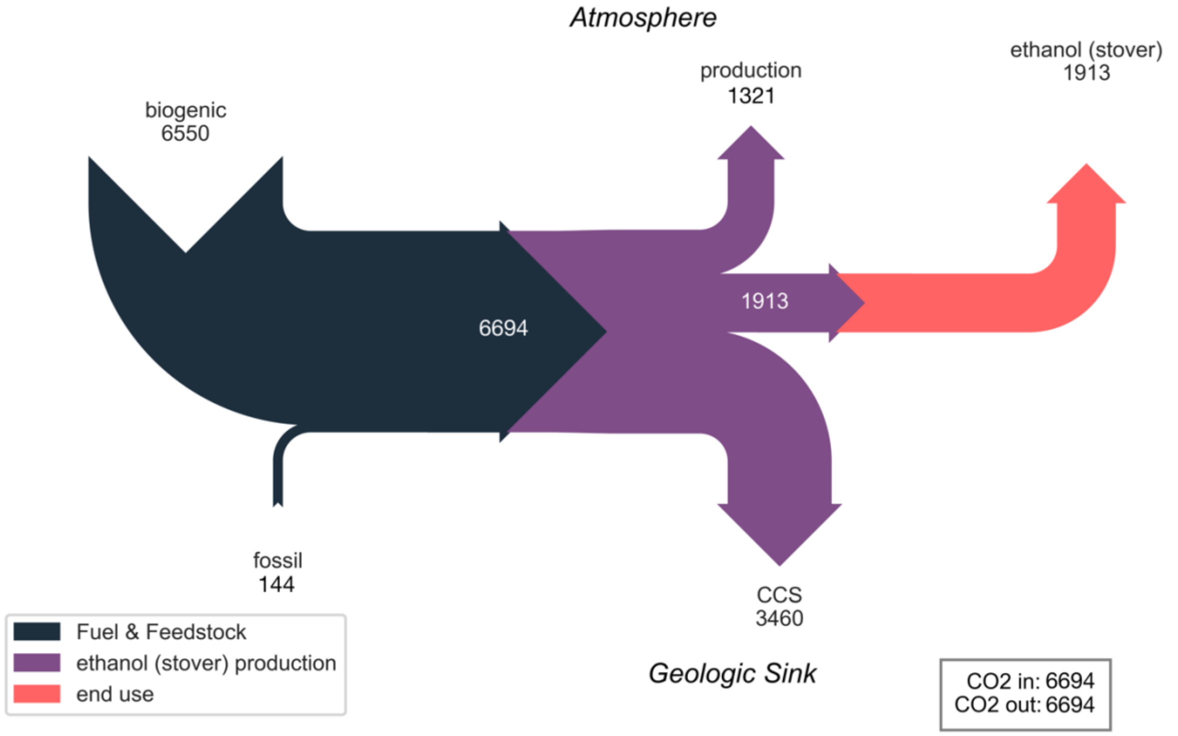


Figure 12. Carbon flow of corn stover bioethanol production and use (including electricity generation, excluding supply chain carbon), full bioCCS case, in kg CO_2eq/t bioethanol

3.3 Ammonia

The reference case of ammonia production, using natural gas as the hydrogen source without any CO_2 storage was estimated to generate 2 tonnes of fossil CO_2 per tonne of ammonia, over 60% of which is from carbon separated from the natural gas feedstock during hydrogen production. A further 25% is from fuel combusted in the reformer burners, and the remainder is primarily upstream emissions from the natural gas supply chain. As there is no atmospheric CO_2 removal in the reference case, this is also the net CO_2 of the reference case, as shown in **figure 13**, 2.0 t CO_2/t NH_3 (2.4 t CO_2/t N).

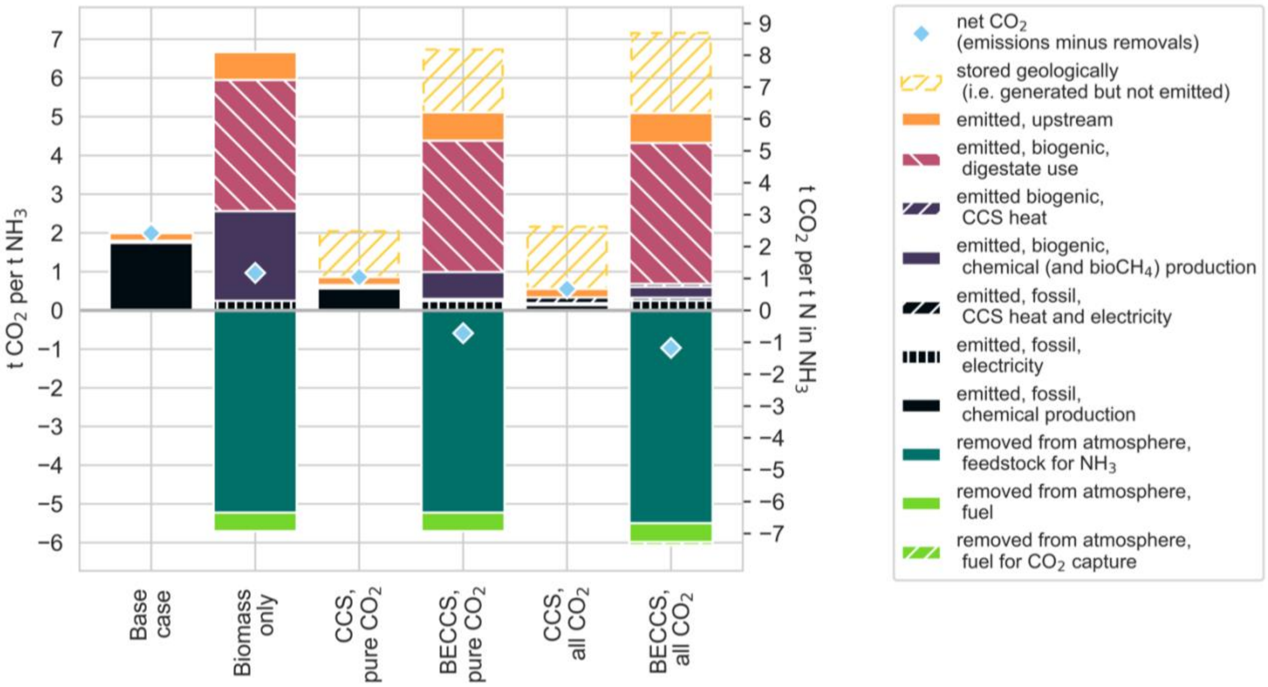


Figure 13. **Life cycle net CO_2 emissions and removals for ammonia in different cases of biomass and CCS use**

The use of biomethane as feedstock triples the amount of carbon in the system, over half of which ends up as residual digestate from biomethane production (**figure 15**). However, the 5.8 t of biogenic CO_2 emissions results from the reuse of carbon in food and agricultural waste that is part of the short-term carbon cycle. Therefore, the net CO_2 of the biomass-only case is the sum of estimated fossil emissions from electricity production, infrastructure, transport, and upstream supply chains, 0.3 t CO_2/t NH_3. Biomethane use also reduced net CO_2 more than CCS alone. For fossil-based production, full CCS resulted in a net CO_2 of 0.6 t CO_2/t NH_3 (0.7 t CO_2/t N), with the storage of 1.6 t of CO_2 and an additional 0.2 t/t NH_3 of CO_2 emissions attributable to CCS.

Both bioCCS cases had negative net CO_2, -0.6 t CO_2/t NH_3 for the capture of high-purity CO_2 streams from ammonia and biomethane production and -1.0 t CO_2/t NH_3, in the full bioCCS case. Approximately, 0.5 t CO_2/t NH_3, is biogenic CO_2 captured during the production of the biomethane feedstock, representing 25% of stored CO_2 in the full bioCCS case.

150 Mt of ammonia was produced in 2019 [25]. If all NH_3 was as in the bioCCS case modelled here, the result would be 270 Mt of negative CO_2 emissions. However, it would require 930 Mt of food and agricultural wastes for anaerobic digestion. The total availability of biowaste is difficult to quantify, but [26] estimates than an annual 1.3 billion tonnes of food is lost or wasted in pre- and post-consumer supply chains, suggesting that, even if agricultural and food waste is currently a main source of commercial biogas production, a completely food waste based NH_3 industry is unrealistic, and that dedicated energy crops would likely be be needed.

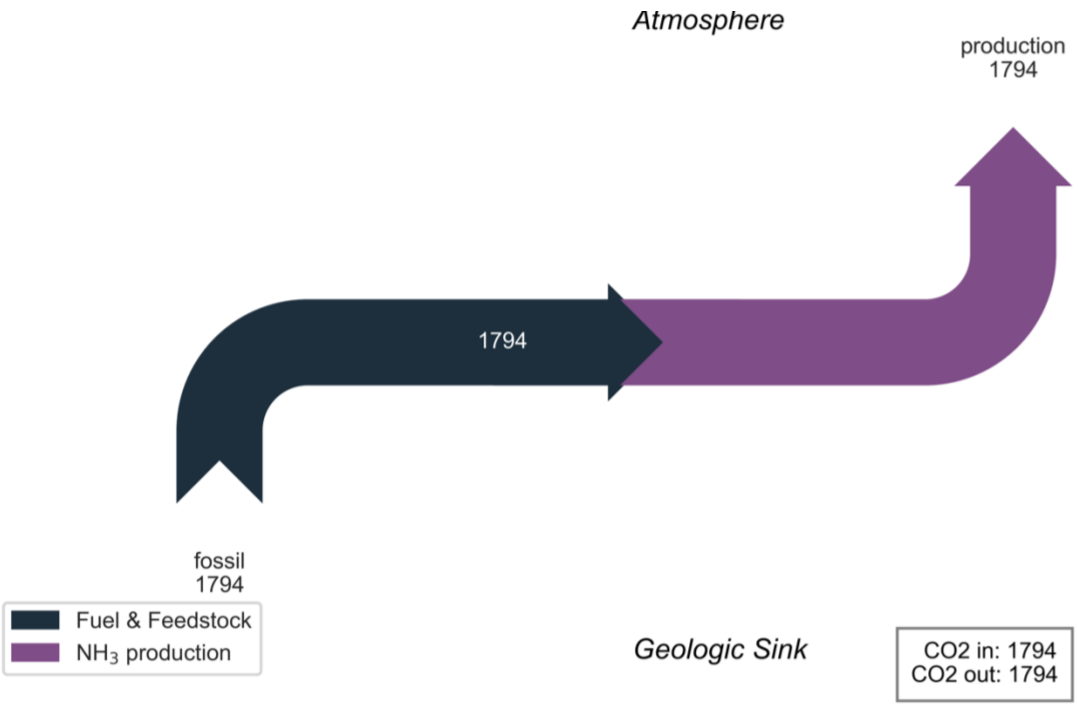


Figure 14. Carbon flow of ammonia production and use (including electricity generation, excluding supply chain carbon), reference case, in kg CO_2eq/t ammonia

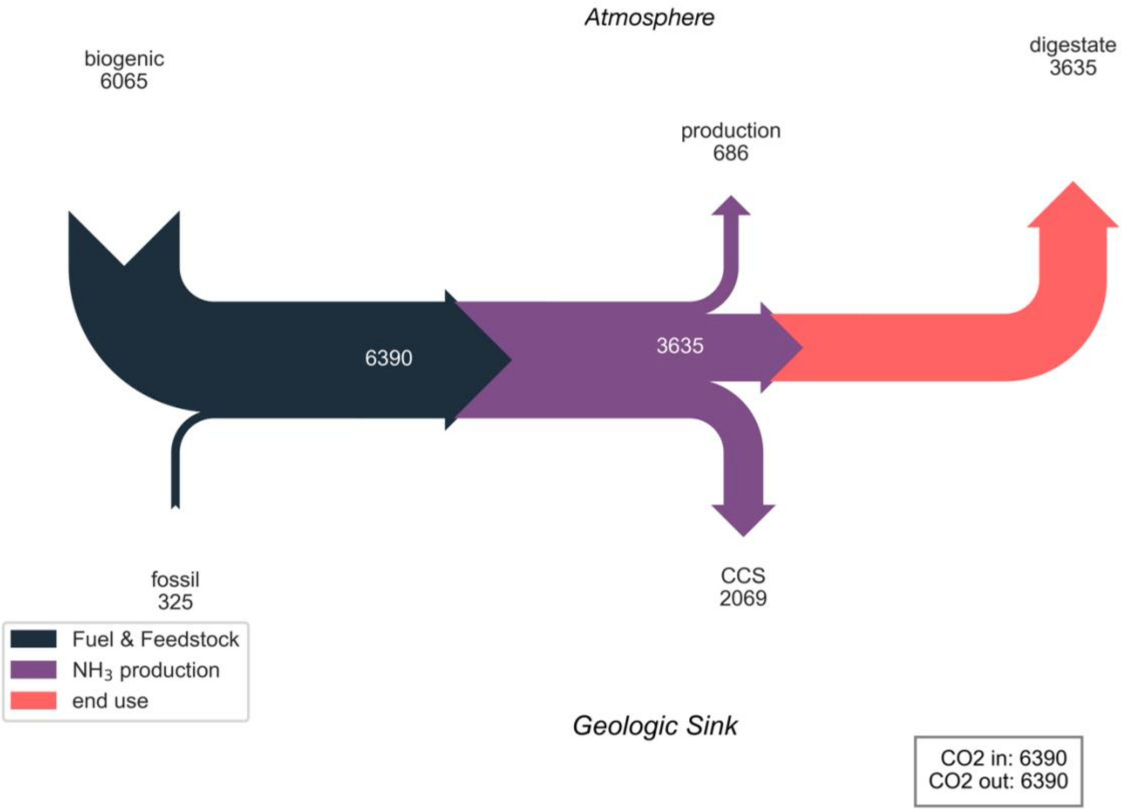


Figure 15. Carbon flow of ammonia production and use (including electricity generation, excluding supply chain carbon), full bioCCS case, in kg CO_2eq/t ammonia

3.4 Urea

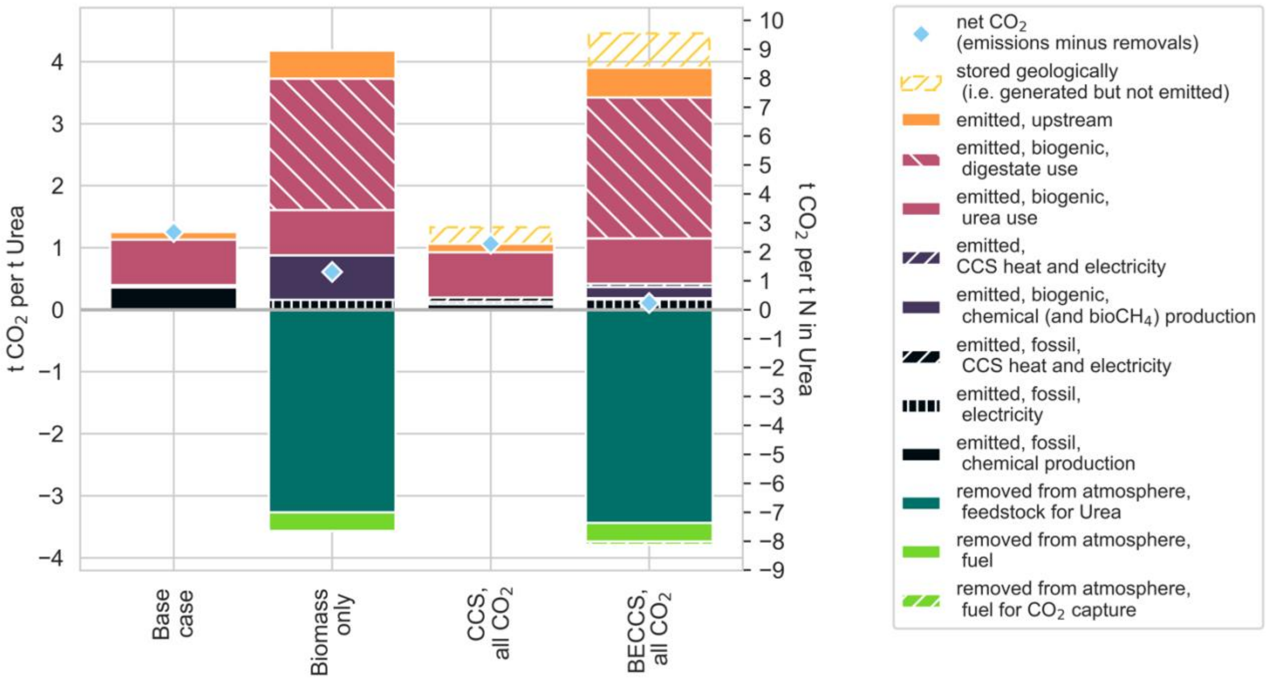


Figure 16. Life cycle net CO_2 emissions and removals for urea in different cases of biomass and CCS use

Roughly 20% of global ammonia production was used to produce 54Mt of urea in 2019 [18]. The production of 1 tonne of urea requires 570 kg of NH_3 and 730 kg of CO_2, for which the CO_2 separated during ammonia production was assumed to be used. This CO_2 is then later emitted to the atmosphere during urea use and is therefore unabatable. As shown in **figure 16**, the reference case was modelled to have a net CO_2 of 1.3 t CO_2/t urea (2.7 t CO_2/t N).

As urea production requires all CO_2 separated from syngas during NH_3 production, CCS alone can only store 0.3 t of CO_2 from the steam methane reformer burners flue gas. The remainder is embodied, and then released from the urea itself (**figure 17**). Thus, the net CO_2 in the urea CCS-only case is 1.1 t CO_2/t urea (2.3 t CO_2/t N), only slightly lower than the reference case. The replacement of natural gas with biomethane is significantly more effective in lowering net CO_2 emissions, resulting in a net CO_2 of 0.6 t CO_2/t urea (1.3 t CO_2/t N).

The full bioCCS case results in near carbon-neutral net CO_2, at 0.1 t CO_2/t urea (0.2 t CO_2/t N) with the stored biogenic CO_2 from biomethane production offsetting the emissions from urea use as a fertiliser (**figure 18**). While carbon-neutrality was not seen, it seems likely that low-CO_2 urea is achievable.

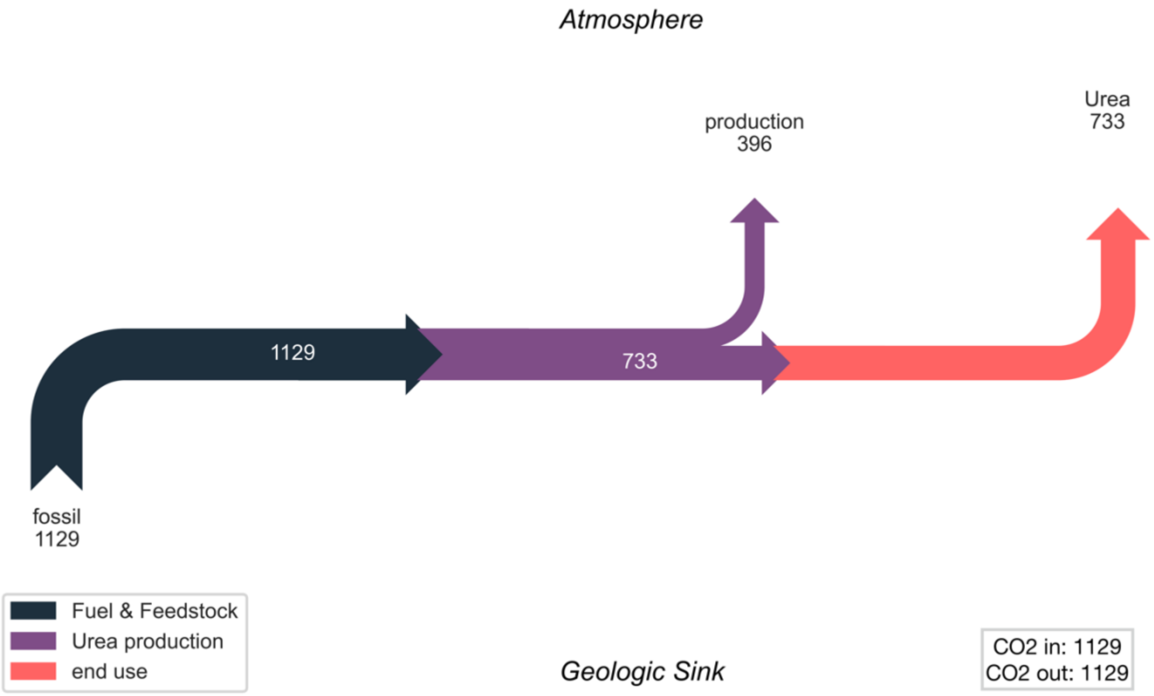


Figure 17. Carbon flow of urea production and use (including electricity generation, excluding supply chain carbon), reference case, in kg CO_2eq/t urea

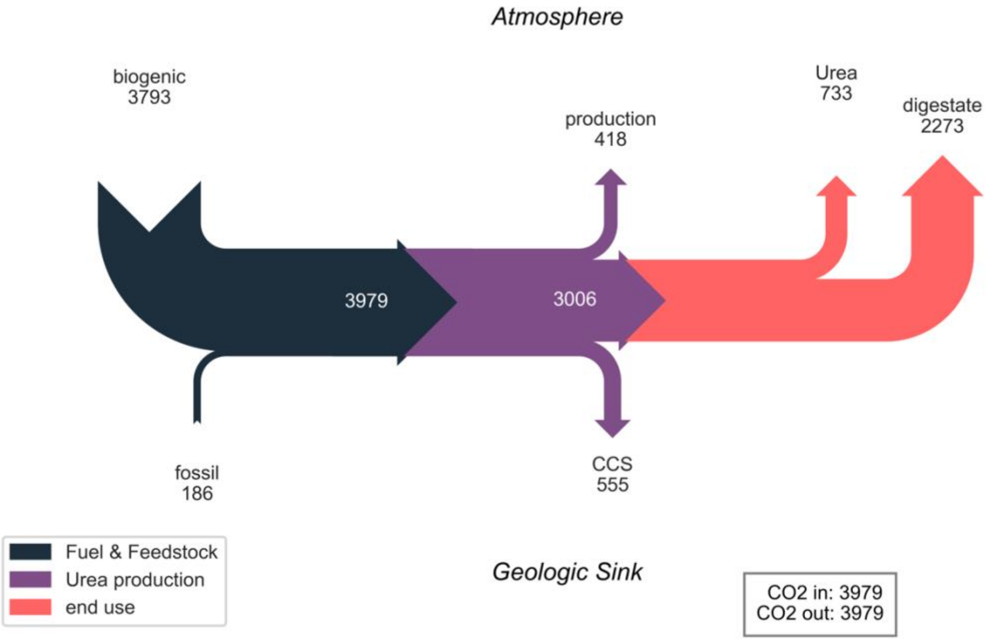


Figure 18. Carbon flow of urea production and use (including electricity generation, excluding supply chain carbon), full bioCCS case, in kg CO_2eq/t urea

3.5 Hydrogen

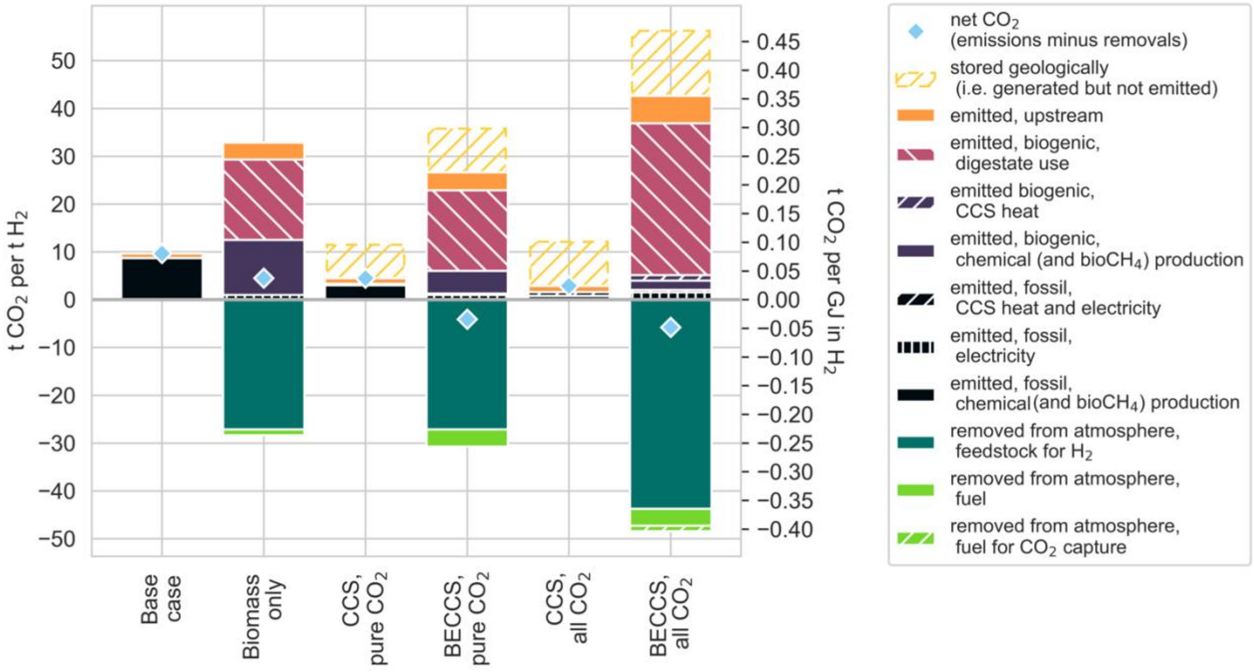


Figure 19. **Life cycle net CO_2 emissions and removals for ammonia in different cases of biomass and CCS use**

In the reference case, the net CO_2 is 9.7 t CO_2/t H_2 (80 g CO_2/MJ) of which 7.4 tonnes are CO_2 separated from the feedstock natural gas, 1.2 t are emitted by natural gas combusted for fuel (**figure 19**). A further 1.2 t CO_2/ t H_2 were estimated to be emitted upstream, primarily in the natural gas supply chain.

Replacing the natural gas fuel and feedstock with biomethane reduces net CO_2 by half, though the amount of CO_2 emitted increases by a factor of three to 28.4 t CO_2/t H_2. This is offset by the assumed CO_2 reuptake by biomass, but it important to emphasise that this is a much more resource intensive system and will be very sensitive to both the biomass source and transportation in any real-world configuration. Full CCS, applied to both high purity and dilute CO_2 streams, without biomass use, was estimated to reduce net CO_2 to 2.9t CO_2/t H_2 (25g CO_2/MJ), with the syngas separation CO_2 stream being triple the volume of CO_2 from reformer burner flue gas.

In the bioCCS cases, the high-purity CO_2 streams from biohydrogen and biomethane production alone results in a net CO_2 of -4.1 t CO_2/t H_2 (-35 g CO_2/MJ), though the carbon in the production system increases sixfold (**figures 20** and **21**). When both dilute and high purity streams are captured, the net CO_2 lowers to -5.8 t CO_2/t H_2 (-50 g CO_2/MJ). Of the roughly 70Mt of hydrogen estimated to have been produced in 2018, approximately 8Mt was produced as merchant hydrogen [27]. At this scale, full bioCCS hydrogen production would be estimated to deliver 65 Mt of negative CO_2 emissions. However, as with ammonia, large quantities of suitable biomass are required, in this case 45 tonnes of food and agricultural waste converted to biomethane per tonne of hydrogen, which may not be feasible on a large commercial scale.

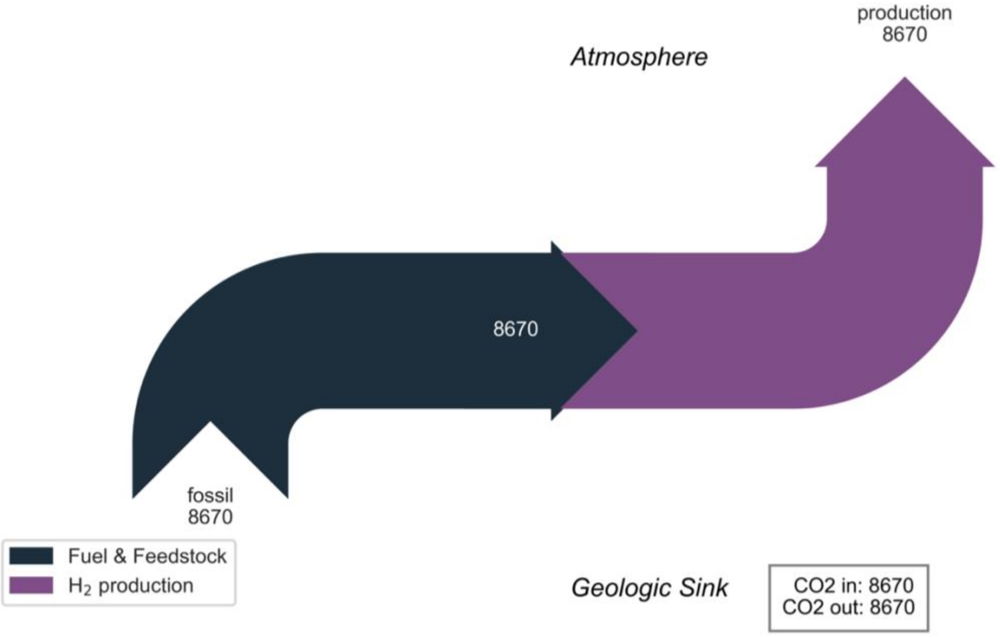


Figure 20. Carbon flow of hydrogen production and use (including electricity generation, excluding supply chain carbon), reference case, in kg CO_2eq/t hydrogen

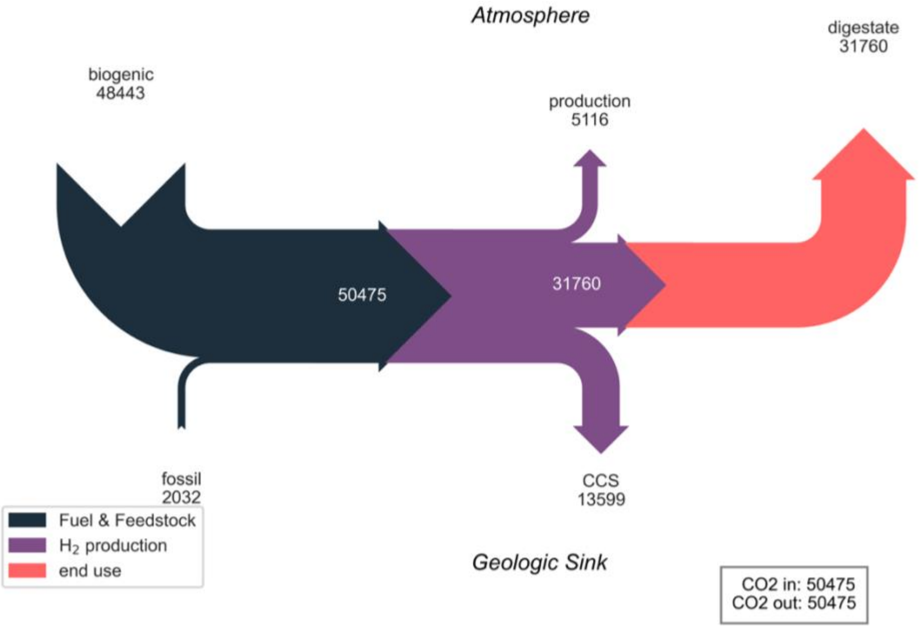


Figure 21. Carbon flow of hydrogen production and use (including electricity generation, excluding supply chain carbon), full bioCCS case, in kg CO_2eq/t hydrogen

4 Conclusions

This chapter modelled the production of bioethanol, ammonia, urea, and hydrogen considering different configurations of biomass use and CCS use, focusing on currently commercialised technology options, estimating life cycle net CO_2 balances for each case. This chapter considered only emissions and removals of CO_2, without allocation for co-products and without making assumptions regarding possible avoided emissions, to provide a clearer picture of CO_2 flows for bioCCS configurations in each chemical production pathway. For brevity, uncertainty analyses were not extensively reported in this initial exploratory paper.

In the scope and specific supply chain configurations considered, aggressive use of bioCCS was modelled to result in CO_2 negative chemical production for all chemicals, except urea. Also except in urea production, the use of CCS as a single intervention had a greater decarbonisation potential than the use of biomass for feedstock and energy without CCS.

The potential for negative CO_2 emissions was greater in chemical pathways where more CO_2 is capturable during chemical production, with net CO_2 in the full bioCCS cases ranging from -0.8 t CO_2/t maize bioethanol to -5.8 t CO_2/t hydrogen. Notably, without CCS or additional bioenergy, stover bioethanol had higher estimated net CO_2 than maize bioethanol, 70 g CO_2/MJ versus 45 g CO_2/MJ respectively, primarily attributable to the high upstream emissions of enzyme production for stover ethanol. However, in the full bioCCS case, stover ethanol had a lower net CO_2 (-50 g CO_2/MJ versus -30 g CO_2/MJ) due to the large quantity of CO_2 capturable from the combustion of lignin used for cogenerated electricity.

On an energy basis, hydrogen with full bioCCS was estimated to have similar negative CO_2 potential as stover ethanol but requires capture of CO_2 from both biomethane production and hydrogen reforming. The production of ammonia, without associated urea production, was modelled to achieve a net CO_2 -1.2 t CO_2/t NH_3 in the full bioCCS case. However, this requires that the large stream of high purity CO_2 be permanently stored and not used for urea production or other applications where it will be eventually emitted to the atmosphere. If the CO_2 is used in urea, even with a full bioCCS installation, with biomethane replacing natural gas as a feedstock, and CCS applied to both the ammonia plant and biomethane plant, resulted in a final net CO_2 of 0.1 t CO_2/t urea (0.2 t CO_2/t N), due to the high fraction of unabatable CO_2 emitted by the urea.

It must be emphasised that negative net CO_2 does not automatically imply a negative global warming potential. To determine if a specific bioCCS configuration has a negative global warming potential, emissions of other greenhouse gases, and other factors influencing global warming need to be considered for the specific system under consideration. Additionally, competition for limited biomass resources must not be allowed to strain carbon sinks through land use change or the disruption of soil carbon. Of note, this chapter focused on short-rotation biomass options, in line with existing routes of bioethanol and biomethane production. If instead, long-rotation energy crops (e.g., trees) were used, this would delay carbon reuptake, increasing the global warming potential of the biogenic CO_2. Nevertheless, we conclude that the integration of bioCCS using current commercially available technology is an option for deep decarbonisation for existing installations of bioethanol, ammonia, urea, and hydrogen, providing compensation for unabatable emissions from product and residue end use and upstream supply chains. For the cases here considered in this chapter, the upper-bound potential for negative CO_2 emissions is 340 Mt/year, which scales the net CO_2 for the full bioCCS cases of the combined production of maize-stover bioethanol, non-urea ammonia production, and merchant hydrogen production to the global demand of these products.

Evaluating the CO_2 balance of chemical production must be specific to the system under consideration and include all upstream and downstream emissions, including product use and disposal, and production of chemical inputs, as well as inputs and energy use for biomass and bioenergy provision and the full carbon capture, compression, transport, and storage, chain. Negative emissions are not a panacea for finite resources, nor reduce the urgency of society wide deep decarbonisation. However, they do provide a decarbonisation option for sectors where CO_2 production is stoichiometrically inevitable and carbon is embodied in short-lived products.

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Appendix

Properties of carbon-based materials

Table 1. Properties of Fuels and Feedstocks

Fuel and feedstock properties	LHV (MJ/kg)	CO_2 emission factor (kg CO_2/kg)	Composition notes	Source
Natural gas	48.0	2.7	93% CH_4	[1]
Maize (dry)	n.a.	n.a.	45% C	[2]
Corn stover (dry)	18.0	1.8	49% C	[3], averaged value
Stover lignin	20.5	2.2	n.a.	[4]
Food waste	n.a.	n.a.	50% H_2O, 27% C	[3], averaged value of organic wastes
Biomethane	42.2	2.8	95% CH_4	[5]

Model parameters

Table 2. Model parameters for ethanol production from maize [2]

Parameter	Value	Unit
Biomass-ethanol conversion efficiency	374.3	kg ethanol/t dry biomass
Wastewater production	301.0	kg/t dry biomass
Distiller grains production	311.0	kg dried distiller grains/t dry biomass
Enzyme demand	5.5	kg/t ethanol
NH_3 demand	6.5	kg/t ethanol
Yeast demand	0.7	kg/t ethanol
Water demand	994.5	kg/t ethanol
Electricity demand	228.0	kWh/t ethanol
Steam demand	6219.7	GJ/t ethanol
Sulphuric acid demand	6.5	kg/t ethanol

Table 3. Model parameters for ethanol production from corn stover [4]

Parameter	Value	Unit
Biomass-ethanol conversion efficiency	247.3	kg/t dry biomass
Wastewater production	269.1	kg/t dry biomass
Lignin separated	301.4	kg/t dry biomass
Enzyme demand	134.5	kg/t ethanol
NH_3 demand	78.1	kg/t ethanol
Yeast demand	2.3	kg/t ethanol
Electricity demand	768.0	kWh/t ethanol
Steam demand	8.6	GJ/t ethanol
Sulphuric acid demand	101.1	kg/t ethanol

Table 4. Model parameters for ammonia production via the haber-bosch process [6]

Parameter	Value	Unit
Methane demand	635.9	kg/t NH_3
Electricity demand	139.0	kWh/t NH_3
Water demand	392.9	kg/t NH_3

Table 5. Model parameters for urea production [6]

Parameter	Value	Unit
NH_3 demand	567.1	kg NH_3/t Urea
CO_2 demand	732.6	kg CO_2/t Urea
CO_2 losses	17.0	kg CO_2/t Urea
Electricity demand	19.2	kWh/t Urea

Table 6. Model parameters for hydrogen production via steam methane reforming [6]

Parameter	Value	Unit
Methane demand	3160.3	kg/t H_2
Electricity cogeneration	1278.6	kWh/t H_2
Water demand	6637.8	kg water/t H_2
Wastewater production	1534.4	kg/t H_2

Table 7. Model parameters for biogas production via anaerobic digestion [7, 8]

Parameter	Value	Unit
Feedstock demand	5000	kg/t biogas
Heat demand	0.8	GJ/t biogas
Electricity demand	111.1	kWh/t biogas
CH_4 concentration	60	% CH_4 in biogas
CO_2 concentration	40	% CO_2 in biogas
CH_4 loss	10	kg/t biogas
CO_2 loss	10	K_g/t biogas
Digestate production	6000	kg/t biogas

Table 8. Model parameters for biogas upgrading to biomethane via regenerative water scrubbing [5, 8]

Parameter	Value	Unit
Biogas CH_4	60	% CH_4 of biogas
Biogas CO_2	40	% CO_2 of biogas
CH_4 losses	3	% to flue
CO_2 losses	3	% to flue
Electricity demand	613.3	kWh/t CH_4

Table 9. Model parameters for CO_2 capture via monoethanolamine adsorption [6]

Parameter	Value	Unit
CO_2 capture efficiency	90	%
Electricity demand	22.0	kWh/t CO_2 captured
Heat demand	3.2	GJ/t CO_2 captured
Solvent demand	1.0	kg/t CO_2 captured
H_2O demand	611.1	kg/t CO_2 captured
NH_3 demand	9.8	kg/t CO_2 captured

Table 10. Model parameters for CO_2 compression

Parameter	Value	Unit	Source
Electricity demand	80	kWh/t CO_2	[9]

Table 11. Model parameters for CO_2 transport via pipeline and injection into onshore geologic storage

Parameter	Value	Unit	Source
Losses	1.5	%	[10]
Pipeline distance	100	km pipeline	own assumption
Electricity demand	8.0	kWh/t CO_2 stored	[11]

Life cycle inventory data

Table 12. **Lifecycle CO_2 inventory values for upstream supply chains**

Parameter	Value	Unit	ecoinvent process [12]
Ammonia	1.9×10^{3}	kg/t NH_3	market for ammonia, liquid RER
Corn stover	2.2×10^{2}	kg/t corn stover (dry)	corn stover, production, average, US, 2022/kg/RNA
Enzymes	9.8×10^{3}	kg/t enzymes	market for enzymes GLO
Food waste	1.2×10^{2}	kg/t food waste (50% H_2O)	market for municipal waste collection service by 21 metric ton lorry GLO
Maize	2.2×10^{2}	kg/t maize (dry)	market for maize grain, Swiss integrated production GLO
Monoethanolamine	2.6×10^{3}	kg/t monoethanolamine	market for monoethanolamine GLO
Natural gas	2.9×10^{2}	kg/t natural gas	market group for natural gas, high pressure Europe without Switzerland
Sulphuric acid	9.79×10^{1}	kg/t sulfuric acid	market for sulfuric acid RER
Wastewater treatment	4.5	kg/t wastewater treated	market for wastewater, average
Water	3.5×10^{-1}	kg/t water	market for tap water Europe without Switzerland

Table 13. **Lifecycle CO_2 inventory values for infrastructure use**

Parameter	Value	Unit	ecoinvent process (or other source)
CO_2 capture	2.4×10^{-5}	kg/t CO_2 captured	[13]
Electricity generation	1.3×10^{-1}	kg/GJ electricity	gas power plant construction, combined cycle, 400MW electrical, RER
Ethanol plant	1.7	kg/t ethanol	ethanol fermentation plant construction, CH
Steam Generation	2.2×10^{-1}	kg/GJ steam	heat, central or small-scale, natural gas, Europe without Switzerland
Biogas plant	4.1	kg/t biogas	anaerobic digestion plant construction, for biowaste, CH
Hydrogen plant	2.5×10^{1}	kg/t hydrogen	chemical factory, organics, RER, construction
Ammonia plant	5.7×10^{1}	kg/t ammonia	chemical factory, organics, RER, construction
Urea upgrading	5.7×10^{1}	kg/t urea	chemical factory, organics, RER, construction
Biogas upgrading	5.7×10^{1}	kg/t biomethane	chemical factory, organics, RER, construction
CO_2 pipeline	5.3×10^{-2}	kg/tkm CO_2 transported	market for transport, pipeline, long distance, natural gas RER

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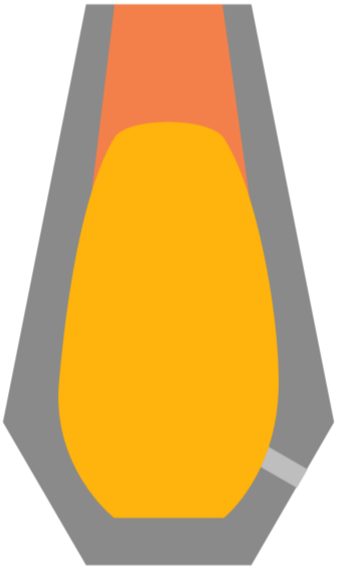
Chapter 5:

Comparing the potential for bioCCS in different steelmaking technologies



This chapter explores the potential of achieving negative emissions in steelmaking by introducing bioenergy with carbon capture and storage (BECCS, or bioCCS) in multiple steelmaking routes, including blast furnace and HIsarna smelt reduction, and Midrex and ULCORED direct reduction. Process modelling and life cycle assessment were used to estimate CO_2 *balances for 45 cases.*

Without bioenergy or CCS, the estimated life cycle CO_2 *emissions for steelmaking were 1.3-2.4 t* CO_2*/t steel. In our model, aggressive bioCCS deployment decreased net* CO_2 *to the order of −0.5t to 0.1 t* CO_2*/t steel. CCS showed a larger mitigation potential than bioenergy, but combined deployment was most effective.*

As bioCCS use increased, CO_2 *from background supply chains became more relevant. In the high bioCCS cases, if decarbonised electricity is assumed, net* CO_2 *estimates decreased by 400-600 kg* CO_2*/t steel. Conversely, at 700 g* CO_2*/kWh, all cases appeared to be net* CO_2*-positive. Accounting for the "carbon debt" of biomass, beyond biomass supply chain emissions, increased net* CO_2 *estimates by approximately 300 kg* CO_2eq*/t steel.*

We conclude that CO_2*-negative steel is possible, but will require significant interventions throughout the production chain, including sustainable biomass cultivation; efficient steel production;* CO_2 *capture throughout steel and bioenergy production; permanent storage of captured* CO_2*; and rigorous monitoring.*

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Note: Spellings, formatting, and abbreviations have been standardised throughout the dissertation.

1 Introduction

Preventing catastrophic climate change requires the rapid and immediate decarbonisation of human activities, to sharply reverse the current trajectory of increasing greenhouse gas emissions, likely even beyond carbon neutrality [1]. Indeed, all scenarios limiting global warming to 1.5°C in the IPCC special report entailed global net negative greenhouse gas emissions within the next 50 years [2]. Negative emissions are intended to both remove historic CO_2 from the atmosphere and to compensate for continued residual emissions. In the IPCC 1.5°C scenarios, these negative emissions result from agriculture, forestry, and land use change, as well as from the use of bioenergy and carbon capture and storage (BECCS, or bioCCS).

As illustrated in **figure 1**, bioCCS involves the uptake of atmospheric carbon by biomass, which is later combusted for energy, and the resulting biogenic CO_2 is captured and sent to permanent storage. Achieving negative CO_2 emissions requires the physical removal of CO_2 from the atmosphere followed by permanently preventing that CO_2 from re-entering the atmosphere [3 (**chapter 2**)]. Furthermore, any emissions resulting from the process of removal and storage, (e.g., from losses, energy use, biomass production, land use change, infrastructure construction, production of combustible co-products) must be accounted for. To result in a decrease of atmospheric CO_2, the net carbon balance of the entire negative emission technology system must be negative [3].

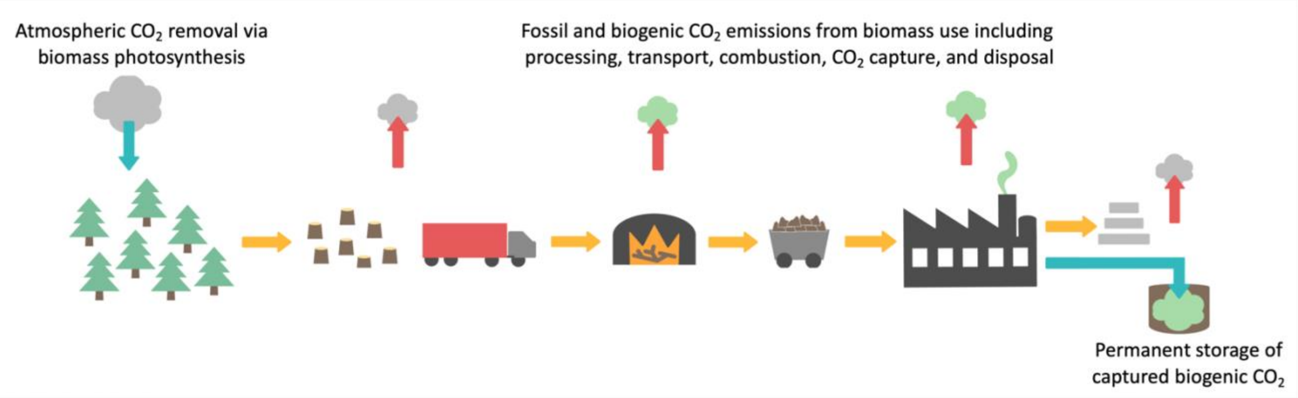


Figure 1. Bioenergy and CCS (bioCCS), simplified. Negative CO_2 emissions can result when the quantity of atmospheric CO_2 removed and stored is greater than the CO_2 emissions of the bioenergy and CCS supply chains. Adapted from [3].

The IPCC 1.5°C scenarios include 100-1100 Gt CO_2 of cumulative negative emissions through the end of the century. The interquartile range of scenarios assume large-scale bioCCS starting in 2030 and scaling up to 7-16 Gt CO_2/yr by 2100. However, the feasible scale of negative emissions is under debate from both biophysical and technoeconomic perspectives (e.g. [4, 5]). Furthermore, top-down decarbonisation scenarios typically do not consider *where* bioCCS could feasibly be incorporated. decarbonisation scenarios allocate bioCCS use to power sector [2] or an unspecified combination of power and industry [1, 6–8] or power and transport fuel production [9]. [10] is more specific, allocating 4.5 Gt of cumulative CO_2 reductions to 2060 from bioCCS use to the industrial sector and 15 Gt to power.

Many studies have focused on the design, economics, and environmental impacts of bioCCS in power (e.g. [11–13]). A demonstration-scale power plant using bioenergy with carbon capture has recently begun operating the UK [14], though the fate of the CO_2 is still undecided. For bioCCS to potentially result in negative emissions, it must be permanently stored. If the CO_2 is reused for short-lived applications, such as fuel, fertiliser, or carbonated beverages, it will be quickly re-emitted to the atmosphere, and therefore cannot result in negative emissions.

In industry, there is already an extant bioCCS installation: an ethanol plant with integrated CCS in Illinois, USA [15]. Industries, such as steel, cement, ethanol, and ammonia emit CO_2 fuel directly processes such as combustion, reduction, calcination, and fermentation. Additionally, these industries are responsible for indirect CO_2 emissions from electricity use, which vary depending on both the level of electrification of the specific industrial installation and the CO_2 intensity of the electricity provision. Further CO_2 emissions arise in both upstream and downstream supply chains. Therefore, the technical viability of bioCCS or other negative emission technologies must be evaluated for individual industrial configurations.

Steel is the largest industrial emitter of CO_2, directly emitting 2.1 Gt of CO_2 globally in 2010 [16], primarily from the combustion of 1000 Mt of coal [17]. decarbonisation options for steel include increasing the efficiency of existing carbon-based iron-reduction [18], iron reduction with hydrogen, or electrolysis of iron using renewable energy [19], all of which could move steelmaking towards carbon neutrality. However, bioCCS is the only substantive way to integrate atmospheric carbon removal and storage into steelmaking, and thus the only substantive opportunity to produce carbon-negative steel. Steelmaking slag does contain an alkali fraction that could be carbonated by atmospheric CO_2, but due to the uncertain and relatively low carbon storage potentials (0.1 - 0.6 kg CO_2/kg slag) [20, 21], slag carbonation was not included in this chapter.

There is currently little knowledge available on the use of bioCCS or other negative emission technologies in the steel industry. However, bioenergy and CCS use are both existing concepts in steel production. The partial replacement of blast furnace coal with charcoal is an established procedure in Brazilian steelmaking [22]. Charcoal has also been shown to be a viable partial replacement for fuel used in ore agglomeration and coke making processes [23].

The use of carbon capture at steel mills is in early commercialisation, with approximately 1.0 Mt of fossil CO_2 per year captured at Emirates Steel in the United Arab Emirates, ArcelorMittal in Belgium, and Shougang Steel in China, though in all cases, the CO_2 is destined for reuse in other industries [24]. Reuse of captured CO_2, also called CO_2 utilisation or carbon capture and utilisation (CCU) can reduce CO_2 emissions by displacing the need to produce fuels or materials by other means, but unless it results in long-term storage, CO_2 reuse will result in net positive CO_2 emissions.

As of April 2020, the only publicly available research studying specific bioCCS configurations in steel production is [25]. The authors consider cost-optimised bioCCS scenarios for 30 blast furnace steel plants in Europe, concluding that bioCCS could be used to achieve carbon neutrality within the boundaries of the steel mill itself. However, as the paper notes, a gate-to-gate CO_2 assessment is not sufficient to determine whether negative emissions can be achieved. Our work further fills this knowledge gap by including steelmaking technologies beyond blast furnace steelmaking, as well as expanding the system of consideration to encompass the cradle-to-grave supply chains of steel, bioenergy, and CCS.

This chapter estimates a first-order decarbonisation potential of bioCCS in steel production. This chapter is part of a larger research project investigating the scale on which carbon-negative industries could contribute to global decarbonisation. The intention is not to provide a comprehensive or optimised assessment of bioCCS-in-steel configurations, but rather to explore bioCCS possibilities for commercial and emerging steelmaking technologies, so to assess the possibility and scale of negative emissions in steelmaking.

This chapter considers the integration of bioCCS into several steelmaking technologies, including the commercial technologies blast furnace ironmaking with basic oxygen furnace steelmaking (BF-BOF) and Midrex direct reduction of iron with electric arc furnace ironmaking (DRI-EAF), as well as the novel technologies of BF-BOF steelmaking with top gas recycling, HIsarna ironmaking with BOF steelmaking, and ULCORED DRI-EAF. For each technology, we estimated life cycle CO_2 emissions for nine cases of wood-based bioenergy use and CCS. To allow for a more equal basis of comparison, all technologies were modelled as if they are available on a commercial scale, regardless of their current state. Each case assumed that the steel mill was situated in a generic western European region. This “what if 2050 technology were available today” scenario ignores potential changes in the background supply chains (such as biomass production or electricity generation) that may also occur towards 2050, to reduce the potential confounding effects of additional uncertainty in these systems. A series of sensitivity analyses explore the significance of these background systems, and other model assumptions, to understand what changes may need to be made if bioCCS in steel is to be implemented on a large scale in the decades to come.

2 Methodology

To achieve carbon negative steel, three things must occur:

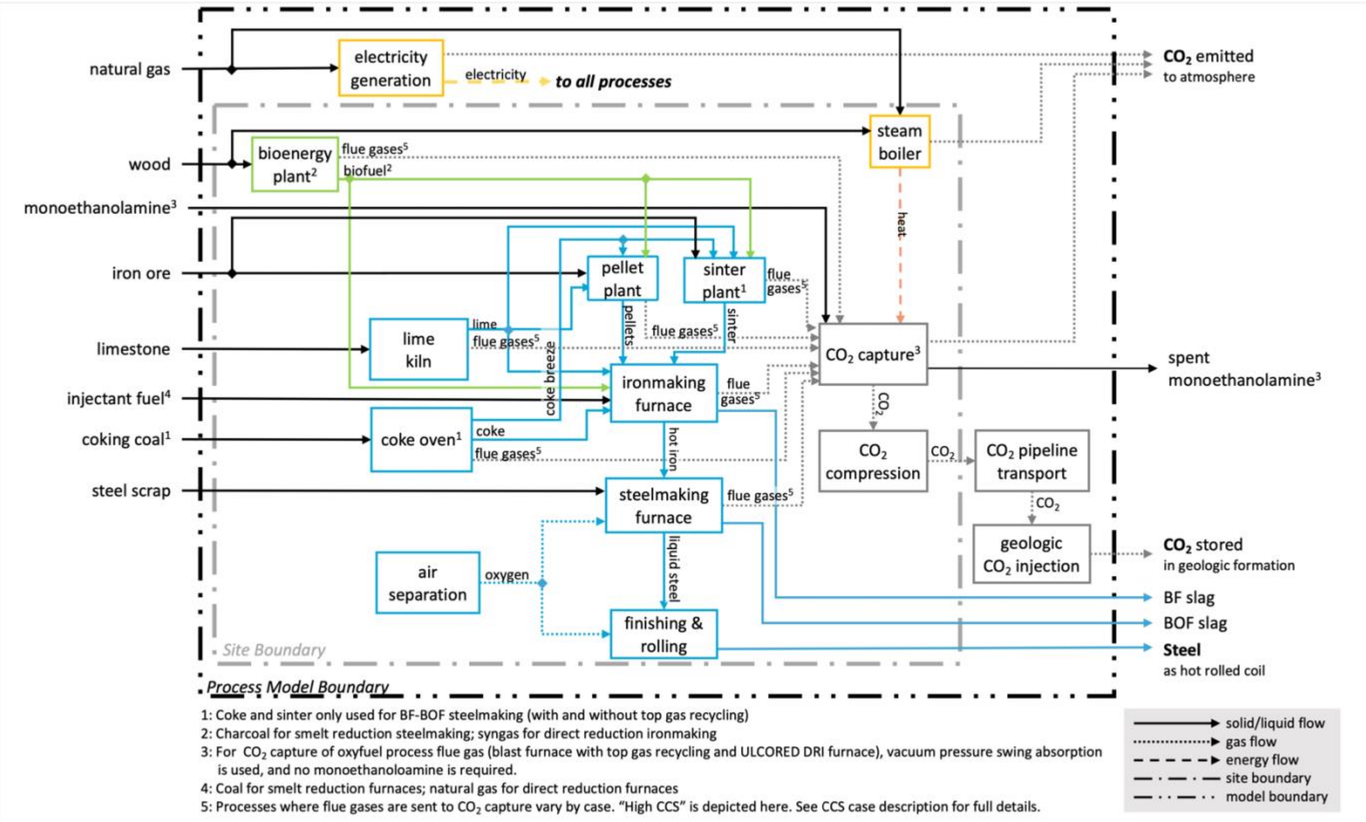
1. Fossil sources of carbon must be replaced with atmospheric sources of carbon. E.g., biomass carbon removed from the atmosphere via the by photosynthesis of biomass.
2. The removed atmospheric carbon must be permanently prevented from returning to the atmosphere. E.g., by the capture and geologic storage of CO_2 produced from the combustion of biomass.
3. CO_2 emissions elsewhere in the supply chains of steelmaking, atmospheric carbon removal, and CCS cannot exceed the atmospheric carbon removed and permanently stored.

Thus, to explore the possibility of negative CO_2 emissions, it is necessary to consider the carbon balance over the complete life cycle of the technology under consideration. Therefore, we constructed a process model that included steel production, biofuel processing, CO_2 capture and storage, and electricity generation. We used the resulting mass balances to estimate upstream and downstream CO_2 removals and emissions using generalised data from a life cycle inventory database. Together, the CO_2 removals and CO_2 emissions of the cradle-to-grave steel life cycle were used to estimate the overall CO_2 balance for each technology.

Our process models were designed to estimate the material and energy inflows, product and waste outflows, and direct CO_2 emissions for each case of steelmaking technology, bioenergy, and CCS use. The boundaries and flows of the model are summarised in **figure 2**. These models included the iron and steel furnaces, steel rolling plant, electricity generation and, as needed lime kilns, coke ovens, ore agglomeration, and/or air separation. Biofuel processing was included in the bioenergy cases, and the CCS cases additionally included CO_2 capture, compression, transport, injection into geological storage, and associated energy production.

Initially, each of the five steelmaking technologies were modelled without any bioenergy or CCS as a reference case. Then, for each technology, we considered cases of limited and high bioenergy use and limited and high CCS. The results of these 45 cases were analysed to explore the impact of steelmaking technology, bioenergy and CCS use, on the CO_2 balances. A series of sensitivity analyses further explored key assumptions in the model, including electricity generation, steam boiler efficiency, CO_2 transport distance, methane emissions, carbon debt of biomass, steel composition, and biofuel production efficiency.

Section 2.1 describes the process models for each steelmaking technology. Section 2.2 describes the bioenergy and CCS cases with their relevant model changes. Finally, section 2.3 describes the life cycle CO_2 accounting methods. **Appendix A** at the end of this dissertation describes the modelling software used.



1: Coke and sinter only used for BF-BOF steelmaking (with and without top gas recycling)
2: Charcoal for smelt reduction steelmaking; syngas for direct reduction ironmaking
3: For CO_2 capture of oxyfuel process flue gas (blast furnace with top gas recycling and ULCORED DRI furnace), vacuum pressure swing absorption is used, and no monoethanoloamine is required.
4: Coal for smelt reduction furnaces; natural gas for direct reduction furnaces
5: Processes where flue gases are sent to CO_2 capture vary by case. "High CCS" is depicted here. See CCS case description for full details.

Figure 2. Process model with system boundaries, including bioenergy and CCS use

2.1 Steelmaking process models

For each case, process models were built to simulate the production of hot rolled coil of carbon steel. A custom python model was built to calculate mass balances for each of the unit processes shown in **figure 2**, linking the process flows to generate mass balances for the steel plant as a whole. The models used fixed ratios of inputs and outputs based on pre-existing literature models, as detailed in the technology descriptions below. The models focus on flows of metal, carbonaceous materials, and energy carriers. While they do not extensively account for chemical reactions or enthalpy flows within individual processes, they allow for a standardised comparison of a greater number of configurations. It is not the intention of this chapter to provide technical or economic optimisation of bioCCS-in-steel, but rather to explore the impacts of different system configurations on the overall CO_2 balance of emerging technologies.

For each technology, the models assumed commercial-scale production, using efficiencies from modern western European steel production. The iron-making process is unique for each technology and they are described in section 2.1.1. As much as possible, auxiliary processes, detailed in section 2.1.3, used the same data sources and assumptions for all technologies, to increase the comparability of the results. Similarly, the energy content and emission factors of fuels were standardised, using factors from [26], shown in **table 1**. Fuel was assumed to be fully combusted. Similarly, limestone and other carbonated fluxes were assumed to be fully calcinated. In all cases, the reference data and assumptions were verified with additional literature, as noted throughout the following sections.

Modern steel mills recycle waste heat and combustible offgases to satisfy the heat demand of endothermic processes, with blast furnace gases typically providing 5 GJ/t HRC [27]. Commonly, the offgas energy exceeds the process heat demand, and is used to co-generate electricity or exported [27-28]. To maintain the comparability of the models, the reuse of offgases in steel mill processes was kept in alignment with the reference models, but co-generation of electricity or export heat was disregarded. All electricity was assumed to be imported from the grid. The integration of bioenergy and CO_2 capture was assumed to not impact the existing heat integration. Any additional heat required by CO_2 capture or bioenergy processes was assumed to be produced via an independently-fired steam boiler.

Table 1. Energy contents and emission factors of fuels used in this model. [26]

Fuel type	GJ/t	kg CO_2/GJ
Coking coal	28.2	94.7
Bituminous coal	25.8	96.1
Natural gas	48.0	56.0
Charcoal	29.5	111.9

2.1.1 Ironmaking technologies

The primary characteristics of the five ironmaking technologies are summarised in **table 2**, including inflows of fuels, electricity, iron ore pellets and sinter, oxygen, and flux. The features of each technology are discussed in this section. Full reduction of iron was assumed, followed immediately by conversion to steel in a steelmaking furnace with an inflow of 83% hot metal and 17% scrap steel. The unalloyed liquid steel is then cast and rolled, exiting the steel mill as hot rolled coil.

2.1.1.1 Blast furnace ironmaking

In the modern blast furnace steelmaking process, powdered iron ore is agglomerated into pellets and/or sinter. The agglomerated ore is combined in the blast furnace at 1600-2000°C with coke as the primary energy source and reducing agent. Fluxes of lime, limestone and/or dolomite are used to remove impurities, such as sulphur. Pulverised coal injection (PCI) and supplemental oxygen are commonly used to increase productivity and reduce coke demand. Less commonly, natural gas, oil, waste plastic, or charcoal is injected instead of pulverised coal [27]. The resulting liquid iron, containing 3-5% of carbon, is sent to a basic oxygen furnace (BOF) for steelmaking. The BF-BOF process is responsible for 70% of global steel production, with CO_2 emissions of 2.0-3.0 t CO_2/t steel [29]. Our BF-BOF model is based on the reference design in [28], whose parameters are summarised in **table 2**. Fuel use aligns with the average EU BF fuel consumption reported in the *Best Available Techniques Reference Document for Iron and Steel* [27], though [30] and [31] report fuel use of 300 kg coke and 200 kg pulverised coal. The ore burden is also in line with the [27], and both higher and lower factions of pellet use is reported in [31].

Table 2. Summary of input parameters for ironmaking furnace models

	Blast Furnace	Blast Furnace with top gas recycling	HIsarna	Midrex	ULCORED
Process identifier	BF-BOF	BF-BOF with TGR	HIsarna-BOF	Midrex DRI-EAF	ULCORED DRI-EAF
Furnace type	Smelt reduction	Smelt reduction	Smelt reduction	Direct reduction	direct reduction
Current status[1]	Fully commercialised	Pilot plant	Demonstration plant	Fully commercialised	Pending pilot testing
Characteristics	Dominant ironmaking technology worldwide	Recycling of blast furnace gas, increasing energy efficiency and concentration of CO_2 in flue gas stream. Can be retrofitted into existing blast furnaces.	Oxygen-fed Multistage furnace, allowing the use of lower-grade coal and iron fines	Dominant DRI technology. Efficient on smaller scales than blast furnaces. Uses gaseous fuel as energy source and reducing agent.	Oxygen-fed DRI, with high-purity CO_2 flue gas stream
Steelmaking process	Basic oxygen furnace	Basic oxygen furnace	Basic oxygen furnace	Electric arc furnace	Electric arc furnace
Fuel demand, per tonne of iron	355 kg coke and 150 kg pulverised coal (13.9 GJ)	253 kg coke and 150 kg pulverised coal (11.1 GJ)[2]	610 kg bituminous coal (15.7 GJ)[3]	244 kg natural gas (11.7 GJ)	173 kg natural gas (8.3 GJ)
Flux demand[2], per tonne of iron	7 kg CaO	3 kg CaO	14 kg CaO	none	none
Iron ore burden, per tonne of iron	352 kg pellets and 1120 kg sinter	353 kg pellets and 1096 kg sinter	1700 kg pellets	1640 kg pellets	1330 kg pellets
Oxygen demand, per tonne of iron	69 kg	361 kg	1070 kg	0 kg	228 kg
Electricity demand, per tonne of iron	104 kWh	35 kWh	104 kWh4	130 kWh	60 kWh
Data sources	[28]	[28]	[32], Interview with Tata Steel research manager (26 August 2019)	[33]	[34]

1: In this chapter, all technologies are modelled as if fully commercialised today.

2: Flux may enter the furnace in the form of CaO, $CaCO_3$, and/or $CaMg(CO_3)_2$, but has been here standardised to CaO using calcination CO_2 equivalences.

3: Based on the estimated consumption of a 1 Mtpa HIsarna plant [32].

4: Actual electricity use unknown. Assumed to be the same as in the base blast furnace.

2.1.1.2 Blast furnace ironmaking with top gas recycling

Top gas recycling (TGR) is an emerging technology to reduce the demand for fresh coke and coal in a standard blast furnace by recycling its offgases back into the furnace, supplemented with oxygen to increase combustion efficiency. The offgases contain uncombusted CO and H_2 and typically have an energy content of 2.7-4.0 MJ/Nm^3 [27]. Their reinjection can reduce the demand for fresh coke and coal. In pilot tests at Tata Steel in IJmuiden, the Netherlands, the use of TGR reduced blast furnace coke demand from 360 to 230 kg per tonne of iron [35] The parameters in the commercial-scale model of TGR ironmaking in [28], shown in **table 2**, were used in this model and are aligned with the pilot test results in [35].

2.1.1.3 HIsarna smelt reduction of iron

HIsarna ironmaking uses a multi-stage furnace with an oxygen-environment and counterflow of combustible gases to maintain smelting temperatures. This allows for the use of low-quality coal and iron pellets or fines, rather than coke and sinter. The resulting liquid iron is essentially the same as from a blast furnace and can be processed to steel in a basic oxygen furnace [32]. The HIsarna model in this chapter is based on the published results of pilot testing [32], as well as communication with a research manager at Tata Steel. The pilot tests were conducted with a 40 kt/yr furnace at Tata Steel in IJmuiden, Netherlands. The construction of a 500 kt/yr HIsarna demonstration plant in Jamshedspur, India was announced at the end of 2018 [36].

In the pilot tests, 750 kg coal was needed per tonne of iron, as the small furnace size led to energy losses of 26% [32]. A commercial 1 Mt/yr HIsarna furnace is expected to reduce heat loss to 11% [32], and this higher efficiency was used in this model. The oxygen demand and iron ore demand, in **table 2**, have been kept the same as in the pilot testing, with additional data from Tata steel.

2.1.1.4 Midrex direct reduction of iron

Direct reduction of iron (DRI) accounted for 7% (89 Mt) of global steel production in 2017 [37]. DRI reduces iron ore without liquefaction, producing a porous solid form known as sponge iron. DRI requires less energy than blast furnace iron reduction [18], but sponge iron is unstable and is typically processed to steel in electric arc furnaces (EAF).

Globally, over 60% of DRI uses the Midrex process [38], which typically uses natural gas or a syngas produced from coal or other steelmaking offgases. The fuel gas is converted into an H_2 and CO rich reducing gas via a reformer, which is also used to recycle furnace gases. The Midrex model in this chapter, whose primary parameters are in **table 2**, was based on the Midrex model from [33]. This model has slightly higher energy use (< +1.0 GJ/t iron) than [18] or [27], but was the most complete reference model available.

2.1.1.5 *ULCORED direct reduction of iron*

ULCORED is a proposed DRI furnace with an oxygen environment and used partial oxidation to prepare the furnace gas. The offgases from the ULCORED furnace are expected to be nearly pure CO_2, and CO_2 removal is integrated into the design of the gas recycling process. This technology has been modelled in simulation. Pilot testing has been proposed by ULCOS and LKAB but has not begun [10]. The model in this chapter follows the ULCORED model detailed in [34], with parameters in **table 2**.

2.1.2 Steelmaking

Liquid iron from smelt reduction ironmaking is converted into steel in a basic oxygen furnace (BOF). Sponge iron from DRI is sent to an electric arc furnace (EAF), which melts it prior to its conversion into steel. In both BOF and EAF furnaces, oxygen is injected to reduce the steel's carbon content. Steel scrap, iron, and/or fluxes are added to the steelmaking furnace to control the composition. In all models a 17% steel scrap rate was assumed, following [28]. Afterwards, the liquid crude steel is sent for alloying and shaping. Our study assumes the production of pure carbon steel, without any alloying metals, produced into hot rolled coil. The parameters of steelmaking and finishing are given in **table 3**.

Table 3. **Summary of steelmaking process model parameters**

Parameter	Value
BASIC OXYGEN FURNACE STEELMAKING	*per tonne of liquid steel*
Liquid iron demand	901 kg
Steel scrap demand	190 kg
Flux demand (as CaO)	76 kg
Oxygen demand	75 kg
Electricity demand	20 kWh
Data source	[29]
ELECTRIC ARC FURNACE STEELMAKING	*per tonne of liquid steel*
Sponge iron demand	901 kg
Steel scrap demand	190 kg
Flux demand (as CaO)	12 kg
Oxygen demand	15 kg
Fuel demand (natural gas)	21 kg
Electricity demand	698 kWh
Data source	[33]
STEEL FINISHING AND ROLLING	*per tonne of hot rolled coil*
Steel losses	74 kg
Flux demand (as CaO)	5 kg
Oxygen demand	10 kg
Electricity demand	141 kWh
Data source	[28]

2.1.3 Auxiliary Processes

The model for each steel production route included the production of coke, pellets, sinter, lime, oxygen, and electricity. In reality, steel mills may purchase some or all of these products rather than produce them on-site. However, our model internalised all of these processes to understand their influence on the system. **Table 4** lists the parameters used for the auxiliary processes. Values from [28] were used for as many processes as possible, to increase standardisation between cases. The exceptions are the DRI-specific pellet production and EAF steelmaking (**table 3**), which are from the Midrex DRI model source, [33], verified with [18]. The heat demand of these auxiliary processes was assumed to be satisfied via heat integration at the steel mill, in accordance with the reference models. Therefore, the exact distribution of heat was not modelled. It was assumed all electricity was produced using natural gas in a combined cycle power plant. The emission intensity of electricity provision was explored in a sensitivity analysis.

Table 4. Summary of auxiliary process parameters (from [28] unless otherwise noted)

Parameter	Value
SMELT FURNACE PELLET PRODUCTION	**per tonne of pellet**
Fuel demand (bituminous coal)	0.72 GJ
Flux demand	19 kg
Electricity demand	75 kWh
DRI FURNACE PELLET PRODUCTION based on [33]	**per tonne of pellet**
Fuel demand (natural gas)	1.3 GJ
Flux demand	14 kg
Electricity demand	70 kWh
SINTER PRODUCTION	**per tonne of sinter**
Fuel demand (coke breeze)	1.77 GJ
Flux demand	75 kg
Electricity demand	32 kWh
LIME PRODUCTION	**per tonne of lime**
Electricity demand	30 kWh
COKE PRODUCTION	**per tonne of coke**
Coking efficiency	78%
Electricity demand	35 kWh
OXYGEN PRODUCTION	**per tonne of O_2**
Electricity demand	385 kWh
ELECTRICITY GENERATION	
Fuel type	natural gas
Generation efficiency	56.5%
STEAM HEAT GENERATION	**per GJ of steam**
Fuel type	natural gas
Combustion efficiency (LHV)	90%
Electricity demand	5 kWh

2.2 BioCCS cases

The decarbonisation potential of bioCCS in steelmaking is largely unknown, though several options of biofuel use [39] and carbon capture [40] are available. Therefore, for each technology, nine cases of bioenergy use and CCS were explored. These included the use of bioenergy alone, the use of CCS alone, the use of both bioenergy and CCS, and a reference case of no bioenergy or CCS. Cases of both “limited” and “high” bioenergy use and “limited” and “high” CCS use were included. The “limited” cases considered only bioenergy use and/or CCS at the iron-making furnace, which is the largest source of CO_2 emissions in the steelmaking process. The “high” cases consider highly ambitious but still technologically feasible uses of bioenergy and CCS. The bioenergy cases are summarised in **table 5** and the CCS cases in **table 6**.

Table 5. Cases of bioenergy use cases considered in this chapter

Steelmaking Technology	Limited Bioenergy Use (LB)	High Bioenergy Use (HB)
BF-BOF	Replacement of PCI with pulverised charcoal, and 100% replacement of steam boiler natural gas with wood chips	As LB, plus 5% charcoal replacement of coking coal, and 50% replacement of agglomeration coal with charcoal
BF-BOF with TGR	Replacement of PCI with pulverised charcoal, and 100% replacement of steam boiler natural gas with wood chips	As LB, plus 5% charcoal replacement of coking coal, 50% replacement of agglomeration fuel with charcoal, and 100% replacement of steam boiler natural gas with wood chips
HIsarna-BOF	20% replacement of furnace coal with charcoal, and 100% replacement of steam boiler fuel with wood chips	45% replacement of furnace coal with charcoal, 50% replacement of agglomeration fuel with charcoal, and 100% replacement of steam boiler natural gas with wood chips
MIDREX DRI-EAF	50% replacement of DRI natural gas with wood biosyngas, and 100% replacement of steam boiler natural gas with wood chips	100% replacement of DRI fuel with wood biosyngas, 50% replacement of agglomeration fuel with charcoal, and 100% replacement of steam boiler natural gas with wood chips
ULCORED DRI-EAF	50% replacement of DRI natural gas with wood biosyngas, and 100% replacement of steam boiler natural gas with wood chips	100% replacement of DRI fuel with wood biosyngas, 50% replacement of agglomeration fuel with charcoal, and 100% replacement of steam boiler natural gas with wood chips

Table 6. Cases of CO_2 capture considered in this chapter

Steelmaking Technology	Limited CCS (LC)	High CCS (HC)
BF-BOF	Capture of blast furnace gas only	Capture of all steelmaking flue gas streams (and of all flue gas streams at charcoal production)
BF-BOF with TGR	Capture of blast furnace gas only	Capture of all steelmaking flue gas streams (and of all flue gas streams at charcoal production)
HIsarna-BOF	Capture of HIsarna furnace gas only	Capture of all steelmaking flue gas streams (and of all flue gas streams at charcoal production)
Midrex DRI-EAF	Capture of pure CO_2 streams only from DRI (and of pure CO_2 biosyngas production)	Capture of all steelmaking flue gas streams (and of all flue gas streams at biosyngas production)
ULCORED DRI-EAF	Capture of pure CO_2 streams only from DRI (and of pure CO_2 biosyngas production)	Capture of all steelmaking flue gas streams (and of all flue gas streams at biosyngas production)

2.2.1 Bioenergy use

In BF-BOF steelmaking, the replacement of coal and coke with biofuel is limited by the need to maintain certain mechanical properties to control the burn rate of the fuel. This chapter assumes the use of charcoal using replacement rates that likely allow for the quality of the product to be maintained without significant alteration to the production process [39]. For HIsarna steelmaking, bioenergy use cases were based on discussions with a research manager from Tata Steel. All charcoal was assumed to be produced in hot tail kilns, which are used for the charcoal produced for the steel industry in Brazil [41]. The model parameters for charcoal production are summarised in **table 7**.

For the DRI steelmaking models, a wood-based biosyngas replaced natural gas as the reducing agent in the DRI furnace. Theoretically, Midrex DRI can use 100% syngas; this is already seen with syngas derived from coal, coke oven gas, and other steelmaking offgases [42]. In theory, any fuel gas with a quality ratio of $(\%\ CO + \%\ H_2) / (\%\ CO_2 + \%\ H_2O) > 2$ can be used for DRI, but in practice, a ratio of 11 or higher is desired [43]. Therefore, a high-purity and high-energy biosyngas was assumed, based on a model of commercialised production of biosyngas intended for Fischer-Tropsch fuel synthesis [44], using the model parameters are summarised in **table 7**.

Table 7. Summary of model parameters for biofuel production

Parameter	Charcoal production (hot tail kiln)	Biosyngas production (Fischer-Tropsch synthesis quality)
Feedstock demand (per tonne of biofuel)	1520 kg wood chips (dry basis)	2930 kg wood chips (dry basis)
CO_2 production (per tonne of biofuel)	1382 kg (flue gas)	1240 kg (pure) 193 kg (flue gas)
Other inputs (per tonne of biofuel)	n.a.	192 kg O_2 2 kg MEA
Biofuel energy content (per tonne of biofuel)	29.5 GJ	21.5 GJ
Biofuel CO_2 emission factor (per GJ of biofuel)	112 kg	65 kg
Data source	[41]	[44]

2.2.2 Carbon capture and storage

The model parameters of CO_2 capture are summarised in **table 8**. For each steelmaking technology, the CO_2 capture technology and energy use were chosen to align with the differences in process, CO_2 concentration, and available literature, based on [40]. Vacuum pressure swing absorption (VPSA) was used for high-concentration CO_2 streams from the oxygen-environment furnaces in BF-BOF with TGR, HIsarna, and ULCORED DRI ironmaking, as well as for biosyngas production. For flue gases from all other processes, MEA-based amine scrubbing was used. The limited CCS cases considers only the capture of offgases from the ironmaking except for the DRI cases with bioenergy use, where CO_2 capture is also applied to the high-purity CO_2 stream in biosyngas production, as it only requires compression and transport. In the high CCS cases, all flue gas streams of steel and biofuel production are captured, except those from electricity and steam generation.

Some processes, such as the ULCORED DRI gas recycling and biosyngas production produce pure CO_2 streams as part of their process design. In the “no CCS” cases, these pure CO_2 streams are assumed to be vented or used in short-lived products. Therefore, all CO_2 produced within the system boundaries in the “no CCS cases” is treated as emissions.

For all CCS cases, the captured CO_2 was compressed to supercritical conditions, using 90 kWh per tonne of CO_2 [28]. The compressed CO_2 was transported 100km by long-distance pipeline to onshore geologic storage, with a fugitive emission rate of 1% of CO_2 transported [45], and assuming electricity use of 7 kWh/t CO_2 for repressurisation and injection [46].

In this chapter, no specific steel mill location was assumed. As access to suitable storage can vary widely, a sensitivity analysis was performed to explore the influence of CO_2 transport distances.

Table 8. Summary of model parameters for CO_2 capture, per tonne of CO_2 captured

Parameter	BF-BOF	BF-BOF with TGR	HIsarna	Midrex DRI	ULCORED	Auxiliary Processes1
Capture type	MEA-based amine scrubbing	VPSA	VPSA	MEA-based amine scrubbing	VPSA	MEA-based amine scrubbing
CO_2 capture rate[2]	90%	90%	90%	90%	90%	90%
Electricity demand	136 kWh	172 kWh	127 kWh	136 kWh	Included[3]	136 kWh
Heat demand	3.0 GJ	*n.a.*	*n.a.*	3.0 GJ	Included[3]	3.0 GJ
Monoethanolamine demand	1.0 kg	*n.a.*	*n.a.*	1.0 kg	Included[3]	1.0 kg
Data source	[28]	[47]	[47]	[28]	[34]	[29]

1: In the high-capture case, streams of lower-concentration CO_2 from auxiliary processes (e.g., the coke oven, lime kilns, and charcoal production) were modelled to be processed using MEA-based amine scrubbing, with the same parameters as that of BF-BOF flue gas assumed. For biosyngas production, VPSA was assumed.

2: Percentage of CO_2 in flue gas that is captured

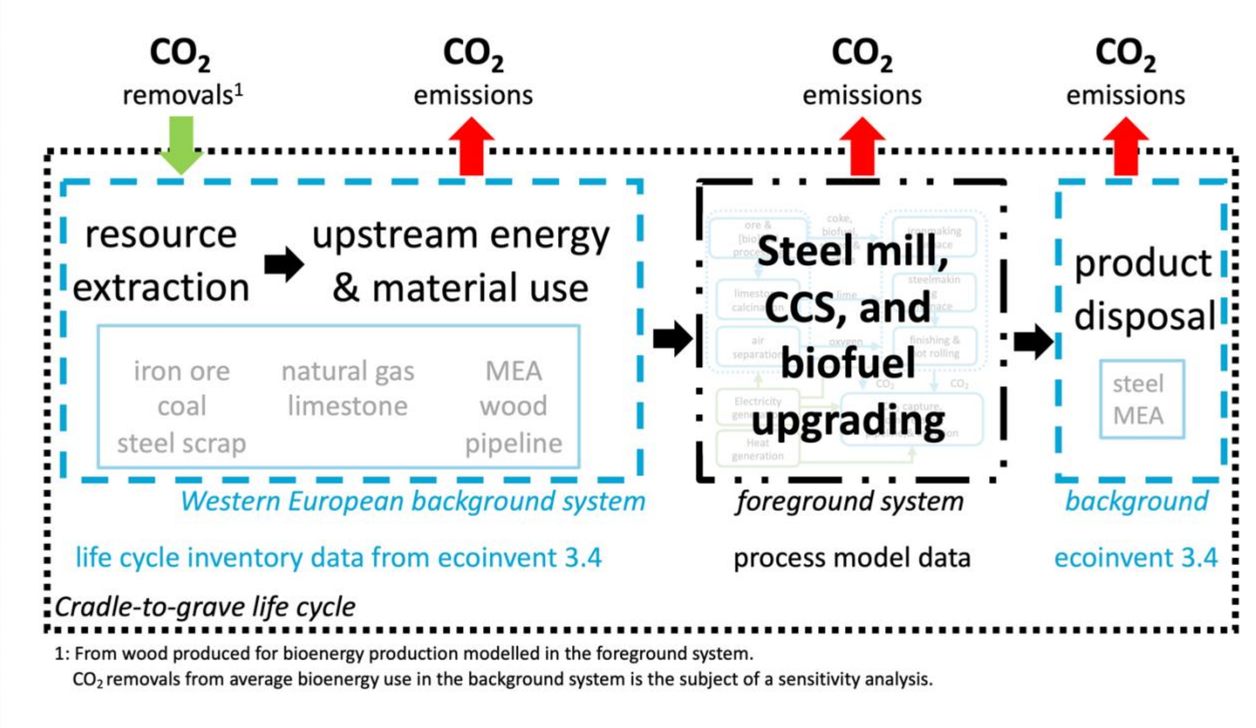
3: VPSA already integrated into ULCORED process, so no additional energy use is required

2.3 Life cycle CO_2 emissions

The process models described above estimate the direct CO_2 emissions of the steel mill, as well as CO_2 capture and bioenergy upgrading. To provide a more complete picture of the CO_2 emissions of each case, the emissions of upstream and downstream supply chains were estimated for the system summarised in **figure 3**. The CO_2 emissions of the background system was estimated using life cycle inventory data from ecoinvent 3.5 [48].

This chapter included the emissions of CO_2 from fossil and biogenic sources and CO_2 emissions attributed to land transformation. Removals of CO_2 from the atmosphere were also included for all processes. Outside of the biomass produced for production of charcoal and biosyngas, CO_2 removals primarily result from bioenergy use throughout the upstream supply chain. **Table 9** summarises the main upstream CO_2 inventory data used for this chapter. The influence of CH_4 emissions was considered in a sensitivity analysis, as is the impact of delayed carbon reuptake for biogenic CO_2 emissions, so called carbon debt. These and a number of other sensitivity analyses explore the influence of the configuration assumptions in the outcomes of this chapter.

Figure 3. **LCA System Boundaries**



1: From wood produced for bioenergy production modelled in the foreground system.
CO_2 removals from average bioenergy use in the background system is the subject of a sensitivity analysis.

Figure 3. LCA System Boundaries

Table 9. Upstream LCI data, from ecoinvent 3.5 [48].

Substance	CO_2 Emissions
Iron ore	63 kg CO_2/t iron ore
$CaCO_3$	5 kg CO_2/t $CaCO_3$
Steel scrap	121 kg CO_2/t steel scrap
Hot rolled coil, disposal	9 kg CO_2/t hot rolled coil
Coal, bituminous	201 kg CO_2/t coal
Coal, coking	241 kg CO_2/t coal
Natural gas	356 kg CO_2/t natural gas
Wood chips[1]	38 kg CO_2/t wood (dry)
Dry cleft timber[1]	33 kg CO_2/t wood (dry)
Monoethanolamine	4581 kg CO_2/t MEA
CO_2 transport	0.1 kg CO_2/tkm
Substance	**Atmospheric CO_2 Removals**
Wood chips	1810 kg CO_2/t wood (dry)
Dry cleft timber	1810 kg CO_2/t wood (dry)

1: Excludes carbon debt

3 Results

This chapter modelled the life cycle CO_2 balances of two commercialised and three emerging steelmaking technologies considering different cases of bioenergy use and carbon capture with permanent storage. The main results, using the initial model parameters are presented first, follow by the sensitivity analyses. Numerical results of CO_2 production, emissions, removals, and storage for all cases are available in the supplemental information of the published version of this chapter.

For clarity, throughout this section, only net life cycle CO_2 is presented in t CO_2/t HRC, and all other quantities are presented in kg CO_2. All quantities are rounded to the nearest 100 kg (0.1 t) to maintain a consistent level of detail.

3.1 Overall results

Figure 4 presents the estimated life cycle CO_2 balances for each case of technology, bioenergy, and CCS modelled using our base assumptions. Without any bioenergy use or CCS, BF-BOF steelmaking estimated life cycle emissions of 2.4 t CO_2/t HRC, of which 1400 kg were from the blast furnace. The addition of TGR to the BF-BOF model decreased estimated furnace emissions to 1100 kg CO_2 and life cycle emissions to 2.0 t CO_2/t HRC. For HIsarna-BOF, which has fewer auxiliary processes, life cycle emissions were 2.1 t CO_2/t HRC, of which 1500 kg were furnace emissions.

For DRI-EAF steelmaking, estimated life cycle CO_2 emissions without bioenergy or CCS were 1.5 and 1.3 t CO_2/t HRC for Midrex and ULCORED. Ironmaking furnace emissions accounted for 500-600 kg CO_2/t HRC. In both cases, approximately 400 kg CO_2/t HRC resulted from electricity use, primarily for the electric arc furnace. Overall, electricity use was 1150 kWh/t HRC, compared to 300-400 kWh/t HRC for smelt reduction technologies.

For all technologies, upstream emissions were between 200-300 kg CO_2/t HRC, accounting for approximately 15% of the life cycle emissions of smelt reduction steelmaking and 20% for DRI steelmaking. Fuel production was responsible for roughly half of upstream emissions in all cases.

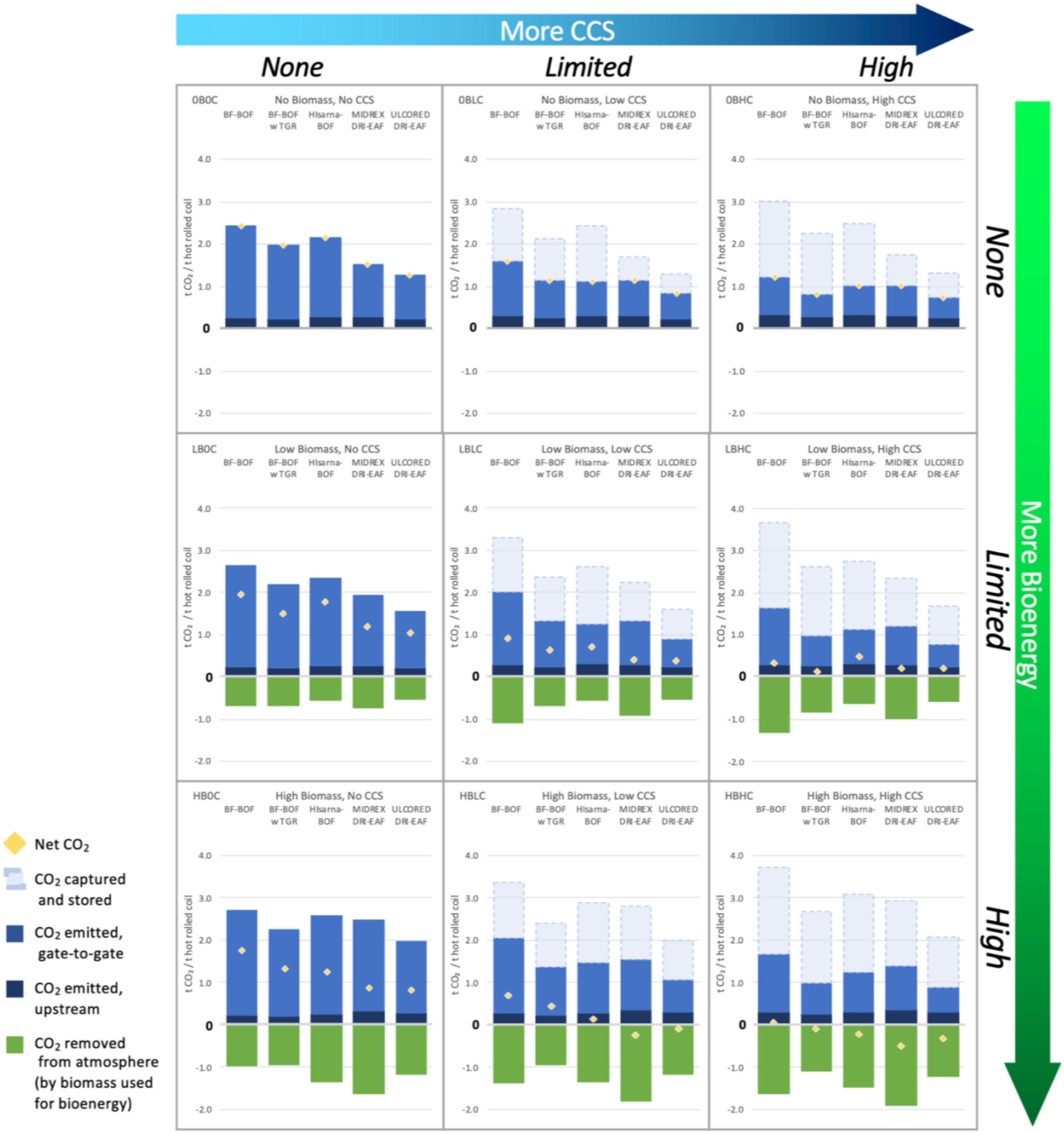


Figure 4. **Estimated life cycle CO_2 of steelmaking by technology and case of bioenergy use and CCS use**

The CO_2 emissions for the reference cases are within 85 - 99% accordance with the reference literature, when considered from the same system boundaries, despite having a coarser level of detail. Most of the difference is attributable to the use of harmonised emission factors and auxiliary process efficiencies, which may differ slightly from the reference literature. A comparison of the results of this chapter with the reference literature and an explanation of the differences are available in the supplemental information of the published version of this chapter.

Next, we considered cases of CCS use without bioenergy. CCS alone results in permanent CO_2 storage, but without the removal of atmospheric carbon, cannot result in negative emissions. In smelt reduction steelmaking, the limited CCS cases only captured CO_2 from the ironmaking furnace and resulted in 1000-1300 kg CO_2/t HRC sent to permanent storage. However, CO_2 production increased by 10% from the reference case, due to the energy demand. The net result was a 35-50% lower life cycle CO_2 to 1.6 t CO_2/t HRC for BF-BOF and 1.1-1.2 t CO_2/t HRC for BF-BOF with TGR and HIsarna-BOF. For DRI-EAF steelmaking, approximately 500 kg CO_2/t HRC was captured and stored, with total CO_2 production increasing 1-3% and life cycle CO_2 emissions decreasing 25-35% to around 1.0 t CO_2/t HRC.

The high CCS cases, applied CO_2 capture to all flue gas streams except steam and electricity production, only significantly further reduced the net life cycle CO_2 for BF-BOF steelmaking, which decreased to 1.2 t CO_2/t HRC without TGR and to 0.8 t CO_2/t HRC with TGR. In all other cases, life cycle CO_2 did not decrease more than 100 kg CO_2/t HRC compared to the limited CCS case. The energy demand of CCS accounted for approximately 90% of additional CO_2 produced. The remaining sources of increased CO_2 production include the transport and storage of CO_2 and the production and disposal of MEA. Overall, the high CCS cases show a 5% increase in CO_2 production for ULCORED steelmaking, a 15% increase MIDREX DRI, BF-BOF with TGR, and HIsarna steelmaking, and a 25% increase for BF-BOF steelmaking; all directly correlated with the throughput of the CO_2 capture unit. However, the full integration of CCS into the steel mills was modelled to reduce gate-to-gate CO_2 emissions by 40-70%, with total life cycle CO_2 emissions decreasing 30-40% for DIR steelmaking and 50-60% for smelt reduction.

In the cases of bioenergy use alone, CO_2 is removed from the atmosphere via the photosynthesis of biomass, but that CO_2 is returned to the atmosphere after the biomass is combusted. Thus, bioenergy use can reduce CO_2 emissions but cannot by itself result in negative emissions. Without CCS, the limited bioenergy cases resulted in 20-25% reduction in net life cycle CO_2 from the reference (BF-BOF: 2.0, TGR: 1.5, HIsarna: 1.8, Midrex: 1.2, ULCORED: 1.0 t CO_2/t HRC) and 30-40% reduction in the “high” cases (BF-BOF: 1.7, TGR: 1.3, HIsarna: 1.3, Midrex: 0.9, ULCORED: 0.8 t CO_2/t HRC). Total CO_2 emitted increased 100-500 kg CO_2/t HRC for smelt reduction steelmaking and 300-1000 kg CO_2/t HRC for DRI steelmaking. In all cases the increase in CO_2 production resulted primarily from the transformation of raw biomass (wood) into a suitable high-energy biofuel: charcoal for smelt reduction and biosyngas for DRI.

BioCCS combines both atmospheric CO_2 removal and permanent CO_2 storage and can theoretically result in negative emissions, if the total amount of atmospheric carbon removed and stored is higher than the amount of CO_2 emitted across the complete life cycle systems of steel, bioenergy, and CCS. In all cases, bioCCS led to both higher total CO_2 generation and lower net CO_2 than the use of bioenergy or CCS alone. Within the assumptions and boundaries in this model, six cases were estimated to be carbon neutral or carbon negative. Additionally, for all five technologies, the "limited bioenergy, limited CCS" case resulted in lower net CO_2 than either the "no bioenergy, high CCS" or "high bioenergy, no CCS" cases. For both bioenergy use and CCS, the limited uses cases already included interventions at the iron furnace, which is the largest consumer and emitter of carbon in all cases. Therefore, the high uses cases saw small marginal reductions in CO_2, compared to the limited use cases.

For BF-BOF steelmaking, with and without TGR, the net CO_2 estimates for the "limited bioenergy, high CCS" case is 300-400 kg / t HRC lower than the "high bioenergy, limited CCS" cases, due to the stricter limits on bioenergy use in the blast furnace arising from the need to maintain the mechanical properties of the fuel. In the "limited bioenergy, high CCS" case, BF-BOF with TGR approaches carbon neutrality (0.1 t CO_2/t HRC). This is in contrast HIsarna and DRI steelmaking, all of which are near or below carbon-neutral (–0.3 to 0.1 t CO_2/t HRC) in the "high bioenergy, limited CCS" case, but at 0.2-0.5 t CO_2/t HRC in the "limited bioenergy, high CCS" case. The HIsarna and DRI pathways have fewer point sources of emissions, as well as higher viable bioenergy use potentials, thus allowing for higher marginal decarbonisation potentials from bioenergy use in the ironmaking furnace.

In the "high biomass, high CCS" case, the estimated CO_2 balance of all technologies approach or exceed net carbon neutrality, with CO_2 production between 2000-4000 kg/t HRC, CO_2 emissions of 900-1700 kg/t HRC, and CO_2 removal between 1100-1700 kg/t HRC. Only BF-BOF steelmaking remained carbon-positive at 0.1 t CO_2/t HRC. The net CO_2 of BF-BOF with TGR was only slightly lower, at –0.1 t CO_2/t HRC, but the reduced fuel consumption resulted in 1000 kg/t HRC less CO_2 produced than in BF-BOF alone.

For DRI-EAF steelmaking, the life cycle CO_2 emissions in the "high biomass, high CCS" case were net negative, estimated at –0.5 t CO_2/t HRC for Midrex and –0.3 t CO_2/t HRC for ULCORED. In DRI steelmaking, CO_2 captured from biosyngas production was over half of the total CO_2 captured. In comparison, charcoal CO_2 accounted for 20-30% of CO_2 captured from smelt reduction technologies.

3.2 Sensitivity analyses

The above results consider the use of bioCCS in different steelmaking technologies under a specific set of assumptions of technological configuration, emission accounting, and the efficiency of background systems. Below we explore the impact of some of these assumptions, including the carbon intensity of electricity, CO_2 transport distance, steam boiler efficiency, methane emissions, charcoal kiln efficiency, carbon debt, and the use of alloying metals. The supplemental information of the published version of this chapter contains the numerical results of the sensitivity analysis as well as the results of sensitivity analyses that had little impact on the results, including the inclusion of upstream emissions of factory and equipment use; atmospheric CO_2 removal in the background supply chain; and the HIsarna burden composition.

3.2.1 Electricity production

The base model assumed that electricity was generated using natural gas, resulting in an electricity emission factor of approximately 400g CO_2/kWh. If electricity is produced from coal, the carbon intensity can be up to 850-1020 g CO_2/kWh, depending on coal type [49], decarbonisation of electricity is a central component the EU's ambition to be carbon-neutral by 2050 [50]. **Figure 5(A)** shows the impact of a CO_2 emission factor of electricity between 0-1000 g CO_2/kWh in the "high bioenergy, high CCS" cases as the high bioCCS cases are those with the highest electricity demand, and thus highest sensitivity to its emission factor.

Without bioenergy or CCS, the reduction of electricity's carbon intensity from 400 g CO_2/kWh to 0 g CO_2/kWh results in 100-200 kg /t HRC less CO_2 for smelt reduction steelmaking and 500 kg t/HRC less CO_2 for DRI-EAF steelmaking. The use of bioenergy has little impact on electricity use, and the difference in electricity demand between the baseline and high bioCCS case results almost entirely from CCS.

At a CO_2 intensity of around 700 g CO_2/kWh, slightly above the average carbon intensity of electricity production in China in 2017 [49], net CO_2 estimates are positive for all technologies. At 300 g CO_2/kWh, similar to that of the EU grid in 2018 [49], all net CO_2 balances are negative. Full decarbonisation of electricity decreases the net CO_2 of the high bioCCS cases by 400 kg CO_2/t HRC for smelt reduction steelmaking and 600 kg CO_2/t HRC for the more electricity-intensive DRI-EAF steelmaking.

3.2.2 Boiler efficiency

In the baseline model, a 90% boiler efficiency is assumed for the provision of heat for CO_2 capture. Depending on size and configuration, boiler efficiency may be lower, particularly for high-moisture fuels, such as wood chips. Overall, boiler efficiency had a noticeable yet limited impact on net CO_2. As shown in **figure 5(D)** for the high bioCCS cases, with a 30% decrease in boiler efficiency increasing the net CO_2 of any case by no more than 100 kg CO_2/t HRC.

3.2.3 CO_2 transport distance

In the reference case, a CO_2 transport distance of 100km is assumed. [25] identified CO_2 pipeline routes between 30 steel plants and off-shore storage aquifers, with pipeline distances ranging from 1 - 799 km. Therefore, our sensitivity analysis considered pipeline distances of

0-1000 km, as shown in **figure 5(C)** for the high bioCCS cases. For all technologies, increasing the transport distance from 100 km to 1000 km increases net life cycle CO_2 emission by less than 100 kg CO_2 /t HRC.

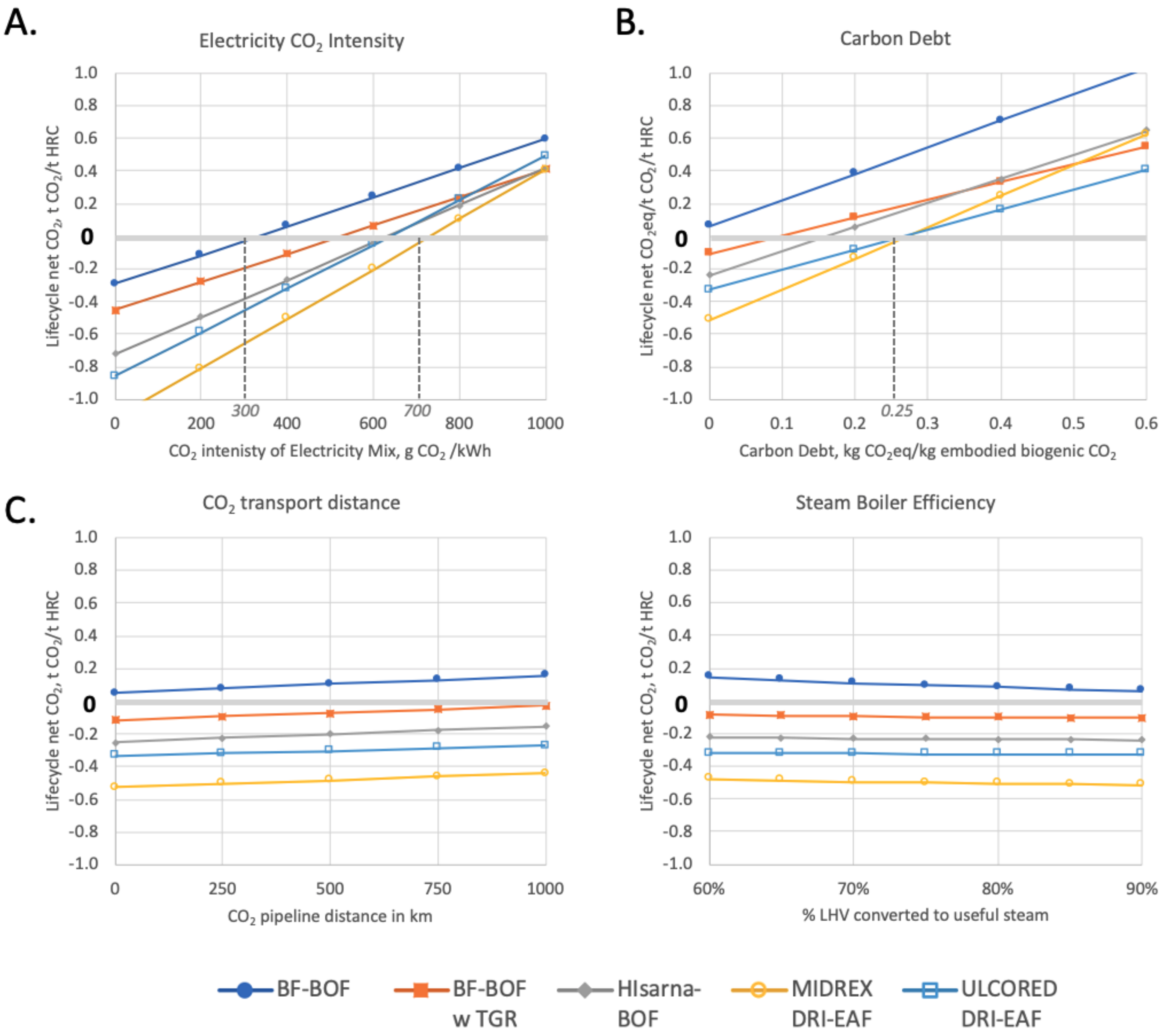


Figure 5. Sensitivity analysis of electricity carbon intensity (A), biomass carbon debt (B), CO_2 transport distance (C), and steam boiler efficiency (D) in the cases of high bioenergy and high CCS use for all technologies.

3.2.4 Carbon debt

CO_2 from bioenergy combustion is emitted all at once, but the equivalent (re-)uptake of atmospheric CO_2 by biomass takes a number of years dependent on the rotation period of the crop. Even when the biomass is sustainably grown, with attention to replanting and land use change, as is assumed in our model, the delay in CO_2 reuptake and changes in soil carbon, bacterial activity, and albedo occurring as a response to biomass harvest increase the global warming potential of biogenic CO_2 emissions [51]. These factors, collectively known as the "carbon debt" of biomass, represent the greenhouse gas emission reduction that the use of biofuel must provide to be carbon neutral [53]. The "carbon debt" is independent of other CO_2 emissions in the biomass supply chain, such as those from fertiliser use or equipment and energy use in harvest and transport.

[53] calculate "GWP_{bio}" factors, estimating the global warming potential of these processes in kg CO_2-eq per kg of biogenic CO_2 emitted. These factors are relative to the rotation period of the biomass and the time horizon of the study. At the 100-year time horizon, annual crops having a negligible "GWP_{bio}" factors (0.003 kg CO_2eq/kg biogenic CO_2), but long-rotation crops, such as hardwood timber with a 100-year rotation period are estimated to a "GWP_{bio}" factor of 0.44 kg CO_2eq/kg biogenic CO_2.

Figure 5(B) shows the impact of these GWP_{bio} factors on the net CO_2 emissions of the high bioenergy cases. At a GWP_{bio} factor of 0.25 (on a 100-year time horizon), corresponding to a rotation period of roughly 60 years, all technologies have a net-positive CO_2 balance.

3.2.5 Methane emissions

The base model only considered CO_2, which is responsible for 90% of the global warming potential of steel production [54]. A full greenhouse gas accounting was outside the scope of this chapter, but **figure 6** shows the influence of including the estimated methane emissions of charcoal production in the hot tail kilns and the methane emissions of upstream processes from ecoinvent. Methane emission data was not available for the biosyngas production. As in the base model, all fuel carbon used for steelmaking is assumed to be fully combusted.

For cases without charcoal use, methane emissions added 100-200 kg CO_2eq/t HRC, over 90% of which results from fossil fuel supply chains. In the high biomass cases for smelt reduction, methane produced for charcoal added an additional 200 kg CO_2eq/t HRC, leading their net life cycle CO_2-eq estimates to increase to a net positive 0.2-0.4 t CO_2eq/t HRC.

While outside the scope of this chapter, methane leakage could play an important role in the greenhouse gas balance of steelmaking, due to its high global warming potential, and the formation of methane in both the steelmaking gases, and the energy supply chains. For the high bioCCS case of MIDREX DRI-EAF, which had the lowest net CO_2, only 20 kg/t HRC of methane leakage are necessary anywhere in the supply chains of steel, bioenergy, and/or CCS for the system to have a net positive global warming potential (on a 100-year time horizon).

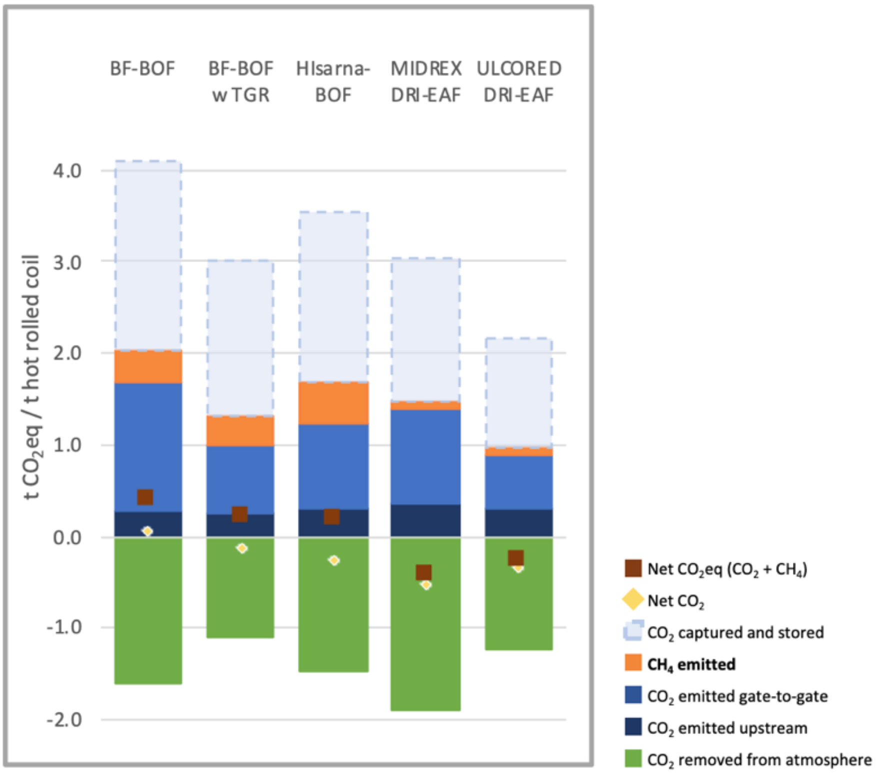


Figure 6. **Estimated impact of CH_4 emissions from charcoal production and background systems on life cycle CO_2-eq (100-year time horizon).**

3.2.6 Charcoal production

The base model assumed that charcoal was produced in industrial hot tail kilns with CO_2 emissions of 1400 kg CO_2/t charcoal. Hot tail kilns are used to produce charcoal for the steel industry in Brazil, however, they are less efficient than Missouri kilns, which can have CO_2 emissions between 400-700 kg CO_2/ t charcoal [41]. Without CCS, use of a highly efficient kiln with CO_2 emissions of 500 kg CO_2/t charcoal reduced the estimated net CO_2 by 200 kg CO_2/t HRC. In the high bioCCS cases, CO_2 capture is assumed to be applied to charcoal production, so while CO_2 production is reduced by a similar amount and total net CO_2 is lower than without CCS use, more efficient kiln usage decreases net CO_2 emissions by less than 100 kg CO_2/t HRC as compared to a hot tail kiln. Further results of the charcoal kiln efficiency analysis are available in the supplemental information of the published version of this chapter, including for less efficient kilns, although these are unlikely to be used on an industrial scale.

3.2.7 Alloying metals

The base model assumed the production of unalloyed carbon steel. Using data from ecoinvent [48], the use of the small amounts of nickel, chromium, and magnesium in “low alloyed” steel added an additional 500 kg CO_2/t HRC to the life cycle net CO_2 in all cases. The sourcing of the much larger amounts of chromium and nickel in 18/8 stainless steel added an additional 3600 kg t CO_2/t HRC to life cycle net CO_2, requiring that any possibility of carbon negative stainless steel address the CO_2 emissions of the chromium supply chain. However, the decarbonisation of the chromium supply chain is outside the scope of this chapter, but as the steel industry is the primary consumer of chromium [55], it is an aspect that requires further attention.

4 Discussion

This chapter considered a “tomorrow’s technology today” scenario where current and emerging steelmaking technologies were considered on an equally commercialised basis, as they might exist in 20-40 years’ time, while using present-day CO_2 emission data for the background supply chains. This reduced the uncertainty in the model and limited the changes in net CO_2 to changes in the steelmaking supply chain. However, the data quality is thus inherently unequal between the different technologies and is more uncertain particularly for HIsarna-BOF and ULCORED DRI-EAF steelmaking.

In the reference BF-BOF case, which most closely represents the current dominant steelmaking technology, over 80% of life cycle CO_2 production resulted directly from the steelmaking process. However, in the high bioCCS case, CO_2 from steelmaking increases slightly, but its relative share drops to 60% of life cycle CO_2 production, as CO_2 from bioenergy production and CCS energy use increase. Similarly, for DRI-EAF, direct emissions from steelmaking represent 50% of life cycle CO_2 production in the reference case and 30% in the high bioCCS case. In a bioCCS-in-steel system, the carbon intensity of the background sectors, particularly for energy sourcing including biomass production, fossil fuel extraction, and electricity generation, become more important, and therefore require greater rigour when estimating the CO_2 balance of a specific bioCCS-in-steel implementation. The influence of the composition of the steel, including both recycled scrap content and alloying metals (see section 3.1.5) also deserve great attention.

It is important to emphasise that negative CO_2 emissions do necessarily imply negative global warming potential. Though CO_2 is responsible for over 90% of steelmaking’s global warming potential [54] the impact of additional greenhouse gases, such as methane, nitrous oxide, and fluorocarbons, are not accounted for in this chapter, though the impact of methane from charcoal productions was briefly discussed (section 3.1.3).

Below, we briefly address some further considerations of bioCCS-in-steel beyond our model, including the practicality of implementation, inefficient negative emissions, and resource use.

4.1 Implementation considerations

The CO_2 emissions of steel production are dominated by those emitted during the steelmaking process, with the ironmaking furnace being the single largest source of CO_2 emissions for all technologies. The choice of ironmaking method affects not only the CO_2 emissions in the reference case, but also the effectiveness of bioCCS.

4.1.1 Bioenergy use

In BF-BOF steelmaking, the replacement of coal with charcoal has been limited by the need to maintain the mechanical properties of the fuel to maintain consistent furnace parameters, and therefore maintain the quality of the iron. In DRI-EAF steelmaking, the use of a gas fuel theoretically allows for complete replacement with biosyngas, and in this model showed a greater potential for negative emissions than BF-BOF steelmaking. However, there is current commercial use of charcoal in blast furnaces, but no commercial DRI plant currently uses biosyngas. The production of charcoal is also an established commercialised process that produces a homogenous end product, whereas the production of high-quality biosyngas is an emerging industry with heterogenous feedstocks and products. This lack of experience may prove a greater hurdle to widespread bioenergy use in DRI-EAF than in BF-BOF steelmaking, even if the decarbonisation potential for bioenergy in DRI-EAF steelmaking is greater.

The bioenergy supply chain has complex impacts on global warming, as captured partially in carbon debt factors in 3.1.2, related to land use change, albedo, soil carbon disruption, and the delay between CO_2 (re)uptake and biomass combustion. Wood-based bioenergy is of particular concern for European biomass production, as spruce and pine can have rotation periods as long as 100-150 years northern European countries [56], with a carbon debt factor of 0.4 kg CO_2eq/kg biogenic CO_2 [53]. In contrast, Eucalyptus in equatorial regions can have a rotation period as short as 5-10 years [56], implying a carbon debt factor of < 0.1 [53]. However, if used in European steelmaking, equatorial biomass adds the additional complexities of long distance transport and multiregional supply chain governance. In our model, biosyngas was assumed to be produced from wood, but biosyngas can also be produced from annual crops (e.g. [57, 58]), which could substantially decrease the carbon debt burden.

4.1.2 CO2 capture

In contrast, CO_2 capture has been commercially applied to DRI-EAF steelmaking, where gas cleaning and reforming is an integrated process. In BF-BOF steelmaking, which produces offgases with more contaminants, CO_2 capture is not yet commercialised.

Top gas recycling theoretically allows for easier CO_2 capture at a blast furnace by increasing to the CO_2 concentration of the offgases, but this technology is still under development. Top gas recycling can also increase the fuel efficiency of iron production, but it reduces the available energy from the blast furnace offgases, so if less energy is then available for the previous use of the offgases (e.g., heat in other steelmaking processes or electricity export), additional energy may be needed to satisfy those processes, thus potentially generating additional CO_2 emissions elsewhere.

In the high CCS cases, we assumed that all flue gases were processed for CO_2 capture, except those of electricity and steam generation. While technologically possible, this may prove economically or spatially impractical, requiring extensive ductwork, and trade-offs between combining and transporting flue gases of different pressures, temperatures, and CO_2 concentration, or CO_2 capture, units at multiple point sources [59]. However, integrated steel mills typically extensively redirect combustible off gases, and therefore are likely to have the expertise necessary to design gas transport solutions for CO_2 capture.

4.2 "Inefficient" negative emissions

The lower net CO_2 emissions of HIsarna over BF-BOF with TGR and of Midrex DRI-EAF over ULCORED DRI-EAF in the high bioCCS case illustrates a counterintuitive phenomenon wherein a bioCCS system with lower energy efficiency can result in a lower CO_2 balance than a bioCCS system with a higher energy efficiency. This is due to the larger quantity of CO_2 which is removed from the atmosphere to supply bioenergy and then is subsequently captured and permanently stored, resulting in more negative CO_2 emissions. In the more efficient systems, the lower bioenergy demand leads to less CO_2 removal from the atmosphere and subsequently less storage of removed atmospheric CO_2.

Such "inefficient" systems can generate more negative CO_2 emissions by using more resources (e.g., wood, electricity) for the same quantity of steel production. However, this necessarily increases costs, as well as competition for limited resources. Unless negative emissions are themselves sufficiently economically valued, the "inefficient" generation of negative emission will not be appealing. This concept of "inefficient" production to increase negative emissions has been explored for power generation in [13].

4.3 Resource demand

The change in demand for energy resources—biomass, fossil fuels, and electricity— from the reference cases to the high bioCCS case is summarised in **table 10**. While the high bioCCS cases decrease net CO_2 by 1500-2400 $kgCO_2/t$ HRC from the reference cases, it increases total primary energy demand by an average of 6 GJ/t HRC including an average of 500 kWh of final electricity demand and 800 kg (dry mass) of wood per tonne of steel.

A first estimate indicates that if all blast furnace steel production in Europe (100 Mt/year) was fitted with top gas recycling and implemented the high bioCCS cases, annual European steel industry CO_2 emissions would decrease by 260 Mt, and the net CO_2 balance of European BF-BOF steelmaking would be −10 Mt CO_2/year, under the assumptions and system boundaries here considered. This case also requires an addition 52 Mt/year of dry wood (dry basis), which is 15% of the total European forestry harvest [60, 61], as well as an additional 50 TWh/year of electricity, increasing European industrial electricity usage by 5%. This increased demand is also expected to compete with the electrification and decarbonisation efforts in other industries and the power sector, compounding pressure on available renewable energy resources.

Table 10. Resource use of bioCCS (high bioenergy, high CCS case compared to reference case)

	Unit	BF-BOF only	BF-BOF with TGR	HIsarna-BOF	Midrex DRI-EAF	ULCORED DRI-EAF
Net CO_2 (change from reference case[1])	t/t HRC	0.1 (−2.3)	−0.1 (−2.1)	−0.3 (−2.5)	−0.5 (−2.0)	−0.3 (−1.6)
Primary energy demand[2] (change from reference case[1])	GJ/t HRC	28 (+10)	20 (+6)	26 (+7)	27 (+5)	20 (+2)
Biomass demand[3]	kg dry wood/t HRC	890	600	820	1030	660
Coal (change from reference case[1])	kg t HRC	460 (−210)	340 (−200)	350 (−290)	0 (0)	0 (0)
Natural gas[4] (change from reference case[1])	kg/t HRC	0 (0)	0 (0)	0 (0)	130 (−140)	120 (−170)
Electricity[5] (change from reference case[1])	kWh/t HRC	890 (+520)	860 (+550)	1200 (+790)	1530 (+390)	1360 (+220)

1: Without bioenergy or CCS

2: Including fossil fuel and biofuel used in steel making, auxiliary processes, and electricity generation. Includes losses.

3: There is no biomass demand in the reference case

4: Excluding for electricity generation

5: Final electricity demand. Excludes losses.

5 Conclusions

In this chapter, 45 cases of steelmaking technology, bioenergy use, and CCS use were modelled to explore the impact of bioCCS on the net life cycle CO_2 of steelmaking. Each case was modelled using a fixed-ratio input-output process model for the production of steel, auxiliary inputs, bioenergy, and CCS, at a commercial-scale modern integrated steel mill in Western Europe. The results of the process model were used to estimate the emissions of the upstream and downstream supply chains. As this chapter focused on exploratory work, the systems were not optimised, and a number of parameters were explored in non-stochastic sensitivity analyses.

In our model, the use of CCS alone resulted in higher net CO_2 reductions than the use of bioenergy alone, but the combination of bioenergy and CCS resulted in greater net CO_2 reductions than the sum of separate interventions. In particular, the use of both bioenergy and CCS at the ironmaking furnace showed greater decarbonisation potential than site-wide deployment of either bioenergy or CCS alone. Aggressive deployment of both bioenergy and CCS in the high bioCCS case resulted in estimates of near-neutral net CO_2 for BF-BOF steelmaking with and without top gas recycling (0±0.1 t CO_2/t HRC), and slightly negative net CO_2 (−0.2 to 0.3 t CO_2/t HRC) for HIsarna-BOF, Midrex DRI-EAF, and ULCORED DRI-EAF. This required the use of bioenergy both for ironmaking and some auxiliary processes, as well as CO_2 capture on all flue gases from steelmaking and bioenergy production, followed by permanent storage.

A series of non-stochastic sensitivity analyses explored the role of the carbon intensity of electricity, CO_2 transport distance, steam boiler efficiency, methane emissions, charcoal kiln efficiency, carbon debt, and the use of alloying metals on the life cycle CO_2 estimates. Net CO_2 estimates were particularly sensitive to the carbon intensity of electricity, the use of alloying metals, and the role of biomass carbon debt. In this chapter, a decarbonised electricity sector was shown to reduce net CO_2 by approximately 500 kg CO_2/t HRC in the high bioCCS cases. However, the high bioCCS cases also increases electricity use by approximately 500 kWh/t HRC, primarily from the CO_2 capture system.

Furthermore, this chapter assumed that the biomass was sustainably harvested and regrown, but the delay in carbon reuptake, along with other impacts of biomass production, can increase the global warming potential of biogenic CO_2, which is highly dependent on the rotation period of the biomass. Slow-growing tree species, such as Norwegian spruce or Scots pine could have additional global warming impacts that negate the carbon removal benefit of bioenergy use, when considered within a 100-year time horizon. Emissions of biogenic methane from bioenergy production, nitrogen emissions from biomass production, and other greenhouse gases, also deserve further attention, to better estimate whether negative CO_2 steel production results in negative global warming potential.

It is our initial assessment that negative life cycle CO_2 emissions in the production of carbon steel are possible through aggressive use of bioenergy paired with the capture and permanent storage of CO_2 from both steelmaking and bioenergy production, if rigorous attention is paid to ensure the sustainability of the energy and biomass supply chains. The use of decarbonised electricity, short-rotation biomass, and efficient bioenergy production increase the likelihood of a net negative CO_2 balance. Real-world implementation of bioCCS in steelmaking requires a thorough life cycle assessment for the specific technological configuration and supply chain choices to determine if negative emissions can be achieved.

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Chapter 6:

Timing of removals and emissions for bioCCS and accelerated carbonation in concrete

The decarbonisation of concrete production will require a multi-pronged approach including the abatement of CO_2 emissions from cement production as well as storage of CO_2 within concrete itself. This chapter explores the decarbonisation potential of combining bioenergy and carbon capture and storage (CCS) during cement production with the accelerated carbonation of fresh concrete and the natural carbonation of demolished concrete for the life cycle net CO_2 of 30MPa ordinary Portland concrete. As both biomass and concrete reuptake CO_2 over time, the timing of CO_2 emissions and removals is explicitly accounted for. At current technology levels, the combination of bioenergy and CCS in cement production combined with the carbonation of demolished concrete was seen in our model to allow for net CO_2-negative concrete. However, the concrete is CO_2-positive until the CO_2 of production is reabsorbed by biomass regrowth and the carbonation of demolished concrete at end-of-life. In our model, accelerated carbonation was, by itself, an inefficient CO_2 storage mechanism, due to the penalty of energy use and injection losses. However, if it led to a gain in concrete strength, accelerated carbonation could result in lower CO_2 via reduced resource demand and cement production.

This chapter was originally published as Tanzer, S. E., Blok, K., Ramirez, A. (2021). *Curing time: A temporally explicit life cycle CO_2 accounting of mineralisation, bioenergy, and CCS in the concrete sector.* Faraday Discussions, 0–11. doi:10.1039/d0fd00139b. Tabularised versions of the results are available in the publication's supplemental information. The raw dataset for this chapter is available at doi: 10.5281/zenodo.6369423.

Note: Spellings, formatting, and abbreviations have been standardised throughout the dissertation.

1 Introduction

Concrete is the most abundant manmade material. Worldwide, 30 billion tonnes are produced annually [1]. The production of 4.1 billion tonnes of cement [2], the binding agent in concrete, was itself responsible for 2.4 Gt of carbon dioxide emissions in 2019, representing 26% of all industrial CO_2 emissions [3]. To abate the worst impacts of the climate crisis, decarbonisation of concrete is critical.

The cement and concrete industries anticipate that full decarbonisation will require a multi-pronged approach, encompassing increases in energy efficiency, the use of wastes and biomass as fuel, and recycling; carbon capture and storage; as well as the decarbonisation of transport and electricity [4, 5].

Cement production is the most carbon-intensive element of the concrete supply chain, requiring 3-4 GJ of thermal energy per tonne of cement, energy which today is provided mostly by fossil fuels [6]. Yet, about 60% of CO_2 emitted during cement production is from the calcination of limestone ($CaCO_3$) into calcium oxide (CaO). Once in concrete, however, CaO reabsorbs CO_2 from the atmosphere, recarbonating into limestone. Estimates suggest that CO_2 reuptake by in-stock concrete offsets 20% of CO_2 emissions from current annual cement production [7]. Over a 50 to 100-year service life, concrete may reabsorb 10-30% of the CO_2 released during calcination of its constituent cement [8, 9]. At the end of its service life, demolition greatly increases the exposed surface area of concrete, providing an opportunity for rapid recarbonisation [10], but less than 1% of demolished concrete is estimated to be recycled in an exposed environment [7].

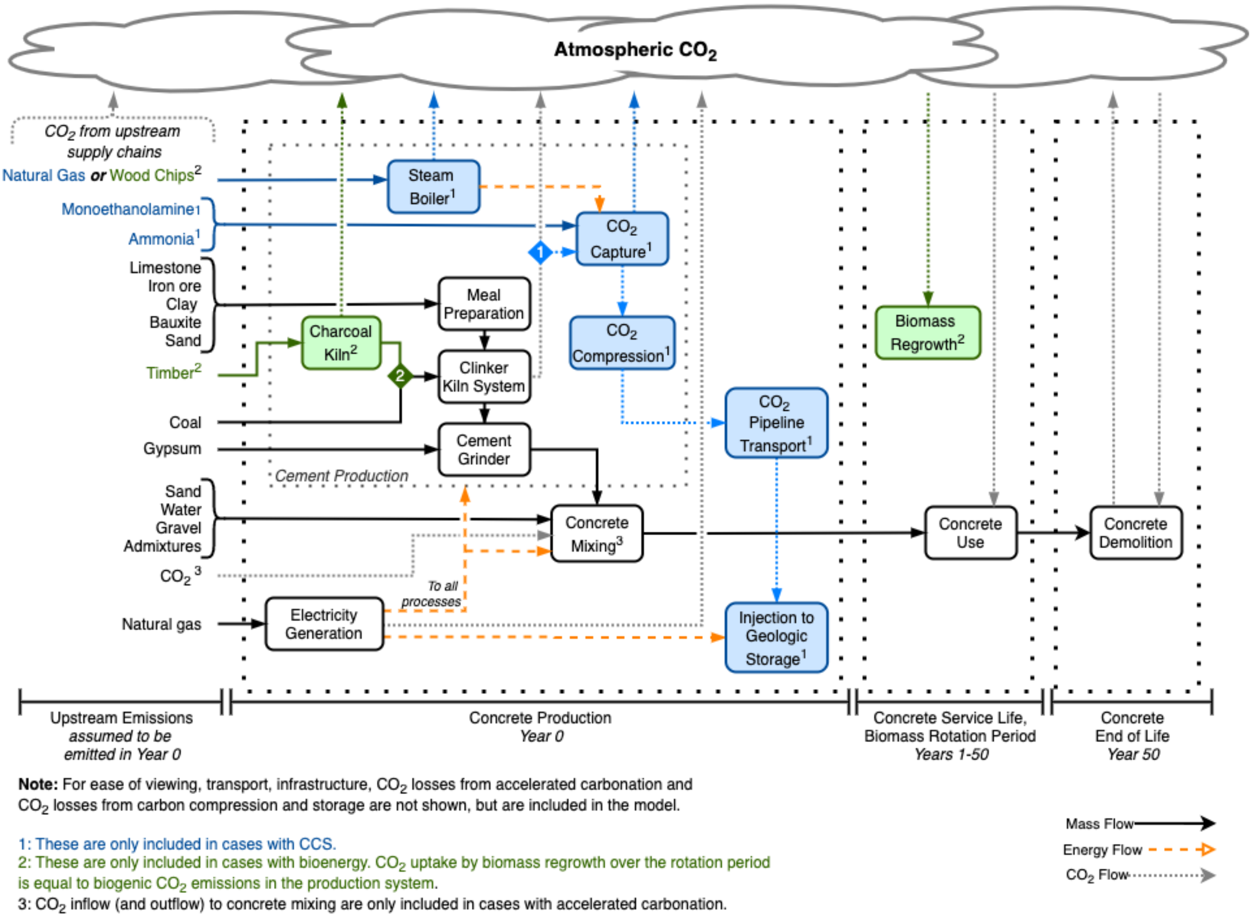
Another pathway to concrete carbonation is to inject CO_2 into fresh concrete. This "accelerated carbonation" was studied in the 1970s [11, 12], as a method to increase the early strength and setting speed of concrete. Recently, several accelerated carbonation products have come to market claiming a reduced carbon footprint [13–15]. However, variation in product type and concrete recipe makes it difficult to quantify the decarbonisation potential of accelerated carbonation by itself.

Furthermore, the concrete life cycle includes the sourcing of sand, aggregate, chemical additives, water, and energy; demolition at end-of-life; and transport of bulk materials. All of these must be accounted for when assessing the CO_2 footprint of concrete.

This chapter explores the combination of decarbonisation technologies to understand their impact on the lifecycle CO_2 balance of ordinary Portland concrete. We focus on accelerated carbonation, bioenergy use in cement kilns, and the capture and storage of CO_2 from cement kilns. As both concrete and biomass uptake CO_2 over time, we chart the balance of CO_2 emissions and removals over time. Additionally, the impact of strength gain from accelerated carbonation, sourcing of accelerated carbonation CO_2, carbonation of demolished concrete, biomass rotation period, and the decarbonisation of electricity and transport are considered.

2 Methods

This chapter is a temporally explicit life cycle CO_2 accounting of concrete production, with and without the use of accelerated carbonation, bioenergy, and/or carbon capture and storage, based on the system in **figure 1**. The unit of analysis (functional unit) is the production of one cubic meter of 30 MPa ordinary Portland concrete, formed into a 20 cm deep exterior wall segment with a 50-year service life. Production and use were assumed to occur in northwest Europe, which is reflected in the choices of technological efficiencies and supply chain data, but the system is otherwise geographically generic.



Note: For ease of viewing, transport, infrastructure, CO_2 losses from accelerated carbonation and CO_2 losses from carbon compression and storage are not shown, but are included in the model.

1: These are only included in cases with CCS.
2: These are only included in cases with bioenergy. CO_2 uptake by biomass regrowth over the rotation period is equal to biogenic CO_2 emissions in the production system.
3: CO_2 inflow (and outflow) to concrete mixing are only included in cases with accelerated carbonation.

Figure 1. System considered in this chapter

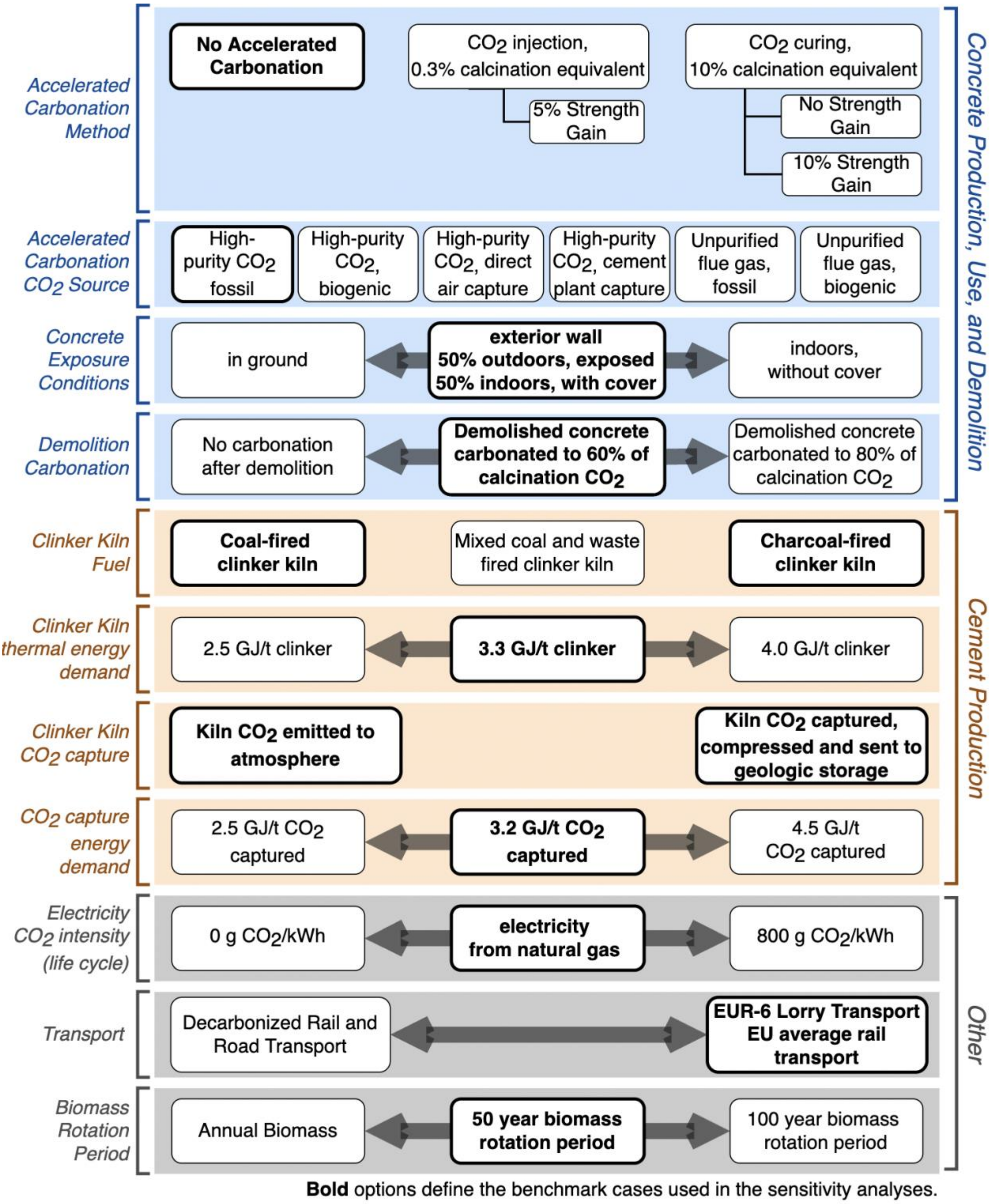


Figure 2. Cases considered in this chapter

The cases considered in this chapter are summarised in **figure 2**. For all cases, the net life cycle CO_2 was estimated, as was the cumulative balance of CO_2 emissions and removals over time. CO_2 emissions from concrete production and upstream supply chains of production inputs were assumed to be emitted in “year 0”. Afterwards, CO_2 is removed from the atmosphere by the natural carbonation of concrete and by replanted biomass. CO_2 emissions and removals associated with demolition occur in the year after the end of the concrete's service life.

The process modelling and CO_2 balances were facilitated by a custom Python3 model, which is described in **appendix A** of the dissertation. For CO_2 emissions in upstream supply chains, life cycle inventory data from ecoinvent 3.6 [16] were used. Tabular data for the model input parameters are available in the **appendix** to this chapter.

2.1 Technology scenarios

Four scenarios of production technology and background systems were considered in this chapter, whose main parameters are summarised in **table 1**:

- **Current, average**, using cement production efficiencies and kiln fuel composition from the European Cement Association, CEMBUREAU [4] and average cement production electricity use [20], and EU-average grid electricity [23]. At end-of-life, the concrete was assumed to be demolished and immediately re-used in a subsurface application, without further carbonation. This scenario is designed to represent current production conditions.

- **Current, benchmark**, assuming "state-of-the-art" cement production [17, 21]. To provide a clearer picture of parameter influence in the sensitivity analysis, simplified energy provision was assumed, with the clinker kiln using only one type of fuel (coal in fossil cases, charcoal in biomass cases), and with electricity provided by a natural gas combined cycle power plant with an efficiency of 56.6%. It is also assumed that at end-of-life, concrete rubble is exposed until 60% of the calcination CO_2 has been recarbonated, as this is an immediately implementable decarbonisation option.

- **Future, conservative**, with improvements in kiln and CO_2 capture efficiencies, increased use of waste, and partial decarbonisation of transport and electricity. Additional electricity efficiency improvements, beyond current state-of-the-art were not considered, due to their negligible impact.

- **Future, optimistic**, with a "practical minimum" clinker kiln efficiency [18], increased use of wastes [4], and fully decarbonised electricity and transport sectors, as envisioned to be available no later than 2050 in the EU [24].

Table 1. Main model parameters, by technology scenario.

Parameter	Unit	Current, average	Current, benchmark	Future, conservative	Future, optimistic
Clinker kiln, thermal energy demand	MJ/kg clinker	3.7 [4]	3.3 [17]	3.0[1]	2.8 [18]
Waste fraction of clinker kiln fuel	% kiln fuel (LHV)	46% [4]	0%	60% [19]	90%[4]
Biomass fraction of waste[2]	% waste	16% [4]	*n.a.*	40% [19]	50% [4]
Meal grinding electricity demand[3]	kWh/t meal	23 [20]	12 [21]	12 [21]	12 [21]
Clinker kiln electricity demand[3]	kWh/t clinker	26 [20]	23 [21]	23 [21]	23 [21]
Cement mixing electricity demand[3]	kWh/t cement	40 [20]	16 [21]	16 [21]	16 [21]
Total carbonation, after demolition	% calcination CO_2	*n.a.*	60%[1]	60%[1]	75%[1]
CO_2 capture, thermal energy demand	MJ/kg CO_2	3.2	3.2 [22]	3.0[1]	2.5[1]
CO_2 capture, electricity demand	MJ/kg CO_2	38	15	15	15
Direct CO_2 intensity of electricity	g CO_2/kWh	269 [23]	360	54[1]	0
Upstream CO_2 of electricity[4]	g CO_2/kWh	90 [16]	48	18	0
CO_2 intensity of road transport (life cycle basis)	g CO_2/tkm	84 [16]	84	34	0
CO_2 intensity of rail transport (life cycle basis)	g CO_2/tkm	51 [16]	51	20	0

1: Assumption.

2: Assumed to be 50% short-rotation biomass (e.g., agricultural wastes), and 50% long-rotation biomass (e.g. sawdust).

3: Electricity efficiency beyond current state-of-the art was neglected due to its negligible impact in scenarios with decarbonised electricity.

4: Upstream CO_2 was approximated by subtracting the direct CO_2 intensity of electricity generation [23] from the total life cycle CO_2 in the ecoinvent 3.6 [16] process for European average electricity generation. Decarbonisation of upstream emissions was assumed to occur at the same rate as for direct CO_2 emissions of electricity generation.

2.2 Concrete production

This chapter considered the production of ordinary Portland concrete (OPC), with a 28-day compressive strength of 30 MPa. 25-35 MPa concrete represents 60% of the 255 million m^3 of ready-mixed concrete produced by members of European Ready Mixed Concrete Organisation in 2018 [25]. This chapter used the concrete recipe in **Table 2**, taken from the ecoinvent 3.6 process for 30-32 MPa ready-mix concrete [16]. The use of cement replacers, such as fly ash or slag, is outside the scope of this chapter.

In the benchmark case, inputs to concrete production were assumed to be transported 200km by heavy lorry to the construction site, where concrete mixing occurs with water available on-site. To minimise variation between cases, accelerated carbonation was assumed to happen on the site of concrete production and use.

Table 2. Recipe for 30 MPa concrete used in this chapter [16].

Ingredient	kg per m^3 concrete
CEM I Portland cement	344
Sand	859
Gravel	960
Water	207
Admixtures	1.2

2.3 Accelerated carbonation

Two cases of accelerated carbonation were considered:

- **0.3% calcination CO_2 injection into the concrete mixer,** based on commercially available technology [26]. A small quantity of CO_2 is injected, equalling approximately 0.3% of the calcination CO_2 emitted during the production of the concrete's constituent cement. This has been shown to increase the strength of the concrete, allowing for approximately a 5% reduction in cement [26]. Therefore, in these cases, each m^3 of concrete has 17kg less cement, with an additional 14 kg of sand to maintain volume.

- **CO_2 curing to 10% of embodied calcination CO_2** where the concrete is exposed to a high-CO_2 atmosphere in a pressurised environment. The literature of CO_2 curing of cement and concrete varies widely in product recipe, curing environment, observed CO_2 uptake, and change in concrete properties. In particular, both strength gain and strength loss has been reported. A comparison of several CO_2 curing studies is included in the ESI. In our model we assumed that two hours of CO_2 curing in a constant pressure environment of 150 kPa resulted in a CO_2 uptake of 10% of calcination CO_2, with no change in strength.

In the benchmark cases, the CO_2 used for accelerated carbonation was assumed to have been captured from industrial flue gas, purified to 95%, and transported via lorry to the concrete production site. CO_2 uptake efficiency was initially assumed to be 60% [27], with unabsorbed CO_2 emitted to the atmosphere. As accelerated carbonation was assumed to occur at a construction site, dedicated pipeline transport of the CO_2 was assumed to be unrealistic.

The study also considered cases where CO_2 curing led to a 10% strength gain, with a corresponding reduction of cement use. In combination, we also explored the impact of four other sources of the CO_2 used in accelerated carbonation:

- 95%-purity CO_2 from the system's own cement plant, when outfitted with CCS.
- 95%-purity biogenic CO_2 from the production of bioethanol, assumed to only require compression and transport.
- 95%-purity atmospheric CO_2 from an on-site direct air capture (DAC) unit [28]. This includes an electricity demand of 366 kWh/t CO_2 and a thermal energy demand of 5.9 GJ/t CO_2, supplied by natural gas. In the DAC CO_2 capture process, approximately 95% of CO_2 from natural gas use is also captured [28].
- raw flue gas (10-20% CO_2), both fossil and biogenic, that has only been cleaned of SO_2 and NO_X.

2.4 Natural carbonation

Concrete absorbs CO_2 as the CaO in the cement recarbonates, first at the surface then gradually penetrating into the concrete mass with decreasing uptake over time as in **figure 3**. The carbonation rate depends on the composition of the concrete, exposed surface area, and exposure environment. It is typically modelled using Fick's diffusion law, and calculated with the equation:

$$\mathrm{CO_2\ uptake} = \sum(k_i \times DOC_i \times A_i) \times \left(\sqrt{t} \div 1000\right) \times U_{tcc} \times C \qquad [9]$$

where

CO_2 uptake, in kg, is the total CO_2 carbonated over period **t**.
k_i in mm/year$^{0.5}$, is the carbonation rate factor, based on the concrete strength and exposure conditions [9].
DOC_i, in percent, is the maximum degree of carbonation specific to the exposure conditions [9].
A_i, in m^2 is the surface area of the exposed concrete.
t, in years, is the length of exposure. **U_{tcc}**, in kg CO_2/kg cement is the maximum theoretical uptake of CO_2, equal to the CO_2 released during calcination. For the CEM I Portland cement, the value is 0.49.
C, in kg cement/m^3 concrete, is the cement content of the concrete.

Concrete use was assumed to be as an exterior wall, with a depth of 20cm, with an external surface exposed to rain, and a painted interior surface. In cases with accelerated carbonation, natural carbonation was assumed to begin from the level of carbonation already present in the concrete. E.g., if 5% of calcination CO_2 was carbonated by accelerated carbonation in 18MPa concrete exposed to rain, carbonation during concrete service life was assumed to continue as if 20 years of natural carbonation had already occurred (as if starting at the year 20 point in **figure 3**).

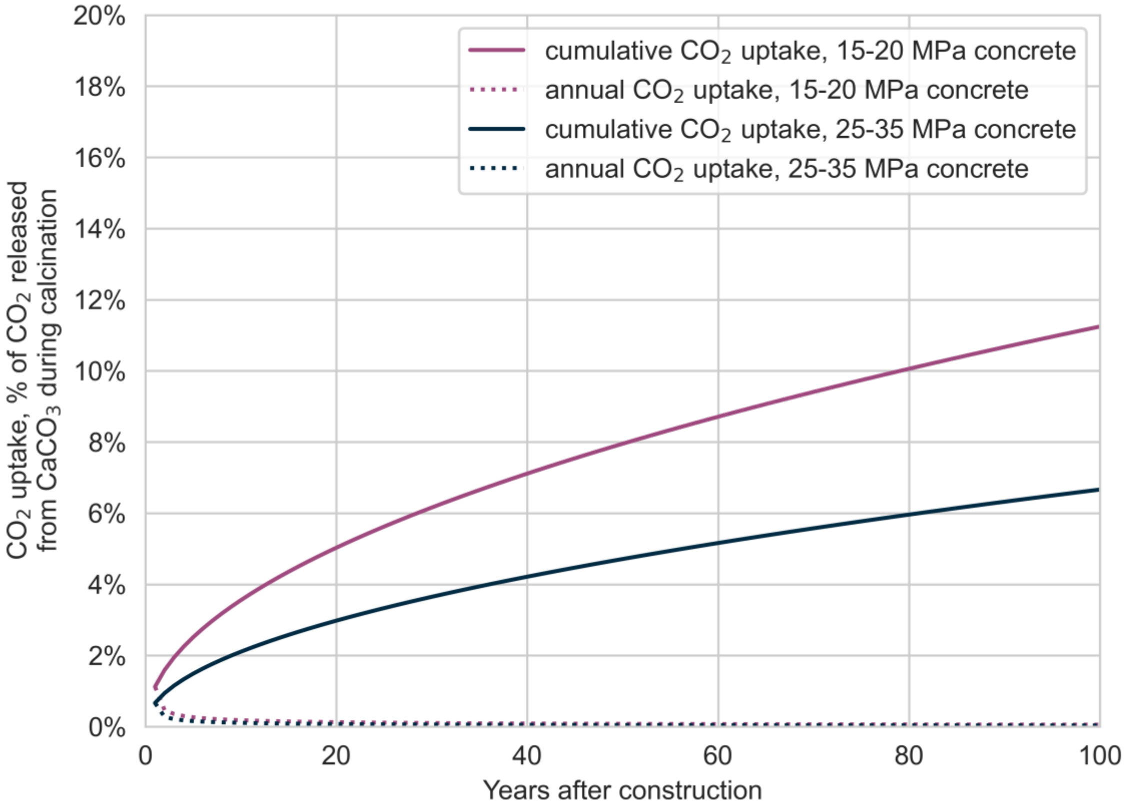


Figure 3. Example of natural carbonation rate of concrete exposed to rain

2.5 End-of-life

At its end of its 50-year service life, the concrete was assumed to be demolished with a life cycle CO_2 footprint of 9 kg/t demolished concrete [16]. As concrete rubble has a large surface area, natural carbonation up to 60-80% calcination CO_2 can be achieved by leaving rubble exposed to air for several weeks [29].

While the EU reports an 90% average recovery rate for construction waste [30], the predominant fate of recovered concrete is road underlayers or backfilling [7], where it is not exposed. Therefore, no additional carbonation of demolished concrete was assumed in the "Current, Average" scenario. Deliberate carbonation of demolished concrete was included in the benchmark scenario and both future scenarios.

2.6 Cement production

Cement production was modelled for CEM I Portland cement [31–33]. Losses from kiln dust or conveyance between processes, were neglected. Production inputs were assumed to travel 200 km by rail to the cement plant. The energy demand for cement production is provided in **table 1**.

Meal preparation: The raw ingredients for cement production consist primarily of limestone, with smaller fractions silicon, aluminium, and iron, typically provided by clay, sand, bauxite, and/or iron ore. This chapter assumed a meal composition based on a real-world mix of a cement plant in Norway [32] that is 77% limestone. The meal was assumed to be crushed and ground in a ball mill.

Clinker kiln system: The pulverised meal is fed into a kiln, which is heated in stages to 1300-1500°C. Between 450-900°C, CO_2 is released from the limestone ($CaCO_3$) during calcination. In this model, a modern short dry kiln with a preheater and a precalciner was assumed. The non-waste fraction of kiln fuel was assumed to be coal, which is replaced with charcoal in the bioenergy cases. The benchmark scenario assumed that no waste was used as fuel.

Cement mixing: The cooled clinker is mixed with gypsum and/or other additives to form cement. CEM I Portland cement consists of 95% clinker and 5% gypsum and was assumed to be processed in a roller mill.

2.7 Carbon capture and storage

In cases with carbon capture and storage (CCS), flue gas from the cement kiln was sent to a post-combustion CO_2 capture unit using a monoethanolamine (MEA)-based absorption process. Flue gas cleaning using ammonia and limestone to remove SO_2 and NO_X was assumed [31], as this is current practice. While there are more advanced solvent-based capture systems, and demonstrations of calcium-looping-capture and direct separation technologies for cement production are underway [34, 35], MEA-based capture is a mature and commercially available technology, so was chosen for the benchmark scenario, which focuses on currently available technologies.

In the benchmark scenario, the CO_2 capture process was assumed to require 3.2GJ/t CO_2 of low-temperature steam [22], provided by a dedicated boiler with a 90% efficiency. The boiler was assumed to use natural gas or, in cases with bioenergy use, wood chips. CO_2 from the boiler was emitted to the atmosphere. No heat integration with the cement plant was assumed in the benchmark case, as available heat can vary widely and may be in use for other purposes.

After capture, the CO_2 was assumed to be compressed to 110bar, requiring 96kWh/t CO_2 [32] transported 200km by pipeline, and injected into geologic storage, with an injection electricity demand of 8 kWh/t CO_2 [36] It was assumed that 1% of CO_2 was lost during transport and injection.

2.8 Bioenergy use and biomass regrowth

In cases with bioenergy use, charcoal replaces coal in the cement kiln and wood chips replace natural gas in the steam boiler for CO_2 capture. Charcoal was selected to ensure that the kiln would be able to reach sufficiently high temperatures, and for usability in existing kilns accustomed to coal-firing. Charcoal was assumed to be produced in industrial Missouri-style kilns, with 69% carbon recovery and ancillary CO_2 emissions of 543 kg/t charcoal [37]

Timber for charcoal production was assumed to have a rotation period of 50 years, as was the long rotation fraction of biogenic wastes. CO_2 reuptake by biomass over time was modelled using a Gaussian distribution, as visualised in **figure 4**, following the equation:

$$\mathrm{CO_2 \text{ uptake in year } \tau} = (2\pi\sigma^2)^{0.5}\, e^{(\tau-\mu)^2} / 2\,\sigma^2 \qquad [38]$$

where $\boldsymbol{\mu}$ is the rotation period halved and $\boldsymbol{\sigma}$ is $\mu/2$.

Biomass replanting was assumed to occur in year 0, and at the end of the biomass rotation period, 100% of biogenic CO_2 emitted during cement production is reabsorbed.

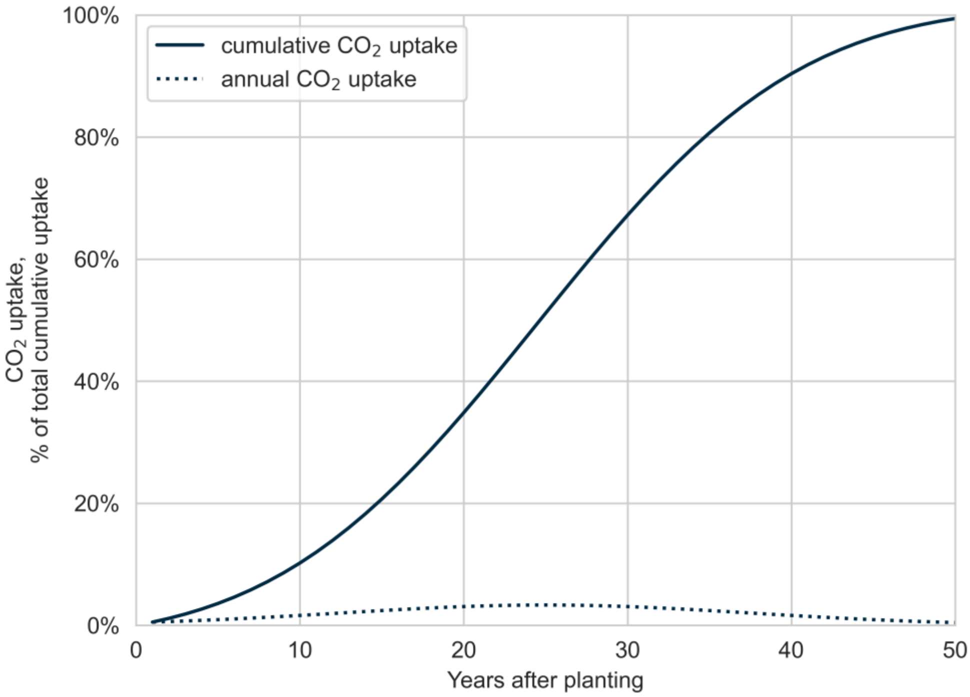


Figure 4. Example of atmospheric CO_2 uptake by biomass growth

2.9 Upstream CO_2

Upstream CO_2 emissions were included for the inflows in **figure 1**, as well for transport of materials to the cement plant and the concrete production site, transport of CO_2 to geologic storage, and infrastructure use for all processes. For this background system, life cycle inventory CO_2 data from ecoinvent 3.6 [16] was used, including the emissions of biogenic, fossil, and direct land use change CO_2. Indirect land use change was not considered. The specific ecoinvent processes and CO_2 factors are provided in the appendix, as are energy content and emissions factors of fuels used in this model [39, 40].

3 Results and discussion

Full tabular results are available in the supplementary information of the published version of this paper. Net CO_2 has been rounded to the nearest 10 kg CO_2/m^3 concrete.

3.1 Technology scenarios overview

Figure 5 summarises the net life cycle CO_2 for each concrete type and production scenario, before considering accelerated carbonation. The net CO_2 of the "current, average" scenario is 380 kg CO_2/m^3 concrete. Excluding end-of-life carbonation, the benchmark scenario, without bioenergy or CCS, has a 15 kg/m^3 lower net CO_2 due to increased production efficiencies. Carbonation of demolished concrete accounts for a further reduction of 85 kg CO_2/t m^3. For embodied cement production, the net CO_2 of the benchmark scenario is 880 kg CO_2/t cement versus 920kg/t for the "current, average" production scenario. All bioCCS cases, as well as the "optimistic" future cases with CCS only (which have a 45% biogenic kiln fuel mix), resulted in CO_2-negative concrete, with atmospheric CO_2 removals during biomass regrowth and concrete recarbonation, exceeding CO_2 emitted during the concrete's life cycle. However, as **figure 6** illustrates, the net CO_2 refers to the CO_2 balance at concrete's end-of-life; 50 years after most CO_2 emissions occur. In all cases, the concrete system is CO_2-positive for at least 40 years. Additionally, all cases with bioenergy use have higher net CO_2 than their fossil counterparts for 15-25 years after concrete production until sufficient atmospheric CO_2 is reabsorbed by sustainably replanted biomass. This is seen in **figure 6** where the lines of the bioenergy cases cross those of their fossil counterparts. The future scenarios' bioenergy cases have flatter curves, attributable to high proportion of both annual biogenic wastes and fossil-based wastes as fuels for cement production. Finally, significant carbonation of demolished concrete is required to reach CO_2 negativity in all but the "future, optimistic" bioCCS cases.

The benchmark cases are discussed below, followed by an assessment of accelerated carbonation options, then sensitivity analyses on natural carbonation, biomass CO_2 uptake, and production efficiencies.

3.2 Benchmark cases

The benchmark case was estimated to emit 380 kg CO_2/m^3 concrete during production and upstream processes, of which 280kg are direct emissions from cement production (175 kg from calcination, 105 kg from fuel use). A further 70 kg/m^3 were emitted upstream, of which 60% were from transport of bulk materials. In our model, 20 kg CO_2/m^3, or 11% of calcination CO_2, were removed by natural carbonation over the 50-year service life of the concrete. Finally, carbonation of the demolished concrete removed an additional 85 kg CO_2/m^3 from the atmosphere, while the demolition process was responsible for 20 kg/m^3 of CO_2 emissions.

Without accelerated carbonation or CCS, the use of bioenergy in cement production decreased net CO_2 by 110 kg/m^3 concrete. CO_2 emissions of production increased by 30 kg CO_2/m^3, but 140 kg/m^3 of CO_2 were reabsorbed by biomass regrowth. In contrast, the use of CCS in cement production alone reduced both CO_2 emitted and net CO_2 by 165 kg CO_2/m^3, corresponding to 250 kg CO_2 sent to geologic storage minus 85 kg CO_2/m^3 emitted as a consequence of CCS, of which 65 kg were direct emissions from energy provision for the capture unit.

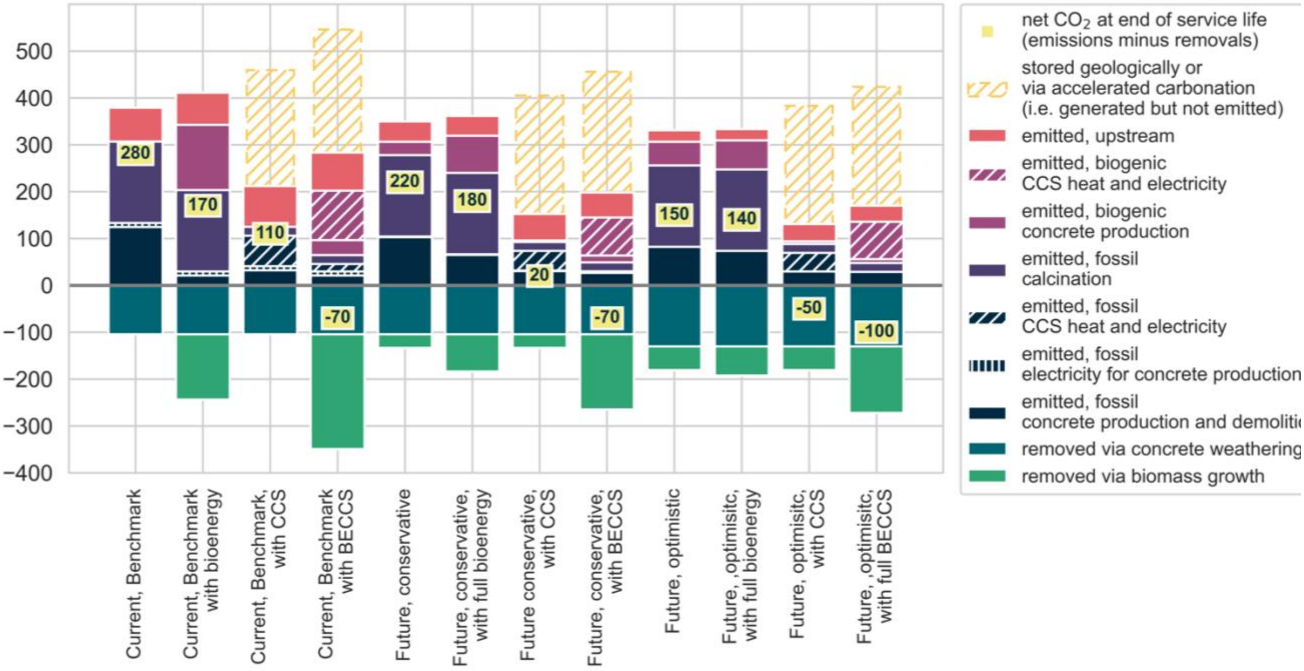


Figure 5. **Life cycle net CO_2 for 30 MPa ordinary Portland concrete in different technology scenarios. Note that the net CO_2 is the CO_2 balance at end-of-life. Concrete was assumed to be used as a 20 cm exterior wall. In all scenarios besides “Current, average”, concrete was assumed to be left to recarbonate following demolition.**

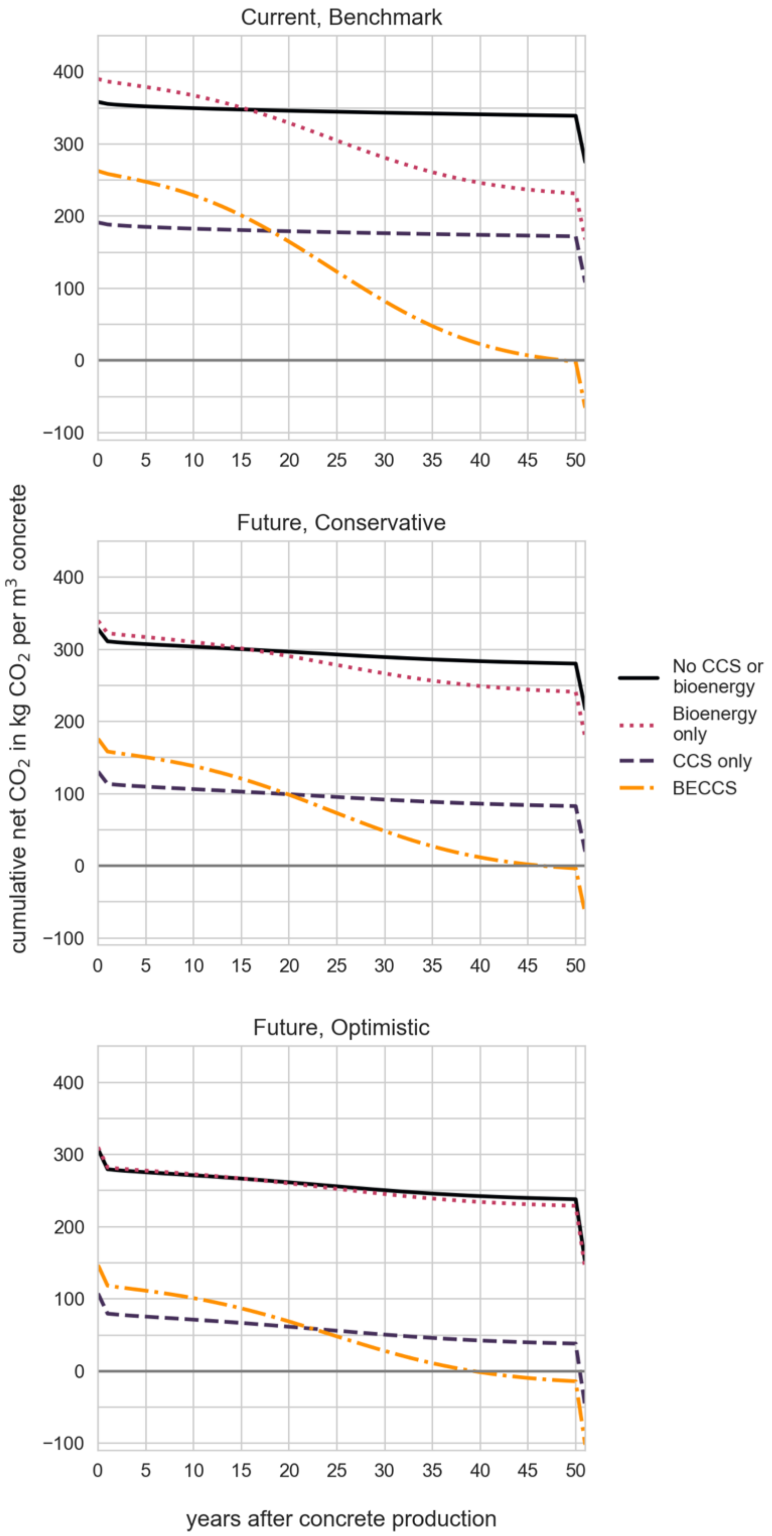


Figure 6. Cumulative life-cycle net CO_2 of 30 MPa ordinary Portland concrete over time, in different technology scenarios, assuming a 50-year concrete service life, and a 50-year rotation period for long-rotation biomass. Concrete was assumed to be used as a 20 cm exterior wall and left to recarbonate following demolition.

The combination of bioenergy and CCS in cement production generates 70 kg more CO_2/m^3 concrete than CCS alone, stores 15 kg more CO_2, and approximately 350 kg of CO_2/m^3 are removed by biomass. The net effect is that concrete produced with bioCCS cement was modelled to be net CO_2-negative, at approximately -70 kg/m^3, but only at end-of-life, after CO_2 reuptake by biomass and the carbonation of concrete both during service life and after demolition.

3.3 Accelerated carbonation

Table 3 summarises the modelling results for accelerated carbonation, in relation to the benchmark case without bioenergy or CCS use. The injection of 0.3% CO_2 during concrete mixing was the only case that resulted in a decrease of net CO_2, (of 9 kg CO_2/m^3), though less than 1 kg CO_2/m^3 concrete was stored in the concrete. The 5% reduction in cement demand due to the increase in concrete strength reduced CO_2 emissions by 15 kg CO_2/m^3, partially offset by decreased natural carbonation from the lower cement content of the concrete. Additionally, as the CO_2 intensity of cement production decreases, so does the apparent decarbonisation benefit of reduced cement use. In the bioCCS case, the net CO_2 of 0.3% CO_2 injection is actually 8 kg CO_2/m^3 higher than without, as increased use of CO_2-negative cement decreases the net CO_2 of the concrete. However, the benefits of decreased resource use are pertinent, even if they are outside the scope of this chapter. If scaled to the 2018 EU production of 250 million m^3 ready-mix concrete [25], 0.3% CO_2 injection would store only 0.1 Mt of CO_2, but a 5% reduction cement demand would reduce cement sector emissions by 3.5 Mt CO_2/year.

As modelled, OPC cured with the equivalent of 10% of calcination CO_2, without strength gain, increased net CO_2 by approximately 40 kg CO_2/m^3 relative to the benchmark case without accelerated carbonation, bioenergy, or CCS. This increase was due to additional emissions from capture, transport, and injection of CO_2; CO_2 lost during the injection process; and reduced natural carbonation. However, this process also stored 17 kg of fossil CO_2 into the concrete. If these avoided emissions are included in the net CO_2 of the concrete system, net CO_2 only increased by 23 kg CO_2/m^3. Even if CO_2 injection was assumed to be 100% efficient, with no losses of CO_2, the net CO_2 would still be higher than without accelerated carbonation.

However, "avoided emissions" reflect a reduction in CO_2 emitted from preventing the release of CO_2 and are not a physical removal of CO_2 from the atmosphere, though both reduce the amount of CO_2 that would have been in the atmosphere than if the "avoided" CO_2 would have been emitted. Thus, for accelerated carbonation, claiming avoided emissions requires that the CO_2 stored in the concrete would have otherwise been emitted, and not sent to geologic storage or otherwise abated. If the CO_2 would have been otherwise abated, there are no avoided emissions. Finally, it is important that avoided emissions are not double counted. In other words, they should only be accounted once—either in the system of CO_2 generation or in the system of CO_2 storage (here, accelerated carbonation), but not both.

Figure 7 shows the impact of CO_2 origin and concrete strength gain for 10% CO_2 curing. The 0.3% CO_2 injection case is not included, as the CO_2 quantity is too small. Assuming there is no gain in concrete strength, only CO_2 curing using a raw flue gas from a biogenic source had a net CO_2 on par with the benchmark case, if avoided emissions can be counted. A 10% gain in strength was sufficient to offset the emissions associated with accelerated carbonation and achieve a small net CO_2 reduction (5-20 kg/m^3) relative to the benchmark case when accelerated carbonation used raw flue gas, pure biogenic CO_2, or CO_2 from direct air capture (if the CO_2 from DAC fuel use is also captured). However, in the cases where raw flue gas is used for curing, a 10% strength gain may be unrealistic, as the lower CO_2 concentration in the curing environment is likely to result in lower CO_2 uptake [41]. Lastly, using CO_2 from a CCS-equipped cement plant only increased CO_2 emissions, compared to the more efficient option of sending the CO_2 to geologic storage, as, in our model, the net impact of increased energy use and decreased natural carbonation from accelerated carbonation exceeds the CO_2 emissions of sending the CO_2 to geologic storage.

Table 3. Impact of accelerated carbonation on net CO_2, with main contributing factors, in kg CO_2/m^3 concrete*

Parameter	**No AC**	**0.3% CO_2 injection**	**10% CO_2 curing**	**10% CO_2 curing, 10% strength gain**
		as change from reference case		
Net CO_2[a]	*280*	-9	+42	+19
CO_2 stored in concrete	0	+<1	+17	+15
CO_2 emitted, AC CO_2 capture and transport	0	+<1	+11	+10
CO_2 emitted, carbonation losses	0	< 1	+11	+10
CO_2 emitted, cement production and upstream	*295*	-15	0	-29
CO_2 removed by natural carbonation, service life	*20*	-1	-10	-11
CO_2 removed by natural carbonation, demolition	*85*	-4	-6	-14
total electricity use (kWh/m^3 concrete)	*24*	-1	+10	+7

1: Avoided emissions from stored CO_2 are not included in the net CO_2.

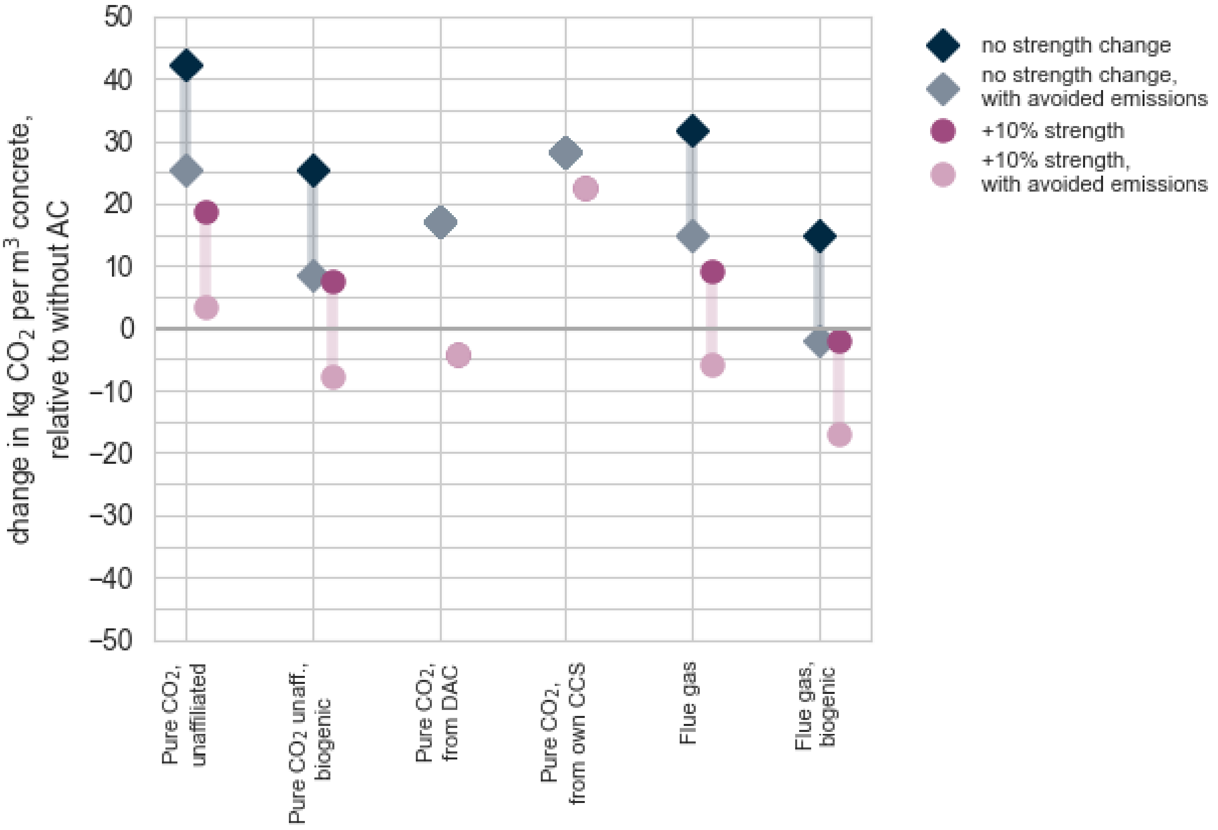


Figure 7. Impact of CO_2 source on life cycle net CO_2 of concrete subject to CO_2 curing to equivalent of 10% of embodied calcination CO_2, considered with and without strength gain and avoided emissions.

3.4 Natural carbonation

3.4.1 Exposure conditions

Uptake of CO_2 by natural carbonation decreases over time, as the CO_2 must continually penetrate deeper into the concrete to achieve additional carbonation. In our model, for a 50-year service life, 50% of natural carbonation occurs by year 12, and 75% by year 28. At the benchmark exposure conditions, carbonation decreased to less than 0.3 kg CO_2/m^3 concrete/year after year 20. Doubling the concrete service life increases CO_2 removal by only 8 kg CO_2/m^3 over years 51-100.

As shown in **figure 8**, in the most favourable conditions for concrete carbonation—outdoors, uncovered, and sheltered from rain—OPC was estimated to absorb 23% of calcination CO_2 over 50 years, double that of the benchmark case. Indeed, an equivalent amount of carbonation occurs in 12 years in these conditions as in 50 years in the benchmark case. In contrast, OPC in ground, such as in road sub-layer applications, or otherwise unexposed, was estimated to absorb less than 5% of calcination CO_2 over 50 years.

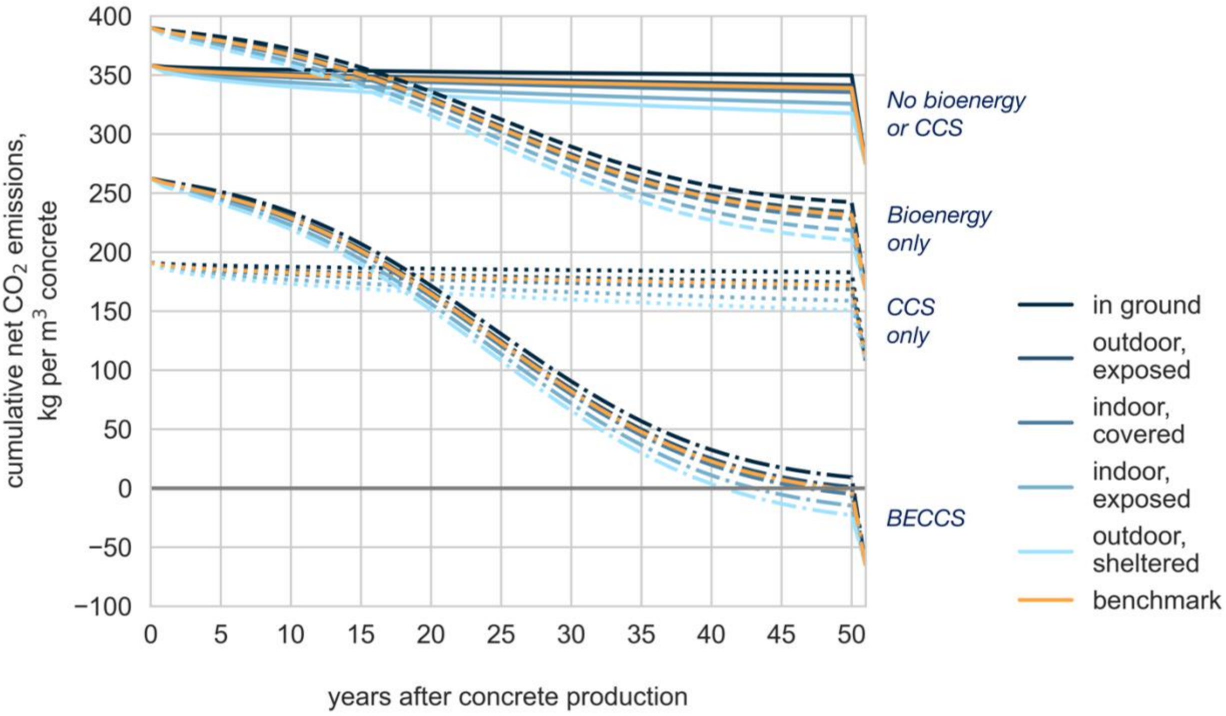


Figure 8. Impact of concrete exposure conditions on net CO_2

3.4.2 End-of-life carbonation

Alone, natural carbonation after demolition has the potential to abate 20-30% of life cycle CO_2 emissions of OPC. The additional energy use could be negligible, if there is no additional transport distance, e.g., left to carbonate for several weeks at the site of demolition or reuse/disposal [29]. However, this abatement only occurs at the very end of the concrete's service life, 50 years after the emissions of its production.

Though end-of-life carbonation is a long-term decarbonisation option for new concrete production, 368 Mt of mineral construction and demolition waste, mostly concrete, was generated in the EU in 2018 [30]. Assuming that, as in our benchmark case, 85 kg CO_2/m^3 concrete is absorbed by end-of-life carbonation, current demolition wastes could remove on the order of 30 Mt of CO_2 per year from the atmosphere, or over 25% of direct CO_2 emissions from EU cement production [6].

3.5 Biomass use

Figure 9 show the impact of biomass rotation period on the net CO_2 of concrete over time. In all cases, the net life cycle CO_2 of the bioenergy cases does not decrease below that of the corresponding fossil energy case until approximately halfway through the biomass rotation period. Until then, the amount of CO_2 in the atmosphere from the production of the concrete is higher than without bioenergy.

This model assumed the use of charcoal for clinker kiln fuel in the bioenergy cases to provide a clear picture of the use of long-rotation biomass. However, from a resource perspective, this is overly simplistic. If all 180 Mt/year of EU-28 cement production (2018) [6] was charcoal-fired, it would require over 40Mt/year of timber, nearly a quarter of current annual European forestry production [42, 43]. If all cement production also installed CCS, a further 20 Mt/year of wood chips would be needed to supply energy for the CO_2 capture reboiler. Instead, energy demand for CCS and part of the energy demand for the kiln could be provided by low-grade fuels, such as agricultural residues or dedicated annual energy crops, which would also decrease the average rotation period of the biomass.

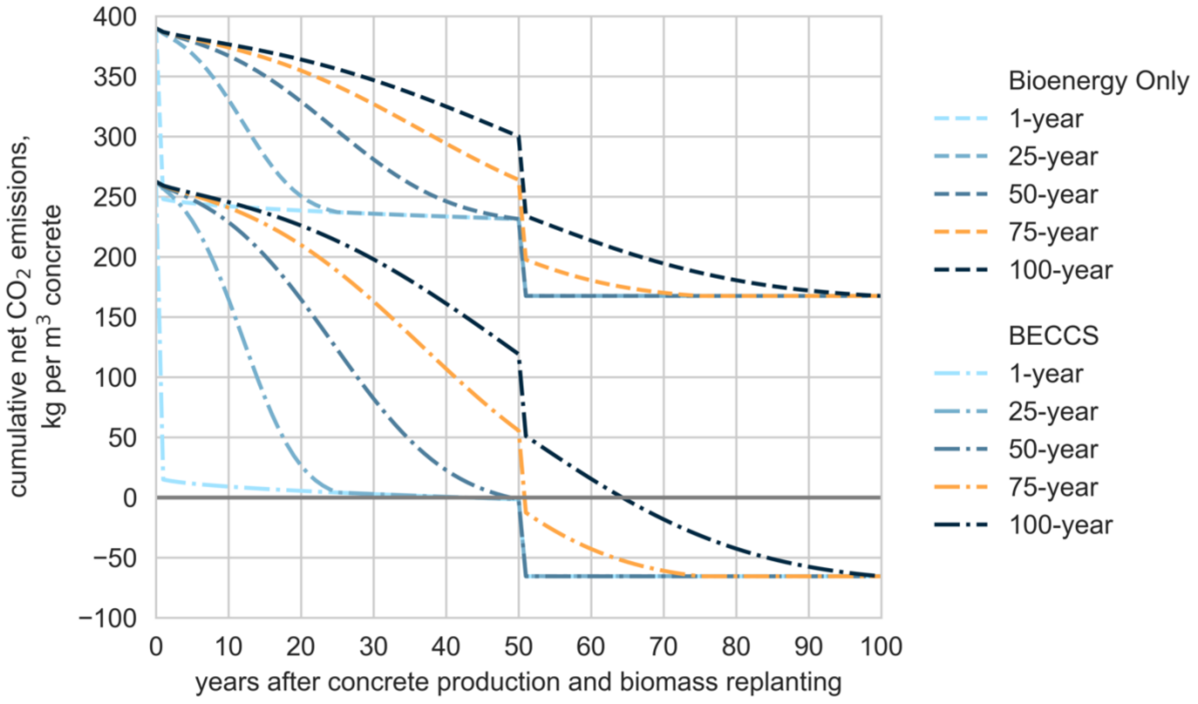


Figure 9. Impact of biomass rotation period on net CO_2

3.6 Efficiencies of production and background supply chains

3.6.1 Electricity

The benchmark scenario assumes electricity with a life cycle CO_2 intensity of approximately 400 g CO_2/kWh. In cases without CCS, full decarbonisation of the electricity supply chain decreased CO_2 emissions by less than 10 kg CO_2/m^3 concrete in the benchmark case. This increases to 25 kg CO_2/m^3 in the CCS case, as the use of CCS in cement production doubles the electricity intensity of concrete production. If CCS is applied to the 180 Mt of 2018 EU cement production [6], it would require 22 TWh of electricity, or over 2% of total current industrial electricity use [30].

3.6.2 Transport

The production of concrete requires the transport of large quantities of bulk materials, and correspondingly the life cycle CO_2 of concrete is sensitive to transport assumptions. Our model assumed 200km of rail transport of minerals to the cement plant and 200km of heavy lorry transport of concrete production inputs, together responsible for over 10% of lifecycle CO_2 emissions in the benchmark case. A decarbonised transport sector could reduce CO_2 emissions by 40 kg CO_2/m^3 of concrete. Conversely, longer transport distances, the use of less efficient or lower-capacity lorries, and/or a reliance on road transport for inputs to the cement plant will rapidly increase the CO_2 intensity of concrete production.

3.6.3 Clinker kiln efficiency

The efficiency of the clinker kiln was most significant to net CO_2 in cases without bioenergy or CCS. An exceptionally efficient clinker kiln, with a thermal energy demand of 2.5 GJ/t clinker would only decrease lifecycle CO_2 emissions by 25 kg CO_2/m^3 concrete compared to the benchmark of 3.3 GJ/t clinker. In the bioCCS cases, a high-efficiency kiln *increased* life cycle net CO_2. This is the phenomenon of "inefficient bioCCS", where decreased energy efficiency increases the amount of biogenic CO_2 that can be captured and stored [44].

3.6.4 CO_2 capture efficiency

In the cases with CCS, increasing energy efficiency of CO_2 capture to 2.5 GJ/t CO_2 decreases CO_2 emissions by approximately 10 kg CO_2/m^3 concrete. While a reboiler duty below 3.0 GJ/t CO_2 is possible with MEA, it is more commonly seen with advanced solvents, and thus may also impact the life cycle CO_2 based on differing upstream impacts of solvent production, though solvent production currently represents less than 0.1% of CO_2 emissions in all cases.

If CO_2 capture is applied to the steam boiler providing heat for the solvent reboiler, a net reduction of 85% of boiler CO_2 could be achieved, 40 kg CO_2/m^3 concrete in the CCS-only case and 90 kg CO_2/m^3 in the bioCCS case. However, the bioCCS case would still take over half the biomass rotation period to reach carbon neutrality.

The use of waste heat, if available, could instead reduce CO_2 emissions by 50 kg or 105 kg CO_2/m^3 concrete for the CCS-only and bioCCS cases, respectively. However, in the bioCCS case, this reduction in CO_2 emissions is offset by the reduced removal of atmospheric CO_2 by biomass, and therefore the reduced emissions and resource use is not reflected in net CO_2.

3.7 Considerations beyond the scope of this chapter

This chapter considered concrete produced with ordinary Portland cement, and the decarbonisation potential of low-CaO cements were outside the scope of this chapter. However, as of 2017, the average clinker ratio of European cement was 77%, with the remainder replaced with other cementitious materials such as fly ash and granulated blast furnace slag [25], and decreasing the proportion of clinker is one of the major avenues of decarbonation proposed by CEMBUREAU, the European cement industry group [4]. The use of ash and slag as clinker replacers lowers CO_2 emissions within the system boundaries of concrete production and may increase the rate of natural carbonation [9]. However, while fly ash and blast furnace slag can reduce the demand for fresh clinker, they are products of the combustion of fossil fuels and calcination of limestone, and therefore, the fate of the CO_2 from their system of origin should be taken into account when assessing their decarbonisation potential. Furthermore, the availability of fly ash is expected to decrease as coal is phased out of the power mix.

Secondly, this chapter did not consider other contributors to global warming potential, such as methane and nitrous oxide emissions or indirect land use, which could be significant for bioenergy-based systems. We also did not consider other global warming impacts from concrete use, though, like avoided emissions, this is specific to the reference system considered, i.e., whether concrete replaces surfaces with lower albedo, (e.g., asphalt) or replaces surfaces that provide evaporative cooling (e.g., grass).

Finally, this was a study on the *marginal* production of 1 m^3 of concrete, and therefore cannot embody the decarbonisation potential of reducing the total production of concrete by improved construction design, increased reuse, or extended concrete service life. In particular, this chapter assumed a concrete use life of 50 years, based on the expected lifespan of modern reinforced concrete structures. This short lifespan for concrete is a modern phenomenon, resulting from the use of iron-based reinforcing bars (rebar). These allow for the construction of very large and strong structures, but corrode and expand as oxygen invades the concrete, causing irreversible structural damage [45]. However, the use life of concrete could be re-extended to multiple hundreds of years if it is unreinforced, or reinforced with non-corroding rebar, such as aluminium bronze [45]. This would greatly decrease the future impacts of the concrete industry, as the concrete stock becomes more durable, reducing overall resource use.

4 Conclusions

This chapter explored the production of ordinary Portland concrete considering different combinations of natural and accelerated carbonation, bioenergy use, and carbon capture and storage. The sensitivity analysis explored the impact of strength gain from accelerated carbonation, the origin of CO_2 used for carbonation; conditions of concrete use and demolition on natural carbonation; biomass rotation period; and efficiencies of electricity, transport, and cement production.

In our model, the aggressive use of bioCCS in cement production and the deliberate natural recarbonation of demolished concrete together resulted in net-CO_2-negative concrete at current technology levels, when considered on a life cycle basis.

However, net CO_2 is the balance of CO_2 emissions and removals for the entire concrete life cycle, measured at the end of the concrete's service life and after all biomass used for bioenergy has been regrown. Depending on the biomass rotation period and the rate of concrete carbonation, CO_2-negative concrete may still have a net-positive CO_2 balance for the entirety of its service life and only reach CO_2 negativity when the demolished concrete is allowed to recarbonate.

Modelled with currently available technology, post-combustion amine-based CCS for cement production reduced life cycle CO_2 of concrete by 40%, and was the single most effective decarbonisation intervention, but alone is insufficient to result in negative emissions. Combined with the use of fully biogenic fuel in the cement kiln, biogenic fuel or waste heat for CO_2 capture, and allowing for carbonation of demolished concrete, bioCCS was estimated to result in a life cycle net CO_2 of -70 kg CO_2/m^3 concrete. However, 280 kg CO_2/m^3 were still emitted during production and in upstream supply chains, more than with CCS alone. It is not until almost halfway through the biomass rotation period that the net CO_2 of bioCCS is lower than in the CCS-only case, assuming that the biomass is indeed sustainably regrown. Using short rotation biomass for cement kiln fuel and encouraging carbonation of current concrete waste can be used to more rapidly decarbonate the concrete sector.

In this chapter, accelerated carbonation of ordinary Portland concrete did not appear to be an efficient method for CO_2 storage on its own. The CO_2 penalty from increased energy use and decreased natural carbonation exceeded the CO_2 stored, though this was highly sensitive to both concrete strength gain and the origin of the CO_2 used for accelerated carbonation. The potential benefit of accelerated carbonation seems to lie not in its ability to directly store CO_2 in the concrete, but rather if it can increase concrete strength and reduce the overall use of cement.

The natural carbonation of concrete is a slow process, and though estimates of total carbonation by global concrete stocks are impressive, the annual CO_2 uptake of in-use concrete is minor relative to the embodied CO_2 of its production. However, increasing carbonation during demolition and recovery by leaving the concrete waste exposed to air for a period of weeks is a promising decarbonisation option that could be implemented in the near term.

Decarbonisation of concrete production is a complex matter, and CO_2 emissions, while important, do not embody the full impacts of the system of concrete production and use. The net CO_2 must be taken in the context of the full specific systems for concrete production. Trade-offs between near-term versus long-term decarbonisation and between decreased CO_2

versus increased energy use must be considered. Even if CO_2-neutral, or CO_2-negative, concrete is achievable, it is very likely to be at the expense of increased resource use. Therefore, the primary decarbonisation priority should always be the reduced use of all concretes via all production methods.

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Appendix

Model parameters

The process model parameters below are provided for the benchmark scenario unless otherwise specified. Please refer to the methods section of the paper for information on differences in other scenarios.

Table 1. Process model parameters.

Parameter	Qty	Unit	Source
MEAL GRINDING, ball mill			
$CaCO_3$ demand	774	kg/t meal	[1]
Sand demand	135	kg/t meal	[1]
Iron ore demand	23	kg/t meal	[1]
Bauxite demand	33	kg/t meal	[1]
Clay demand	3	kg/t meal	Assumed
Electricity demand	12	kWh/t meal	[2]
CLINKER PRODUCTION - short dry kiln with precalciner and preheater			
Raw meal demand	1.57	t/t clinker	
Calcination efficiency	100	%	Assumed
Thermal energy demand	3.3	GJ/t clinker	[3]
Electricity demand	23	kWh/t clinker	[2]
CEMENT MIXING, roller mill			
Clinker demand	950	kg/t cement	
Gypsum demand	50	kg/t cement	
Electricity demand	16	kWh/t clinker	[2]
CONCRETE MIXING			
Cement demand	344	kg/m^3 concrete	[4]
Sand demand	859	kg/m^3 concrete	[4]
Aggregate demand	960	kg/m^3 concrete	[4]
Water demand	207	kg/m^3 concrete	[4]
Admixture demand	1	kg/m^3 concrete	[4]
Electricity demand	5	kWh/m^3 concrete	[4]
Density of concrete	2371	kg/m^3 concrete	[4]

Parameter	Qty	Unit	Source
Transport of raw material	200	km	[4]
Water use (not in concrete)	40	kg/m^3 concrete	[4]
CO_2 INJECTION, CURING - 2 hour, 150kpa			
Electricity demand	3	kWh/t CO_2/hr	[5]
CO_2 uptake	10	% of cement calcination CO_2	assumed, based of literature review
CO_2 uptake efficiency	60	% of CO_2 injected	assumed
CO_2 INJECTION, into mixer			
Electricity demand	37.5	kWh/t CO_2	[6]
CO_2 uptake	0.3	% of cement calcination CO_2	[6]
CO_2 uptake efficiency	60	% of CO_2 injected	[6]
CO_2 CAPTURE, monoethanolamine solvent absorption			
Capture efficiency	90	% of CO_2 in	[14, 15]
Electricity demand	15	kWh/t CO_2 captured	[14]
Heat demand	3.2	GJ/t CO_2 captured	[14]
Solvent makeup demand	1	kg/t CO_2 captured	[15]
Water demand	611	kg/t CO_2 captured	[15]
Ammonia demand	10	kg/t CO_2 captured	[15]
CO_2 COMPRESSION to 110 bar			
Compression ;osses	0	%	assumed
Electricity demand	96	kWh/t CO_2 compressed	[1]
CO_2 STORAGE, geologic			
Losses	1	% of CO_2 stored	assumed, based on [1] and [7]
Transport distance	200	km	assumed
electricity demand	7	kWh/t CO_2 stored	[16]
STEAM BOILER			
Efficiency	90	%	
Electricity demand	5	kWh/GJ steam	[8]

Parameter	Qty	Unit	Source
ELECTRICITY GENERATION - IGCCC			
efficiency	56.6	%	[8]
CHARCOAL PRODUCTION, Missouri-style kiln			
Timber	2.7	t timber (dry mass)/t charcoal	[9]
CO_2 emissions	540	kg/t charcoal	[9]
Carbon efficiency	69	% of C in wood in charcoal	[9]
NATURAL CARBONATION			
Service life	50	years	assumption
Exposure conditions	Outdoors, exposed to rain; Indoors, covered		assumption
Ks	1.6, 4.4	mm/sqrt(year)	[10], Annex BB.1
Surface area	5, 5	m^2	assumption
Degree of carbonation	0.85, 0.4	%	[10], Annex BB.1
Maximum CO_2 uptake	0.49	t CO_2/t concrete	[10], Annex BB.1

Table 2. Energy content of fuels

Parameter	Qty	Unit	Source
Bituminous coal	25.8		[11]
Natural gas	48	GJ/t fuel	[11]
Charcoal	31.5	GJ/t fuel	[9]
Wood chips	20	GJ/t fuel	[11]
Mixed waste, 50% Ca	15	GJ/t fuel	assumption based on [12] and [13]

Table 3. Emission factors of fuels

Parameter	Qty	Unit	Source
Bituminous coal	96	kg CO_2/GJ	[11]
Natural gas	56	kg CO_2/GJ	[11]
Charcoal	112	kg CO_2/GJ	[9]
Wood chips	112	kg CO_2/GJ	[11]
Mixed waste, 50% Ca	122	kg CO_2/GJ	assumption based on [12] and [13]

Table 4. Life cycle CO_2 intensities of upstream supply chains

Parameter	Qty	Unit	Source
Aggregate	5	kg CO_2/t aggregate	[4]: market for gravel, round CH
Ammonia	1913	kg CO_2/t NH_3	[4]: market for ammonia, liquid RER

Bauxite	26	kg CO_2/t bauxite	[4]: market for bauxite GLO
Clay	10	kg CO_2/t clay	[4]: market for clay CH
Coal	169	kg CO_2/t coal	[4]: market for hard coal Europe, without Russia and Turkey
Concrete admixture	1620	kg CO_2/t admixture	[4]: market for chemical, organic GLO
Demolition	9	kg CO_2/t concrete demolished	[4]: treatment of waste concrete, not reinforced, sorting plant Europe without Switzerland
Electricity, current average	25	kg CO_2/GJ electricity	[4]: market group for electricity, medium voltage RER (minus direct CO_2 intensity of generation for data year (2016, 295g CO_2/kWh)
Gypsum	7	kg CO_2/t gypsum	[4]: market for gypsum, mineral RER
Iron ore	8	kg CO_2/t iron ore	[4]: market for iron ore, crude ore, 46% Fe GLO
Limestone	5	kg CO_2/t limestone	[4]: market for limestone, crushed, washed CH
Monoethanolamine	2557	kg CO_2/t MEA	[4]: market for monoethanolamine GLO
Natural gas	286	kg CO_2/t natural gas	[4]: market group for natural gas, high pressure Europe without Switzerland
Sand	11	kg CO_2/t sand	[4]: market for sand RoW
Timber, for charcoal	32	kg CO_2/t timber (dry)	[4]: market for cleft timber, measured as dry mass Europe without Switzerland
Transport, lorry	84	g CO_2/tkm	[4]: market for transport, freight, lorry >32 metric ton, EURO6 RER
Transport, pipeline	53	g CO_2/tkm	[4]: market for transport, pipeline, long distance, natural gas RER
Transport, rail	47	g CO_2/tkm	[4]: market for transport, freight, lorry >32 metric ton, EURO6 RER
Water	0.3	kg CO_2/m^3 water	[4]: market for tap water Europe without Switzerland
Wood chips	58	kg CO_2/t wood chips (dry)	[4]: market for wood chips, wet, measured as dry mass Europe without Switzerland

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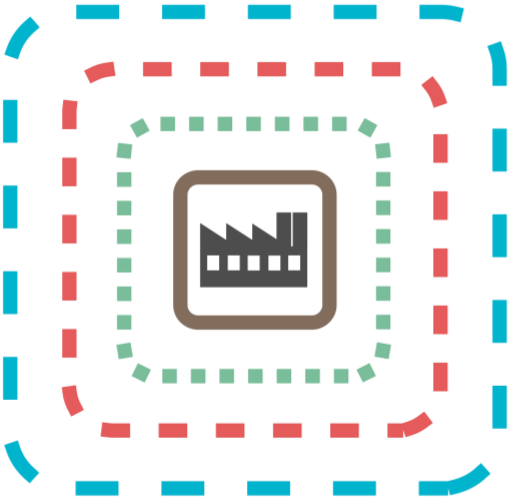
Chapter 7:
Scoping cost and abatement metrics for biomass with carbon capture and storage



Negative emission technologies such as biomass with carbon capture and storage (bioCCS) may become an important instrument to limit global warming. Currently, estimates of CO_2 avoidance cost for bioCCS vary widely. Using a case study of bioCCS in cement, this chapter illustrates how this variance is not only attributable to the assumed configuration of the bioCCS system, but also to system boundary choices made by modellers in accounting for both cost and abatement. Using ranges of system boundaries seen in literature, the avoidance cost for a bioCCS-in-cement plant was estimated to range from 48-321€$_{2017}$/t CO_2(eq) and the net CO_2(eq) from -660 to 16 kg CO_2(eq)/t cement, without any change in the technological model, equipment and input costs, or life cycle emissions assumed. The only change that occurred was change in the system boundaries used for cost and emission accounting, reflecting the different boundaries used across bioCCS literature. Limitations of single-point metrics such as net CO_2(eq) and avoidance cost are discussed, and guidelines are proposed to allow for clearer and more comparable estimates of bioCCS costs. These include always accounting for costs and emissions of both biomass production and the full chain of carbon capture, transport, and permanent storage, as both are fundamental to the role of bioCCS as a potential "negative emission technology". We also advocate for clear decomposition of metrics; separation of virtual "avoided emissions" from physical flows of greenhouse gases; and explicit consideration of the temporality of the bioCCS system. Accounting for these guidelines, the range of avoidance cost of the bioCCS-in-cement shrinks to 157-193€$_{2017}$/t CO_2(eq) for near-term estimates, depending on which greenhouse gases are considered, and to 89-107€$_{2017}$/t CO_2(eq) for longer-term estimates depending on the greenhouse gases and future technology scenario considered.

1 Introduction

The Glasgow Climate Pact reaffirmed a global commitment to limiting global warming to "well below 2°C" (3.6°F), a commitment that requires reducing our annual net emissions of carbon dioxide to zero—or less—within the next few decades [1]. To do so, and thus avoid the most catastrophic outcomes from the ongoing climate crisis will likely require the deployment of massive scale "negative emission technologies" that permanently remove greenhouse gases such as carbon dioxide from the atmosphere [1].

Biomass with carbon capture and storage (BioCCS) is a potential negative emission technology where biomass is used as an energy carrier or feedstock and the resulting biogenic CO_2 is captured and permanently stored, such as in a geologic formation. While large models primarily allocate bioCCS to the power sector or biofuel production [1, 2], it also has the potential to compensate for residual emissions from difficult to decarbonise industrial sectors, where carbon is a necessary element of feedstocks, catalysts, or products [3 (**chapter 3**)]. However, bioCCS combines the complexities of large-scale sustainable biomass use, the high energy demand of CO_2 capture, and the infrastructure demands of transporting and storing captured CO_2.

To understand whether bioCCS can be a viable option for CO_2-neutral industrial production, a fundamental question is how much does bioCCS cost? Cost estimates for industrial bioCCS in recent literature are limited and vary widely, with estimates ranging from 13-388 EUR_{2020}/t CO_2 abated [3], similar to that seen for bioCCS in general [1]. This variance is commonly discussed in terms of technological differences in the system or parametrical assumptions for cost estimates [3, 4]. In particular, whether the CO_2 is diluted or concentrated and the distance between where the CO_2 is captured and where it is stored are major factors in cost estimates.

However, variation in bioCCS abatement cost estimates is also due to variation of scope (**figure 1a**). Cost estimates may be presented per tonne of CO_2 captured; per tonne of CO_2 stored; or per tonne of CO_2 "avoided", which considers the reduction in emissions from the unabated case. Adding further ambiguity, all of these may be referred to as cost of "CO_2 avoided" in literature [5–10]. Furthermore, estimates of CO_2 avoidance potential of bioCCS—including those independent of cost estimates—also embody a wide range of system boundaries (**figure 1b**), and may or may not include CO_2 emissions from supply chains of biomass production, energy use, or chemicals and materials, and/or may or may not include emissions from (by)product use or CO_2 transport and storage. Additionally, in estimates of avoidance cost, the system represented by the cost estimate may not be the same as that represented by the emission avoidance estimate, such as assuming that the captured CO_2 is permanently stored in the emission abatement estimate, but excluding capital costs or emissions associated with transport and storage [6, 8, 11].

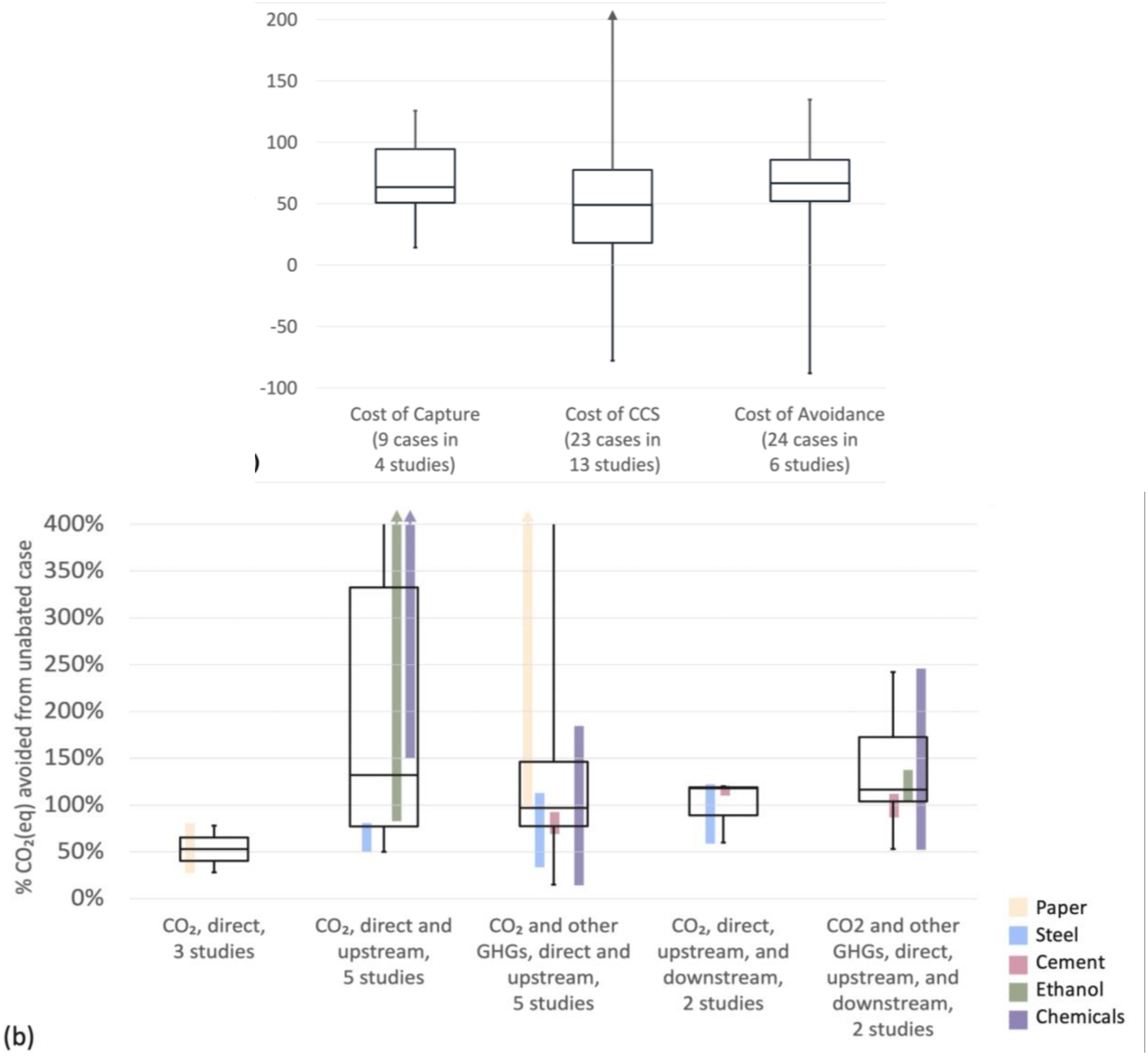


Figure 1. (a) Cost estimates of industrial bioCCS in literature by whether the estimate includes only the cost of capturing the CO_2 at the industrial site ("cost of capture"), the cost of capturing, transporting, and storing the CO_2 ("cost of CCS"), or the cost of reduction in emissions from the unabated case "cost of avoidance". The highest estimate of cost of CCS was 726 $€_{2017}$ and was clipped for clarity. The large range of cost of CCS is due primarily to differences in transport distance between studies. Negative values result from the assumption of a credit or subsidy for stored CO_2 or the sale of CO_2, e.g., for enhanced oil recovery. (b) Estimates of CO_2(eq) avoidance of industrial bioCCS, relative to the unabated production, aggregated by what system boundaries are used for emission accounting. Ranges reported for individual industries are given in the coloured bars. Many studies combined estimates of physical emissions and removals of greenhouse gases with estimates of "avoided emissions", assumed reductions in emissions occurring in other systems from e.g., the sale of cogenerated electricity. Therefore, a relative avoidance > 100% does not necessarily indicate negative emissions (i.e., physical net removal of greenhouse gases from the atmosphere). High estimates of relative avoidance resulted when the unabated system was itself biogenic and the study assumed "carbon neutral" biomass, or included "avoided emissions", leading to an unabated system with a low

or negative net CO_2(eq) estimate. Both figures are based on the literature review conducted in chapter 3.

If bioCCS is to be a lynchpin technology in reaching net-zero in the coming decades, it is critical that there are transparent and comprehensive estimates of both cost and emissions available. Overestimation of avoidance, or underestimation of cost, can lead to miscalculations in investment and policy or misassessment of the possible role of bioCCS. This chapter explores the influence of system boundary choices found in bioCCS studies on estimates of technological cost, potential abatement, and CO_2 avoidance cost.

Instead of assuming a static set of system boundaries and exploring the influence of the assumed configuration of technology, input costs, or geography, this chapter holds the configuration static and explores the influence of system boundaries on the avoidance cost. By this, we explore the relative influence of these choices on avoidance cost and propose guidelines for bioCCS avoidance cost estimates. The boundaries considered are based on those found in other bioCCS literature [3] and, for cost, include operational and capital expenses of capture, transport, and storage, as well as the type of cost-scaling estimation used. For avoidance, system boundary options include direct emissions from the industrial production site, electricity generation, energy supply chains, material and chemical supply chains, CO_2 transport and storage, and product use. Other greenhouse gas emissions and warming impacts of biogenic CO_2 are also considered.

To explore the impact of system boundaries on avoidance costs, this chapter considers the case of a bioCCS retrofit of a cement plant. Cement is the second largest industrial emitter of CO_2, after iron and steel, with 2.4 Gt of CO_2 emissions in 2019 [12], of which approximately 60% were from calcination of limestone ($CaCO_3 \rightarrow CaO + CO_2$), and thus cannot be decarbonised by a fossil-free energy mix. The cement industry itself perceives the need for a decarbonisation approach that requires both CCS and the increased use of biogenic fuels [13, 14]. In 2019, co-fired biomass represented 18% of European cement kiln fuel [15], primarily in the form of biowastes, and the first full-scale retrofit of CO_2 capture into cement production is currently under construction in Brevik, Norway [16]. However, the existing literature on bioCCS-in-cement is sparse [9, 17–19] and as of 2021, there exists, to our knowledge, no dedicated study on the CO_2 avoidance costs of bioCCS-in-cement.

2 Methods

The results of this chapter are based on an *ex-ante* model of the use of biomass and CCS (bioCCS) in cement. It consists of a process model of cement production with and without biomass use and CO_2 capture, transport and storage; a life cycle accounting of greenhouse gas emissions; and an economic assessment of cement production costs. **Figure 2** summarises the boundaries of the complete system that was modelled.

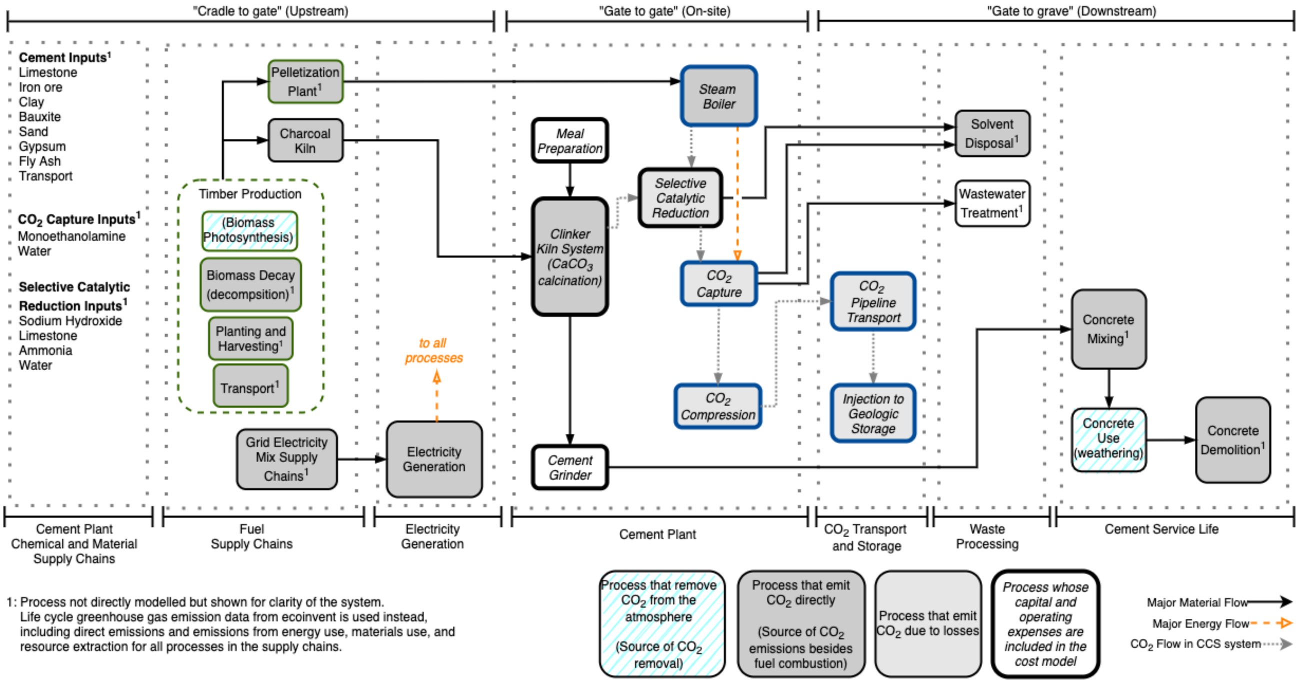


Figure 2. Model overview. For clarity, many material flows are not explicitly connected. Full model parameters are available in the appendix.

As our baseline, a state-of-the-art coal-fired[8] cement plant in northwest Europe was modelled based on [20, 21], in line with the EU Best Available Technology guidelines [22]. To align with the reference models, it has a design capacity of 1.36 Mt/year of CEMII cement (73% clinker), with an average energy consumption of 2.4 GJ and 104 kWh per tonne of cement [20, 21]. For our bioCCS system, the kiln was assumed to be charcoal-fired. All CO_2 emissions from the kiln, from both fuel combustion and limestone calcination, were sent to a post-combustion MEA-adsorption unit, assuming a 90% capture rate and a reboiler duty of 3.2 GJ/t CO_2. The steam demand of CO_2 capture was met by pellet-fired boiler, whose CO_2 was also captured. The captured CO_2 was compressed and transported 100 km by dedicated pipeline for storage in a near-shore legacy gas reservoir. The cement is assumed to be used in the production of an exterior concrete wall with a 50-year lifespan.

2.1 Process model

The process model consisted of connected fixed-ratio black box models for each unit process and was constructed following the methodology described in Chapter 6, using the custom

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[8] Commonly, European cement kilns use a mix of fuels that include fossil and biogenic waste products alongside coal. For the clarity of this case study, the simplification of a single-fuel kiln was used for both the unabated and bioCCS cases.

open-source python modelling library described in Appendix A at the end of this dissertation. The main model parameters are summarised in **table 1** and are provided in full in the appendix. For the use case model, the cement was assumed to be used in 25 Mpa concrete, requiring 200 kg cement/m^3 of concrete [31].

Cement is produced by heating ground limestone and aluminosilicate minerals such as clay or bauxite, which are then to 900-1200°C, allowing the limestone to calcinate ($CaCO_3$ + heat → $CaO + CO_2$) and amalgamate with the other mineral constituents into clinker, the primary component of cement. The clinker is then blended with additives such as gypsum, fly ash, and/or steel slag and pulverised into cement powder. To produce concrete, the cement is hydrated, mixed with aggregates (sand and gravel), and poured into form, where it hardens.

The CCS model included the retrofit of post-combustion CO_2 capture using monoethanolamine (MEA) to separate CO_2 from the clinker kiln flue gases. MEA-based capture was selected as it is a commercially available technology and provides a conservative assumption for the energy demand of capture. Based on a literature review of MEA-based CO_2 capture, a capture rate of 90% with a reboiler duty of 3.2 MJ/kg CO_2 separated was assumed [17 (**chapter 6**)]. It was assumed that no waste heat from the kiln was available for use in the CO_2 capture system. Instead, steam was provided from a dedicated boiler. Flue gases from the steam boiler were also assumed to be sent to CO_2 capture. Captured CO_2 was then compressed to 11 Mpa, transported by pipeline (100 km onshore and 10 km offshore), and injected into a legacy gas reservoir.

In the bioCCS case, charcoal was assumed to replace coal 1:1 as clinker kiln fuel on an energy basis and the steam boiler was assumed to be fired with wood pellets. Charcoal was assumed for the kiln as it has a sufficient energy density to theoretically reach the required temperatures for clinker production [24, 26]. In all cases, electricity was assumed to be provided by the Dutch electricity grid mix with a direct generation intensity of 390 g CO_2/kWh [26]. **Table 2** provides the emission factors and energy contents of fuels used in this model.

Table 1.Main model parameters

Parameter	Qty	Unit	Source
CEMENT PRODUCTION			
Clinker content of cement	737	kg/t cement	[27]
Limestone content of meal	745	kg/t meal	[19]
Clinker kiln thermal energy demand	3.3	GJ/t clinker	[22]
Electricity demand	104	kWh/t cement	[28]
Transport of raw materials, by train	200	km	Assumed
CO_2 CAPTURE AND COMPRESSION			
Capture efficiency	90	% of CO_2 in	
Reboiler duty	3.2	GJ/t CO_2 captured	Assumed, based on literature review of MEA capture [17]
Monoethanolamine makeup	1	kg/t CO_2 captured	Assumed, based on literature review of MEA capture [17]
Electricity demand, capture	38	kWh/t CO_2 captured	[20, 27]
Electricity demand, compression to 110 bar	106	kWh/t CO_2 compressed	[27]
STEAM BOILER			
Efficiency	90	%	assumed
Electricity demand	5	kWh/GJ steam	[17]
CO_2 TRANSPORT BY PIPELINE AND INJECTION TO GEOLOGIC STORAGE			
Transport distance, onshore	100	km	assumed
Transport distance, offshore	10	km	assumed
electricity demand, transport	3	kWh/t CO_2 transported	[29]
electricity demand, injection	7	kWh/t CO_2 stored	[30]
CHARCOAL PRODUCTION – MISSOURI-STYLE KILN			
Carbon efficiency	69	% of C in wood in charcoal	[31]
CO_2 emissions	540	kg CO_2 / t charcoal	[31]
CH_4 emissions	1022	kg CO_2eq / t charcoal	[31]
CONCRETE WEATHERING			
Service life	50	years	assumption
Uptake of CO_2, service life	12%	% of CO_2 removed during calcination	[32], Annex BB.1
Uptake of CO_2, demolition	3%	% of CO_2 removed during calcination	[32]

Table 2. Fuel energy contents and emission factors [31, 37]

	Energy Content (GJ/t)	CO_2	CH_4	N_2O	Unit
Coal	25.8	96.1	<0.001	<0.001	kg CO_2eq/GJ
Natural gas	48.0	56.1	<0.001	<0.001	kg CO_2eq/GJ
Charcoal	31.5	112	0.005	0.001	kg CO_2eq/GJ
Wood pellets (dry)	19.1	112	0	0.002	kg CO_2eq/GJ

2.2 Life cycle assessment

The estimates of net greenhouse gas emissions included cement production, biomass production and use, and CCS. For the emission estimates of upstream supply chains of material and energy carriers, downstream supply chains of waste disposal, and supply chains of transport and building and equipment construction, data from the life cycle inventory database ecoinvent 3.7.1 [33] was used. These include estimates of greenhouse gas emissions as a result of resource extraction, material production, energy use, and transport throughout the supply chains—and the supply chains of the supply chains. The exact database processes used are provided in the appendix. Emissions of greenhouse gases other than carbon dioxide are characterised in their 100-year "CO_2 equivalent" (CO_2eq) global warming potential [34].

The model of life cycle emissions, like the process model itself, relies on generic literature data, and is not meant to represent a specific installation of bioCCS-in-cement, nor determine an optimal system of production, but rather to provide a representative example based on currently available data.

Emissions of CO_2, CH_4, and N_2O from the production of cement, concrete, and biofuels, as well as from steam production, electricity generation, and CO_2 capture, transport, and storage, were also estimated. For N_2O emissions at the cement plant, selective catalytic reduction was assumed to abate 90% of produced N_2O [35].

The wood used for charcoal and pellets was assumed to be from sustainable European forestry with a 100-year rotation period, such as for boreal forestry species of Scots pine or Norwegian spruce [36], after which the total carbon removed and stored in the timber is equal the total carbon embodied in the initial amount of biomass used. The biomass was assumed to be harvested and replanted in the same year of cement production.

Concrete also absorbs biomass over time as the free lime recarbonates into limestone. CO_2 uptake by concrete was modelled for CEMII concrete, assuming that the concrete was used as a 20cm exterior wall with a 50-year lifetime using the calculation method in [32], described in section 2.4 of chapter 6. At end of life, the concrete was assumed to be reused as a road sublayer or other application where it is no longer exposed to air. In total, recarbonation during concrete use and demolition was assumed to result in the uptake of the equivalent of 15% of the CO_2 released during limestone calcination.

2.2.1 System boundaries considered

This chapter considers the net emissions—total emissions of greenhouse gases to the atmosphere minus total removals of CO_2 from the atmosphere—for nine different system boundaries seen in bioCCS literature, listed in **table 3**. Between the different boundary options, configuration and parameters of the model itself do not change, only what elements are included in the estimation of net emissions according to each boundary.

2.3 Economic model

The economic model estimated the cost of cement production with and without bioCCS, building on the results of the process model. The cement plant was assumed to be located in the Netherlands and operate at 91.3% of its design capacity, as was assumed in the main cost model reference [27]. The infrastructure of CO_2 capture and compression were assumed to be retrofitted into an existing cement plant, while the CO_2 pipeline was assumed to be built on unused land. The cost model in this chapter followed the guidelines for CCS cost estimation in [40].

All costs are presented in 2017 Euros, using EPCCI to scale capital costs [41] and historical inflation rates to scale operational costs [42].

Table 3. System boundaries considered in this chapter

	Gate-to-gate CO$_2$	Gate-to-gate CO$_2$ assuming “CO$_2$ neutral” biomass	Gate-to-gate CO$_2$ and electricity	Cradle-to-gate CO$_2$, energy supply chains only	Cradle-to-gat” CO$_2$	Cradle-to-grave CO$_2$, excluding use	Cradle-to-grave CO$_2$, including use	Cradle-to-grave CO$_2$eq including biogenic CO$_2$ GWP	Cradle-to-grave CO$_2$eq, all GHGs and biogenic CO$_2$ GWP	Data source
CO$_2$ emitted at cement plant from cement kiln	X	X	X	X	X	X	X	X	X	[20, 27, 37]
CO$_2$ emitted at cement plant, from CO$_2$ capture system	X	X	X	X	X	X	X	X	X	[20, 75]
CO$_2$ emitted at by fuel combustion at electricity generation site			X	X	X	X	X	X	X	[26]
CO$_2$ emitted in supply chains of coal, charcoal, wood pellets, and electricity				X	X	X	X	X	X	[33]
CO$_2$ emitted in supply chains of non-energy material and chemical inputs					X	X	X	X	X	[33]
CO$_2$ emitted in supply chains of cement and CO$_2$ capture infrastructure						X	X	X	X	[33]
CO$_2$ emitted by CO$_2$ transport and storage and its energy and infrastructure supply chains						X	X	X	X	[33, 38]
CO$_2$ emitted by the disposal of wastewater and waste solvents						X	X	X	X	[33]
CO$_2$ emitted by the production, use and demolition of concrete and their supply chains							X	X	X	[33]
GWP of biogenic CO$_2$ emitted								X	X	[39]
CH$_4$ and N$_2$O emitted during the production charcoal and cement									X	[31, 37]
CH$_4$, N$_2$O, and other greenhouse gases emitted in upstream supply chains									X	[33]
CO$_2$ removed by biomass photosynthesis		X	X	X	X	X	X	X	X	equal to biogenic CO$_2$ produced
CO$_2$ removed by concrete weathering							X	X	X	[32]

2.3.1 Capital expenses

The capital cost models used equipment scales derived from the material and energy flow estimates in the process models. For CO_2 capture, compression, and steam production, equipment costs were scaled using **equation 1**. The CO_2 capture system also included 500m of stainless steel ducting for flue gas transport within the cement plant, using cost estimates from [40]. The size of the CO_2 transport pipeline was calculated assuming an inlet pressure of 11 Mpa, outlet pressure of 8 Mpa, and a pressure drop of 50 m/second, using the method presented in [29]. **Table 4** presents the estimated equipment scales, costs, and data sources used.

$$\text{Cost}_{scaled,2017} = \text{Cost}_{base} \times \left(\frac{\text{Capacity}_{scaled}}{\text{Capacity}_{base}}\right)^{0.7} \times \left(\frac{\text{Cost Index}_{2017}}{\text{Cost Index}_{base}}\right) \qquad (1)$$

Table 4. Equipment costs, installation costs plus maintenance factor

Equipment	Equipment Cost		Base Scale	Scale unit	Installation Factor	Process Contingency	Maintenance	Source
CO_2 absorption (MEA)	7.901	M €$_{2014}$	765	kt CO_2 captured/year	1.76	18%	7%	[27]
CO_2 desorption (MEA)	7.024	M €$_{2014}$	765	kt CO_2 captured/year	2.04	18%	7%	[27]
CO_2 compression	14.857	M €$_{2014}$	765	kt CO_2 captured/year	1.24	23%	7%	[27]
CO_2 dehydration (TEG)	0.228	M €$_{2014}$	765	kt CO_2 captured/year	4.07	23%	7%	[27]
CO_2 dehydration (TEG)	0.228	M €$_{2014}$	765	kt CO_2 captured/year	4.07	23%	7%	[27]
Flue Gas Cleaning Unit (additional capacity)	1.1	M €$_{2013}$ (direct cost)	765	kt CO_2 captured/year	*n.a.*	*n.a.*	3.5%	[27]
Steam boiler	34	M €$_{2013}$	4730	TJ/year	2.08	10%	3.5%	[20]
CO_2 injection	19	M €$_{2010}$ (direct costs)	1000	kt CO_2 stored/year	*n.a.*	*n.a.*	included in storage variable costs	[43]

Starting from equipment costs, scaling factors were used to determine the costs of installation, labour, land and buildings, construction contingencies, financing, insurance, and taxes. As, CO_2 capture is not yet a commercialised technology for cement production, with the first full-scale installation currently under development [16], capital expenses were estimated using factors for a “first of a kind” cost escalation that assumes the need for redundant equipment and substantially larger factors for contingency and supplementary funds to account for the need for “on the job” technological learning and a higher likelihood of unexpected costs and delays. However, as many economic models for bioCCS-in-industry follow the convention of “Nth of a kind” cost estimation, which assumes that all technology components are available and usable as if they were commercialised technologies [3], an "Nth of a kind" estimate was also calculated. The cost escalation factors for both methods are presented in **table 5**.

Table 5. Cost model scaling factor

Cost Factor	Includes		"First of a kind"	"Nth of a kind"
Installed Costs (IC)	Equipment Costs (EC) +	Installation costs	as in Table 4	
Direct Costs (DC)	IC +	Process contingencies		
		Equipment redundancies (for CO_2 capture and compression equipment)	equipment scaled to 3 x 50% of capacity	no redundancies
Total Plant Costs (TPC)	DC +	Owner costs	7% of DC	7% of DC
		Indirect costs	14% of DC	14% of DC
		System contingencies	10% of DC	n.a.
		Project contingency	50% of DC	30% of DC
		Supplementary funds	50% of DC	25% of DC

2.3.2 Capital charge

The capital expenses of the CCS system were annualised into a capital charge assuming a 25-year lifetime (n) and an 8% discount rate (i) using **equation 2**. The capital charge also includes the costs of a three-month shutdown of cement production to retrofit the CO_2 capture system during which time the fixed costs of cement production still occur [40].

$$\text{Capital Charge}_{annual} = \text{Total Plant Costs of CCS} + \text{Fixed Costs during Retrofit} \times \frac{i \times (i+1)^n}{(i+1)^n - 1} \qquad (2)$$

The cement plant was assumed to be extant and paid off and its component capital costs were not estimated and no capital charge was considered. For the purposes of estimating taxes, insurance, and maintenance, a total capital cost of 150.7 M€$_{2017}$ for the cement plant was assumed [27].

2.3.3 Operating expenses

The operating cost model includes the variable costs of material inputs and utilities and the fixed costs of labour, maintenance, insurance, and taxes.

2.3.3.1 Variable costs

The variable costs included the cost of materials, fuels, and utilities needed for the production of cement and operation of CO_2 capture, transport, and storage. The costs and data sources used are presented in **table 6**.

Table 6. Variable costs

Variable Costs	Cost	Unit	Source
ENERGY COSTS			
Electricity	0.06	€$_{2017}$/kWh	[44]
Coal	3	€$_{2017}$/GJ	[45]
Charcoal	10	€$_{2017}$/GJ	[46]
Wood pellets	11	€$_{2017}$/GJ	[46]
UTILITY COSTS			
Water	1	€$_{2017}$/t water	Netherlands regional market average
CEMENT PRODUCTION COSTS			
Raw meal	5.1	€$_{2017}$/t clinker	[27]
Other materials	1.1	€$_{2017}$/t clinker	[27]
CCS COSTS			
MEA	1476	€$_{2017}$/t MEA	[27]
Ammonia	132	€$_{2017}$/t NH_3	[47]
Sodium hydroxide	377	€$_{2017}$/t NaOH	[27]
CO_2 injection, offshore	6.5	€$_{2017}$/t CO_2 stored	[43]

2.3.3.2 Labour

The cement plant was assumed to employ 100 workers and the operation of the CO_2 capture system was assumed to require 20 additional personnel [27]. Labour costs were assumed to be 62 000€$_{2017}$/person/year [48] based on the Dutch manufacturing sector average, with an additional 30% of operational labour costs for administrative labour.

2.3.3.3 Maintenance

Maintenance costs were based on the installed cost of equipment, as indicated in **table 4**.

2.3.3.4 Insurance and taxes

Insurance and taxes were assumed to be 2% of total capital costs per year [27].

2.3.4 System boundaries considered

Cost estimates of bioCCS in literature encompass different components of the CCS system, and therefore different boundaries of cost estimates were considered in this chapter (**table 7**), both from the perspective of "first of a kind" and "Nth of a kind" cost scaling assumptions.

Table 7. **Cost model system boundaries considered**

	without CCS	with marginal cost of CO_2 capture	with full cost of CO_2 capture	with full cost of CO_2 capture and marginal cost of CO_2 transport and storage	with full cost of CO_2 capture, transport, and storage	data sources
Cement production materials	x	x	x	x	x	[27]
Cement production fuel and electricity (including kiln fuel switching from coal to charcoal)	x	x	x	x	x	[44–46]
Cement production labour, maintenance, insurance, and taxes	x	x	x	x	x	[27, 48]
CO_2 capture materials		x	x	x	x	[27, 47]
CO_2 capture and compression fuel and electricity		x	x	x	x	[44, 46]
CO_2 capture system labour, maintenance, insurance, and taxes		x	x	x	x	[27, 48]
Annualised capital expenses of CO_2 capture system			x	x	x	[27]
CO_2 transport and storage energy use				x	x	[44]
CO_2 transport and storage labour, maintenance, insurance, and taxes				x	x	[43]
Annualised capital expenses of CO_2 pipeline and injection					x	[29, 43]

2.4 CO_2(eq) avoidance cost

The CO_2(eq) avoidance cost is the cost per unit reduction in net CO_2(eq) emissions from one system configuration and another, calculated as in Equation 3. In this chapter, it is the cost in €$_{2017}$/t CO_2(eq) of reducing the greenhouse gas emissions of cement by retrofitting bioCCS into the system of production. As the cost of CO_2(eq) avoidance depends on both the estimated net emissions and estimated costs, it is presented for each combination of cost and emission system boundaries.

$$\mathrm{CO_2(eq)\ avoidance\ cost} = \frac{\mathrm{Cost\ of\ production\ with\ abatement} - \mathrm{Cost\ of\ unabated\ production}}{\mathrm{CO_2(eq)\ of\ unabated\ production} - \mathrm{CO_2(eq)\ of\ production\ with\ abatement}} \qquad (3)$$

3 Results and discussion

3.1 Accounting for costs

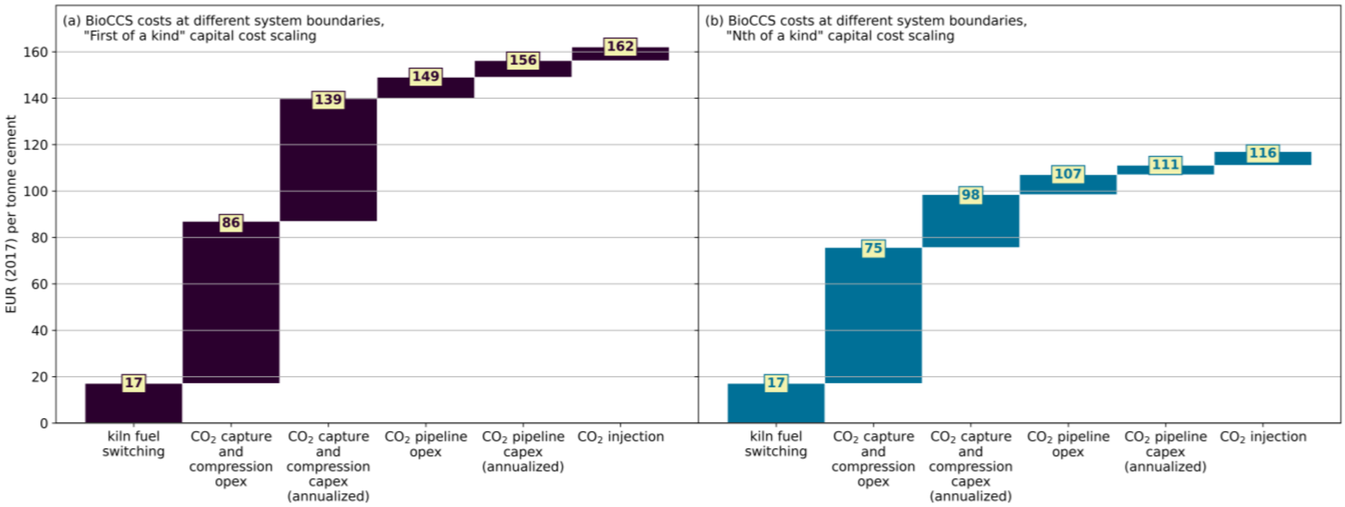

Figure 3. Cost of bioCCS, separated by system component. These costs are in addition to the baseline costs of cement product (33€$_{2017}$/t cement).

The first component of avoidance cost estimates is the cost of production relative to the unabated system, in this case, the cost of cement with bioCCS in comparison to without. In our model, the cost of unabated cement production was 33€$_{2017}$/t cement. While the production of charcoal and monoethanolamine-based capture systems are themselves commercialised technologies, they are not commercialised in the cement industry. Therefore, we considered two cost scaling options: "First of a kind" (FOAK) cost scaling (**figure 3a**), which assumes greater contingencies and equipment redundancies for a near-term installation where technological learning is still needed; and "Nth of a kind" (NOAK) cost scaling (**figure 3b**), which assumes that bioCCS is available as a fully commercialised technology. Most available studies on bioCCS use NOAK scaling, focusing on the question of what bioCCS *could* cost, once it is fully developed [40]. FOAK estimates instead consider the question of what bioCCS *would* cost if implemented in the near term.

In our model, the cost of bioCCS costs were dominated by "on-site" costs of fuel switching and CO_2 capture. The most expensive element is the marginal cost of capture (75€$_{2017}$ (63 NOAK) / t CO_2 captured), of which half is the purchase of wood pellets to supply the energy needed for the capture unit, 20% is electricity, and the remaining is chemicals, labour, maintenance, and other operating costs. The difference in operating costs between the FOAK and NOAK cases is explained by the difference in estimated capital costs, as the FOAK capital cost estimate is double that of the NOAK estimate. This, in turn, leads to a 51% higher total "cost of capture" at 133 vs 88€$_{2017}$/t CO_2 captured for FOAK and NOAK, respectively.

Cost of transport and storage was a smaller factor in this model, which assumed 100 km of dedicated pipeline transport, at 24€$_{2017}$ (22 NOAK)/t CO_2 transported and stored, of which half are capital expenses. However, transport costs accounted for the majority of variability in the studies reviewed in **chapter 3**, ranging from 5-368€$_{2017}$/t CO_2, depending on distance and mode of transport. In our model, doubling transport distance to 200 km increased costs by 11€$_{2017}$ (8 NOAK)/t CO_2, primarily from additional pipeline construction, but also 1.5€$_{2017}$ from additional electricity needed for recompression and pumping.

3.2 Accounting for abatement

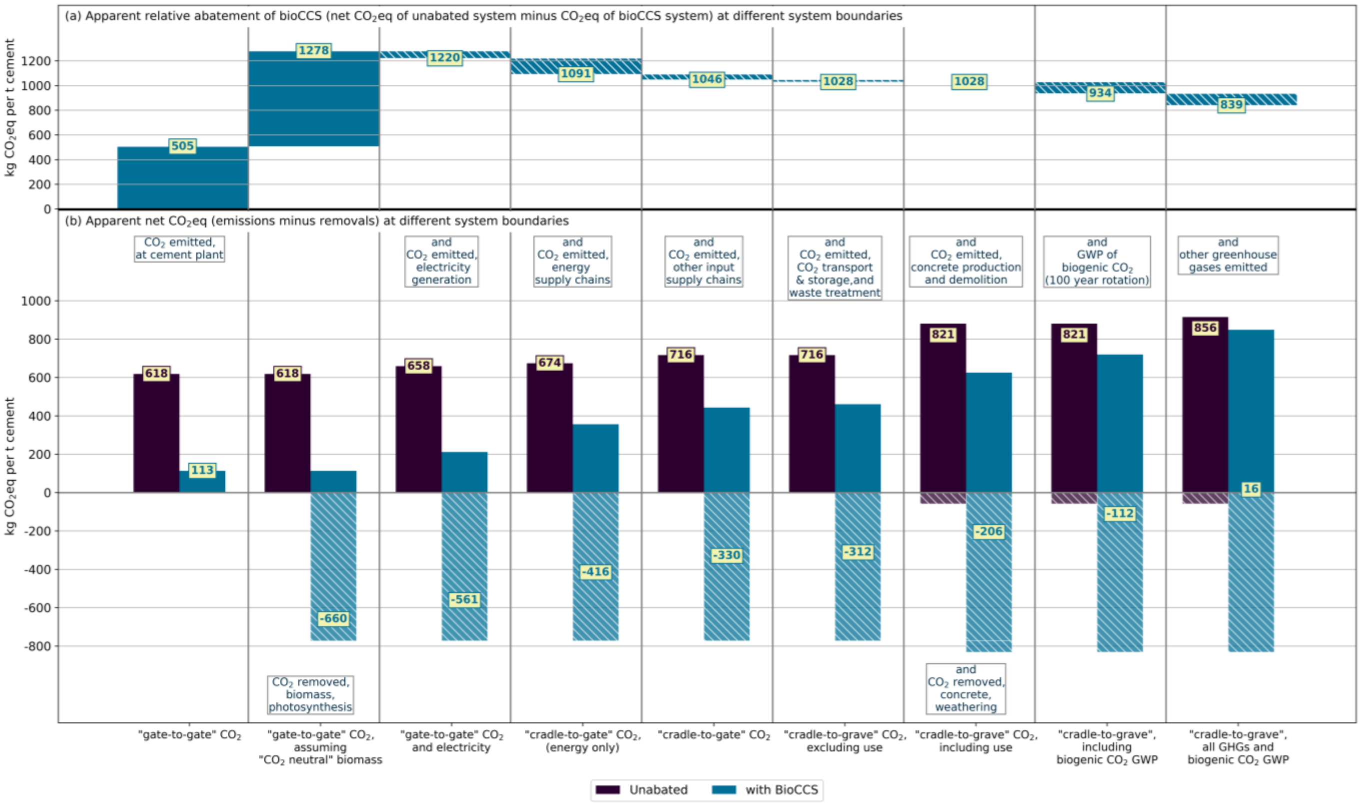

Figure 4. (a) Relative abatement, the metric used in CO_2 avoidance cost, is the difference in emissions from unabated cement production to cement production with bioCCS. The change in apparent relative abatement as system boundaries expand is the difference in how much the apparent net CO_2eq of each case changes as additional parts of their life cycle system are accounted for. As the emissions of cement use are the same for both systems, the relative abatement does not change in that instance, even though both their net CO_2eq increases. (b) Here net CO_2(eq) is the net total of modelled emissions and (permanent) removals of greenhouse gases in the atmosphere. The only thing that changes between the different instances is what emissions and removals are accounted for; the model itself is static. Biogenic CO_2 is assumed to have a 100-year GWP of 0.44kg CO_2eq/kg Co_2bio [39] and other greenhouse gases are also characterised by their 100-year CO_2eq GWP [34].

The other component of avoidance cost estimates is the change in net emissions from the unabated case. In this case, the estimated abatement potential is the net emissions of a present-day cement plant minus the net emissions of that cement plant with bioCCS, which is the “relative abatement” presented in **figure 4a** at different system boundaries. Beneath, **figure 4b** presents the apparent net emissions and removals, and the resulting net CO_2eq, of both the unabated and bioCCS cement plants for each system boundary.

In the unabated case, CO_2 emitted at the cement plant accounts for over 80% of the total CO_2 emissions estimated for production, supply chains, and cement use and disposal. Upstream supply chains account for 99 kg CO_2/t cement, of which 41 kg is from electricity and 15 kg is from coal.

Studies that focus on the technological cost of bioCCS often only account for CO_2 emitted at the industrial plant itself. At this “gate-to-gate” boundary, bioCCS results in an 80% decrease in estimated CO_2 emitted. If the model also assumed that the biomass is “CO_2 neutral”, without accounting for other impacts, the bioCCS case appears to be deeply “CO_2 negative”. However, in the bioCCS case most emissions occur outside the cement production gates. Besides the 84 kg CO_2 from supply chains of the material and electricity demand of cement production, the biofuel supply chains emit 243 kg CO_2/t cement, and the electricity demand of CO_2 capture and compression is also responsible an additional 64 kg CO_2. Downstream, emissions from the transport and storage of CO_2 are less significant in this model, accounting for less than 15 kg CO_2/t cement, though this may not be true for systems that use more carbon-intensive truck transport, e.g., as in [49, 50].

From a “cradle-to-grave” perspective that incorporates upstream and downstream emissions of CO_2 in the bioCCS and cement production chains, bioCCS has an apparent avoidance potential of 1028 kg CO_2/t cement, or 144%. However, this metric of relative avoidance does not provide information about the absolute magnitude of net emissions. In bioCCS systems, the combination of emissions and removals also means that the “net CO_2” metric also obscures this information. Furthermore, the total carbon intensity of the system is further obscured if the amount of stored CO_2—here 907 kg/t cement— is not reported. Despite its lower net CO_2 emissions, the bioCCS system *produces* 652 kg/t cement more CO_2 than the unabated system. If the fate of CO_2 was not permanent storage, but instead reuse in short-term products or otherwise re-emitted, the net CO_2 of the bioCCS system would be 595 kg CO_2/t concrete, just 120kg lower than the unabated case.

If bioCCS is to allow for “CO_2 neutral” (or negative) cement production, then it must also produce enough “negative emissions” to compensate for emissions for use and end-of-life of the cement, as well as direct and upstream emissions. Expanding the system to also include downstream emissions of cement use in concrete increases net CO_2 by 115 kg/t cement. Since this is the same for both systems, relative abatement, the metric considered in avoidance cost, remains unchanged.

In studies considering "CO_2 neutral biomass", CO_2 reuptake by biomass is assumed to perfectly offset biogenic CO_2 emissions. However, emitted biogenic CO_2 contributes to global warming during its temporary residence in the atmosphere. For long rotation biomass, this impact can be significant in the short term, with a bioCCS system contributing more CO_2 to the atmosphere than its fossil counterpart in the first third of rotation period [17]. In this model, we assumed that the timber for charcoal and pellet production has a 100-year rotation period. Accounting for this by using a 100-year global warming potential factor of 0.44kg CO_2eq/kg biogenic CO_2 [39] increases the net CO_2eq of the BioCCS case by 89 kg CO_2eq/t cement.

BioCCS can also have substantial non-CO_2 greenhouse gas emissions. In the unabated system, other greenhouse gases increase the net CO_2eq estimate by 34 kg CO_2eq/t cement—half being fugitive methane from fossil fuel supply chains. In contrast, the net emissions of the bioCCS system increase by 128 kg CO_2eq, of which 60% is CH_4 from charcoal production and 25% is from electricity supply chains.

At the broadest system boundaries considered in this chapter, bioCCS in cement production is no longer "CO_2eq negative". That is, the removal and storage of biogenic CO_2 is insufficient to compensate for the life cycle greenhouse gas emissions of cement production and use, bioenergy production, and CCS. At 16 kg CO_2eq/t cement, the net CO_2eq estimate of this more complete system is 676 kg/t cement higher than estimating only the net emissions from "gate to gate CO_2 with carbon neutral biomass". The estimated relative abatement changed less, decreasing by 439 kg CO_2, as the net CO_2eq of the unabated system also increased with the expanding boundaries, adding 238 kg CO_2eq/t cement. Nothing in the cement or bioCCS system has changed, only how comprehensively the emissions and removals are estimated.

3.3 The avoidance cost possibility space

Combining the different system boundaries of near term FOAK cost estimates with those of the "present day" avoidance potential estimates results in avoidance cost "possibility space" in **table 8a** ranging from 68-321€$_{2017}$/t CO_2(eq) avoided. For each abatement boundary considered, expanding the costs considered from marginal cost of fuel switching and CO_2 capture to the full operating and capital expenses of bioCCS leads to a doubling in the cost of avoidance. In our model capital costs of capture and compression account for 30% of avoidance costs and transport and storage 15%.

	Increase in cement production cost (EUR$_{2017}$/t cement)	gate-to-gate CO_2	gate-to-gate CO_2 assuming "CO_2 neutral" biomass	gate-to-gate CO_2 and electricity	cradle-to-gate CO_2, energy supply chains only	cradle-to-gate CO_2	cradle-to-grave CO_2	cradle-to-grave CO_2eq, including biogenic CO_2 GWP	cradle-to-grave CO_2eq, all GHGs and biogenic CO_2 GWP
(a) using "First of a kind" cost scaling and baseline model parameters									
Apparent abatement of bioCCS system (kg CO_2eq/t cement)		*505*	*1279*	*1220*	*1091*	*1047*	*1029*	*934*	*839*
Kiln fuel switching and CO_2 capture opex	*87*	172	68	71	80	83	85	93	104
...and CO_2 capture capex (annualized)	*140*	277	109	115	128	134	136	150	167
...and CO_2 pipeline transport and storage opex	*149*	295	117	122	137	142	145	160	178
...and CO_2 pipeline transport and storage capex (annualized)	*162*	321	127	133	149	155	158	174	193
(b) using "Nth of a kind" cost scaling, and baseline model parameters									
Apparent abatement of bioCCS system (kg CO_2eq/t cement)		*505*	*1279*	*1220*	*1091*	*1047*	*1029*	*934*	*839*
Kiln fuel switching and CO_2 capture opex	*76*	150	59	62	69	72	74	81	90
...and CO_2 capture capex (annualized)	*98*	195	77	81	90	94	96	105	117
...and CO_2 pipeline transport and storage opex	*107*	212	84	88	98	102	104	115	128
...and CO_2 pipeline transport and storage capex (annualized)	*117*	232	91	96	107	112	114	125	139
(c) using "Nth of a kind" cost scaling and assuming modest improvement in kiln and CO_2 capture efficiencies, and 60% decarbonization of transport and electricity									
Apparent abatement of bioCCS system (kg CO_2eq/t cement)		*494*	*1182*	*1180*	*1066*	*1039*	*1026*	*942*	*903*
Kiln fuel switching and CO_2 capture opex	61	124	52	52	58	59	60	65	68
...and CO_2 capture capex (annualized)	83	167	70	70	77	79	80	88	91
...and CO_2 pipeline transport and storage opex	90	183	76	77	85	87	88	96	100
...and CO_2 pipeline transport and storage capex (annualized)	99	200	84	84	93	95	96	105	110
(d) using "Nth of a kind" cost scaling and assuming optimistic improvement in kiln and CO_2 capture efficiencies, and 100% decarbonization of transport and electricity									
Apparent abatement of bioCCS system (kg CO_2eq/t cement)		*494*	*1064*	*1064*	*971*	*952*	*940*	*872*	*855*
Kiln fuel switching and CO_2 capture opex	51	103	48	48	53	54	54	59	60
...and CO_2 capture capex (annualized)	70	141	65	65	72	73	74	80	81
...and CO_2 pipeline transport and storage opex	77	155	72	72	79	81	82	88	90
...and CO_2 pipeline transport and storage capex (annualized)	85	172	80	80	88	89	91	98	100

Figure 5. Avoidance costs by system boundaries of cost and net emission estimates, €$_{2017}$/t CO_2 abated. The future scenarios considered in (c) and (d) are based on those used in [17 (chapter 6)]. Graphs of the net CO_2(eq) and relative abatement for the future scenarios are available in the appendix.

The highest avoidance cost estimates are seen when only gate-to-gate abatement is considered. These are higher than the "cost of capture" and "cost of CCS" seen in section 3.1, as more CO_2 is captured (619 kg/t cement) than is abated (505 kg/t cement), as steam provision for CO_2 capture also generates CO_2 that is captured. In contrast, expanding the system boundary to also consider CO_2 removed by biomass—without considering any other impacts outside the cement plant— results in the lowest avoidance cost estimates, 60% lower than the gate-to-gate estimates.

Expanding the system boundaries from "gate-to-gate with CO_2 neutral biomass" to "cradle-to-grave, CO_2 only" increases avoidance costs by 24%, though CO_2 emissions accounted for in the bioCCS system quadrupled, from 113 kg to 460 kg CO_2/t cement. In contrast, including the global warming potential of biogenic CO_2 and other greenhouse gases also increases cradle-to-grave avoidance costs by 24% from the CO_2-only metric even though the estimated net CO_2eq of the bioCCS system increases by only half as much, 223 kg CO_2eq/t cement from the CO_2-only metric. This apparent incongruity is because the change in CO_2 avoidance cost is not linked with absolute net CO_2(eq) emitted but with the difference in emissions between the abated and unabated system. Thus, it does not necessarily reflect the magnitude of changes in accounted absolute emissions or removals.

The avoidance costs discussed above consider near-term estimates for both costs and abatement potential. The few other literature estimates for avoidance costs bioCSS-in-industry typically consider Nth-of-a-kind costs paired with abatement potential estimates that consider present-day efficiencies and background systems [e.g., 8, 9, 11]. This creates a "tomorrow's technology today" scenario, which is not necessarily intuitive to interpret, as it can both underestimate the avoidance cost of near-term implementation and overestimate the avoidance cost of future implementation.

Table 8b shows the avoidance cost estimates using NOAK costs and near-term abatement potential, which are 13-30% lower than the corresponding FOAK estimates, depending on which costs are included. As the cost scaling primarily effects capital costs, the impact is lower for cases that do not fully include annualised capital costs. In contrast, these estimates are 13-27% and 20-40% higher than those shown in **table 8c** and **8d** respectively. These contain avoidance cost estimates using NOAK costs and abatement potentials that include projections of increased efficiencies of cement production and CO_2 capture and decarbonisation of electricity and transport, based on a conservative and optimistic scenario of future technological development.

Though the net CO_2(eq) of the optimistic scenario is lower than those of the conservative scenario (see appendix), the estimated CO_2(eq) abated is also lower. Partly, this is because the unabated system also has lower estimated net CO_2 in the optimistic scenario. It is also due to the phenomenon of "inefficient bioCCS"; since the clinker kiln optimistic scenario was assumed to be more efficient and therefore require less (bio)fuel, it resulted in less CO_2 being removed from the atmosphere from biomass production [51]. While the optimistic scenario has lower overall resource use, this is not embodied in either the metric of net CO_2(eq) or CO_2(eq) avoidance cost.

3.4 Accounting for virtual abatement

The model in this chapter only accounted for direct, physical emissions and removals of greenhouse gases in the bioCCS system. However, estimates of abatement potential and avoidance cost sometimes consider changes in emissions outside the system of consideration, such as those that are assumed to be displaced by the use of a (by)product from the system, often referred to as "avoided emissions" [e.g., 9, 10, 19, 52–54]. A common example is to assume that excess electricity cogenerated at an industrial production site replaces electricity produced by the grid and therefore the grid-average emissions of that amount of electricity is deducted from that system's net CO_2.

Avoided emissions are typically accounted for by subtracting them from the net CO_2 in the same manner as for physical removal of atmospheric CO_2. However, avoided emissions do not represent a physical reduction in atmospheric CO_2, but rather a virtual reduction in the CO_2 accounted for within the system of consideration. When such virtual flows are accounted for in the same metric as physical flows, it can lead to a "negative" net CO_2(eq) estimate without physical removals of CO_2 exceeding physical emissions [55 (**chapter 2**)]. This can be particularly confusing for technologies such as bioCCS, as it can lead to apparent negative emissions without physical negative emissions occurring. Therefore we advocate that avoided emissions should always be separated from the net CO_2(eq) metric for physical emissions and removals.

When such virtual abatement results from the sale of a by-product, it also adds complication to CO_2(eq) avoidance cost, as it changes both the estimate of abatement potential and net cost. Care should be taken to align the assumptions used for abatement and cost of these virtual flows. Returning to the example of excess cogenerated electricity, if it is assumed to displace grid-average generation, then grid-average pricing should be assumed. If it is assumed to be sold at a premium, or receive a credit, for being low-carbon, then it should also be assumed to displace comparable low-carbon electricity. Similarly, for electricity in particular, it should be considered whether the profile of the cogenerated electricity would be more likely to replace constant base load or variable peaking electricity generation and apply the costs and emission factors appropriate for that type of generation. Otherwise, inconsistent assumptions can further decrease the accuracy of the avoidance cost estimates.

4 Conclusions

BioCCS is a complex carbon-intensive technology system involving the permanent removal of CO_2 from the atmosphere. The primary goal of bioCCS is to result in "negative emissions", a net decrease in atmospheric CO_2. The use of bioCCS in carbon-intensive industries has the potential to allow for "CO_2(eq) neutral" or "CO_2(eq) negative" production, if the negative emissions produced via bioCCS is sufficient to compensate for CO_2 emitted in the life cycle of the industrial product. Clear and comprehensive metrics of the abatement potential and cost of bioCCS are needed to make informed decisions of when bioCCS is an effective abatement option and when can it result in negative emissions.

In this chapter, we evaluated the the case of bioCCS integration into cement production under different system boundaries to understand the impact of these modelling choices on estimates of net greenhouse gas emissions and costs. Therefore, the model of the cement and bioCCS system was held static and only the boundaries of the system of estimated greenhouse gas emissions and cost components were varied, based on boundaries seen in bioCCS literature. Depending on the system boundaries considered, estimates for net greenhouse gas emissions for a near-term retrofit of bioCCS into a cement plant ranged from -660 to 16 kg CO_2eq/t cement; cost estimates ranged from 87 to 162€$_{2017}$/t cement; and CO_2(eq) avoidance cost from 68 to 321€$_{2017}$/t CO_2(eq) abated.

In the case of unabated coal-based cement production, 72% of all emitted greenhouse gases occurred at the cement plant itself. However, for the bioCCS case, 82% of emissions—and all CO_2 removals—occur outside of the cement plant gates. Ignoring CO_2 removal from biomass can lead to overly modest estimates of abatement. In contrast, if CO_2 removals are considered without also considering the emissions from supply chains of biomass and other inputs, the net CO_2 estimate can appear to be deeply "net negative", even though the full system, accounting for upstream and downstream emissions of CO_2, other greenhouse gases, and the global warming potential of biogenic CO_2 has a positive net CO_2(eq). Additionally, as the net CO_2 metric contains both emissions and removals and does not consider CO_2 stored, it obscures that the bioCCS case is more carbon intensive than the unabated case.

Relative abatement, as used in CO_2 avoidance cost estimates, further obscures the emission intensity of both the bioCCS and unabated cases, and small change in avoidance cost can hide large changes in net CO_2(eq), as it only the difference between the unabated and bioCCS systems is considered. Avoidance cost can also obscure misalignment between cost and abatement estimates, such as when "Nth of a kind" cost estimates are paired with a "present day" abatement estimate, leading to an underestimate of near-term avoidance cost and, potentially, an overestimate of future avoidance cost, if the efficiencies of the technology and background systems improve over time.

To increase the comparability and usefulness of bioCCS avoidance cost estimates, we propose the following guidelines to ensure that estimates maintain a minimum level of completeness and transparency and align with the nature of bioCCS as a potential negative emission technology:

1. Estimates of abatement potential should include emissions throughout both the chain of biomass production and CO_2 transport and storage. As the crux of bioCCS the removal and permanent storage of atmospheric CO_2, the full impacts of both these processes must be included.
2. Only physical emissions and removals of greenhouse gases should be included in the "net CO_2(eq)" metric of bioCCS; virtual abatement, such as avoided emissions from by-product sales, should always be accounted for separately. This prevents the potential appearance of "net negative CO_2(eq)" without physical net removal of CO_2.
3. The fate of the captured CO_2 should always be explicitly stated and estimates of bioCCS costs must include the transport of CO_2 to permanent storage, as this is a fundamental component of bioCCS's abatement potential.
4. Emissions of other greenhouse gases, and the global warming potential of long-rotation biomass should always be explicitly treated. If they are excluded from the

study, this should always be mentioned, and the conclusions should be limited accordingly.

5. The temporality of the study should be explicitly stated, with costs and abatement potentials both aligned to the timeframe considered.
6. Assessment of "carbon neutral" industrial production should also include the full life cycle emissions of the industrial product considered.

Applying these guidelines to the bioCCS case in this chapter, the range of avoidance cost estimates would shrink from 48-321€$_{2017}$/t CO_2(eq) avoided to 157-193€$_{2017}$/t for near-term estimates, depending on which greenhouse gases are considered, and to 89-107€$_{2017}$/t for longer-term estimates depending on the greenhouse gas and future technology scenario considered.

Furthermore, given the limitations of single-point metrics of net CO_2(eq) and CO_2(eq) avoidance cost, we propose that studies on costs or abatement of bioCCS always also provide clearly decomposed metrics. For costs, we recommend that the cost of CO_2 capture, transport, and storage be presented separately—as is also recommended by [38]—as well as the cost of fuel switching, if relevant. These should be presented prior to the inclusion of any assumed taxes, subsidy, credit, or by-product sales to clarify the technological cost from assumptions of broader economic circumstances. For emissions, we recommend the independent presentation of on-site and off-site CO_2(eq) emissions; CO_2 removals by biomass; CO_2 permanently stored; and virtual abatement of CO_2(eq). This will allow for easier comparison between studies as well as a clearer assessment of the carbon intensity of bioCCS systems, which is obscured in the net CO_2(eq) metric.

BioCCS, as a negative emission technology, is intended to reduce the concentration of greenhouse gases in the atmosphere. However, whether bioCCS results in net removals depends on the specific combination of technology, biomass, logistics, and efficiencies in a given system. The emissions and removals of greenhouse gases of this system occur regardless of whether we chose to measure them. Strategic choices in system configuration—the type of biomass use, the method and distance of transport, the efficiency of CO_2 capture or the industrial production, the system of electricity generation—can decrease the net emissions of a bioCCS system. Similarly, technological choices and technological learning will reduce costs of implementation. However, without comprehensive, transparent, and comparable estimation, it is not possible to understand the significance of those choices.

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Appendix

Abatement potential of future cement product scenarios

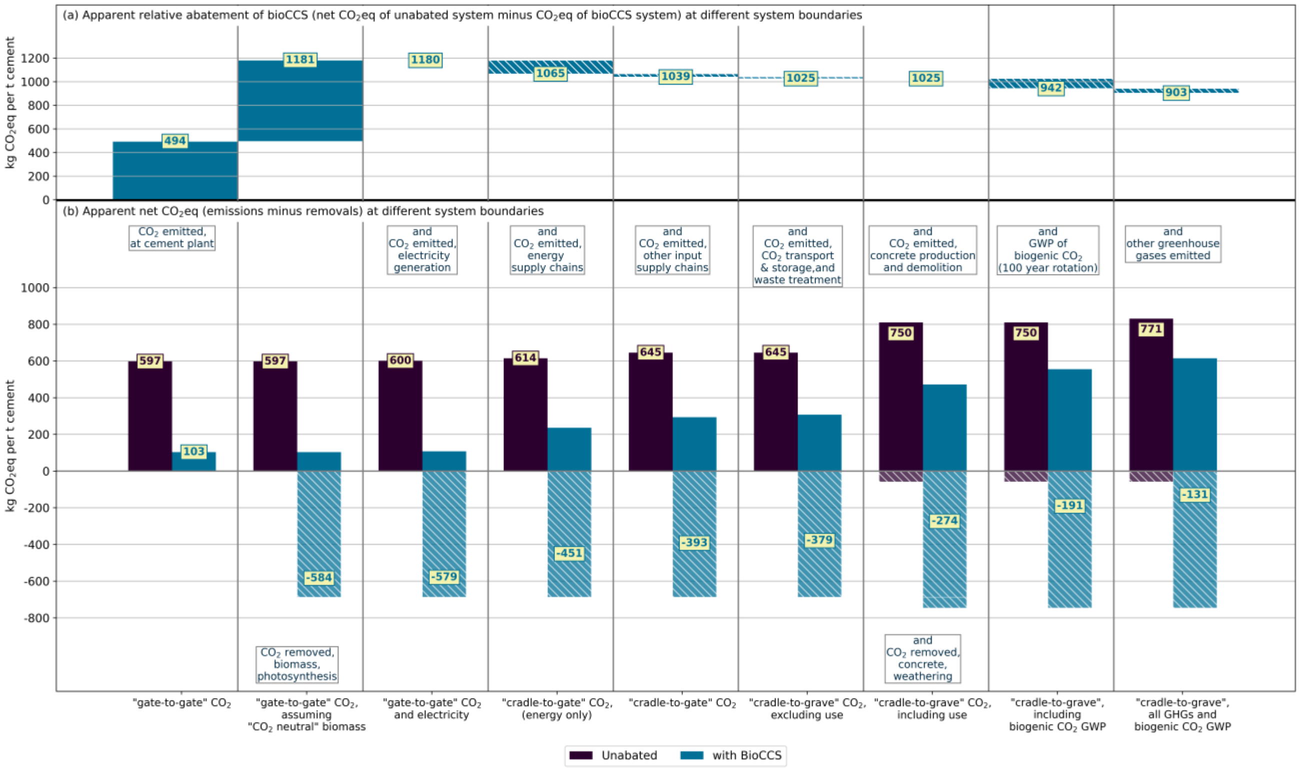

Figure 1. Net CO_2 of future cement production from different system boundaries, conservative scenario [1]

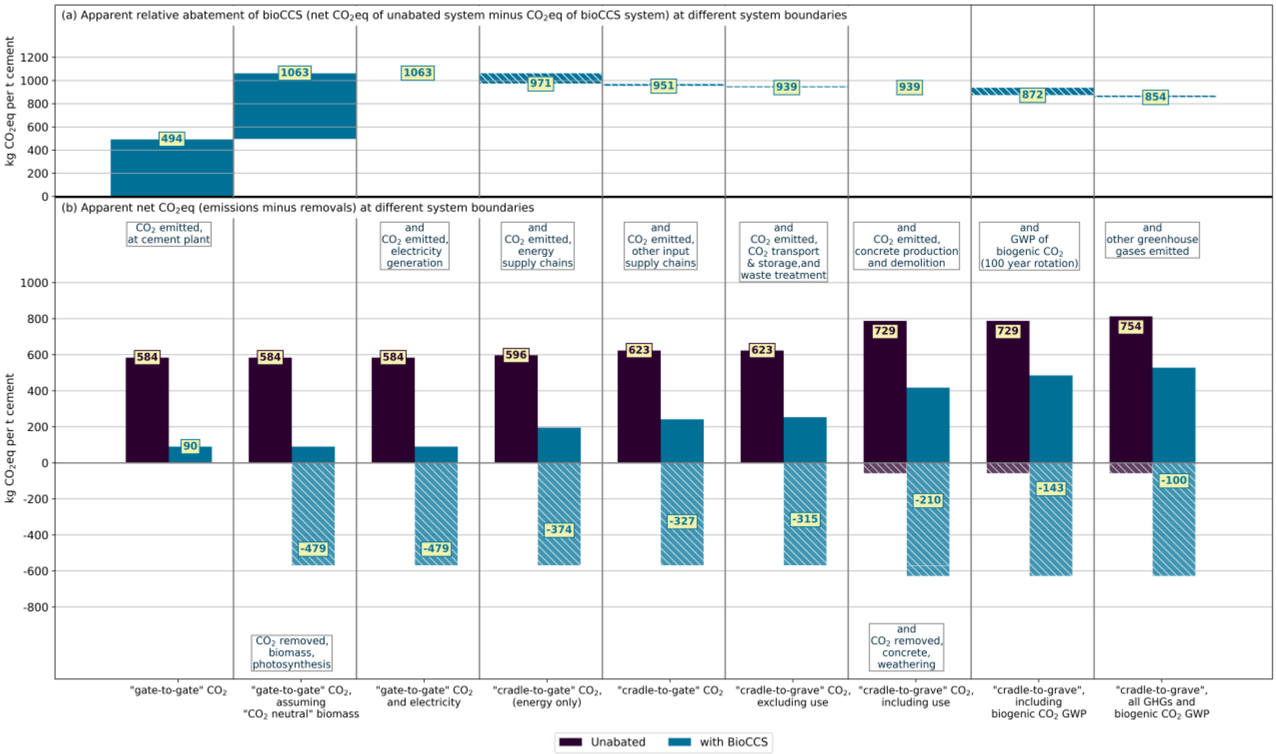

Figure 2. Net CO_2 of future cement production from different system boundaries, optimistic scenario [1]

Model parameters

Table 1. Process model parameters

Parameter	Qty	Unit	Data source
MEAL GRINDING, ball mill			
Limestone demand	745	kg/t meal	[2]
Clay demand	135	kg/t meal	[2]
Iron ore demand	23	kg/t meal	[2]
Bauxite demand	33	kg/t meal	[2]
Clay demand	3	kg/t meal	Assumed (to close mass balance)
Electricity demand	12	kWh/t meal	[3]
Transport of raw materials, by train	200	km	Assumed
CLINKER PRODUCTION, short dry kiln with precalciner and preheater			
Raw meal demand	1.57	t/t clinker	
Calcination efficiency	100	%	Assumed
Thermal energy demand	3.3	GJ/t clinker	[4]
Electricity demand	23	kWh/t clinker	[3]
CEMENT MIXING, roller mill			
Clinker demand	737	kg/t cement	[5]
Gypsum demand	50	kg/t cement	Assumed
Fly ash demand	213	kg/t cement	Assumed
Electricity demand	16	kWh/t clinker	[3]
CO_2 CAPTURE, monoethanolamine solvent absorption			
Capture Efficiency	90	% of CO_2 in	
Electricity demand	38	kWh/t CO_2 captured	[5]; [6]
Heat demand	3.2	GJ/t CO_2 captured	Assumed, based on literature review of MEA capture
Solvent makeup	1	kg/t CO_2 captured	Assumed, based on literature review of MEA capture
Water demand	473	kg/t CO_2 captured	[5]
CO_2 COMPRESSION, to 110 bar			
Compression losses	0	%	assumed
Electricity demand	106	kWh/t CO_2	[5]

Parameter	Qty	Unit	Data source
		compressed	
Heat demand	2.6	MJ/t CO_2 compressed	[5]
STEAM BOILER			
Efficiency	90	%	
Electricity demand	5	kWh/GJ steam	[7]
FLUE GAS CLEANING			
Electricity demand	11.4	kWh/t CO_2 in flue gas	[8]
Water demand	9.2	kg/t CO_2 in flue gas	[8]
NH_3 demand	6	kg/t CO_2 in flue gas	[8]
NaOH demand	0.1	kg/t CO_2 in flue gas	[8]
CaCO3 demand	1	kg/t CO_2 in flue gas	[8]
CO_2 TRANSPORT BY PIPELINE AND INJECTION TO GEOLOGIC STORAGE			
Losses	1	% of CO_2 stored	assumed, based on [2] and [9]
Transport distance, onshore	100	km	assumed
Transport distance, offshore	10	km	assumed
Electricity demand, transport	3	kWh/t CO_2 transported	[10]
Electricity demand, storage	7	kWh/t CO_2 stored	[11]
CHARCOAL PRODUCTION, Missouri-style kiln			
Timber demand	2.7	t (dry mass)/t charcoal	[12]
CO_2 emissions	540	kg CO_2/t charcoal	[12]
CH_4 emissions	1022	kg CO_2eq/t charcoal	[12]
Carbon efficiency	69	% of C in wood in charcoal	[12]
NATURAL CARBONATION			
Service life of concrete	50	years	assumption
Exposure conditions	Outdoors, Exposed to rain; Indoors, Covered		assumption
Ks	1.76, 4.84	mm/sqrt(year)	[13], Annex BB.1
Surface area	5, 5	m^2	assumption

Parameter	Qty	Unit	Data source
Degree of carbonation	0.85, 0.4	%	[13], Annex BB.1

Life cycle inventory emissions

Upstream CO_2	CO_2 only	Total GHGs (CO_2eq, 100year GWP)	Unit	Data source
Aggregate	5.1	5.2	kg CO_2eq/t	Gravel, round {CH},market for gravel, round, [14]
Ammonia	2535	2723	kg CO_2eq/t	Ammonia, anhydrous, liquid {RER},market for ammonia, anhydrous, liquid, [14]
Bauxite	25.7	26.7	kg CO_2eq/t	Bauxite {GLO},market for bauxite, [14]
clay	9.6	10	kg CO_2eq/t	Clay {CH},market for clay, [14]
Coal	168	358	kg CO_2eq/t	Hard coal {Europe, without Russia and Turkey},market for hard coal, [14]
Concrete, production	132	132	kg CO_2eq/t	Concrete, normal {CH},unreinforced concrete production, with cement CEM II/B (excluding cement production)
Concrete, demolition	8.3	8.3	kg CO_2eq/t	Waste concrete {Europe without Switzerland},market for waste concrete, [14]
Electricity, direct	390	*n.a.*	kg CO_2eq/MWh	[15]
Electricity, upstream	26	57	kg CO_2eq/MWh	market group for electricity, medium voltage RER, [14] , minus direct CO_2 intensity of NL 2017
Gypsum	7.2	7.6	kg CO_2eq/t	Gypsum, mineral {RER},market for gypsum, mineral, [14]
Iron ore	47	49	kg CO_2eq/t	market for iron ore, crude ore, 46% Fe GLO, and Iron ore, crude ore, 63% Fe {GLO},market for iron ore, crude ore, 63% Fe, [14] (50/50 split)
Limestone	4.8	5.1	kg CO_2eq/t	Limestone, crushed, washed {CH},market for limestone, crushed, washed, [14]
Monoethanolamine	2878	3254	kg CO_2eq/t	Monoethanolamine {GLO},market for, [14]
Monoethanolamine disposal	1956	1975	kg CO_2eq/t	Spent solvent mixture {Europe without Switzerland},treatment of spent solvent mixture, hazardous waste incineration, [14]
Natural gas	285	458	kg CO_2eq/t	Natural gas, high pressure {Europe without Switzerland},market group for, [14]
Sand	10.9	11.4	kg CO_2eq/t	Sand {RoW},market for sand, [14]
Sodium hydroxide	1212	1337	kg CO_2eq/t	Sodium hydroxide, without water, in

Upstream CO_2	**CO_2 only**	**Total GHGs (CO_2eq, 100year GWP)**	**Unit**	**Data source**
				50% solution state {GLO},market for, [14]
Timber, for charcoal	44	47	kg CO_2eq/t	Cleft timber, measured as dry mass {Europe without Switzerland},market for, [14]
Transport, lorry	0.1	0.1	kg CO_2eq/tkm	Transport, freight, lorry >32 metric ton, euro6 {RER},market for transport, freight, lorry >32 metric ton, EURO6, [14]
Transport, pipeline, onshore	0.05	0.06	kg CO_2eq/tkm	Transport, pipeline, onshore, long distance, natural gas {RER},market for transport, pipeline, onshore, long distance, natural gas, [14]
Transport, pipeline, onshore	0.05	0.06	kg CO_2eq/tkm	Transport, pipeline, offshore, long distance, natural gas {RER},market for transport, pipeline, offshore, long distance, natural gas, [14]
Transport, rail	0.05	0.05	kg CO_2eq/tkm	Transport, freight train {Europe without Switzerland},market for, [14]
Water	0.3	0.4	kg CO_2eq/t	Tap water {RER},market group for, [14]
Wastewater, disposal	0.3	0.4	kg CO_2eq/t	Wastewater from concrete production {CH},market for wastewater from concrete production, [14]
Wood pellets	695	709	kg CO_2eq/t (dry)	Wood pellet, measured as dry mass {RER},wood pellet production, [14]
Infrastructure use - charcoal kiln	5.1		kg CO_2eq/t	!!extracted from...
Infrastructure use - pellet plant	2.4	2.5	kg CO_2eq/t	extracted from Wood pellet, measured as dry mass {RER}, wood pellet production
Infrastructure use - Cement production	3.2	3.2	kg CO_2eq/t	extracted from Cement, alternative constituents 6-20% {Europe without Switzerland}, production
Infrastructure use - CO_2 capture	<0.001	<0.001	kg CO_2eq/t	[16]
Infrastructure use - selective catalytic reduction	<0.001	<0.001	kg CO_2eq/t	[16] (assumed to be the same as for CO2 capture)
Infrastructure use - steam boiler	0.5	0.5	kg CO_2eq/GJ	extracted from heat, district or industrial, other than natural gas {CH} heat production, softwood chips from forest, at furnace 1000kW

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Chapter 8:
Conclusions

1 Overview

This dissertation explored the decarbonisation potential of integrating negative emission technologies (NETs) into industry, focusing on biomass with carbon capture and storage (bioCCS). It began by investigating how the term "negative emissions" is used in literature (**chapter 2**), which highlighted the importance of system boundary choices in NET modelling. **Chapter 3** provided an overview of existing bioCCS-in-industry literature and identified sectors where bioCCS could be a viable decarbonisation strategy. **Chapters 4-7** explored the decarbonisation potential of integrating bioCCS into the production of ethanol, hydrogen, ammonia, urea (**chapter 4**), iron and steel (**chapter 5**), and cement and concrete (**chapters 6** and **7**). **Chapter 6** also considered the accelerated carbonation of concrete and addressed the timing of carbon dioxide emissions and removals. Finally, the costs of bioCCS-integrated cement production were explored in **chapter 7**, highlighting the impact of system boundary choices estimates of CO_2 avoidance cost.

This final chapter synthesises the findings of this dissertation, answers the research questions presented in **chapter 1**, reflects on the methods uses and outcomes obtained, and provides recommendations for future research on, and implementation of, NETs in industry.

2 Research outcomes

The overarching research question of this dissertation was

To what extent and under what conditions could negative emission technologies (NETs) play a role in the decarbonisation of the industrial sector?

This section presents first the answers to each of the subquestions asked and then to the main research question.

Subquestion 1: What criteria must negative emissions technologies meet to result in a net decrease of atmospheric greenhouse gases?

As shown in **chapter 2**, the term "negative emissions" has been used to refer to not only the removal of atmospheric greenhouse gases but also to the relative reduction in greenhouse gases emissions from one system to another ("avoided emissions"), or simply an amount of greenhouse gas stored, regardless of origin. Not all uses of "negative emissions" specified the removal of atmospheric greenhouse gas or considered whether the greenhouse gases would be reemitted later. Therefore, we proposed four minimum criteria to be used to evaluate whether a proposed “negative emission technology” could potentially fulfil the oft-stated goal of negative emissions: to reduce the net amount of greenhouse gases in the atmosphere. These criteria are:

1. *Physical greenhouse gases are removed from the atmosphere.*
2. *The removed gases are stored out of the atmosphere in a manner intended to be permanent.*
3. *Upstream and downstream greenhouse gas emissions associated with the removal and storage process, such as biomass origin, energy use, gas fate, and co-product fate, are comprehensively estimated and included in the emission balance.*
4. *The total quantity of atmospheric greenhouse gases removed and permanently stored is greater than the total quantity of greenhouse gases emitted to the atmosphere.*

However, these criteria are not necessarily sufficient to result in a reduction of global warming. For example, a system that results in negative CO_2 emissions can still increase global warming due to the emissions of other greenhouse gases, or the timing of removals after emissions, both of which are discussed further below.

Also of importance is ensuring that the origin and fate of all carbon are accounted for. This includes clarifying which carbon comes from fossil sources and which comes from atmospheric sources, and that only the latter is counted towards carbon removal. The carbon must then be followed until it returns to the atmosphere, is injected into geologic storage, or enters another long-term sink; including accounting for the final fate of carbon embodied in products. Furthermore, adding together physical and non-physical flows of carbon (e.g., “avoided emissions”) should be avoided, as this can lead to a “negative” global warming potential that does not represent the actual emissions and removals of the system.

The goal of negative emissions is, in a way, the same as the goal of emission reduction: that, ultimately, there will be less atmospheric greenhouse gases—and thereby less global warming—than there would have been otherwise. Therefore, it might seem counterproductive to make the distinction between “fossil CO_2 not emitted” and “CO_2 removed from the atmosphere”, as the latter includes prior emissions of CO_2. However, CO_2 reduction is not CO_2 removal, as CO_2 reduction cannot compensate for CO_2 that has already been, or will be, emitted. At the same time, CO_2 removal is not CO_2 reduction, which remains the necessary cornerstone of mitigating catastrophic climate change. CO_2 removal, as shown throughout the case studies in this dissertation, is resource, energy, and carbon intensive, and cannot compensate for the sheer amount of CO_2 we currently emit. Making the distinction between the reduction of greenhouse gas emissions and negative emissions is critical for understanding what role negative emissions can play in the pathway towards “net zero”.

Subquestion 2: To what extent could NETs reduce the CO_2 footprint of carbon intensive industrial processes?

Of the different NETs available, this dissertation focused primarily on bioCCS for its applicability to a range of carbon-intensive industries. **Chapter 3** identified three types of industries that were particularly suited for bioCCS: existing biobased industries, such as ethanol and paper, industries that are fossil fuel intensive, such as steel and cement, and chemical industries that involve CO_2 separation and whose feedstock could be biobased, such as ammonia or transport fuels. Each of these industry types was explored in this dissertation: ethanol in **chapter 4**, steel in **chapter 5**, cement in **chapter 6** and **7**, and hydrogen-ammonia-urea in **chapter 4**.

As summarised in **table 1**, bioCCS using biomass from sustainable agriculture and forestry and using pipeline transport to deliver captured CO_9 to dedicated geologic storage has the potential to allow many of the explored industrial processes to reach or exceed CO_2-neutrality, even when accounting for the life cycle emissions of the both bio-CCS and industrial production systems.

The cases where bioCCS did not result in CO_2-neutrality were those where either an insufficient amount of fossil fuel or feedstock could be replaced with biomass or an insufficient amount of CO_2 was capturable during production to compensate for CO_2 emitted elsewhere in the system. The urea system in **chapter** 4 is an example of the latter, with more carbon embodied in, and then released from, the urea product than can be captured during urea production. In an example of the former, while the vast majority of CO_2 from steelmaking is released at the steel plant, present-day blast furnaces rely on the structural properties of coke, so only a limited amount can be replaced by biomass. Thus, the blast furnace cases in **chapter 5** without modifications to the furnace did not result in negative emissions.

The potential for bioCCS to result in CO_2 negativity can be increased with other changes in the system of industrial production. As discussed in **chapter 6** and **7**, the co-capture of CO_2 emitted during steam generation for the CO_2 capture unit itself can further decrease the net CO_2 of the system, particularly if the steam boiler is biofuel-fired. For steelmaking, production technologies with less or no coke demand allow for higher biomass replacement rates. For cement systems, leaving demolished concrete exposed to air could allow it to absorb CO_2 from the atmosphere, recarbonating the lime that was calcinated during cement production.

However, CO_2 negativity by itself does not ensure a decrease in global warming. If the bioCCS system relies on long-rotation biomass, as the steel and cement systems in this dissertation assume, "carbon negativity" is delayed, and the temporary atmospheric residence of biogenic CO_2 has a warming effect until it is reabsorbed by regrown biomass. Furthermore, non-CO_2 emissions can be significant for biomass supply chains, such as methane emissions, particularly from charcoal production or nitrous oxide emissions, particularly, from biomass production. In the steel and cement cases where they were considered, these other global warming impacts meant that the bio-CCS systems were not "global warming negative", though they do result in a significant abatement of net global warming impact.

Beyond bioCCS, the natural and accelerated carbonation of concrete was also explored in **chapter 6**. When the CO_2 from limestone calcination is captured, the natural carbonation of concrete will absorb approximately 5-20% of the calcined CO_2 over the concrete's use life, depending on exposure conditions. Accelerated carbonation, which injects pressurised CO_2 into concrete, can result in a transfer of CO_2 from the atmosphere to long term storage—mineralised in concrete— if the CO_2 is biogenic or from direct air capture. However, in the model used here, the CO_2 emissions from the energy demand and injection losses of the accelerated carbonation process were greater than the amount of CO_2 estimated to be stored. The potential advantage of accelerated carbonation was not the storage of CO_2 in concrete but rather its potential to increase concrete strength, thus reducing cement demand and thereby CO_2 emissions associated with its production.

Table 1.Summary of decarbonisation potential of bioCCS-in-industry, high bioCCS cases, per unit

Industrial product	**Production technology**	**CO_2 only**			**also including other GHGs**		**Unit**
		Unabated	BioCCS,	BioCCS, including GWP of biogenic CO_2[1]	Unabated	BioCCS	
Carbon steel[2,3]	BF-BOF	2.43	0.12	0.51	2.57	0.87	t CO_2eq/t hot rolled coil
	BF-BOF with Top Gas Recycling	1.98	-0.05	0.08	2.09	0.41	
	Hisarna-BOF	2.16	-0.15	0.01	2.29	0.45	
	Midrex DRI-EAF	1.53	-0.52	-0.32	1.64	-0.22	
	ULCORED DRI-EAF	1.28	-0.33	-0.28	1.36	-0.19	
Portland cement (CEMI, 95% clinker)[3,4]	dry process clinker kiln with precalciner and preheater	0.88	-0.11	0.07	n.m.	n.m.	t CO_2eq/t cement
Portland concrete[4]		0.36	0.02	0.14	n.m.	n.m.	t CO_2eq/m^3 concrete
Portland concrete, with recarbonation of demolished concrete[4]		0.28	-0.07	0.06	n.m.	n.m.	
CEMII cement[3,4,5]		0.72	-0.33	-0.23	0.74	-0.09	t CO_2eq/t cement
CEMII concrete[4,5]		0.16	-0.04	-0.02	0.17	0.01	t CO_2eq/m^3 concrete
Bioethanol	fermentation from maize	1.21	-0.78	n.a.[6]	n.m.	n.m.	t CO_2eq/t ethanol
	fermentation from stover	0.07	-1.36	n.a.[6]	n.m.	n.m.	
Hydrogen	steam methane reforming	9.65	-5.79	n.a.[6]	n.m.	n.m.	t CO_2eq/t hydrogen
Ammonia	Haber-Bosch process	2.94	-0.97	n.a.[6]	n.m.	n.m.	t CO_2eq/t NH_3
Urea	Bosch–Meiser process	5.72	0.11	n.a.[6]	n.m.	n.m.	t CO_2eq/t Urea

n.m.: not modelled

1: assuming a rotation period of 100 years and a GWP of 0.44 kg CO_2eq/kg bioCO_2

2: Steel scenarios here contain upstream emissions of infrastructure use. “Other greenhouse gases” only included methane

3: Product use and disposal excluded

4: The reference cases for cement and concrete assumed a 100% coal-fired kiln.

5: The BECCS cases for CEMII cement/concrete assumed co-capture of emissions from the boiler that provides steam for CO_2 capture, unlike the other cases

6: Chemical production cases included only annual biomass

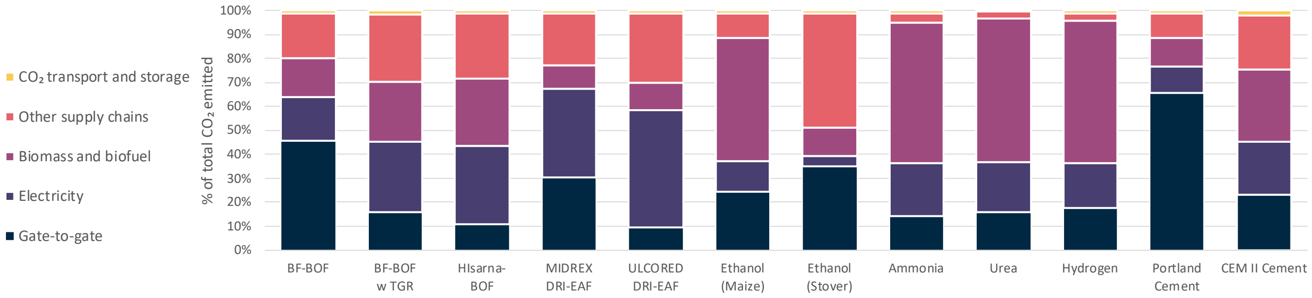
Subquestion 3: What aspects of integrating NETs into industrial processes have the greatest influence on their potential decarbonisation performance and costs?

Aspects influencing decarbonisation performance

The environmental performance of bioCCS is intimately tied to the conditions under which biomass is produced, transported, and transformed. Beyond the fossil CO_2 emitted throughout the cultivation, transport, and processing of biomass, methane emissions from the conversion of raw biomass to biogas, charcoal, or pellets can be a notable contributor to greenhouse gas emissions. For example, in the steelmaking cases, charcoal production in hot tail kilns were modelled to emit 1320 kg CO_2/t charcoal, or 230-330 kg CO_2/t steel. In the cement cases, the charcoal was assumed to be produced in Missouri kilns, which emit less than half of the carbon dioxide as hot tail kilns, but still emit 1020 kg CO_2/t charcoal, or 80 kg CO_2/t cement. **Chapter 7** also considered other greenhouse gases, the largest contributor of which electricity supply chains, accounting for 20 kg CO_2eq/t cement. And, as discussed above, the global warming potential of emitted biogenic CO_2 could also be significant in cases with long rotation biomass.

Beyond biomass, the emissions from other energy supply chains were typically the most influential factor on the net greenhouse gas emissions of bioCCS-in-industry, as is evident in **figure 1**. As the CCS models in this dissertation typically used an electrical CO_2 compression technology (as opposed to steam-powered compression), CO_2 emissions from electricity typically accounted for 20-40% of system emissions, with an electricity CO_2 intensity of 300-400 g CO_2/kWh. However, this means that as electricity grids decarbonise, thus lowering the CO_2 intensity of electricity generation, the net CO_2 of CCS should also decrease.

Whether bioCCS could fully compensate for the CO_2 emissions of industrial production, allowing for "CO_2-neutral" or "CO_2-negative" products also depended on the supply chains of the industrial process under consideration. For concrete, which requires the movement of large quantities of mineral and mineral by-products, the type and efficiency of transport accounted for over half of the non-biofuel upstream emissions shown in **figure 1**. For the model of cellulosic ethanol production in **chapter 4**, upstream enzyme production accounted for 30% of total system CO_2 emissions. And for steel production, while bioCCS was capable of leading to carbon-negative carbon steel, the CO_2 emissions resulting production of the alloys used in stainless steel were more than double the amount of CO_2 produced in the total carbon steel system itself.



Total CO_2 emitted (shown in graph)	1730	1050	1330	1480	990	1230	3280	1460	900	10890	600	450	kg CO_2/t product
Biogenic CO_2 GWP (100 year rotation)	390	90	100	200	50	0	0	0	0	0	180	90	kg CO_2eq/t product
Total other GHGs emitted	360	330	440	100	90	0	0	0	0	0	100	140	kg CO_2eq/t product
CO_2 removed via photosynthesis	1610	1090	1480	1900	1220	5320	6550	6070	3790	48440	710	770	kg CO_2/t product
CO_2 Stored	2060	1700	1860	1590	1220	1700	3460	2070	560	13600	760	910	kg CO_2/t product

Figure 1. Sources of CO_2 emissions for bio-CCS cases considered in this dissertation. Tabular results are in kg CO_2(eq)/t product

Aspects influencing cost

Chapter 3 reviewed recent technoeconomic studies on bioCCS-in-industry. Costs were highly variable across studies and industries, ranging from 13-388€$_{2020}$/t CO_2 avoided. This range embodies the variation in assumptions about biomass prices (0-15.4€$_{2020}$/GJ), CO_2 capture (3-30€$_{2020}$/t CO_2 for pure CO_2, 41-110€$_{2020}$/t CO_2 for amine-based capture of dilute CO_2), CO_2 transport (5-380€$_{2020}$/t CO_2), taxes on fossil CO_2 emissions, and subsides for CO_2 storage. The wide range of transport estimates, in particular, also embodies assumptions about who is responsible for providing CO_2 transport infrastructure. In several studies, it was assumed that CO_2 pipelines would be constructed by an external party and therefore the assumed transport costs were only "pay-for-use" costs, and ranged from just 5-17€$_{2020}$/t CO_2. If the infrastructure was included in the cost of the bioCCS system, its expense was dependent on both the volume and distance of CO_2 transported. The widest range was seen in estimates for sugarcane biorefineries in Brazil, mostly located several hundred kilometres from the offshore Campos basin, which was most commonly assumed as the storage site.

Chapter 7 considered the economic performance of a bioCCS-in-cement system. The primary contributor to the cost of bioCCS was the higher demand for energy and the higher price of biofuel compared to fossil fuel, which together increased energy costs from 14€$_{2017}$/t cement in the reference case to 75€$_{2017}$/t cement in the bioCCS case. The annualised capital expenses of the CCS system were also a major contributor, dominated by the CO_2 capture and compression system, accounting for 53€$_{2017}$/t cement for a near term "first of a kind" retrofit. Overall, the operating and capital costs of CO_2 capture and compression represented 85% of the total bioCCS-in-cement costs. However, the literature review in **chapter 3** shows that the CO_2 transport scenario can be highly influential on total system costs, particularly if the transport distances required exceeded several hundred kilometres, pipeline transport is not available, or if the amount of CO_2 transported is small.

In contextual terms, near term deployment of bioCCS-in-cement, using a first-of-a-kind costing estimate, was modelled to increase the cost of cement production from 33€$_{2017}$/t to 195€$_{2017}$/t. Based on an analysis by [1], this could translate into an 80-90% increase in the price of concrete and a 3-15% increase in the cost on construction, depending on the type of building.

Subquestion 4: How do modelling choices affect the perceived environmental and economic performance of selected industrial NETs?

Chapter 1 of this dissertation highlighted the necessity of inclusive system boundaries to determine whether a proposed negative emission technology results in the net removal of greenhouse gases from the atmosphere. In **chapter 3**, we see the particular importance of including not only upstream emissions of supply chains, but also downstream emissions of product use and disposal, particularly for carbon-containing products such as fuels and fertilisers. For example, if a bioCCS-in-urea system only considers production and the upstream supply chains, the system appears to be deeply negative at -623 kg CO_2/t urea. However, when emissions from urea decomposition—which occur regardless of whether they are accounted for—are included, the net CO_2 of the system increases to 110 kg CO_2/t urea.

Chapter 7 illustrates that inclusive system boundary choices are of particular importance for bioCCS systems, by assessing the abatement and costs of a static model of bioCCS-in-cement from the different system boundaries found in bioCCS literature. For the reference case of unabated cement production, 80% of all CO_2 emissions, and 70% of all greenhouse gas emissions occurred at the site of cement production. However, for the bioCCS, case less than 25% of CO_2 emissions (and less than 20% of total greenhouse gas emissions) occurred on-site. Including the CO_2 emissions from upstream supply chains and concrete production and use increases net CO_2 from -660 to -206 kg/t cement. Including the global warming potential of biogenic CO_2 from long rotation biomass and other greenhouse gases increased net emissions to 16 kg CO_2eq/t cement, no longer resulting in net negative emissions.

In the cost estimates, on-site costs dominated, with the largest cost of a near-term retrofit using “first of a kind” cost scaling being the marginal cost of capture (69€$_{2017}$/t cement), followed by the annualised capital costs of the CO_2 capture system (+53€$_{2017}$/t cement), and the marginal and capital costs of CO_2 pipeline transport and storage (+23€$_{2017}$/t cement). When combined with the wide range of estimated greenhouse gas emissions this create a “possibility space” of CO_2 avoidance cost estimates 68-321€/t CO_2 for the same bioCCS-in-cement system. This range helps explain the wide variation in CO_2 avoidance cost estimates seen in the literature review in **chapter 3**. While it is partly due to differences in technology choice, cost assumptions, system configuration and regional considerations, a major contributor is the fact that few of these studies assume the same system boundaries when calculating either cost or greenhouse gas emissions.

This dissertation also investigated the limitations of the standard single point metric of "life cycle net CO_2" in **chapter 6**, decomposing it into a curve of net emissions and removals over the life cycles of concrete, biomass, and CCS. This highlighted the potential for delay between CO_2 emissions, which typically occur over a short time span during production, and CO_2 removals, which occur as biomass is regrown, and depends on the rotation period of the biomass. This also showed that the single-point metric obscures that bioCCS results in more CO_2 emitted at the start of life than fossil-based production with CCS, and it takes over a third of the biomass rotation period for the bioCCS system to reach parity with the fossil CCS system. The metric also obscures the carbon-intensity of the system, as it does not account for CO_2 that is produced but not emitted.

To what extent and under what conditions could negative emission technologies (NETs) play a role in the decarbonisation of the industrial sector?

The integration of bioCCS into existing industrial production pathways can significantly reduce the net greenhouse gas emissions of steel, cement, ethanol, and fertiliser, potentially via retrofitting existing facilities with CO_2 capture and switching to bio-based fuels and feedstocks, thus avoiding stranded assets.

Whether bioCCS can allow for CO_2-neutral or CO_2-negative production depends on the specific characteristics of both the industrial production system and the bioCCS system. In particular, bioCCS is most effective when the majority of the emissions of the industrial system are emitted at the site of production where bioCCS is implemented. BioCCS cannot abate CO_2 that is emitted by the product itself, such as the CO_2 emitted by the use ethanol or urea. Likewise, if the supply chains are carbon-intensive, bioCCS at the production site will be insufficient to compensate for those upstream emissions, such as from fuel production, transport, alloying metals, or enzyme production.

BioCCS is carbon-intensive and substantially increases the amount of greenhouse gases emitted outside of the production site. Effective bioCCS requires a sustainable source of biomass, preferably fast-growing, long-term storage of captured CO_2, and minimisation of greenhouse gas emissions from biomass production and processing. The emission intensity of available electricity and transport options for both biomass and CO_2 also play a prominent role in determining whether bioCCS can result in negative emissions. As these, and other relevant sectors, decarbonise, bioCCS becomes more effective both by emitting less greenhouse gases and needing to compensate for fewer emissions from the system of industrial production.

Table 2. **Global decarbonisation potential of bioCCS-in-industry and resource use**

Industrial product	Production technology	Change from reference case[1] to full bioCCS case					Global scale				
		Net CO_2	Thermal energy demand	Electricity demand	Biomass demand	Biomass type	Current production[2]	Decrease in CO_2 emissions	Potential negative CO_2	Total additional electricity demand	Total additional biomass demand
		t/t product	GJ/t product	kWh/t product	GJ/t product		Mt/year	Mt/year	Mt/year	TWh/year	EJ/year
Carbon steel	BF-BOF with top gas recycling	-2.5	1.7	550	12	Timber (for charcoal and wood chips)	1341	-3324	-65.7	738	16
	Midrex DRI-EAF	-2.1	3.2	390	21		111	-228	-57.7	43	2
CEMII xement	Dry process clinker kiln with precalciner and preheater	-1.0	2.5	158	3	Timber (for charcoal and wood pellets)	886	-927	-288.8	140	3
Bioethanol	Fermentation from stover	-2.6	7.7	404	9		86	-221	-116.7	35	1
Hydrogen	Steam methane reforming	-15.4	43.1	5733	450	Agricultural Wastes (for biomethane via anaerobic digestion)	8[3]	-124	-46.3	46	4
Ammonia	Haber-Bosch process	-3.9	2.6	769	55		136[4]	-531	-131.8	105	8
Urea	Bosch–Meiser process	-5.6	1.6	435	35		81	-455	*n.a.*	35	3

1: Reference cases are the corresponding unabated cases from table 1 except for the BF-BOF with Top Gas Recycling, where the reference case is the unabated BF-BOF case, and stover bioethanol production, where the reference case is unabated bioethanol produced from maize.

2: BOF Steel: 2019 [4], DRI: 2019 [5], Cement: 2019 [6], Bioethanol: 2019 [7], Hydrogen: 2018 [9], Ammonia: 2019 [10], Urea: 2016 [11].

3: Estimated merchant hydrogen fraction of 10 Mt European and 87 Mt global total production.

4: Subtracting that needed for the estimated urea production.

BioCCS is also resource intensive. As an illustration, **table 2** imagines that one bioCCS configuration for each industry is implemented globally following the same assumptions used in this dissertation, it would reduce industrial CO_2 emissions by 6 Gt/year, including 0.8 Gt of "negative CO_2 emissions". Such a global implementation would require 36 EJ of biomass, which is approximately 65% of current global bioenergy production [2], and 1100 TWh of electricity, or 12% of present-day industrial electricity use [3]. However, this example assumes no optimisation of production or capture methods and is therefore likely to be conservative.

3 Reflections on methods and outcomes

In the course of this dissertation, I not only learned about negative emission technologies, but also about the process of doing research, the limitations of "single-factor" metrics, and many aspects of NET systems which I was unable, due to limits of time and scope, to consider. Some of these are presented here.

3.1 Building tools for comparative black box modelling

Industrial processes can be modelled at different levels of detail. At the most technical, the mass and energy flows for every chemical reaction in every piece of equipment are calculated, accounting for the specific conditions and compositions of the processes. This level of detail, while highly instructive, was not suited for the exploratory modelling conducted in this dissertation. Instead, this dissertation used "black box" modelling, which evaluates processes based on fixed ratios of inflows and outflows, one for each industrial unit process (e.g., the iron furnace, lime kiln, coke oven, CO_2 capture unit, steam boiler, &c). This simplification made it easier to focus on the energy and carbon carriers and other material inputs of the processes.

The most common method of creating black box model is via spreadsheets or hardcoded scripts that calculate each flow individually. Several software libraries do exist to facilitate the parameter optimisation of individual black box models, which was not the focus of this dissertation. These tools lacked the modularity needed to facilitate the comparisons of many cases, particularly when both the configuration of unit processes and coefficients of the calculations change.

Therefore, this dissertation also includes a bespoke software library for comparative black box modelling called blackblox.py. Unit processes are described in two spreadsheets, one for the calculations and one for coefficients that allowed for multiple cases to be defined, and a third specifies how the unit process are connected. Python scripts are then used to solve the models, exporting results to spreadsheets, and also generating diagnostic model diagrams. This modularisation allowed for rapid construction of many models, the reuse of existing unit processes, and the ability to generate results for many configurations and scenarios without having to retool each individual model parameter.

Blackblox.py has been released open access on GitHub as part of this dissertation, along with the data and models used in each of the preceding chapters. An introduction to the software, which does not require learning Python to use its basic functionality, along with information on where to find it, is provided in **appendix A**. I hope at least one of you will find it useful in your own research.

3.2 Limitations of single point metrics for bioCCS

As discussed throughout this chapter, the metric of "net CO_2(eq)" can obscure the choice of system boundary, delays between emissions and removals, and the carbon-intensity of bioCCS systems. While any study that relies on such a single-point metric suffers from similar limitations, the nature of bioCCS systems makes this reliance particularly problematic.

Foremost, bioCCS, and other negative emission technologies, involve both the emission and removal of greenhouse gases. In a fossil-based system, all contributions to "net CO_2" would be strictly additive[9], with a one-to-one correlation between change in net CO_2 and the CO_2 intensity of the system. In a bio-based system without CCS, CO_2 removals cannot exceed emissions, which limits the possible range of net CO_2 estimates to values above zero. In a bioCCS system, the net CO_2 metric can be either positive or negative and provides no information about the amount of carbon in the system. In **chapter 5**, blast furnace steelmaking the bioCCS cases with and without the addition of top gas recycling had net CO_2 estimates within 180 kg CO_2 of each other. However, the top gas recycling system was modelled to emit 700 kg CO_2/t steel less than the unmodified blast furnace system, which removes and stores more biogenic CO_2 due to its higher energy use. However, the carbon intensity of a bioCCS system is not revealed in the net CO_2 metric.

The obtuseness of the net CO_2 metric is further compounded when it is used as the denominator of the "CO_2 avoidance cost" metric. CO_2 avoidance cost ostensibly provides the change in cost per unit decrease of CO_2 intensity. However, the estimate of cost is also subject to variability in system boundary—which may not align with the boundaries used for the net CO_2 estimate—as well as its own set of uncertainties and assumptions.

The purpose of metrics such as "net CO_2" and "CO_2 abatement cost" is to provide a point of comparability that decision makers can use to assess possible options. However, between studies, these metrics are not typically directly comparable, a point that is rarely emphasised and may not be obvious to non-modellers.

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[9] assuming that only physical flows are being accounted for.

3.3 Considerations beyond the scope of this dissertation

This dissertation encompassed four years of research, which was sufficient to learn how insufficient four years is to understand systems as complex as negative emission technologies. Therefore, while this dissertation provided both new insights on the influence of system boundaries for modelling negative emission technologies and foundational research on the decarbonisation potential of bioCCS in specific industrial sectors, there are many important considerations that it could not cover.

Foremost among these is that global warming potential is only one of many impacts that a technology system has. This dissertation, like too many other studies on negative emissions, did not consider impacts of land use, water use, biodiversity loss, acidification, eutrophication, which are all particularly pertinent to technologies that rely on biomass. Nor was it able to consider issues of governance, employment, or social acceptability of increased biomass use or CCS.

Secondly, this dissertation did not encompass the optimisation of resource use or the technology options considered. Monoethanolamine was used as the primary CO_2 capture solvent considered due to the availability of data, but it is not the most efficient option commercially available. Likewise, the assumption of a 90% capture rate is a literature standard, but is fundamentally arbitrary, and higher capture rates are technically possible. The use of coppice forestry or dedicated fast-growing biomass crops were not considered, instead focusing on existing forestry options. Therefore, the estimates presented in this dissertation may be on the conservative side.

Finally, it was not the intention of this dissertation to consider whether bioCCS is the best method to decarbonise these sectors or, more broadly, to decarbonise the functions that these industrial products fulfil. For example, while the use of bioCCS in concrete production was compared to the production of concrete with only biomass, only CCS, or unabated, and with and without accelerated carbonation, it did not consider whether concrete is the best material to use to build a bridge or a house, or, even more broadly, whether the bridge or house in question needed to be built. However, these larger questions of what is best fit-for-purpose should be the foundation of change-focused decision making.

4 Recommendations

4.1 Recommendations for future research

As emphasised throughout this dissertation, and particularly in **chapters 1** and **7**, research on the decarbonisation potential of negative emission technologies must strive to be as inclusive as possible in its system design and transparent as possible in its reporting. In particular, estimates of greenhouse gas emissions or costs should be presented in a disaggregated manner, e.g., in addition to or instead of "cost of bioCCS", reporting the "cost of biomass use", "cost of CO_2 capture", "cost of CO_2 transport", and "cost of CO_2 storage". This will increase the ability of the reader to understand the scope of the study and where studies agree or differ most in design and results. Furthermore, avoided emissions should always be accounted for separately from physical emissions and removals in NET research. Stacking avoided emissions with physical emissions and removals can lead to a "negative" global warming potential that is not indicative of negative emissions.

Clear and comprehensive reporting of system boundary choices and assumptions is vital to avoid misinterpretation of results and is particularly critical as policies for negative emissions are established. Misunderstanding of when negative emissions occur could potentially lead to financial instruments or other policies that incentivise technologies that increase atmospheric greenhouse gases under the label of negative emissions. Preventing this requires that not only that the system of consideration is presented as clearly as possible, but also that the NET technologies are evaluated on comprehensive life cycle accounting of greenhouse gases, including the long-term fate of the removed CO_2.

Research on negative emission technologies in industry is still nascent and there is much work to be done. However, to let you reach the end of this dissertation, I will list only three topics for future research on industrial NETs, in addition to those implied above in "Considerations beyond the scope of this dissertation".

First, the representation of industrial bioCCS in integrated assessment modelling should be improved based on the growing research, particularly with regard to the non-CO_2 systemic impacts of bioCCS, and competition for sustainable biomass. To aid this, more research is needed on the role of industrial bioCCS in an economy with increasing demand for biomass and negative emissions but a limited supply of sustainable biomass. Furthermore, as bioCCS is resource and carbon intensive, evaluation criteria are needed to assess when the use of bioCCS could provide needed time for novel production technologies to develop and when does it prevent the deployment of low-carbon industrial production methods by extending the life of old technologies.

Second, this dissertation also focused on the geologic storage of captured CO_2 and did not consider the potential storage of CO_2 in products beyond the carbonation of concrete. While some research exists on the global warming potential of atmospheric carbon stored in products [e.g., 12], research is needed on specific implementations, such as the use of atmospheric CO_2 in long-lived plastics, or the use of biomass as a concrete aggregate (with or without accelerated carbonation), confronting the issues of storage (im)permanence and responsibility for product end-of-life.

Finally, research is needed on the feasibility of industrial NETs that account for more specific geographies, and particularly those outside of Europe and North America. This includes not only the local availability biomass, electricity, and potential CO_2 storage, but also infrastructure needs, political climate, and availability of finance, and designing the NET system to minimise burden-shifting of impacts.

4.2 Recommendations for policy and implementation

For those tasked with regulating and incentivising negative emission technologies, I caution that negative emissions technologies are not a panacea and cannot replace the rapid and radical reduction of greenhouse gas emissions. It will always be less resource intensive, less logistically complicated, and less risky to prevent the emission of greenhouse gases in the first place rather than to attempt to remove them from the atmosphere later.

The success of bioCCS is dependent on the availability, and acceptability, of CO_2 capture, transport, and storage, which will likely require substantial public investment. To incentivise bioCCS specifically, encouraging CCS in existing biobased industries is the most logical place to start. This would require either the taxation of biogenic CO_2 emissions and/or a credit for stored (biogenic) CO_2, as currently there is no mechanism to reward going beyond carbon neutrality.

When attempting to implement or regulate a negative emission technology system, the most important consideration should be comprehensive and continual accounting of emissions and removals of greenhouse gases that occur not only at the site of industrial production, but throughout the complete system of carbon removal, use, storage, and all associated supply chains. It should also not be assumed that negative emissions occur instantaneously, but timing of emissions and removals and permanence of storage should be taken into account. To minimise these temporal issues, the use of residual or sustainability grown short-rotation biomass should be prioritised over long-rotation forestry biomass.

With regards to industrial bioCCS in particular, the judicious use of “partial bioCCS”—where biomass and CO_2 capture are used only for the most carbon-intensive parts of production, e.g., on the blast furnace of a steel plant, could help provide deep decarbonation while minimising the complexity of technological change needed, and could be more effective than more widespread use of either biomass or CCS. However, the decarbonisation potential of bioCCS is highly sensitive to the specific system design, and also requires that infrastructure for CO_2 transport and storage is available. BioCCS is also expensive and will likely require governmental support for capital investment or operation, particularly for early implementation, as well as strict taxation of fossil emissions and incentives for stored (biogenic) CO_2 to encourage industries to decarbonise past the point of carbon neutrality.

The ongoing climate crisis is an “all hands on deck” situation for decarbonisation options. The judicious use of negative emission technologies has the potential to be a valuable asset to that arsenal, allowing for the compensation of residual emissions in an otherwise “net zero” society. However, negative emission technologies will never be a substitute for reducing emissions or for absolute dematerialisation. The only way for human society to become sustainable is for us to honestly confront the hard limits of our resource-constrained world.

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Appendix A: blackblox.py

1 What is blackblox.py?

Blackblox is an open source python3 library for constructing and solving “black box” process models created by S.E. Tanzer, with contributions for refinement and release from João Pizani Flor, a software developer in the Department of Engineering Systems and Services at the Faculty of Technology, Policy, and Management, at the Delft University of Technology. Blackblox was used to produce the process models used in **chapters** 4-7 of this dissertation. Its particular strength is allowing for rapid comparison of different scenarios of parameters. Input data can be stored in Excel workbooks or tab or comma delimited text files. Results are returned as dictionaries for use within python and written to Excel workbooks. Results can also be displayed on the terminal console.

The basic “block” of blackblox is the **UnitProcess**, which contains the relationship between the inflows and outflows of a process or subprocess under evaluation. This block can be connected with others to form a linear **ProcessChain**. Units and chains can be further connected in a linear or branching fashion to form a **Factory** model.

For each level of model, two main functions are available:

- `balance()`: which evaluates the model based on one scenario, given a quantity and a specific flow to balance on.

- `run_scenarios()`: which evaluates the model on multiple scenarios, returning comparative results in a single table.

Each of these return the results as dictionaries or Pandas DataFrames, which can be used for further process within Python. Results can be written to the console or saved to an Excel file.

Additionally, the **Factory**-level model has support for single-variable sensitivity analysis.

2 Unit Processes

Unit processes are the smallest "block" in blackblox. Each unit process has a set of inflows and outflows and a set of specified fixed-ratio (linear) relationships between the process flows. Then, given a quantity for one inflow or outflow, the quantities of the remaining inflows and outflows can be calculated.

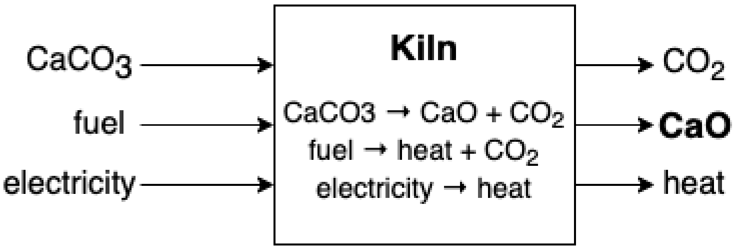

Figure 1. Example Unit Process

A unit process is defined by two tables, a **calculations table** that specifies relationships between flows, and a **variables table** that specifies the numerical values for the variable parameters used in the calculations. Extra data about unit process flows can be stored in **lookup tables**. All unit processes need to be listed in a **unit library** table, which tells blackblox where to find them.

Figure: Representation of a unit process

2.1 Calculations Tables

The calculation tables specify the relationships between flows in a unit process. Each row in the table:

- names two flows that have a relationship
- designates whether each flow is an inflow, outflow, or a flow internal to the process
- specifies the type of calculation that would generate the quantity of the second substance if the quantity of the first substance is known
- specifies the variable parameter (if any) used in that calculation that is the same name as a column in the variables table.

Table 1 provides an example of a calculation table for a simplified cement kiln.

Table 1. Example of blackblox calculation table for a clinker kiln

KnownQty	k_QtyFrom	UnknownQty	u_QtyTo	Calculation	Variable
clinker	outflow	CaO	tmp	Ratio	CaO_in_Clinker
CaO	tmp	CaCO3	inflow	MolMassRatio	1
CaCO3	inflow	clay	inflow	Remainder	CaCO3_in_Meal
CaCO3	tmp	CO2	emission	MolMassRatio	
clinker	outflow	fuel	tmp	inflow	fuelDemand
fuelDemand	inflow	energy_from_fuel	outflow	Combustion	combustEff
clinker	outflow	electricity	inflow	Ratio	elecDemand

- **KnownQty** (k) and **UnknownQty** (u) contain the two substances with a known relationship.
- **k_QtyFrom** and **u_QtyTo** specify the location of each flow.
  - **inflow** or **i**: inflow of unit process. Each inflow must be uniquely named and represent a unique calculation.

  - **outflow** or **o**: outflow of unit process. Each outflow must be uniquely named
  - **tmp** or **t**: a process-internal dictionary. This can be used for intermediate calculations and does not appear in inflows or outflows.
  - **emission** or **e**: outflow of unit process, which does not have to be unique. E.g., In our example table, both the calcination of limestone and the combustion of fuel will generate CO_2. By specifying CO_2 as the *u_QtyTo* location, the CO_2 from both calculations can be added together. Emission flows cannot be used to balance the unit process.
  - **coinflow** or **c**: inflow of unit process, which does not have to be unique. Coinflows cannot be used to balance the unit process.
- **Calculation** specifies the type of calculation performed between the two flows. The calculation types must be those available in the program's calculator library, some of which are described in **Table 2**.

- **Variable** specifies the column in the Variables Table where to find the value of the variable parameter used in the calculation. It is also possible to define substance names that are "lookup variables" that allow the substance to be defined in the variables table and also have properties defined elsewhere. By default, in blackblox, the flow name "fuel" is designated as a lookup variable, corresponding to the "fuelType" column in the Variables Table.

By default, flow names that begin or end with "energy", "heat", or "electricity" are assumed to be energy flows. All other flows are assumed to be mass flows.

Table 2. Calculation Types. (k = KnownQty, U = UnknownQty, V = variable)

Calculation Name	Variable Parameter Type (v)	Relationship (k:u)	Description
Ratio	float	k * var = u	Multiples the known flow quantity by the variable parameter
Remainder	float	k * (1 - var) = u	Multiples the known flow quantity by (1 - the variable parameter). Only works if the variable parameter is less than one.
Returnvalue	float	k = u	Returns the known flow quantity.
MolMassRatio	float or none	(Molar Mass of U / Molar Mass of K) * v = U	Multiples the known flow quantity by the molar mass ratio of the unknown flow to the known flow. Requires both flows names to be valid chemical formulas. If no value for the variable is provided, it defaults to 1.
Combustion	float	k (mass) * (MJ/kg of K) * v = u (energy) OR k (energy) / MJ / (1-v) = u (mass)	Using data from a separate “fuels” table, generates the energy provided by combusting a specified amount of fuel, or the fuel required to generate a specified amount of energy, as well as any specified emissions from the combustion. The variable parameter is the combustion efficiency, with the waste heat retuned as a separate flow.
		e.g. [k OR u] (energy) * (1-v) = wast heat (as emission)	Combustion also adds the heat remaining after the combustion efficiency is applied as “waste heat” to the dictionary.
		e.g. [k OR u] (mass) * (kg CO_2/kg fuel) = CO_2 (as emission)	Combustion can also look up emission factors from a separate “fuels” table (here shown for CO2, but any emission desired can be added).
Addition	none	k + k2 = u	Adds two known flow quantities together. Requires two columns to be added to the calculations table: `2nd Known Substance`, `2Qty Origin`
Subtraction	none	k - k2 = u	Adds two known flow quantities together. Requires two columns to be added to the calculations table: `2nd Known Substance`, `2Qty Origin`

2.2 Variables tables

The **variables table** provides the values of the variable parameters specified in the calculations table. There is one column for each of the variables named in the **calculation table**. The column name must be exactly the same as the specified variable name. Each row is a set of variable parameter values, identified by a **scenario** name.

Separating the values into their own table allows for the same unit process to be easily evaluated for multiple scenarios of parameter values (e.g., different production efficiencies or fuel types). (Note: rows whose scenarios begin with "meta" are ignored by blackblax and can be used for notes. This is also true for columns whose name begins with "meta".)

Table 3. Example of a blackblox variables table for a hypothetical clinker kiln.

scenario	fuelDemand	fuelType	CaO_in_Clinker	CaCO3_in_Meal	combustEff	elecDemand
meta-units	(mj /t clinker)	name	(t/t)	(t/t)	(%)	(mj/t clinker)
default	3	coal	0.65	0.8	1	0.1
EU-old	3.6	coal	0.75	0.8	1	0.2
EU-bat_bio	3	charcoal	0.65	0.8	1	0.1
EU-typical	3.2	coal	0.67	0.8	1	0.1

2.3 Lookup tables

Blackblox.py has the ability to get data about flows stored in other spreadsheets. By default, a "fuels" table is a lookup table that is recognised by the combustion calculation to get information about energy content and emission factors.

Table 4. Example fuels table

fuel type	LHV	CO2__fossil	CO2__bio	meta-source
meta-units	(GJ/dry tonne)	(t/t combusted)	(t/t combusted)	
heavy fuel oil	40.4	3.127	0	IPCC emission factor database
coal	25.8	2.4794	0	IPCC emission factor database
natural gas	48	2.6928	0	IPCC emission factor database
charcoal	29.5	0	3.304	IPCC emission factor database
coke	28.2	3.0174	0	IPCC emission factor database

2.4 Unit library

The unit library is a table that lets blackblox know what unit processes exist and how to identify them. It requires the columns:

- **id**: a unique identifier for the unit.
- **display name**: what the unit is called in the resulting output.
- **product**: the default product of the unit. This is the flow the unit will be balanced on if no other is specified.
- **productType**: ("inflow" or "outflow"). Whether the default product is an inflow or outflow of the unit process.

Table 5. Example unit library

id	display name	product	productType
clinker_kiln	kiln	clinker	outflow
cement_blender	blender	cement	outflow
meal_mixer	mixer	meal	outflow
electricity	electricity generation	electricity	outflow
gas_scrubber	SCR flue gas cleaning	flue gas	inflow

2.5 Storing data

By default, blackkblox looks for data in a `data/` subfolder of the working directory. Data can be in tab or comma delimited text files (.txt, .tsv, .csv) or in the sheets of an Excel workbook (.xls, .xlsx). If Excel workbooks are used, variable tables and calculation tables must be in separate workbooks.

The data folder needs to contain the unit process library, `unitlibrary.xlsx` (or .csv, .tsv, .txt) and any lookup tables, e.g. `fuels.csv`.

Blackblox will look for unit process data in the `data\` folder and its immediate subfolder. It identifies files that contain variable or calculation tables using a filename prefix. By default, unit variable data files need to begin with `var_` (e.g., `var_cement.xls)` and calculation data files need to begin with calc_(e.g., `calc_cement.txt`).

If using delimited text files to store unit process data, the file name after the prefix should be the same as the unit id listed in the Unit Library. For example, a process with the id `clinker_kiln` would require the files `var_clinker_kiln.csv` and `calc_clinker_kiln.csv`.

If using excel files to store unit process data, the excel file must begin with the file identifiers (e.g., `var_units.xlsx` and `calc_units.xlsx`) and the sheet names of each file must align with the unit ids from the unit library (e.g., `clinker_kiln` should appear as a sheet in each `var_units.xlsx` and `calc_units.xlsx` with the relevant variable table and calculation table, respectively).

So, a valid file structure for blackblox could look something like this:

```
project/
├─ config.yaml
├─ data/
│  ├─ unitlibrary.csv
│  ├─ fuels.csv
│  ├─ units/
│  │  ├─ var_units.xlsx
│  │  ├─ calc_units.xlsx
│  │  ├─ var_unitA.csv
│  │  ├─ calc_unitA.csv
```

2.6 Creating and balancing a unit process

Balancing a unit process calculates the quantity of all inflows and outflows of that unit process for a given set of variable parameters. To balance a unit process, the following information is needed:

- the quantity of one inflow or outflow. (if not specified, defaults to 1.0)
- the name of that inflow or outflow substance (if not specified, defaults to the product listed in the unit library)
- whether the substance is an inflow or outflow (if not specified, defaults to the productType listed in the unit library)
- the name of the scenario to use from the variables table (if not specified, defaults to `default`.)

After calculating all flows based on the user input, blackblox then checks for whether the mass and energy flows are balanced, and either raises an exception or, by default, creates an 'UKNOWN' balancer flow where needed. When processing combustion-type calculations, blackblox writes an "energy in combusted `fuelType`" flow to the inflows dictionary, to balance the energy of combustion. A similar balancer flow is provided for the combustion emissions.

2.7 Balancing a unit process in python

Unit Processes need to be created and then can be balanced. In python, the unit process can be created via:

```
blackblox.unitprocess.UnitProcess(unit_id)
```

So, to create and balance the clinker kiln of our example, you could create it using:

```
kiln = blackblox.unitprocess.UnitProcess(clinker_kiln)
```

and then balance it with:

```
kiln.balance(scenario='EU-typical', qty=100)
```

The results are returned as a dictionary and can be displayed in the console using `write_to_console=True` or written to an excel sheet using `write_to_xls=True`.

To compare multiple scenarios, the function `run_scenarios` can be used. By default, the results are written to an Excel file.

```
kiln.run_scenarios(scenario_list=['EU-typical', 'EU-old', 'EU-bat_bio'])
```

3 Process chain

A process chain is a linear set of connected unit processes, where an outflow of a preceding unit process is an inflow of the following unit process.

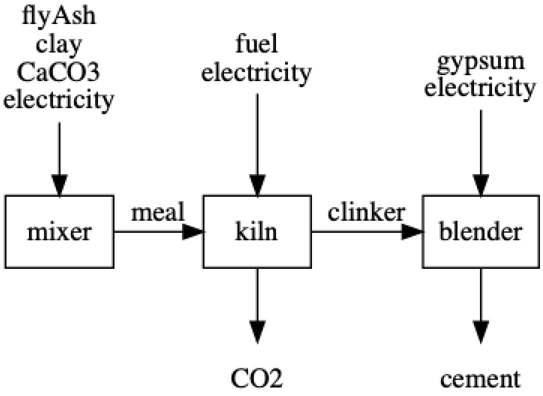

Figure 2. Example of a process chain

3.1 Defining a chain

A process chain is defined by a table with a list of unit processes with an inflow and outflow to each, where the outflow of a unit process must be the inflow into the next unit process, as shown in **table 6**.

Table 6. Example chain table

Inflow	Process_ID	Outflow
CaCO3	mixer	meal
meal	kiln	clinker
clinker	blender	cement

When the process chain is first used, an initialisation process creates each of the unit processes, if they do not already exist, and verifies that the inflows and outflows specified in the chain table exist for the corresponding unit processes.

A process chain can be defined by specifying the location of the chain table either in an Excel workbook or delimited text file using

```
chainName = processchain.ProcessChain(chain_data='path/to/chaindata.xlsx',
                                       xls_sheet='sheet name')
```

(`xls_sheet` is only required if using an Excel file with multiple sheets)

3.2 Balancing a chain

Balancing a chain calculates the quantity of all inflows and outflows of each unit process in the chain, either from first inflow to last outflow or from last outflow to first inflow. To balance a chain, the following arguments are used:

- `qty`: the quantity of one inflow or outflow somewhere in the chain
- `product`: the name of that inflow or outflow substance. (Attempts to default to product of last unit process if an outflow. If not, will attempt to default to the product of the first unit process, if an inflow.)
- `i_o`: whether the product is an inflow or outflow (defaults to the default product location)
- `scenario` the name of the configuration scenario to use from the variables table. (defaults to “default”)
- the name of the unit process in the chain, if the specified flow is not an inflow into the first process or an outflow of the last process.

All arguments besides the quantity can be optional if a default product can be determined.

Balancing a chain returns a dictionary with both the calculated inflows and outflows for each unit process, as well as the overall inflows and outflows. The results can also be written to the console or an excel file.

3.3 Generating a chain diagram

After a chain has been defined, a process flow diagram of the chain can be generated automatically using

```
processName.diagram()
```

This worka only if Graphviz is installed to your computer’s path. If not, this will fail with a hopefully useful error message.

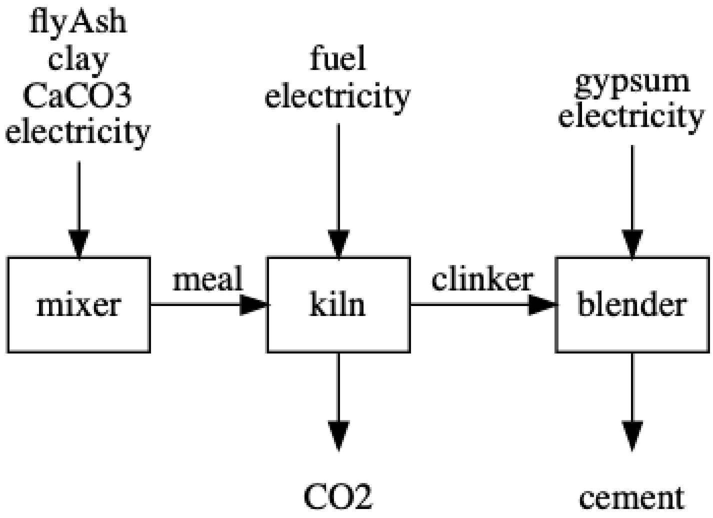

Figure 3. This ProcessChain diagram was autogenerated

4 Factory

A factory is a collection of one or more connected process chains, where the inflow of outflow of any unit process within any chain can be the inflow or outflow of any other chain. A factory has a single main chain, and zero or more auxiliary chains. By specifying an input or output quantity to the main chain, it is possible to calculate the inflows and outflows of all processes within the chain.

4.1 Defining a factory

A factory is defined by two tables: one that lists the chains (and their file locations) and another that lists the connections between the chains. Factory data can be in separate tab delimited text files or in a single excel workbook.

4.1.1 Factory chains table

This table specifies the location of the process chain data used in the factory, as well as the primary product and whether that product is an inflow or outflow of the chain.

The first chain of the chain list is assumed to be the main product chain, but otherwise it doesn't matter. The chains can either be in the same file, if an excel workbook, or in a separate file.

Table 7. Example of a Factory Chains Table

ChainName	ChainProduct	Product_IO	ChainFile	ChainSheet
cement	cement	outflow	here	Cement Chain

CO2capture	CO2	inflow	here	CO2 Capture
power	electricity	outflow	here	Power Chain

4.1.2 Factory connections table

The factory connections table specifies how the chains connect, including the origin process chain and unit process, the destination chain, the connecting product, and whether the product is an inflow or outflow of the origin and destination. The connections table is order dependent and the first chain should always be the main chain.

Table 8. Example of a Factory Connections Table

OriginChain	OriginProcess	Product	Product_IO_of_Origin	Product_IO_of_Destination	DestinationChain
cement	kiln	CO2	outflow	inflow	CO2capture
cement	all	electricity	inflow	outflow	power
CO2capture	all	electricity	inflow	outflow	power

If the destination is not the start or end of a chain, an optional `destination process` column can be used. This column is also used when specifying recycling connections. Currently two types of recycling are usable:

- *1-to-1 replacement*: allows for a recycle flow to replace a flow in another unit process, given that is a 1-to-1 correspondent and does not affect other flows in unit process.
- *energy replacing fuel*: allows for recycled energy to replace energy generated from a fuel in a unit process that has a combustion calculation. The emission outflows (e.g. CO_2) and inflow of oxygen are also recalculated.

If there is a greater quantity of the recycled flow than is used in the destination process, the remainder is treated as a normal output. A single flow can be recycled into multiple units, by specifying each connection as a new line in the table. If there is insufficient recycling flow to fully replace the existing flow, it is only partially replaced.

4.2 Balancing a factory

By default, the factory is balanced on a quantity of the product of the main ProductChain, and uses a single scenario of variables for the whole factory. `factory.balance()` takes the same parameters as `productchain.balance()`. Additionally, the factory can be balanced in other products from the main chain by passing `product`, `product_unit`, `product_io` parameters. A factory cannot be balanced on auxiliary chains.

Balancing a factory returns a nested dictionary with the inflows and outflows of the factory, every chain, and every unit process. By default, the results are saved to an Excel workbook with sheets for:

- Total factory inflows and outflows
- A matrix for all inflow from every unit process
- A matrix for all outflows from every unit process
- A list of all internal flows/connections, both within chains and
- Inflows for each chain unit process and chain totals (per chain)
- Outflows for each chain unit process and chain totals (per chain)

By default, the data is divided into mass and energy flows, if energy flow name signifiers are provided, with totals for both mass and energy flows.

4.3 Generating a factory diagram

After a factory has been defined, a process flow diagram of the factory can be generated. Graphviz must be properly installed for this to work. However, due to the limitations of the diagram rendering software, they are unlikely to be pretty.

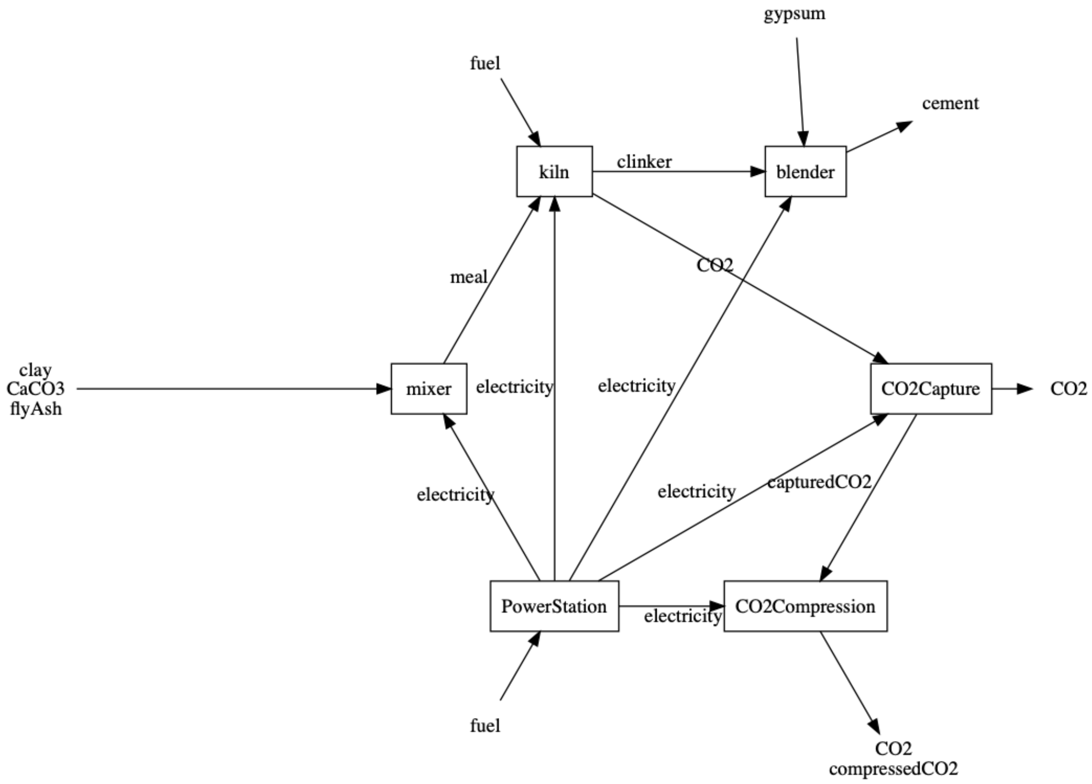

Figure 4. Example of an auto-generated factory diagram

4.4 Sensitivity analyses

A single-variable sensitivity analysis can also be run on a Factory model. The `factory.run_sensitivity()` function takes all the same parameters as `factory.balance()` as well as:

- `unit_name`: (str) the identifier of the **UnitProcess** containing the variable of interest.
- `chain_name`: (str) the **ProcessChain** containing the unit of interest.
- `variable`: (str) the variable of interest, which should correspond to a column in the unit's **Variable Table**.
- `variable_options`: a list of the variables options to be calculated.

The sensitivity analysis returns an Excel workbook with results for each variable option in columns.

5 Where to find blackblox.py

Blackblox.py is a free and open source library released under the GNU General Public License v3 (GPLv3). Blackblox is under active development. Currently available features and documentation may be different than what is provided in this Appendix.

- Full documentation for blackblox can be found at the project homepage: https://concoctions.org/blackblox/
- Source code and data are available on GitHub: https://github.com/concoctions/BlackBlox
- Blackblox can be installed using `pip install blackblox`. Information about the package can be found at PyPi: https://pypi.org/project/blackblox/
- Blackblox can be cited as: S.E. Tanzer. (2021). *BlackBlox.py*. Zenodo. https://doi.org/10.5281/zenodo.5800103

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This dissertation would not have been possible without the support of the wonderful community I've found at TPM, in Leiden, and beyond. It would take another dissertation, the contents of a pastry store, and probably some interpretive dance to comprehensively express my gratitude to all of you, so instead I will limit myself to a few words about a few of the key players in my PhD journey. For the dozens of others who I neglect to mention here, I only hope that I have expressed my gratitude and appreciation clearly in person over the years.

I did not plan to do a PhD. It was Andrea who first encouraged me to consider it during my master's thesis. And Andrea is the reason why I said yes. I knew nothing about the topic or whether I wanted to consider a career in academia, but I knew I wanted to spend four years working with her. I admired her sharpness, her attention to both detail and the big picture, and how she always challenged me with a criticality tempered with kindness. Four years later, my admiration and affection has only grown. Andrea, thank you for pushing me outside of my comfort zone, thank you for bringing me back down to earth when I float away on my ideas, thank you for always being my advocate, thank you for helping me find my voice, thank you for trusting me to do things my way, thank you for four years of insightful feedback and critical discussion and laughter.

A year into my PhD, Kornelis became my co-promotor. I benefitted greatly from his experience; he was a wonderful sounding board and always had suggestions of other perspectives to take and regularly provided me with useful resources.

I couldn't have asked for a better group of colleagues than those in the Energy & Industry section (and beyond) at TPM. It was a warm and supportive atmosphere, with enough space for both seriousness and silliness. And I reserve particular fondness for two office mates, Frank and Jessie. While a promotor transfer and a pandemic kept us a part for the past couple of years, I still couldn't have asked for better people to share a room with.

However, I'd be remiss not to mention the shining jewel of the third floor of TPM, Diones. Though she left us for bigger and better things in Applied Sciences, I am grateful that she was at TPM for so much of my PhD. Diones, thank you for always being there for a kind word or a laugh or a bit of gossip, as well as being so sharp and responsive whenever I actually needed help.

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My parents moved to Leiden halfway through my PhD. I can still hardly believe it. Having them nearby again has been a joy. Thank you, mom and dad, for always encouraging me to always try to be slightly better at whatever I want to do. Thank you for being the kind of people I want to be when I grow up.

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Curriculum Vitae

Samantha Eleanor Tanzer was born on the 14th of June 1988 in Arlington, Virginia, USA. She graduated from Walter Johnson High School in Rockville, MD, USA, in 2006 with a high school diploma and certificate of merit, and from Swarthmore College in Swarthmore, PA, USA in 2010 with a Bachelor of Arts in Economics.

Before obtaining her masters, Samantha worked for 5 years at an independent education software company in Boston, MA, USA, Upward Mobility, reaching the position of Director of Product Development. In 2017, she received a Master of Science in Industrial Ecology jointly from Delft University of Technology and Leiden University, with the honours of summa cum laude. She was also the 2017 Best Graduate of the faculty of Technology, Policy, and Management at Delft University of Technology for her master's thesis, "Plant + Boom = Boat + Vroom: A technoeconomic and environmental assessment of marine biofuel production in Brazil and Scandinavia using residual lignocellulosic biomass and thermochemical conversion technologies".

Samantha is also affiliated with the Kaji Aso Studio for the Arts in Boston, MA, USA, where she was a tea ceremony apprentice from 2012 to 2015 at the studio's traditional Japanese teahouse, the "House of Flower Wind". She is currently the program manager for the studio's virtual art events.

List of publications and conference appearances

Publications

Tanzer, S.E., Blok, K., Ramírez, A., 2021. Decarbonising Industry via BECCS: Promising Sectors, Challenges, and Techno-economic Limits of Negative Emissions. Current Sustainable/Renewable Energy Reports: Deep Decarbonisation: BECCS. doi: 10.1007/s40518-021-00195-3.

Tanzer, S.E., Blok, K., Ramírez, A., 2021. Curing time: A temporally explicit life cycle CO2 accounting of mineralization, bioenergy, and CCS in the concrete sector. Faraday Discussions, 230. doi: 10.1039/D0FD00139B.

Tanzer, S.E., Blok, K., Ramírez, A., 2020. Can bioenergy with carbon capture and storage result in carbon negative steel? International Journal of Greenhouse Gas Control, 100. doi: 10.1016/j.ijggc.2020.103104.

Tanzer, S.E., Ramírez, A., 2019. When are negative emissions negative emissions? Energy & Environmental Science, 12. doi: 10.1039/C8EE03338B.

Conference appearances

2021, November, COP26: Bellona Pavilion (Blue Zone), Glasgow, UK “Guiding principles for Carbon Dioxide Removal” (Panel member; virtual)

2021, April, Carbon Dioxide Utilisation: Faraday Discussion, Sheffield, UK “Curing time: A temporally explicit life cycle CO2 accounting of mineralization, bioenergy, and CCS in the concrete sector” (Oral, one of only 18 selected speakers; virtual)

2021, April, 15th International Conference on Greenhouse Gas Control Technologies, Abu Dhabi, UAE “Negative emissions in the chemical sector: lifecycle CO2 accounting for biomass and CCS integration into ethanol, ammonia, urea, and hydrogen production” (Oral; virtual)

2019, October 16th Netherlands Process Technology Symposium, Eindhoven, Netherlands “Carbon negative steel: Possibilities for negative emissions using bioenergy and CCS in current and future steelmaking technology” (Oral)

2018, May, 1st International Conference on Negative CO2 Emissions, Gothenburg, Sweden “The potential influence of negative emission technologies on the projected cumulative emissions of a decarbonized European industrial sector in 2030 and 2050” (Poster)