

Life Cycle GHG Emissions of Mineralization and Carbon Utilization Research Perspectives:  
Modelling and Stakeholder Elicitation

by

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## Abstract

A recent International Panel on Climate Change (IPCC) report indicates that a rise of global mean surface temperature by 1.5°C is expected to place increased risk on health, livelihoods, food and water security, economic growth, and climate systems. Mitigation options are available for the energy supply and demand sectors. Carbon capture utilization is one mitigation strategy. Carbon capture and utilization (CCU) technologies convert carbon dioxide into value added products and hence have the ability to mitigate greenhouse gas (GHG) emissions. The converted products include fuels, chemicals and materials. The prevalence of CCU technologies has gained considerable momentum in the last decade. Reports have placed the CCU potential to mitigate carbon dioxide (CO<sub>2</sub>) at 15% of global emissions. One such technology is known as mineralization. This involves a reaction between a metal oxide containing mineral and carbon dioxide to form insoluble mineral carbonates. The market for these products includes binders and aggregates for the construction industry.

This research is composed of three components. First, a comparison of the evolution and parallels between CCU and carbon capture and storage (CCS) is made in terms of research and industry. This was carried out using a bibliometric analysis to assess the state of the art in the CCS & CCU field, co-citations, co-authorships, temporal distribution and highly cited publications in CCS & CCU. The bibliometric analysis is combined with a meta-analysis of four topical areas- policy, technology, environment and economy, as well as industry project data. It was determined that CCS research generally shows greater prevalence than CCU. The policy field also shows fewer records compared to other fields. The CCS co-citation network of journals identified the following clusters: energy & fuels, chemical engineering, multidisciplinary sciences, and geochemistry & geophysics. Whereas, the CCU field has the energy & fuels, chemical engineering, and biotechnology & applied microbiology clusters.

The second part of this research used a life cycle GHG framework and model for CCU technologies to develop a consistent methodology to compare across pathways. The effectiveness of GHG reduction potential is often difficult to assess and compare due to differences in conversion processes, boundaries, product streams and other factors. These factors are addressed by this research. Specifically, the CCU technologies are assessed using four assessment metrics: kgCO<sub>2</sub>equivalents per kgCO<sub>2</sub>converted, kgCO<sub>2</sub>equivalents per kg or megajoule (MJ) of product, CO<sub>2</sub> avoided emissions and global emission reduction potential. Compared to other technologies and incumbents, CO<sub>2</sub> mineralization technologies offer the most significant GHG emission reduction potential, that being between -0.68kgCO<sub>2</sub>eq/kgCO<sub>2</sub>Converted and -0.35 kgCO<sub>2</sub>eq/kgCO<sub>2</sub>Converted. Additional sensitivity factors are examined for mineralization technologies including variation in energy sources, life cycle stages, reaction conditions, capture source, and replacement of cement in concrete.

The final part of this research examines the benefits and barriers of mineralization technologies in Alberta. Six stakeholder groups were interviewed: building and construction contractors, CCU technology developers, service providers, governmental organizations, cement and concrete companies, and industry associations. Qualitative research design methods were used in this research for the interviews and analysis. The results were coded to reveal five categories: individual and company background, relevant areas of work, areas of opportunity/improvement, benefits of mineralization, and risks and challenges of mineralization. The results from this analysis produced five themes: carbon emissions, technology development, competition/collaboration, policy/lobbying, and risk & uncertainty. Although mineral carbonation was recognized for its carbon emission reduction potential, risk with lack of

disclosure and challenges with policy structures were identified to be in the top 10 most abundant codes.

The results from this research contribute to modelling and stakeholder elicitation of mineralization. This work builds on the knowledge of parallels and evolution of CCU and CCS fields. It further provides valuable information on the modelling and sensitivities of the GHG emissions of mineralization technologies. Additional comparisons made to other CCU technologies show the importance of using consistent methodologies in LCAs. Moreover, stakeholder and industry elicitation add to the understanding of risks and challenges faced by these technologies.

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## **Chapter One: Introduction and Overall Objectives**

### **1.1 Introduction**

Carbon utilization by mineral carbonation has significant potential to contribute to reducing carbon dioxide emissions in the atmosphere. This is due to its potential to permanently sequester carbon dioxide in rock formations. The products of mineral carbonation can be used for a wide variety of markets. Assessing mineral carbonation technologies and comparing them to other CCU pathways from a life cycle assessment perspective will add to literature by postulating solutions with reduced environmental consequences. Mineral carbonation has the potential to mitigate climate change and reduce adverse effects of rising global average surface temperatures, which needs further investigation.

First, in order to identify parallels and evolution in the CCU and CCS field, a bibliometric analysis was conducted. This examined the statistical importance of books, articles and publications by determining relatedness to identify networks and mapping the development of new and existing fields of research. Few bibliometric papers have examined the broader low-carbon market (Du, et al., 2015), (Wang, et al., 2017) and technology investments (Yu, et al., 2016). Findings from literature also don't discuss CCS independently from CCU. Doing so identified research gaps and served as a starting point for the research concepts presented here. These findings were connected to the prevalence of industrial projects in the field and 4 topical areas of research in CCS and CCU. Following this, a full life-cycle assessment was conducted to compare mineralization technologies to incumbent technologies as well as other CCU technologies. A sensitivity analysis was also conducted showing hot spots for the technology. Additional recommendations to the LCA methodology were suggested to ensure fair comparisons across technologies and processes. An electronic excel based tool (Nishikawa, et al., 2020) was also developed to allow technology developers to assess their technologies against

others and change inputs for process improvements and energy efficiency. Finally, industry engagement of different stakeholders within the mineralization field was carried out to identify benefits and risks for the technology. This highlighted important gaps that were not captured in academic research alone. It also acted to propose solutions to address challenges in technology development as well as in policy supports, as investigated earlier in the bibliometric analysis as well.

## **1.2 Thesis Objectives and Approach**

Mineral carbonate construction products were examined in this research due to their reuse and market expansion capabilities. The long-term objectives of this research are to investigate and compare mineral carbonation technologies to postulate solutions to mitigate climate change and to develop consistent methodologies to assess these technologies.

The main research question is: What are the GHG emission impacts and technological challenges of mineral carbonation technologies?

The specific objectives are as follows:

### ***1.2.1 Objective 1. Performing Bibliometric Analysis to Compare Parallels and Evolution of Carbon Capture Storage (CCS) and Carbon Capture Utilization (CCU) Research***

Identifying gaps in the CCU research field was done by examining the field and comparing findings to similar well-established fields, in this case CCS. Bibliometric analysis examines the statistical importance of terms, sources and authors associated in an area of research. It does this by determining relatedness in publications, journals, authors, keywords and co-citations. Publications, researchers, keyword and journals can be examined for their co-authorship, co-occurrence, citation or co-citation networks. Co-occurrences refer to the number

of times that words occur together in the title or abstract. Whereas co-citation refers to the number of times that two documents are cited together in other documents. This technique illustrated the current state of the art of the field of research and connectedness. In this study, results from the bibliometric analysis are compared to CCS & CCU industry data. Moreover, a meta-analysis of 4 topical areas within is conducted (i.e. technology, policy, economy and environment) to illustrate the evolution and parallels in CCS & CCU research.

VOSviewer software was used in conjunction with the Web of Science Database to create:

1. Co-occurrences network of terms over time
2. Temporal distribution of CCS and CCU publications and terms
3. Co-citation network of journals, as well as citations and link strength to others
4. Findings from highly cited documents
5. Number of CCS documents, CCU documents, CCS facilities and CCU start-up companies
6. Co-authorship relationship of countries
7. Selected meta-review of papers for CCS and CCU topical areas organized by categories

An analysis of bibliometric data was done with the landscape of CCS & CCU industrial projects globally and a meta-analysis of 4 topical areas. The process was used to examine the state of the art of the CCS and CCU fields, and to compare findings with CCS and CCU research and industry for researchers, industry partners and policy makers.

### ***1.2.2 Objective 2. Determining Process Life Cycle GHG Emissions for Mineral Carbonation and Comparison with Other CCU Pathways***

A full life cycle assessment of GHG emissions in CO<sub>2</sub> equivalents was conducted for mineral carbonation technologies and compared with other CCU technologies. Specifically, this included defining and categorizing pathways to their appropriate subcategories. The goal and scope were defined as gate-to-grave, including CO<sub>2</sub> capture. A boundary diagram for the state-of-the-art mineral carbonation process was determined. The boundary of this study was gate-to-grave and provided a detailed analysis of the specific processes and identified potential hot spots as well. This includes the capture and conversion of carbon dioxide, the extraction and mining of raw materials associated with the CCU technology, conversion steps and product end use stages. Material and energy flows were identified for each unit process to determine GHG impacts. Representative cases were selected from literature variability and compared with company data to provide expert/industry elicitation. Rationales were provided for the selection of representative cases. The variability of multiple products of mineral carbonation were compared with different conventional incumbents. Specifically, carbonates can be stored geologically or used to replace products such as cement, concrete and aggregates for the construction industry (Intergovernmental Panel on Climate Change, 2005).

The different mineral carbonation pathways were assessed using four metrics of analysis:

1. Per kilogram product formed
2. Per kilogram CO<sub>2</sub> converted
3. Amount of CO<sub>2</sub> avoided compared to incumbents
4. Global emission reduction potential based on market sizes

Results from this analysis were compared among different mineral carbonation categories, to literature variability within the category, as well as to other CCU options to determine the CO<sub>2</sub> mitigation potential. A sensitivity analysis of all assessed parameters for the selected representative case was performed to determine parameters which impact GHG emissions most significantly.

### ***1.2.3 Objective 3. To Investigate the Benefits, Risks and Opportunities for Mineral Carbonation Technologies***

Mineral carbonation products were assessed for their potential to replace cement, supplementary cementitious material, utilized as additive and as novel cement and concrete. This analysis was conducted using qualitative methods, specifically, stakeholder interviews. A total of 6 stakeholder groups were identified in Alberta: CCU technology developers, governmental organizations, cement and concrete companies, industry associations, service providers, and building and construction contractors. An Alberta specific case-study was conducted on these mineralization technologies. This allowed for a concerted investigation since most standards are jurisdictional and may raise compliance issues/recommendations (The National Academies Press, 2019). The potential emission reduction that arises from these replacements were assessed. This objective investigated the barriers to development of these technologies and postulated solutions for policy makers and other stakeholders on how to address them. This process will assist in shaping potential policies for these products and the broader CCU products as well.

The following aspects were assessed for CO<sub>2</sub> mineralization products for the building material and construction industries:

1. Stakeholder knowledge and company background

2. Industry experience of technology parameters/limitations
3. Mineralization markets identification and deployment
4. Mineralization benefits and opportunities
5. Mineralization risks and challenges

### **1.3 Impact of Research**

Carbon utilization by mineral carbonation has significant potential to contribute to reducing carbon dioxide emissions. This is due to its potential to permanently sequester carbon dioxide in rock formations. The products of mineral carbonation can be used for a wide variety of markets. Assessing mineral carbonation technologies and comparing them to other CCU pathways from a life cycle assessment perspective will add to literature by postulating solutions with reduced environmental consequences. The bibliometrics assessment will contribute to better understanding the linkages within academia and industries of CCS and CCU. Additionally, qualitative analysis will highlight the benefits and challenges that these technologies face in industry. Mineral carbonation has the potential to mitigate climate change and reduce adverse effects of rising global average surface temperatures, which was further assessed in this research.

Aside from contributing to academia and researchers, this research will also assist industry, government, the public, and will allow stakeholders to better understand how CCU technologies, specifically mineralization can be used as a tool to mitigate GHGs. Industry stakeholders, including CCU technology developers, governmental organizations, cement and concrete companies, industry associations, service providers, and building and construction contractors, will be able to identify gaps in the technology development and further develop areas of opportunity. Government organizations will be able to determine appropriate policy and guidelines to address the challenges faced by stakeholders. All groups will also be able to



compare across the lifecycles of CCU pathways using consistent comparison methodologies to determine factors and pathways that produce the lowest GWP impacts when compared to incumbents and other pathways. Additionally, these groups will be able to identify the state of the art of the field to determine gaps as well as factors that assist in CCU and CCS technology development globally.

#### **1.4 Thesis Organization**

This thesis is organized into 6 chapters in which Chapters 3, 4 and 5 act as independent chapters written in paper format. Chapter 2 provides a brief introduction to the background of carbon capture utilization, carbon capture storage, mineralization, life cycle assessment and qualitative analysis techniques. Chapter 3 provides a comparison of the evolution and parallels between research, 4 topical areas and industry data of CCS and CCU using bibliometric analysis techniques. Chapter 4 discusses the life cycle assessment of carbon capture and utilization technologies by providing an in-depth analysis of mineralization technologies compared to other CCU pathways and the incumbent process. Chapter 5 identifies the benefits, risks, opportunities and challenges of mineralization technologies using qualitative analysis techniques involving multiple stakeholders in the field. Chapter 6 ends with overall conclusions and future work from this research.

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## Chapter Two: Literature Review

### 2.1 Sustainability

Sustainable development is defined as using today's natural resources in a manner that doesn't affect the ability to use them for the long term. Specifically, sustainable development is comprised of three pillars that are known as the economic, environmental and social aspects. By achieving harmony among these components one can optimize today's resources in a manner that doesn't affect the future generation's ability to utilize theirs.

The Earth's surface temperature is maintained by greenhouse gases (GHGs), particularly carbon dioxide, methane and water vapour. These gases allow short-wave radiation to enter and prevent long-wave infrared radiation from escaping Earth's atmosphere. This process, also known as the 'greenhouse effect' maintains Earth's surface temperature around 15°C (Boyle, 2012). The Keeling Curve, which measures carbon dioxide accumulations at the Mauna Loa Observatory in Hawaii, currently shows an average atmospheric CO<sub>2</sub> concentration of 420.97ppm (August 2023) (National Oceanic and Atmospheric Administration, 2023). Post industrialization, from 1880-2012 Earth's average land and ocean surface temperature has risen by 0.85°C (IPCC, 2014). At its current rate, global warming is expected to reach 1.5°C warming between 2030 and 2052 (IPCC, 2018). The effects of this 1.5°C is expected to increase land and ocean surface temperatures, hot extremes, heavy precipitation in some areas and drought in others, rise in sea levels, biodiversity and ecosystem species loss and extinction, as well as declines in coral reefs and marine fisheries as a result of ocean acidification and decrease oxygen levels (IPCC, 2018).

The total world population in 2023 according to the United Nations Population Fund was 8.045 Billion (UNFPA, 2023). Economic and population growth are identified as important

drivers for the increase in CO<sub>2</sub> emissions from fossil sources; the increase in population affects depletion of limited global natural resources as well (IPCC, 2014). The 1.5°C scenario is also expected to have adverse human health effects, effects on livelihood and food security, as well as on economic growth by way of changes to gross domestic product (IPCC, 2018).

Globally, in 2020 the following sectors accounted for the largest carbon dioxide emissions: electricity/heat (15.1 Gigatons), transportation (7.29 Gigatons), manufacturing/construction (6.18 Gigatons) and agriculture (5.87Gt) (CAIT Climate Data Explorer - Climate Watch Data, 2023). The highest fossil fuel emissions in 2020 were from China (10.96 Gigatons), the United States (4.72 Gigatons), India (2.45 Gigatons) and Russia (1.62 Gigatons) (Global Carbon Project, 2021). Globally, the burning of coal, natural gas and electricity from fossil sources is the largest contributor of energy-related carbon dioxide emissions (United States Environmental Protection Agency, 2020). Reducing emissions will improve all aspects of sustainable development for future generations.

## **2.2 Carbon Capture Utilization and Carbon Capture Storage**

Considerable efforts are being made to reduce carbon emissions to mitigate potential effects of climate change. One such way is by storing CO<sub>2</sub> underground to sequester it. Carbon capture storage (CCS) differs from carbon capture utilization (CCU), whereby technologies can utilize carbon dioxide as a potential feedstock for many value-added products. Particularly, it can be converted using minerals, used in Fischer-Tropsch reactions, utilized for enhanced oil recovery, and converted into biofuels, among other applications (Cuéllar-Franca & Azapagic, 2015).

CCS involves capture, separation, transport, and storage stages. The capture of CO<sub>2</sub> after combustion is called post-conversion, this typically occurs in coal fired power plants. In pre-

conversion capture, for example when coal undergoes gasification, the generated syngas is converted to hydrogen and CO<sub>2</sub> via the water-gas shift reaction, which is then removed. Finally, in oxy-fuel combustion, oxygen in the place of air is used for the combustion process (Cuéllar-Franca & Azapagic, 2015). Removing CO<sub>2</sub> from flue gas streams occurs with the aid of CO<sub>2</sub> separation technologies. These include absorption by liquid sorbents, which include monoethanolamine (MEA) and diethanolamine (DEA), they have high absorption efficiency and are regenerated using heat. This is the most commonly employed capture method. Adsorption occurs by solid sorbents, such as activated carbon and zeolites, which are regenerated with temperature and pressure swings. Chemical looping technology is where metal oxides are reduced at the fuel reactor side, separate from the air reactor side, to produce a pure stream of CO<sub>2</sub> which doesn't require separation. Additionally, membranes constructed from metals and polymers may also remove CO<sub>2</sub> from flue gas. Cryogenic distillation uses low temperature and high pressure to separate CO<sub>2</sub> from flue gas. Once CO<sub>2</sub> is separated, it is transported to geologic formations for storage into deep saline aquifers, ocean storage, or used for enhanced oil recovery. (Leung, et al., 2014) (Cuéllar-Franca & Azapagic, 2015)

There are many ways in which carbon capture and utilization can take place, namely CO<sub>2</sub> may be converted into fuels, chemicals, materials- such as mineral carbonates, or it can be used for enhanced oil recovery, and as solvents, among other things. The following technologies belong to the technology readiness level (TRL) 7: urea, salicylic acid, polycarbonates, hydrogenation, microbial conversion and microalgae, and mineral carbonation (Tsvetkov, et al., 2019) (Chauvy, et al., 2019). Catalytic CO<sub>2</sub> hydrogenation produces fuels such as methanol and methane, both of which have low unit price and high market volumes (Chauvy, et al., 2019). Urea production is a two-stage process where CO<sub>2</sub> reacts with ammonia to form ammonium

carbamate, this undergoes a dehydration reaction to form urea (Baena-Moreno, et al., 2019). Urea also has a low unit price and high market volume (Chauvy, et al., 2019). Microalgae have the ability to fix CO<sub>2</sub> from flue gas, cultivation of which occurs in raceway ponds and photobioreactors. The biomass content is harvested and dried and biofuel is produced using biochemical conversions (Cuéllar-Franca & Azapagic, 2015).

### **2.3 Mineralization**

Reacting CO<sub>2</sub> with minerals containing magnesium and calcium cations forms magnesite and calcite, as shown in Eq. (1) & (2). In-situ mineralization is a natural process during which CO<sub>2</sub> reacts with different mafic and ultramafic rocks. Basalts and peridotite are targets for in-situ mineralization. However, this process requires long time frames to occur (Sanna, et al., 2014). In contrast, ex-situ mineralization often requires chemical processing in a plant and energy requirements (Intergovernmental Panel on Climate Change, 2005). A variety of mineral and waste materials can serve as the reactive compounds, such as olivine, serpentine, wollastonite, basic oxygen furnace slag, fly ash and air pollution control residues. The reaction of these compounds with CO<sub>2</sub> can occur in a single step (direct mineralization) or multi-step (indirect mineralization), where the reactive compounds are first extracted, followed by carbonation (Sanna, et al., 2014). Two common processes have been developed for direct and indirect mineral carbonation referred to as the National Energy Technology (NETL) process, and the Abo Akademi University (AAU) process, respectively (Giannoulakis, et al., 2013). These processes differ in heat, energy and mineral requirements, efficiencies, pre-treatments and other parameters. There are a variety of different process that use direct mineral carbonation. This can occur without pre-treatment with high pressure CO<sub>2</sub>, or with pre-treatment using mechanical grinding, chemical leaching and thermal as well as mechano-chemical pre-treatments. Pre-

treatment increases carbonation by increasing efficiency and surface area. High pressure CO<sub>2</sub> can also be reacted with aqueous olivine or serpentine in the aqueous carbonation route. Also, CO<sub>2</sub> can be injected into residual brines, rich in magnesium and calcium, such as from oil and gas operations, to form hydrated carbonates (The National Academies Press, 2019). Organic acids can also be used to decrease pH, thereby enhancing dissolution rates (Sanna, et al., 2014). Waste mineral carbonation involves the use of solid wastes from coal plants, solid waste incinerators, cement operations and steel and paper industries. They often require lower pre-treating and less energy intensive operations. The materials include furnace slag, basic oxygen furnace slag, cement kiln dust, municipal solid waste incineration ash, air pollution control residue, coal fly ash, mine tailing and many others (Sanna, et al., 2014) (The National Academies Press, 2019).



There are several products of mineral carbonation. Direct mineral carbonation typically yields a mixture of calcium and magnesium carbonates, silica sand, iron oxides and silicate residues. However, indirect mineral carbonation produces 3 separate streams of silica, carbonates and iron oxides (Sanna, et al., 2012). There are also several common construction markets for mineral carbonation products, these include cement, aggregates, supplementary cementitious materials and novel concrete (Sanna, et al., 2012) (Pasquier, et al., 2018) (The National Academies Press, 2019) (Pan, et al., 2012). Calcium carbonate can also replace limestone in cement (Iizuka, et al., 2004) (CEMCAP, 2017) (ASTM International, 2019). Some other markets

for carbonates include fillers, cement additives and liming agents (Sanna, et al., 2012). Some common mineralization pathways are presented in Figure 2.1.

The life cycle impact assessment stage is used to quantify the environmental impact of elementary flows to and from the environment; CO<sub>2</sub> and CH<sub>4</sub> emissions measure effects on climate change, NO<sub>x</sub> and SO<sub>x</sub> on acidification and eutrophication (Baumann & Tillman, 2004) (Matthews, et al., 2014). Mineral carbonation midpoint indicators for global warming potential (GWP), ozone depletion, acidification, eutrophication, terrestrial and aquatic toxicity, human health, resource depletion and land and water use have been discussed in the literature (Cuéllar-Franca & Azapagic, 2015) (Khoo & Reginald, 2006) (Giannoulakis, et al., 2013) (Kirchofer, et al., 2012) but have not compared NETL and AAU nor the different product streams resulting from these processes using consistent methodologies (Nduagu, et al., 2012). Additionally, the numerous different CCU pathways have also inadequately been compared with each other due to inconsistent boundaries, emission factors and energy sources (Cuéllar-Franca & Azapagic, 2015).

Other forms of ex-situ mineralization technologies also exist based on different feedstock. Namely, carbonation curing occurs where CO<sub>2</sub> reacts with calcium silicates within the ordinary Portland cement (OPC) to form calcium-silicate-hydrate and CaCO<sub>3</sub> (Thonemann, et al., 2022). Additionally, carbonation mixing occurs when CO<sub>2</sub> is introduced during the mixing stage of concrete production (Winnefeld, et al., 2022). This also reacts with the calcium-silicate in OPC to form CaCO<sub>3</sub> (Thonemann, et al., 2022). This process confers to increased compressive strength of the cement, thereby reducing the amount of cement required



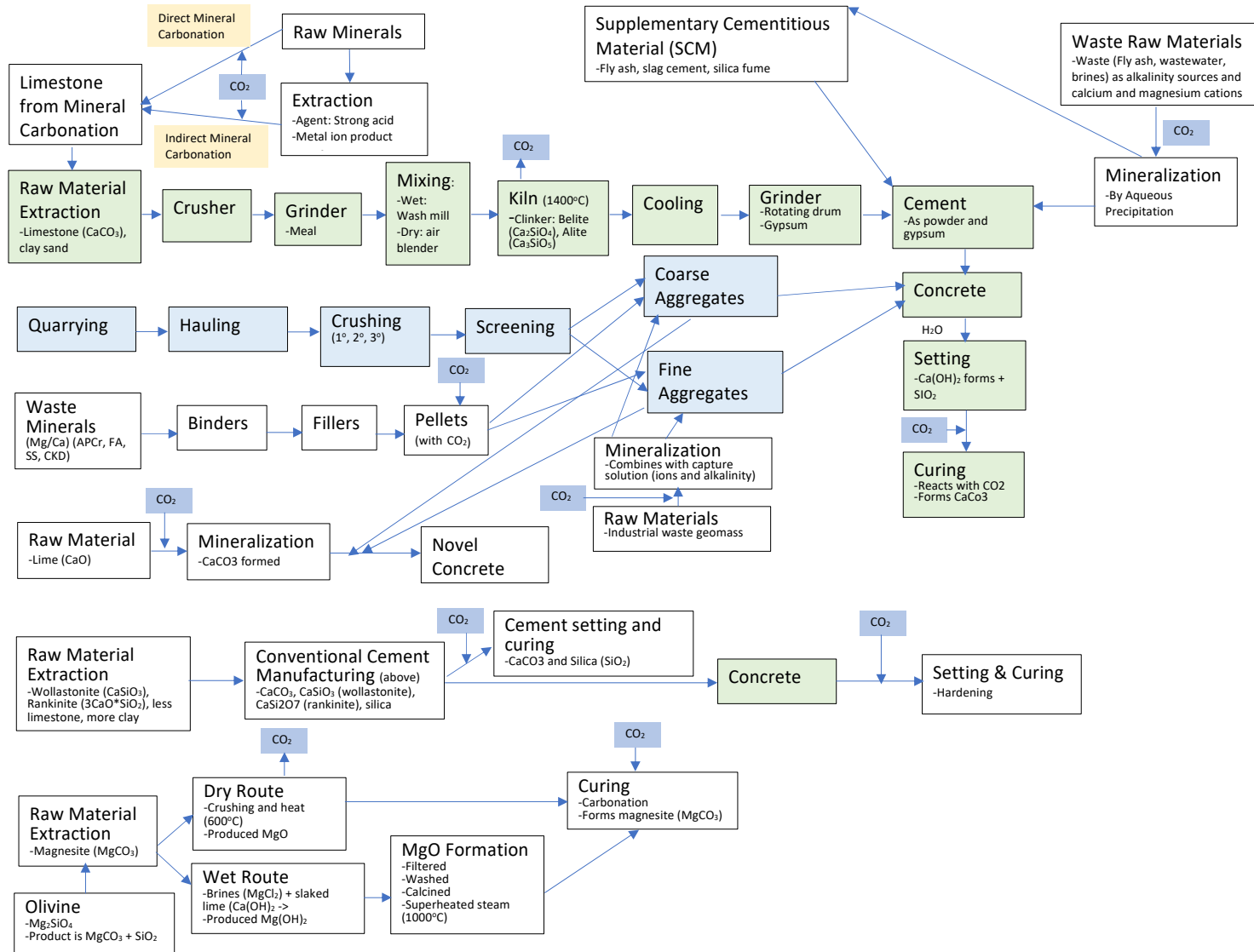


Figure 2.1: Common pathways for converting raw materials into mineral carbonation products. Conventional concrete pathways are shown in green and aggregates pathway are shown in blue

Additionally, industrial by-product feedstock, such as coal ash, steelmaking slag and cement waste can also be used in ex-situ mineralization as well (Ibrahim, et al., 2019). These alternative (non-OPC) binders are referred to as supplementary cementitious materials. They are commonly added to concrete mixes to replace cement (Kazemian & Shafei, 2023). The SCMs react with portlandite ( $\text{Ca}(\text{OH})_2$ ) in hydrated cement to form calcium-silicate-hydrate gel (Krishnan, et al., 2015). Moreover, aggregates act as fillers in concrete that do not take part in enhancing the mechanical strength. Pre-treatment of these aggregates prior to addition into concrete can add an additional source of mineral carbonation for concrete. This can reduce the overall environmental impacts of the concrete mixture (Kazemian & Shafei, 2023).

## **2.4 Life Cycle Assessment**

Life cycle assessment (LCA) allows for comparison across different products and systems. LCA is defined by ISO 14040:2006 to have four stages: goal and scope, life cycle inventory analysis (LCI), life cycle impact assessment (LCIA) and interpretation (International Standards Organization, 2006a). It identifies the strengths, weaknesses and hot spots for technologies based on predetermined boundaries and goals. The goal and scope are where the context of the study, and the methodologies for modelling are defined. This includes items such as defining the system and process by using functional units and reference flow. Any assumptions made for the LCA must also be discussed here. A reference flow is the quantified amount of equivalent products, whereas a functional unit is a quantified unit of the function that the system is expected to perform; all of the flows are described in terms of the functional unit. System boundaries are often technical and describe the cut of criteria for inclusion/exclusion of flows. They can also include geographical and time considerations. Namely, the most inclusive systems boundary is cradle-to-grave, which is a full LCA from resource extraction to disposal/end-of-life. Any

allocation criteria for separating emissions for multi-product systems must also be described in this stage. The life cycle inventory analysis involves the creation of flow charts for each unit process or operation, based on the system boundary, and the goal and scope. Data collection is conducted for the energy, resources, emissions and wastes for each unit process. These output flows are determined per functional unit. (Baumann & Tillman, 2004) (Matthews, et al., 2014)

The life cycle impact assessment stage quantifies the environmental consequences of the inventories defined in the previous stage. There are two types of indicators for life cycle impacts: midpoints and endpoints. The following midpoint (upstream) categories are commonly observed: climate change, acidification, resource depletion, ecotoxicity and human toxicity. While others determine the impacts of endpoint (downstream) indicators: human health, ecosystem and resources (Su, 2020). Once the elementary flows are multiplied by characterization factors (such as the global warming potential- GWP) and divided by normalization factors, they can be characterized as midpoint or endpoint indicators. The midpoint indicators are then converted to endpoint indicators using dimensionless weights. Impact categories can also be grouped, in terms of global or regional impacts, for example (International Standards Organization, 2006b). Several public and commercial databases exist for LCIA data, such as TRACI (Bare, 2012), which characterize midpoint (upstream) indicators while others, such as Eco-Indicator 99 (Ministry of Housing, Spatial Planning and the Environment, 2000), determine the impacts of endpoint (downstream) indicators. A few, such as ReCiPe, also allow both midpoint and endpoint characterizations. These platforms are known as life cycle impact assessment methods, they conduct the classification, characterization, normalization and weighting of the LCI inputs (Su, 2020) (Baumann & Tillman, 2004). For example, ReCiPe LCIA methods have 18 midpoint indicators: climate change, ozone depletion, ionizing radiation, fine particulate matter formation,

photochemical oxidant formation- ecosystem, photochemical oxidant formation- human health, acidification, eutrophication- marine, eutrophication- freshwater, human toxicity- cancer, human toxicity- non-cancer, ecotoxicity- freshwater, ecotoxicity- marine, land use, water use, mineral scarcity and fossil resource scarcity (Huijbregts, et al., 2017). These are described by quantifiable representations, for example: the infrared radiating forcing indicator quantifies the impact for the climate change category, the end result is in units of kgCO<sub>2</sub>equivalents. The specific characterization factor for the climate change impact category is global warming potential. Other characterization factors for other impact categories include: abiotic depletion potential, ozone depletion potential, acidification potential, human toxicity potential, freshwater aquatic ecotoxicity, marine aquatic ecotoxicity, terrestrial ecotoxicity and others (Baumann & Tillman, 2004). Many publicly available LCA software, such as Simapro (PRé Sustainability B.V., 2021), GaBi (Sphera Solutions GmbH, 2021) and OpenLCA (GreenDelta, 2021) create extensive models and visualizations of different processes using various life cycle impact analysis databases. Many of these inventories, databases and software are specific to Europe and the US. This causes a great deal of variability in the results. Unequal boundaries for comparison between two different systems, arbitrary cut-off points and inadequate LCIs are often recognized limitations of LCAs (International Standards Organization, 2006a).

The cumulative global warming impact for a greenhouse gases over a specified time period relative to an equal mass of carbon dioxide (CO<sub>2</sub>) is called global warming potential (GWP).

GWP is calculated as follows (Masters & Ela, 2008):

$$GWP_i = \frac{\int_0^T F_i \times R_i(t) dt}{\int_0^T F_{CO_2} \times R_{CO_2}(t) dt} \quad (1)$$

Where  $F_i$  is the radiative forcing efficiency of a gas  $i$  ( $\text{W/m}^2/\text{kg}$ ),  $F_{\text{CO}_2}$  is the radiative forcing efficiency of  $\text{CO}_2$  ( $\text{W/m}^2/\text{kg}$ ),  $R_i(t)$  is the fraction of gas  $i$  remaining in the atmosphere at time  $t$  (kg),  $R_{\text{CO}_2}(t)$  is the fraction of  $\text{CO}_2$  remaining in the atmosphere at time  $t$  (kg) and  $T$  is the time period for the cumulative effects in years (Masters & Ela, 2008). Both the numerator and the denominator are called the absolute global warming potentials (AGWP). This is defined as the time integrated radiative forcing due to emissions of gas  $i$  (Intergovernmental Panel on Climate Change, 2013). The concept of radiative forcing (RF) is important in that it determines the effects of natural and anthropogenic factors on climate change. RF is the net change in energy balance of the Earth due to a perturbation (Masters & Ela, 2008). This perturbation results in a disruption of the global energy balance thus forcing the Earth to a new equilibrium. For example, increasing GHGs, a form of radiative forcing, due to different stages of the lifecycle of CCU technologies results in increasing the energy balance. The Earth responds to this increase in energy by increasing its temperature to reach the new equilibrium. The GWP of  $\text{CO}_2$  is 1 for all time horizons due to the equivalence of the numerator and denominator.

Therefore, once the GWP has been determined for a specific time horizon, the global warming effect (GWE) in tonnes, can be determined using Equation 2 (Pacca & Horvath, 2002). Where  $\text{GWP}_{i,TH}$  is the global warming potential for each gas for the respective time horizon and  $M_j$  is the amount of GHG released (tonnes). These two equations can be used to determine both time dependent instantaneous (typically one year) as well as cumulative impacts. Determining impacts in discrete time intervals is known as the dynamic LCA approach.

$$GWE = \sum M_i \times \text{GWP}_{i,TH} \quad (2)$$

The last stage, the interpretation stage of LCAs involve checking for completeness, consistency, sensitivity and uncertainty. Results from the LCIA stage are consolidated (Baumann & Tillman, 2004). The findings from the LCI and LCIA stages are discussed together to provide recommendations, limitations and suggestions for future research (International Standards Organization, 2006a).

## **2.5 Qualitative Research Methods**

Qualitative research methods are used when context is required from individuals for a complex and detailed understanding of issues (Creswell & Poth, 2018). There are several goals of using qualitative research: to understand meanings and beliefs of the participants, processes that lead to outcomes, studying unintended phenomena and influences, studying cause and effect situation, developing credible understanding, developing outcomes with the goal to improve practice and policy, and also for a collaborative approach to problem solving, which often involves multiple stakeholders (Maxwell, 2013).

The qualitative approach entails 5 stages: defining the goals, identifying conceptual frameworks, selecting research questions, developing methods to collect and analyze data, and ensuring validity (Maxwell, 2013). There are numerous personal, practical and intellectual goals of qualitative research (described above), which are often shaped by researcher assumptions (Maxwell, 2013) (Creswell & Poth, 2018). There are 5 common qualitative approaches to conducting these studies. These include narrative- collecting stories and experiences from individuals, phenomenological- describing lived experiences of a phenomenon, grounded theory- generating theory or explanations for a process, ethnographic- study patterns and behaviours of a culture or group of people, and case-study- gaining in-depth understanding of a particular case within a context or setting (Creswell & Poth, 2018).

There are several well documented case study types including descriptive, explanatory, exploratory, intrinsic, instrumental and collective case studies (Yin, 2014) (Stake, 1995). Descriptive case studies are often used in sociological investigations to describe a phenomenon in the real-world context. Explanatory studies search for cause-and-effect factors for events, particularly the ‘why’ and ‘how’ behind an event. Exploratory case studies are perhaps some of the more commonly used qualitative methods, as they serve to identify research questions for future studies. (Yin, 2014) (Priya, 2021) (Baxter & Jack, 2018) (Baškarada, 2014)

Additionally, intrinsic case studies identified by Stake (1995) seek to understand the specifics of a particular case or subject under investigation. In this case, the main purpose may or may not to build on a theory. In contrast, instrumental case studies seek to understand something other than the situation or case (i.e. often to refine a theory). Lastly, collective case studies use multiple case studies to formulate new investigations (Ebneyamini & Moghadam, 2018) (Baxter & Jack, 2018).

Once the qualitative approach is selected, appropriate research questions must be developed to frame the study and guide decisions about methods and frameworks. A research question takes into account why the study is being conducted, connections to paradigms and frameworks, what’s known in literature and any predictions about phenomena using tentative theories (Maxwell, 2013). The research questions determine what is being understood, whereas interview questions will generate data for the research questions being investigated (Maxwell, 2013). Four main components are needed in the qualitative methods. These include 1. How the researcher forms relationships with the participants, 2. Setting selection- whom the researcher decides to select as participants and in what setting, 3. Data collection- how the researcher obtains data, and 4. Data analysis- how the researcher make sense of the data. There are 3 ways

of selecting participants: probability sampling, in which samples are selected randomly, versus convenience sampling. The third, more common type of sampling known as purposive sampling selects for a setting, person or activity specific reason. There are 5 goals to purposive sampling: selecting cases to achieve representation, capturing heterogeneity, testing theory, establishing comparisons, and creating the most purposeful relationships. (Maxwell, 2013)

There are several data collection methods: interviews, observations, documents, audiovisuals and focus groups, among others (Maxwell, 2013) (Creswell & Poth, 2018).

Interviews allow for a deeper understanding of the participant's description of events and can be focused on few open ended and guiding questions (Maxwell, 2013). They are chosen to establish common patterns and themes between respondents and phenomena (Warren , 2001). The 7 stages of interviews include: thinking about the topic, designing/planning the research, conducting the interview, transcribing it, analyzing the data, verifying it, and finally reporting it (Warren , 2001). Usually, interviews require a list of interview questions, a fact sheet with the demographical information, an informed consent form, recording device and writing materials (Warren , 2001).

The study presented in this research examines an explanatory case study of mineralization technologies in Alberta. It looks for causal factors to explain the critical factors which determine mineralization technology adoption based on the Rogers diffusion of innovations model. The case study examines Alberta specific benefits and opportunities. The richness of information provided by semi-structured interviews with six stakeholder groups reveal why these technologies face adoption challenges and how they can overcome them.

Once the study is conducted, data analysis of qualitative research is carried out which involves either content analysis or thematic analysis. Thematic analysis is most common as it



details qualitative perspectives of the data, and quantitative counts of codes or other specific associations (Bradshaw, et al., 2017) (Vaismoradi, et al., 2013). Data can be fractured and then rearranged into thematic categories. Developing categories based on what the data and what the researcher deems to be important is defined as open coding (Maxwell, 2013). After the data is analyzed, the findings must be verified. A number of data validity methods exist. Validity is relative to the research context and in relation to the connections of the conclusions to reality (Maxwell, 2013). A number of factors determine validity, and their applicability depends on the context: long-term participant observation, detail and variability of data, respondent validation, level of intervention in the experiments, reporting discrepancies in findings, triangulation- using a range of settings, methods and/or individuals, and determining the amount of evidence sufficient to relay conclusions (Maxwell, 2013). Integration of data from interviews with other sources such as written forms is referred to as methodological triangulation (Roulston & Choi, 2018).

There are many barriers of replacing conventional products with CCU products, such as mineral carbonates, due to engineering/performance and public perception challenges. Studies found that risks associated with CCU disposal, product use and product quality decrease their public acceptance (Arning, et al., 2019) (Arning, et al., 2017) (Heek, et al., 2017). However, these perceptions require further investigation, especially in North America. Construction codes and standards would define acceptable performance for mineral carbonates and their replacement products (The National Academies Press, 2019). This raises additional questions about the properties and performance of these materials for the product and market they intend to replace. Currently, there are no International Panel on Climate Change or International Standards Organization reports for CCU products. There are, however, sections within other reports

focussing on a few CCU technologies (IPCC, 2005) (IPCC, 2000). The commercial viability of these technologies will be conducted in this study by determining gaps in the regulatory and engineering aspects of CCU technologies using semi-structured qualitative interviews with stakeholders along the CCU supply chain. This will lead to policy implications for the utilization of mineral carbonate products.

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## **Chapter Three: Evolution of and Parallels Between Carbon Capture Utilization and Carbon Capture Storage using Bibliometric Analysis Techniques**

### **3.1 Introduction**

Considerable efforts are being made to reduce carbon emissions to prevent the potential effects of climate change. One such way to mitigate emissions is by using carbon Capture and Storage (CCS) which is the process of storing CO<sub>2</sub> underground and in the ocean to permanently sequester it. However, over the past decade, carbon dioxide has begun to be viewed as a potential feedstock for several value-added products. Carbon capture and utilization (CCU) is a suite of technologies and pathways that can offer many creative and innovative options for converting carbon into useful products for application as materials (e.g. calcium carbonate, magnesium carbonate), fuels (e.g. methanol, methane, dimethyl ether), chemicals (e.g. urea, formaldehyde, salicylic acid) and food/feed (e.g. microalgae, animal feed) (Chauvy & De Weireld, 2020) (Garcia-Garcia, et al., 2021). The term “Carbon Capture, Utilization and Storage/Sequestration” (or CCUS) is often used to encompass both CCS and CCU technologies. Some applications, such as CO<sub>2</sub>-enhanced oil recovery (EOR), are often categorized as both CCS (for its ability to sequester carbon) and CCU (for its use in extracting/producing oil & gas reserves) (Gaspar Ravagnani, et al., 2009) (Farjzadeh, et al., 2020).

For both CCS and CCU, different options have been developed to capture CO<sub>2</sub> from various point sources (e.g. power and chemical processing plants) or from the atmosphere using direct air capture (DAC), mainly using technologies that incorporate chemical or physical solvents. Chemical solvents (e.g. monoethanolamine) use absorption and heat regeneration, and physical solvents (e.g. selexol and rectisol) are regenerated with reduced pressure to capture and collect CO<sub>2</sub>. For point sources, carbon dioxide can be captured at three possible points in most processes: 1) after conversion of the fuel (post-conversion), 2) as a co-product of an intermediary



reaction of the fuel (pre-conversion), and 3) as a combustion product from fuel with the use of pure oxygen (oxy-fuel combustion).

The CCS field is far more mature than CCU, and was originally identified to mitigate greenhouse gas emissions in the early 1970s (i.e. the Sharon Ridge oilfield in Texas, USA) (Liu, et al., 2018). The main advantage with CCS is its potential to sequester large quantities of carbon at a specific location and at scale. Commercial applications of CCS already exist in the form of CO<sub>2</sub>-enhanced oil recovery (EOR) and commercial storage of CO<sub>2</sub> in saline aquifers (Ringrose, et al., 2021) (Hosa, et al., 2011). Estimates indicate 8 gigatons of CO<sub>2</sub> can be stored in the USA with EOR; the CO<sub>2</sub> global storage potential is expected to rise to 60 gigatons of CO<sub>2</sub> by 2050 (Núñez-López & Moskal, 2019) (IEA, 2015). Experience in deploying these technologies has resulted in both having a high technology readiness level (TRL 9) (Bui, et al., 2018) (Hepburn, et al., 2019). The decision to implement CCS is driven by economics, and number of technical and socio-political drivers including carbon pricing, minimization of capital costs, reliable monitoring, increasing public acceptance and knowledge, shared transportation and market expansion (Araújo & de Medeiros, 2017) (Aminu, et al., 2017). The major challenge faced by CCS technologies are economic issues. Aside from significant capital costs and the ongoing operational costs required to run and maintain facilities, there are also economic risks in the form of long-term liabilities such as leakage of CO<sub>2</sub>, induced seismicity, contamination, acidification of bodies of water and damage to ecosystems (Anderson, 2017). Much of this is managed at the regional level, where local governments assume long-term liability for storage sites, thereby reducing investor cost and risk (Anderson, 2017) (Rassool, et al., 2020).

Compared to CCS, the development and commercialization of CCU technologies is less mature but has evolved quickly over the past decade. The main advantage of CCU technologies

is its ability to be implemented at a smaller scale, and is less restricted in terms of industry type and geographical location (Zhu, 2019) (Zhang, et al., 2023). In terms of common processes, carbon dioxide can be used as a chemical feedstock when it is converted to syngas by the reverse water gas shift reaction followed by the Fisher-Tropsch reaction to produce hydrocarbons (Ahmad, et al., 2017). It can also be stored as minerals, utilized for enhanced oil recovery, or converted into biofuels (Cuéllar-Franca & Azapagic, 2015) (Torres, et al., 2013). Several CCU technologies are now mature including projects that convert CO<sub>2</sub> into mineral carbonates (Giannoulakis, et al., 2013) (Khoo, et al., 2011) (Sanna, et al., 2014). This can be done using a multi-step process devised at the Åbo Akademi University (AAU) (Nduagu, et al., 2012) as well as the single stage process devised at the National Energy Technology Laboratory (NETL) (Khoo, et al., 2011). A number of mineralization facilities are currently being piloted or demonstrated, and are at the TRL 5-8 stage (Hepburn, et al., 2019) (Chauvy & De Weireld, 2020). Other carbon conversion technologies with similar maturity levels can be found in the beverage industry, and in the production of urea, methanol, microalgae, polycarbonates, and salicylic acid (Bui, et al., 2018) (Chauvy & De Weireld, 2020) (Tcvetkov, et al., 2019). While there has been growing interest in these spaces, CCU technologies have also faced a number of challenges in its implementation including economics, scale, market size and penetration, control of external factors (e.g. beyond producer's value chain), unintended outcomes (e.g. land, water, chemical and fuel use patterns), availability of waste streams, risks with use (e.g. regulatory constraints, possible contamination threat to human health), and lower than expected life cycle greenhouse gas reductions (NAP, 2019) (Al-Mamoori, et al., 2017). As a result, policy drivers and economic incentives are required to increase the feasibility of CCU projects in the long term

(NAP, 2019) (Edwards & Celia, 2018). These drivers and challenges are important research areas to determine the future prevalence and success of CCU and CCS technologies.

In order to better understand research trends in a given discipline or target area, bibliometric analysis has been used by the academic community to help assess trends and gaps in a particular field. Bibliometric analysis is a quantitative tool that is capable of characterizing large volumes of data into decipherable clusters and themes. This is done using commonly employed procedures such as co-occurrence, co-citation, highly linked and cited, and temporal distribution network maps (Donthu, et al., 2021) (van Eck & Waltman, 2023). A co-occurrence network map is a visual representation of connection between terms, and is used to provide an overview of the research field and identify potential gaps between subfields (Ranjbar-Sahraei, & Nagenborn, 2017). A co-citation network map examines cited references, sources and authors to provide an overview of the landscape by which articles of different researchers are co-cited by the same articles. A highly cited network is a visualization of the documents, sources, authors, and countries, which provides an overview of the most influential publications and subareas with higher citation impact in the field. Finally, a temporal distribution shows the evolution of topical areas over time based on average publication year of papers with the corresponding terms (van Eck & Waltman, 2023) (Donthu, et al., 2021).

Bibliometric analysis of the CCS literature has, thus far, mainly examined the CCS field in specific countries such as China (Jiang & Ashworth, 2021) (Wang, et al., 2020) (Wong, et al., 2021), as well as specific results on costs/investment associated with CCS (Li, et al., 2019a) and carbon capture technologies (Omoregbe, et al., 2020) (Yi, et al., 2020) (Naseer, et al., 2022) (Viebahn & Chappin, 2018). CCU bibliometric studies are less numerous, and discuss country prevalence research findings (Nawaz, et al., 2022). Numerous studies are found that present

results on the analysis of carbon capture utilization and storage (CCUS). These studies discuss generalized CCUS trends to compare with other fields, such as other low-carbon technologies (Tapia, et al., 2018). Specific areas within CCUS are also discussed by some studies, such as patent information (Zhu, et al., 2023) and life cycle assessment (de Cruz, et al., 2021). To date, however, no studies compare between CCS and CCU separately using bibliometric analysis techniques to analyze the parallels and evolution of each research area.

The objective of the current paper is to use bibliometric analysis techniques to assess the state of the art with respect to CCS and CCU research and application, and to highlight how these two fields have evolved over time. Using comparative bibliometric analyses of CCS and CCU, research themes and industry trends are identified by evaluating the evolution, temporal and geographical distribution of results. The analysis also identifies knowledge gaps within CCS and CCU research. Research prevalence is compared with the landscape of industrial CCS and CCU projects using country specific data. The comparison of parallels provide suggestions for future research and industry directions. Additionally, based on drivers and challenges, a methodology for a meta-analysis is developed which examine four topical areas within CCS and CCU literature including policy, environment, economy and technology. This process highlights the evolution of research and important themes within these noted topical areas. Consequently, the analysis conducted by this research offers suggestions for future work in CCU and CCS for researchers, industry partners and policy makers.

### **3.2 Methods**

The purpose of this analysis is to assess the evolution and parallels between CCS and CCU to identify research findings and future directions. This section discusses the procedure used to conduct a bibliometric analysis of CCS and CCU and the procedure for selection of papers for 4

topical areas. First, the data mining and search string techniques used in the analysis are presented, followed by the process for developing bibliometric network maps. This is followed with an overview of the methods used for construction of the network of co-occurrence of terms, co-citation relationship of journals, highly linked and cited documents and co-authorship of countries. An overview of the methods for grouping similar data into clusters to identify hot spots in research are also discussed in their respective sections. Next, the methodology for the selection of review papers from each topical research area is presented. Lastly, the industry dataset methods are discussed.

### ***3.2.1 Data Mining and Search String***

Bibliometric data was gathered from the Web of Science (WoS) Core Collection, in February 2023. The results are downloaded as tab-delimited text files. The WoS database encompasses over 21,973 journals, books and conference proceedings, and has over 87 million records and 2 billion cited references dating back to 1900 (Clarivate, 2023b). It is one of the oldest and most widely used reference databases (Singh, et al., 2021) (Birkle, et al., 2020).

For the current analysis, the following search strings are utilized for data capture:

- (1) For CCS: Topic = ("carbon capture" AND storage) OR Topic = ("carbon capture" AND sequestration)
- (2) For CCU: Topic = ("carbon capture" AND utilization)

The timeframe examined for this search was from January 1998 to February 2023. The two search strings above ensure that CCS and CCU results could be studied individually for each category when analyzing trends. The CCS search string includes the terms “sequestration” and

“storage”. The former was more commonly used in earlier CCS literature, but has now been replaced by the latter term.

### **3.2.2 Constructing the bibliometric network maps**

In this study, a number of network maps were created using VOSviewer (Centre for Science and Technology Studies, 2023) which is a publicly available software tool used to create maps from network data for visualizing and exploring bibliometric relationships. VOSviewer uses a distance-based approach to map the similarity between items such as publications, researchers, terms and journals. Each item represents a node, and the distance between nodes represents the strength in similarity. The size of the node depends on the type of map being generated and represents the total number of terms, citations or publications for each individual item in the map. Lines (or edges) between nodes indicate the existence of a relationship (e.g. co-occurrences, co-citations or co-authorships) and the strength of that relationship (Van Eck & Waltman, 2014). The distance between two nodes reflects the level of relatedness between the nodes. In general, smaller distances between the nodes indicate a stronger level of relatedness, which depends on the network being constructed (van Eck & Waltman, 2023).

VOSviewer creates maps using three steps: 1) a similarity matrix is created using the co-occurrence relationships, 2) the maps are constructed using the similarity matrix, and 3) lastly, these maps are translated, rotated and reflected (van Eck & Waltman, 2010). The similarity matrix can be calculated using Equation 1 (van Eck & Waltman, 2010):

$$S_{ij} = \frac{C_{ij}}{W_i W_j} \quad (1)$$

where  $C_{ij}$  is the number of co-occurrences of items  $i$  and  $j$  together, and  $W_i$  and  $W_j$  represents the total number of occurrences/co-occurrences of items  $i$  and  $j$ .

### 3.2.2.1 Co-occurrence Network of Terms

The co-occurrence network of terms maps for both the CCS and CCU search terms were developed using the VOSviewer tool and text data from the relevant publications' titles, abstracts and keyword. Co-occurrence maps of terms were generated for two distinct time periods: 1998-2012 (Jan 1, 1998 to Dec 31, 2012) and 2013-2023 (Jan 1, 2013 to Feb 02, 2023). The intent was to compare and highlight any differences in terminology used in both the CCS and CCU fields for these two time periods. In these network maps, each node represents a commonly found term, and the size of each node (also known as their occurrence) indicates the number of occurrence of terms in publications that have the term in their title, abstract or keyword. The relevance score is defined as the difference between the distribution of co-occurrences of a term relative to the overall distribution of co-occurrences (van Eck & Waltman, 2011). Terms with high relevance scores co-occur together and represent distinct topics in the field, whereas a low relevance score represent generic topics.

The VOSviewer tool was also used to perform cluster analysis of the key terms. Similar nodes that are highly relevant are organized into clusters based on selected resolution parameters. Binary counting method was applied using a minimum of 10 occurrences, this counts the presence or absence of a term in the documents to derive their occurrence and relevance scores. A clustering resolution of 0.9 and 0.7 was used in creating the CCS and CCU co-occurrence network of terms maps, respectively. Commonly found terms in each cluster are noted and used to characterize the cluster. This clustering provides an indication of key subject areas studied for both the CCS and CCU search terms over time.

### 3.2.2.2 Co-citation Network of Journals

The co-citation of journals network for the CCS and CCU search terms were developed using bibliometrics data extracted from Web of Science (WoS). Two publications are said to be co-cited when a single publication cites the two publications. The cited sources or journals are mapped using fractional counting which divides the co-citation relationship by the total number of co-citations it has, whereas full counting counts the total number of co-citations (van Eck & Waltman, 2023) (Van Eck & Waltman, 2014). The minimum number of citations are set to 20, as per default settings. A clustering resolution of 0.7 is selected for both the CCS and CCU fields, with a minimum cluster size of 30 for the CCU field. The top three journals with the highest co-citations for each cluster were determined and represented in the results. The top ten linkages ranked by link strength were also extracted and presented. The areas represented by each cluster are determined using the categories allocated to the journals from the Web of Science's Journal Citation Reports (Clarivate, 2023a). The top three co-cited journals from each cluster were used to label their respective cluster. The cluster categories that could be determined with a high degree of certainty based on this method are selected first and the others are selected by a process of elimination.

### 3.2.2.3 Network of Highly Cited Documents

The VOSviewer tool was also used to generate the most cited references (documents) for both the CCS and CCU search terms using data extracted from WoS. The minimum citation value was set to 300 for the CCS search term, and results were obtained for the top 25 documents in the field. For the CCU search term, the minimum citation was set to 200, and results were obtained for only the top 17 documents in the field non-connected items were not displayed. Clustering was not carried out for this analysis, as specific clusters were not needed for the



references. All documents were characterized as per key areas of contribution, year of publication, number of citations and country.

#### 3.2.2.4 Co-authorship Network of Countries

The co-authorship network of countries map was also developed using the VOSviewer tool using the previously mentioned CCS and CCU WoS search strings. Fractional counting method was applied for this network, and the minimum number of documents for a country was set to five. From this analysis, the top 10 countries in terms of the number of documents is presented along with the total link strength.

#### ***3.2.3 Selection of Review Papers for Topical Areas***

A closer examination of research topics over time was conducted through a sampling of existing review papers for both the CCS and CCU search terms. Due to the rather large number of review papers published in this space, a novel methodology to capture key research themes over time was developed and is presented in Figure 3.1. Overall, four important topical areas were investigated using the following additional search terms: policy, technology, environment and economic. The total number of CCS and CCU records for each research area was filtered by their appropriate search term. The reviews were organized by most cited for 5 time periods: 1998-2002, 2003-2007, 2008-2012, 2013-2017 and 2018-2023. The title and abstract were manually read and organized as highly representing the research area, somewhat representing and not representing. A maximum of five most cited and closely representative reviews per research area were selected. These reviews were categorized by their respective topical areas categories, year of publication and number of citations. Key research themes were identified

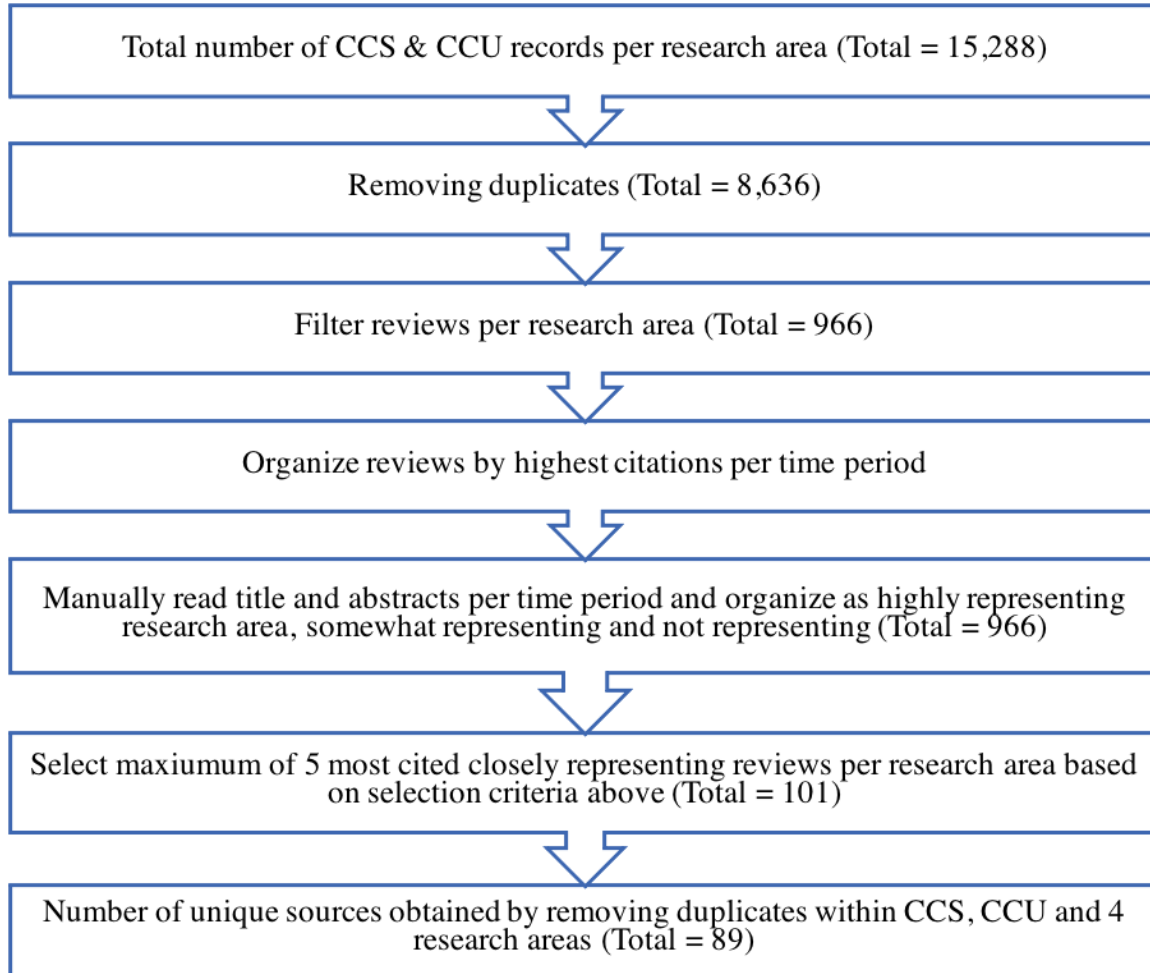


Figure 3.1 Methodology for selection of records for 4 topical areas in CCS and CCU

from these reviews for each of the four key topics and cataloged to highlight changes over time.

### 3.2.4 Industry Dataset

CCS industry results are categorized based on the facility status. Two categories are shown, for facilities that are currently developed (i.e. completed and operational) and other facilities that are under construction (i.e. in construction, early development and advanced development). This includes the number of commercial CCS facilities (i.e. ongoing commercial facilities of CO<sub>2</sub> capture and transport for permanent storage), and pilot and demonstration

facilities (i.e. non-commercial CO<sub>2</sub> capturing facilities for testing, developing or demonstration of CCS technologies and processes). The CCS facility data was obtained from Global CCS Institute, which is an extensive and up to date database for all CCS facilities and projects (Global CCS Institute, 2022). The number of start-up carbon dioxide utilization companies' data was obtained from the Global CO<sub>2</sub> Initiative-University of Michigan, which provide details about CCU startup companies across the globe (Global CO<sub>2</sub> Initiative, 2020). The total CCS and CCU documents for respective countries are also presented in Figure 3.9. Additionally, annual carbon dioxide emissions released by countries in Gigatons of CO<sub>2</sub> equivalents (GtCO<sub>2</sub>) (Ritchie & Roser, 2021) and gross domestic product (GDP) in Trillions of USD (The World Bank, 2021) are presented in Table 3.10. Items in Figure 3.9 and Table 3.10 are sorted by total documents per country.

### **3.3 Results and Discussion**

This paper examines the evolution of research and draws parallels between the fields using bibliometric analysis techniques. The results are described such that bibliometric findings are discussed first, followed by specific results from topical areas. These are discussed in terms of broad areas of CCS and CCU (i.e. policy and technology), followed by relevant topics for specific research areas (i.e. environmental and economic). What follows are results from bibliometric analysis of the co-occurrence of terms, temporal distribution of publications, co-citation network of journals, and highly linked and cited documents. This is followed by a closer investigation of results of a meta-analysis of CCS and CCU search terms from 4 topical research areas: policy, technology, environment and economy. The process of presenting results follows thematic analysis of research findings. Then, the landscape of CCS and CCU projects and co-

authorship of countries is examined to identify parallels. Lastly, suggestions for future research directions are provided.

### ***3.3.1 Temporal Distribution of Publications***

Studying the timeline of research data shows evolution of trends in CCS and CCU. Specifically, a total of 8,790 CCS records and 2,288 CCU records were obtained from the WoS database. Figure 3.2 shows the total number of publications in the CCS and CCU fields. The first observed CCS record from our search was in 1998 (Zepp, et al., 1998) and the first observed CCU record was in 1999 (Benson, et al., 1999). This initial work corresponds to the initial adoption of the Kyoto Protocol in 1997 (UNFCCC, 1998). Between 1998 and 2007 only 2 CCU records are found, both of which discuss biomass and biofuel production (Benson, et al., 1999) (Read, 2002), whereas 111 CCS records are found, this indicates that the CCU research field developed after the CCS field. CCS trends show a sustained increase in data from 2009 onwards, with a rise in records from 87 to 259 from 2008-2009, respectively. This increase in research activity corresponds to the acceptance of the Copenhagen Accord in 2009 by the parties to the United Nations Framework Convention on Climate Change (UNFCCC). Although not legally binding, it recognizes the importance of limiting global temperatures to a maximum of 2°C through setting emission reduction targets for 2020 by the signing countries (UNFCCC, 2009). Overall, the cumulative records of CCS literature compared to CCU indicate that CCS has grown at a much faster rate than CCU literature. CCU literature continues to rise steadily for all years shown. In 2022, CCU reached its peak of 548 records which is more than a 7-fold increase from 2015. For the same period CCS literature rose by approximately 2-fold, to a maximum of 1,047 records in 2022. This shows a gradual and consistent increase in CCU technologies research,

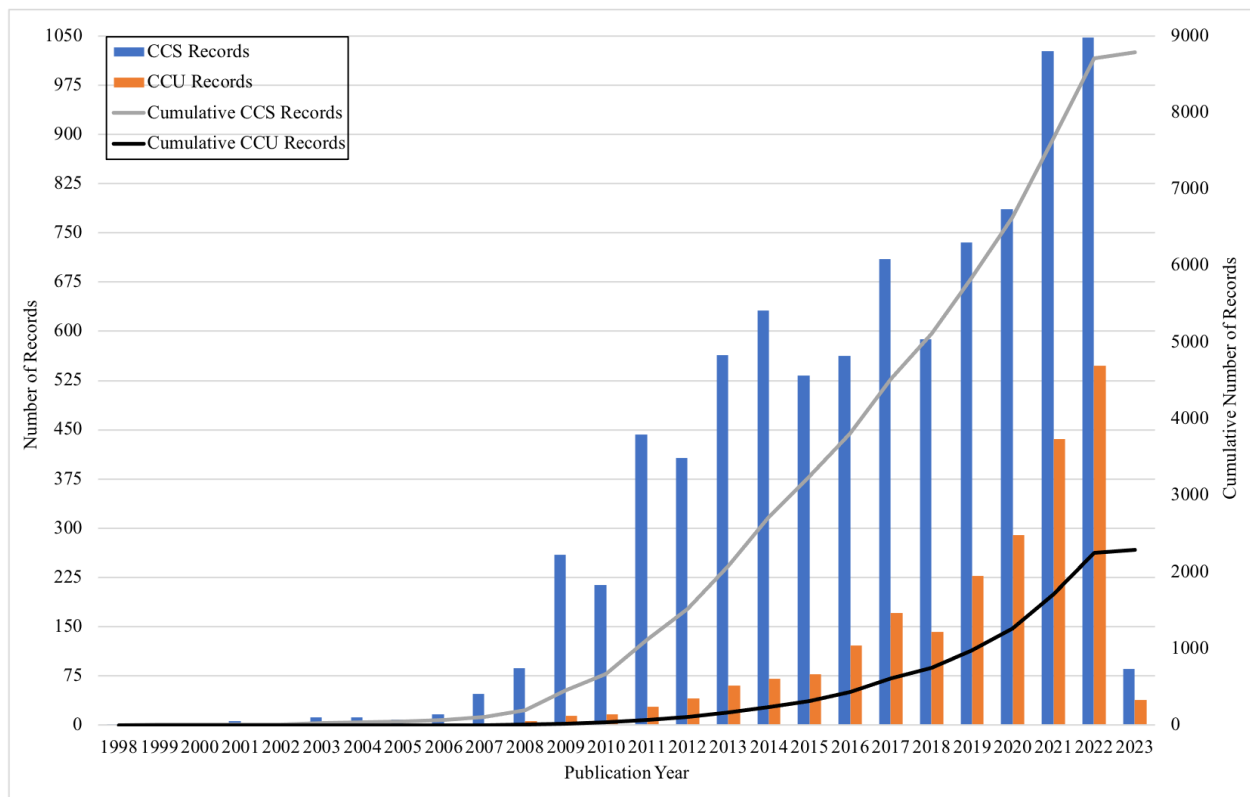


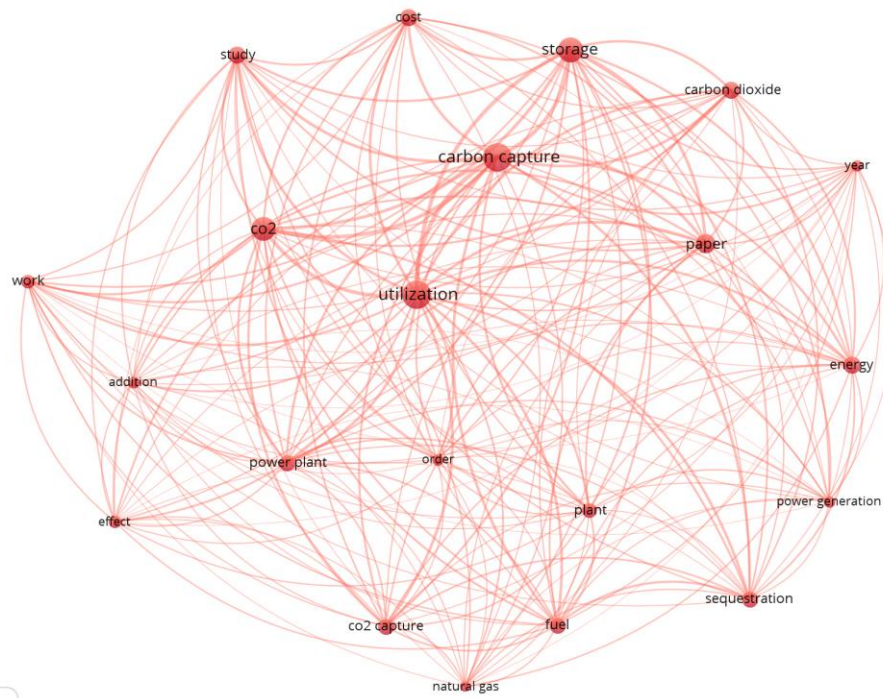
Figure 3.2 Total number of records for CCS (blue bars), CCU (orange bars), and the cumulative increase for CCS (gray line) and CCU (black line) from 1998-2023. Data has been obtained from the Web of Science database.

which expanded from 2016 to 2022 compared to previous years. Both increases in CCS and CCU records also correspond to the implementation of the Paris agreement in 2015 (UNFCCC, 2015).

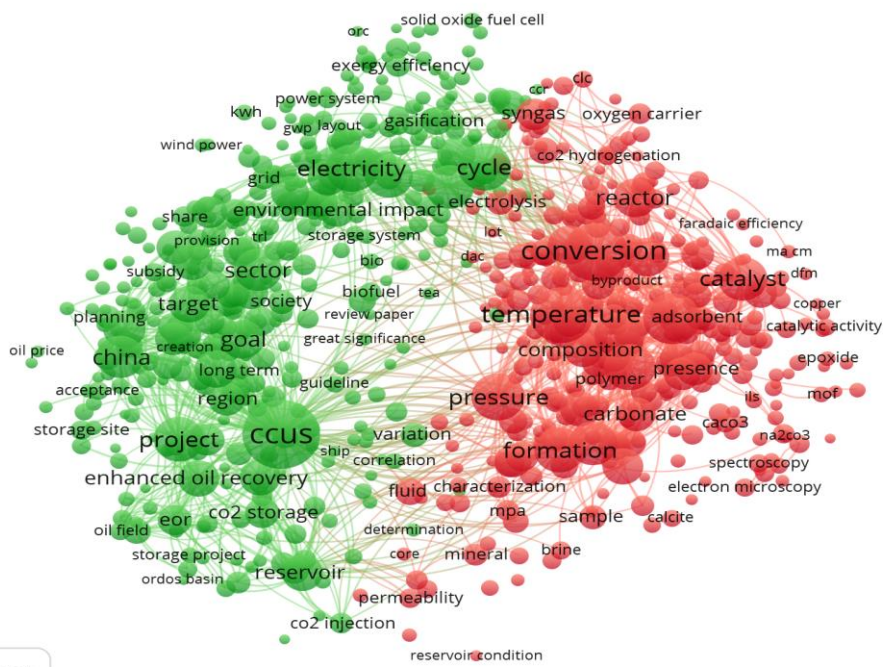
### 3.3.2 Co-occurrence of Terms

The co-occurrence of terms map for CCS depict the state of the art of research, how different research areas are clustered and their interconnections. The terms map evolves from a total of 455 terms from 1998-2012 (Figure 3.3a) to 2,228 terms from 2013-2023 (Figure 3.3b) using the search term for CCS described in the methodology (Section 3.2.1). The number of times that terms co-occur with each other in the title, abstract and keywords is shown by the network





(a)



(b)

Figure 3.4 Visualization of co-occurrence network of terms for (a)CCU from 1998-2012 and (b)CCU from 2013-2023 using VOSviewer version 1.6.19. Extracted text data from titles and abstracts using binary counting, minimum number of 10 occurrences per term and cluster resolution of 0.7.

co-occurrence of terms map (Figures 3.3 and 3.4). The distance between the nodes shows the level of relatedness between them, whereas the size of the nodes indicates the number of citations. Lines reflect the relation between any pair of nodes. Three clusters are visible in the 1998-2012 CCS map (Figure 3.3a). This includes the red cluster- policy and perception area, which includes the 5 policy terms (e.g. policy, climate policy, energy policy), investment and government. The yellow cluster- CO<sub>2</sub> injection and storage field, which include 4 injection terms (e.g. carbon dioxide injection, injectivity), as well as pressure and reservoir. The green cluster- CO<sub>2</sub> capture and power requirements, which includes 13 terms associated with CO<sub>2</sub> capture (e.g. oxy-combustion, post-combustion and pre-combustion). The number of nodes for all of these clusters in Figure 3.3a are fewer than those observed for 2013-2023 (Figure 3.3b). Four clusters are observed in Figure 3.3b. Each representing distinct areas in research. The red cluster describes terms related to the policy and perception area. The following terms can be seen in this cluster: 20 policy terms (e.g. CCS policy, climate policy, energy policy, environmental policy), investment, public perception, government, country and stakeholders. The yellow cluster represents the CO<sub>2</sub> injection and storage field. It includes 20 injection terms (e.g. carbon dioxide injection, fluid injection, gas injection, injection pressure), CCS site, pressure, formation, leak and oil recovery. The green cluster refers to terms associated with CO<sub>2</sub> capture and power requirements. Included in this cluster are the terms amine, membrane and 26 terms associated with CO<sub>2</sub> capture and post/pre combustion (e.g. oxy-combustion, pre-combustion, post-combustion). The blue cluster represents the CO<sub>2</sub> chemistry field and includes the following terms: reaction, mixture, temperature, property and particle (Figure 3.3b). Particle reactions such as with mineralization are also found in this cluster. The green cluster from 1998-2012 in Figure 3.3a, which includes terms associated with the CO<sub>2</sub> capture and power requirements continues to



increase in nodes and eventually forms 2 clusters, that being blue-CO<sub>2</sub> chemistry field and green-CO<sub>2</sub> capture and power requirements, as seen in the 2013-2023 CCS terms map (Figure 3.3b).

A total of 21 and 740 terms were found to fit the selection criteria described in the methodology (Section 3.2.2.1) for the 1998-2012 (Figure 3.4a) and 2013-2023 (Figure 3.4b) CCU maps, respectively. The cluster for CCU terms from 1998-2012 shows one cluster that include both carbon utilization and carbon storage specific terms (Figure 3.4a). This includes terms such as utilization, storage and carbon capture. Two distinct clusters can be observed for the CCU terms in 2013-2023 (Figure 3.4b). The green cluster describes terms associated with carbon capture and storage. It includes themes of policy, environment, energy and economics. Specifically, the following terms can be found in this cluster: enhanced oil recovery (EOR), 10 storage terms (e.g. carbon storage, storage capacity, long-term storage), pre and post-combustion capture, electricity, policy, economic viability and life cycle assessment. The red cluster represents carbon conversions terms associated with carbon utilization. The terms found in this cluster are conversion, mineralization, electrochemical conversion, microalgae, hydrogenation, as well as the technical aspect associated with CCU. The carbon utilization and carbon storage cluster from Figure 3.4a increases in nodes and two clusters become visible in the 2013-2023 terms map (Figure 3.4b). The top three largest nodes by occurrence for CCU terms between 2013-2023 (shown in brackets) are: CCUS (431), conversion (310) and temperature (282). These terms are specific to both CCU and CCS fields, unlike the commonly found CCS terms mentioned which are specific to the CCS field (Figure 3.3b). Namely, the top three largest CCS nodes between 2013-2023 by occurrence (shown in brackets) are temperature (959) in the blue cluster, pressure (778) in the yellow cluster, and electricity (635) in the green cluster. This represents the aforementioned terms related to CO<sub>2</sub> chemistry, CO<sub>2</sub> injection and storage, and

CO<sub>2</sub> capture and power requirements, respectively. The common cluster/area of the 2013-2023 CCS and CCU maps include carbon storage and carbon capture. 118 and 18 terms about the CO<sub>2</sub> molecule also exist in CCS (e.g. CO<sub>2</sub> eq, CO<sub>2</sub> carrier, CO<sub>2</sub> purification, CO<sub>2</sub> compression) and CCU maps (e.g. CO<sub>2</sub> concentration, mt CO<sub>2</sub>, CO<sub>2</sub> reduction, CO<sub>2</sub> stream), respectively.

The number of lines in Figures 3.3 and 3.4 indicate the number of documents in which the two nodes co-occur. In Figure 3.3b, the CO<sub>2</sub> injection and storage (yellow) and CO<sub>2</sub> chemistry (blue) clusters have many more lines connecting them than either do with the policy and perception (red) cluster. Likewise, the most cited terms pressure and temperature have 1,797 and 1,855 links, respectively. These terms are linked to both clusters and the CO<sub>2</sub> capture and power requirements (green) cluster, but the term pressure and temperature only share 1 and 0 link, respectively with the policy and perception (red) cluster. The decrease distance and increased lines between the CO<sub>2</sub> injection and storage (yellow) field and the CO<sub>2</sub> chemistry (blue) fields are reflective of their relatedness in CCS operations, where CO<sub>2</sub> dissolution and precipitation affects the physical, chemical and mechanical properties of formations (Fatah, et al., 2020) (Widdicombe, et al., 2018). The policy and perception (red) cluster's most occurring term-policy, is only linked to 2 nodes of a different cluster- the CO<sub>2</sub> capture and power requirements (green) cluster (i.e. electricity and energy efficiency), this is also the cluster with which it shares most of the lines with max lines set to 1000. This suggests that the physical sciences, represented by the CO<sub>2</sub> injection and storage (yellow), CO<sub>2</sub> chemistry (blue) and CO<sub>2</sub> capture and power requirements (green) clusters, are not highly interconnected by lines with the social sciences, represented by the policy and perception (red) cluster. Other studies have identified the green economy cluster that include policy terms separately from the environmental impact cluster (Santeramo, 2022). Additionally, examining social ecological systems identifies a social/policy

cluster to be distinct from other clusters such as climate change and ecological theories and concepts (Nielsen & Faber, 2021).

In examining Figure 3.4b, it is evident that both clusters share many lines. There is a great deal of relatedness between the carbon conversions associated with CCU (red), and carbon capture and carbon storage (green) clusters. This is due to the fact that the search term involves carbon capture and the associated utilization stage. Carbon utilization, such as mineral carbonation, chemical production and biodiesel production are distinct from carbon storage, which convert carbon into a sequestered product and carbon capture methods, such as solvents, sorbents and membranes (Cuéllar-Franca & Azapagic, 2015) (Kenarsari, et al., 2013). This is depicted by the carbon conversions (red) cluster that shows more lines within the cluster itself than with the carbon capture and carbon storage (green) cluster. However, overall compared to CCS, the co-occurrence of terms of CCU literature shows that clusters are more interconnected as shown by the numerous lines seen between the nodes in Figure 3.4b.

Overall, from these network maps, early research topics for CCS focussed on CO<sub>2</sub> injection, CO<sub>2</sub> capture and policy, while later topics focussed on CCS formation, CO<sub>2</sub> capture, CCS policy and CO<sub>2</sub> reaction chemistry. For CCU, early research topics were quite sparse due to lack of maturity in this space and included key terms such as carbon utilization and capture. Later topics in the CCU area started to show a more mature research landscape (similar to CCS) focussed on CO<sub>2</sub> capture, carbon storage and carbon conversion. However, it can be seen that there is no clear clustering or delineation of specific technologies or pathways which are plentiful in the CCU research space (Chauvy & De Weireld, 2020). It is anticipated that further clustering of research topics will occur over time, as the sector becomes more mature and specific technologies/research areas dominate the literature.

### ***3.3.3 Co-Citation Network of Journals***

Co-citation refers to any two publications that are cited by a different, third publication (Van Eck & Waltman, 2014). The greater the number of publications that cite the two publications, the larger the co-citation link strength between two publications. This is measured in terms of co-citations of journals/sources. The visualization of co-citation networks for CCS and CCU sources is presented in Figures 3.5a and 3.5b. The size of each circle refers to the number of citations received by the journal. Whereas the location of journals relative to others represent the strength of their co-citations. Co-cited journals, which show greater affinity as a result of being cited more frequently appear closer in clusters, as seen in the Figures.

A cluster represents a set of related nodes based on similarity. Each journal in this case is assigned to only one cluster. The specifics of the clustering technique and algorithms can be found in (van Eck & Waltman, 2010). CCS research (Figure 3.5a) shows four clusters, that differ in size and abundance. As discussed in the methodology (Section 3.2.2.2), Journal Citation Reports from the Web of Science Database (Clarivate, 2023a) was used to determine the categories for the respective clusters. The yellow cluster represents journals from geochemistry and geophysics. The red cluster mainly has energy and fuels research. The green cluster represents the chemical engineering area. The blue cluster primarily depicts the multidisciplinary science research area. In contrast, the CCU field (Figure 3.5b) only shows three clusters. The red cluster represents the chemical engineering field. The green cluster shows the energy and fuels field. The blue cluster represents the biotechnology and applied microbiology field. The common clusters in both CCS and CCU are energy and fuels, and chemical engineering (Figures 3.5a and 3.5b).

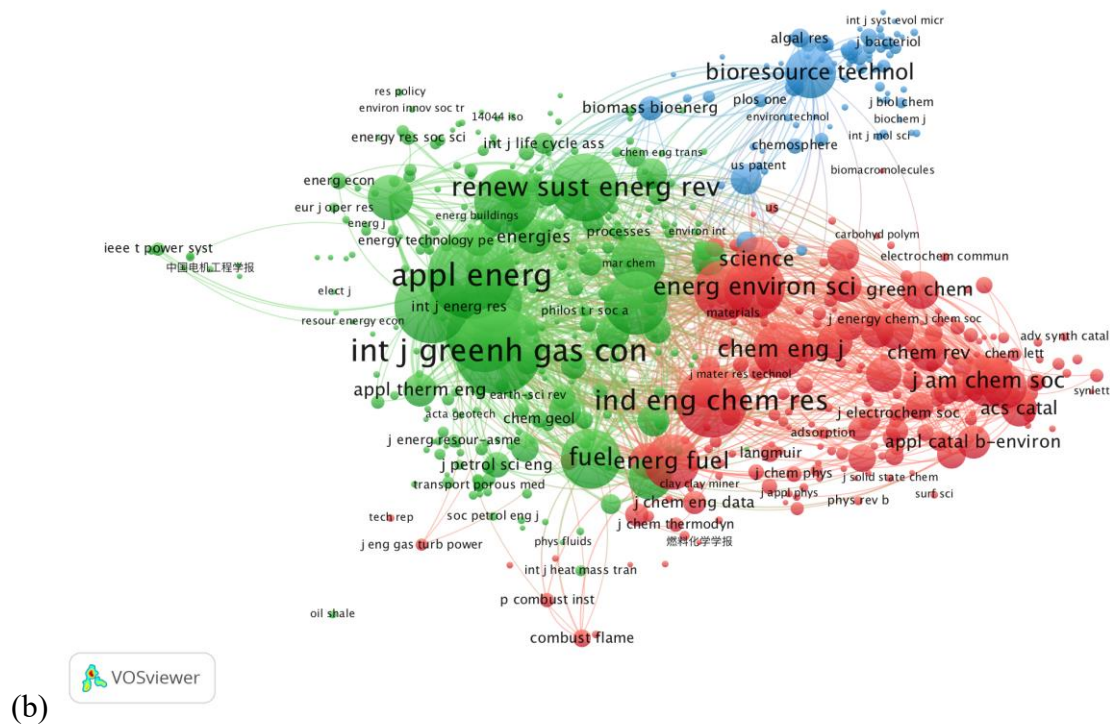
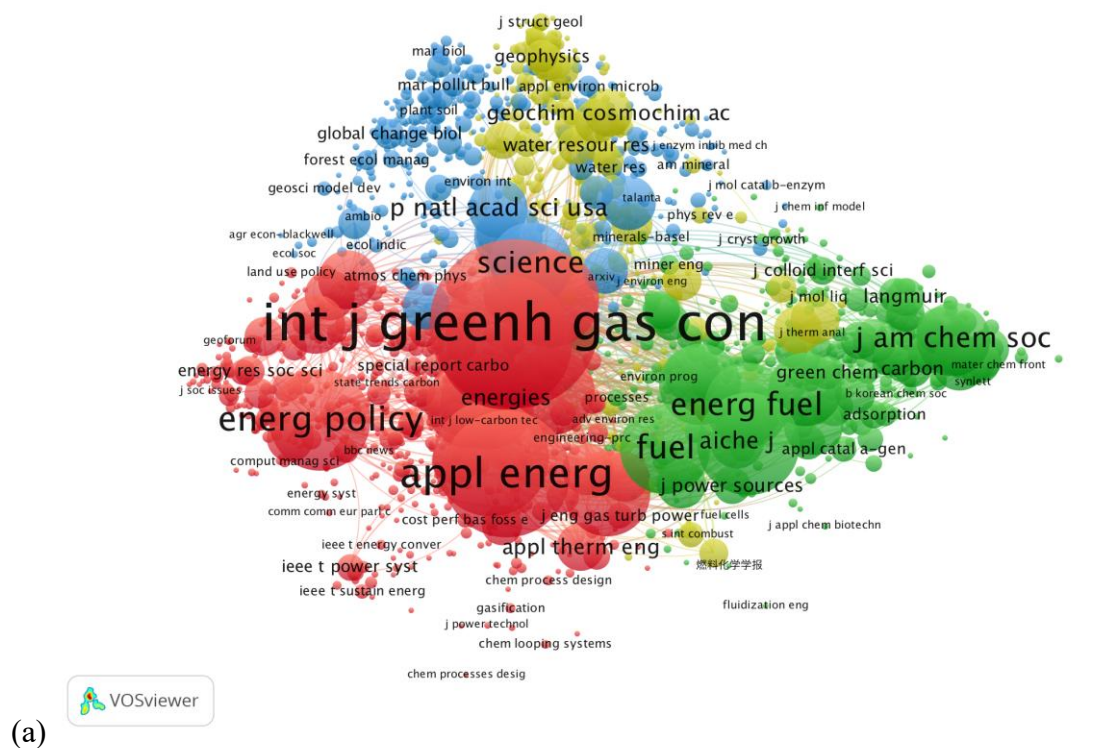


Figure 3.5 Visualization of co-citation network of journals for (a)CCS and (b)CCU using VOSviewer version 1.6.19. Data was extracted using fractional counting methods. Cluster resolution was selected as 0.7 for (a) and (b) with a minimum cluster size of 30 for (b).

The largest number of citations in the CCS field is found for the energy and fuels (red) cluster (Table 3.1). Specifically, *International Journal of Greenhouse Gas Control* has the highest citations, that being 18,577 (Table 3.1). It represents 5.70% of all citations in the CCS field. The second highest citation, in terms of abundance, is chemical engineering (green), followed by the multidisciplinary science (blue) cluster. The CCS cluster with the lowest citations is geochemistry and geophysics (yellow). Like in CCS, the cluster with the highest CCU citations is energy and fuels (green) (Table 3.2). Also, like in CCS, the highest number of CCU citations, were found for the *International Journal of Greenhouse Gas Control*, that being

Table 3.1 Citations, total link strength and citation percentage of clusters for CCS from Figure 3.5(a). Journals are presented based on the three highest citations and total link strengths for each cluster.

Cluster	Journal Name	Citations	Total Link Strength	Citation Percentage (%)
Energy & Fuels (Red)	<i>International Journal of Greenhouse Gas Control</i>	18577	15891.03	5.7
	<i>Energy Procedia</i>	11678	10439.46	3.58
	<i>Applied Energy</i>	10487	9156.1	3.22
Chemical Engineering (Green)	<i>Industrial &amp; Engineering Chemistry Research</i>	6795	6301.38	2.08
	<i>Fuel</i>	5316	4850.3	1.63
	<i>Energy &amp; Fuels</i>	4635	4342.5	1.42
Multidisciplinary Sciences (Blue)	<i>Science</i>	4358	4186.97	1.34
	<i>Nature</i>	2965	2863.53	0.91
	<i>Proceedings of the National Academy of Sciences of the United States of America</i>	2242	2176.96	0.69
Geochemistry & Geophysics (Yellow)	<i>Journal of Chemical &amp; Engineering Data</i>	1544	1413.51	0.47
	<i>Fluid Phase Equilibria</i>	1514	1342.53	0.46
	<i>Geochimica et Cosmochimica Acta</i>	1481	1345.72	0.45
Total				21.95

4,327 (Table 3.2). This journal represents 4.17% of all citations in the CCU field. The second highest cluster in terms of abundance of CCU citations is chemical engineering (red); this is also the second highest cluster in the CCS field. Lastly, the biotechnology & applied microbiology (blue) cluster had the lowest number of CCU citations (Table 3.2).

A total of 1,602 CCS sources matched the selection criteria in the methodology (Section 3.2.2.2) and are shown in Figure 3.5a. The CCS cluster with the most numerous nodes is energy and fuels (red), with 591 nodes. Followed by the chemical engineering (green) cluster, with 398 nodes. Each node represents a journal within the respective clusters. The multidisciplinary sciences (blue) cluster has 318 nodes. And the geochemistry and geophysics cluster has 294 nodes. As depicted, the clusters with the highest citation sources coincide with them having the most nodes; the ranking for the other clusters is identical to their citation ranking as well. A total

Table 3.2 Citations, total link strength and citation percentage of clusters for CCU from Figure 3.5(b). Journals are presented based on the three highest citations and total link strengths for each cluster.

Cluster	Journal Name	Citations	Total Link Strength	Citation Percentage (%)
Energy & Fuels (Green)	<i>International Journal of Greenhouse Gas Control</i>	4327	3800.7	4.17
	<i>Applied Energy</i>	4259	3640.04	4.1
	<i>Energy</i>	3086	2770.89	2.97
Chemical Engineering (Red)	<i>Industrial &amp; Engineering Chemistry Research</i>	2838	2652.63	2.73
	<i>Energy &amp; Environmental Science</i>	2068	1977.56	1.99
	<i>Journal of CO<sub>2</sub> Utilization</i>	2032	1906.08	1.96
Biotechnology & Applied Microbiology (Blue)	<i>Bioresource Technology</i>	1478	1259.62	1.42
	<i>Proceedings of the National Academy of Sciences of the United States of America</i>	570	556.88	0.55
	<i>Biomass &amp; Bioenergy</i>	341	331.13	0.33
Total				20.22

of 595 CCU sources are shown in Figure 3.5b. The CCU energy and fuels, and chemical engineering clusters have 291 and 224 nodes, respectively. The biotechnology and applied microbiology cluster has the remaining 79 nodes. Similar to the findings for the CCS field, the CCU energy and fuels cluster has the most nodes and citations.

Literature on bibliometrics of different areas within low-carbon research corroborates the work presented in this paper. The low-carbon market research papers found that the top three subjects in terms of total publications were energy and fuels, environmental sciences and environmental studies (Du, et al., 2015) (Wang, et al., 2017). Research in low-carbon technology investments also reveal similar patterns, where the most popular subjects are energy fuels, environmental science, ecology and engineering (Yu, et al., 2016). The energy and fuels subject from literature has the largest total publications and is also shown by this research to be the most prevalent. However, this research also identifies additional clusters (e.g. chemical engineering, multidisciplinary sciences, geochemistry & geophysics, biotechnology & applied microbiology) and makes comparisons between CCS and CCU research.

The total link strength is also displayed for each node, which indicates the total strength of the co-citation relationship of one journal (node) to other journals. Stronger co-citation relations result in higher total link strengths. The highest total link strengths for both CCS and CCU coincide with the energy and fuels cluster, indicating that journals in this cluster have the highest frequency of interaction with other journals. The CCS *International Journal of Greenhouse Gas Control* has a 52% higher total link strength than *Energy Procedia*, the second highest journal (Table 3.1).

Table 3.3 shows the co-citation link strength between different journals in the CCS field. The strongest link is found between the *International Journal of Greenhouse Gas Control* and



*Energy Procedia* journals (1449.78) indicating that these two journals are most frequently co-cited and share a high degree of interactions. It is also evident that 9 of the 10 linkages are within the energy and fuels (red) cluster. Indicating that this cluster appears frequently in co-citation relationship. Only one linkage involves the chemical engineering (green) cluster (Table 3.3). This indicates that the strongest CCS co-citation relations are found in one cluster- energy and fuels. 6 out of 10 linkages involve the *International Journal of Greenhouse Gas Control*, which further reiterates this finding in the CCS field.

Nine of the ten CCU linkages involve the energy and fuels (green) cluster as well (Table 3.4). Only one linkage involves the chemical engineering (red) cluster. The strongest co-citation link strength was determined to be between the *International Journal of Greenhouse Gas Control* and *Energy Procedia* journals (315.31) (Table 3.4). These trends are similar to those observed for the CCS field. They further corroborate the finding the strongest CCU co-citation relationships are also in the energy and fuels cluster.

Table 3.3 Link Strengths for top 10 linkages of journals presented for CCS from Figure 3.5(a).

Node 1	Node 2	Link Strength
<i>International Journal of Greenhouse Gas Control</i> (Red)	<i>Energy Procedia</i> (Red)	1449.78
<i>International Journal of Greenhouse Gas Control</i> (Red)	<i>Applied Energy</i> (Red)	671.2
<i>Applied Energy</i> (Red)	<i>Energy</i> (Red)	551.51
<i>International Journal of Greenhouse Gas Control</i> (Red)	<i>Energy</i> (Red)	493.89
<i>International Journal of Greenhouse Gas Control</i> (Red)	<i>Environmental Science &amp; Technology</i> (Red)	420.18
<i>Energy Procedia</i> (Red)	<i>Applied Energy</i> (Red)	405.02
<i>International Journal of Greenhouse Gas Control</i> (Red)	<i>Energy Policy</i> (Red)	386.23
<i>International Journal of Greenhouse Gas Control</i> (Red)	<i>Industrial &amp; Engineering Chemistry Research</i> (Green)	384.63
<i>Energy Procedia</i> (Red)	<i>Energy</i> (Red)	349.48
<i>Applied Energy</i> (Red)	<i>Renewable and Sustainable Energy Reviews</i> (Red)	327.78

### 3.3.4 Network of Highly Linked and Cited Documents

The evolution of work in CCS and CCU is examined using highly linked and cited documents to assess their similarities, prevalence and interconnectedness. The minimum citation was set to 300 and 200 for CCS and CCU, respectively, due to CCS having more records. The top 25 nodes for CCS and 17 nodes for CCU with the selection criteria discussed in Section 3.2.2.3 are shown in Figures 3.6a and 3.6b. Nodes that are not connected are not depicted in these images. The number of citations, publication year, key findings and country of the source for CCS and CCU are shown (Table 3.5). Out of the literature shown, 17 belong to CCS, 9 to CCU and 8 for both the CCS and CCU fields (denoted as CCUS). A total of 34 sources are shown in Table 3.5.

Table 3.4 Link Strengths for top 10 linkages of journals presented for CCU from Figure 3.5(b).

Node 1	Node 2	Link Strength
<i>International Journal of Greenhouse Gas Control (Green)</i>	<i>Energy Procedia (Green)</i>	315.31
<i>Applied Energy (Green)</i>	<i>Energy (Green)</i>	236.78
<i>International Journal of Greenhouse Gas Control (Green)</i>	<i>Applied Energy (Green)</i>	226.22
<i>Energy (Green)</i>	<i>Energy Conversion and Management (Green)</i>	185.47
<i>Applied Energy (Green)</i>	<i>Renewable &amp; Sustainable Energy Reviews (Green)</i>	150.8
<i>Energy Procedia (Green)</i>	<i>Applied Energy (Green)</i>	147.76
<i>International Journal of Greenhouse Gas Control (Green)</i>	<i>Industrial and Engineering Chemistry Research (Red)</i>	143.99
<i>International Journal of Greenhouse Gas Control (Green)</i>	<i>Energy (Green)</i>	142.76
<i>Energy Conversion and Management (Green)</i>	<i>Applied Energy (Green)</i>	140.16
<i>Applied Energy (Green)</i>	<i>Journal of Cleaner Production (Green)</i>	133.24

The most frequently cited documents present findings related to carbon capture, which is a component of both CCU and CCS (Table 3.5). The highly cited publication has 1,909 citations, it reviews solid adsorption capture systems for application of CCS (Choi, et al., 2009). Carbon capture is applicable to CCS due to its potential for utilization with power plants using CCS. There are many separation technologies such as separation with solvents/sorbents, membrane separation and cryogenic separation. Capture with chemical absorption using monoethanolamine is the most common method (Cuéllar-Franca & Azapagic, 2015) (Boot-Handford, et al., 2014). Another highly cited publication, has 1,569 citations. This discusses CCU transformations and CCS, it is commonly cited for both fields (Mikkelsen, et al., 2010) (Table 3.5). A common theme in the most cited documents is that of capturing CO<sub>2</sub> for transformation applications (Table 3.5). Such transformations include chemical, electrochemical, photochemical, biological, reforming, mineralization, EOR, fuels and others (Cuéllar-Franca & Azapagic, 2015) (Mikkelsen, et al., 2010). The majority of sources for this appear in the last decade, as discussed previously CCU is a fairly new and actively developing field. Another commonly discussed theme is power plants and their potential to employ CCS (Table 3.5). This is an important area of research for CCS because different capture options affect net plant outputs and project economics; both are required in promoting CCS for new and existing plants (Boot-Handford, et al., 2014) (Rao & Rubin, 2002). Studies also discuss the challenges with implementing carbon capture and storage, such as cost of capture, site monitoring and site capacity estimates (Haszeldine, 2009) (Aminu, et al., 2017). Improvements in technologies associated with implementing CCS, and innovation of low-cost methods of capturing CO<sub>2</sub> are needed to improve industrial outlook. Overcoming the environmental and techno-economic hurdles of CCS and CCU are also important considerations. Safety and risks of CCS are discussed in Table 3.5 as concerns to the public, especially with

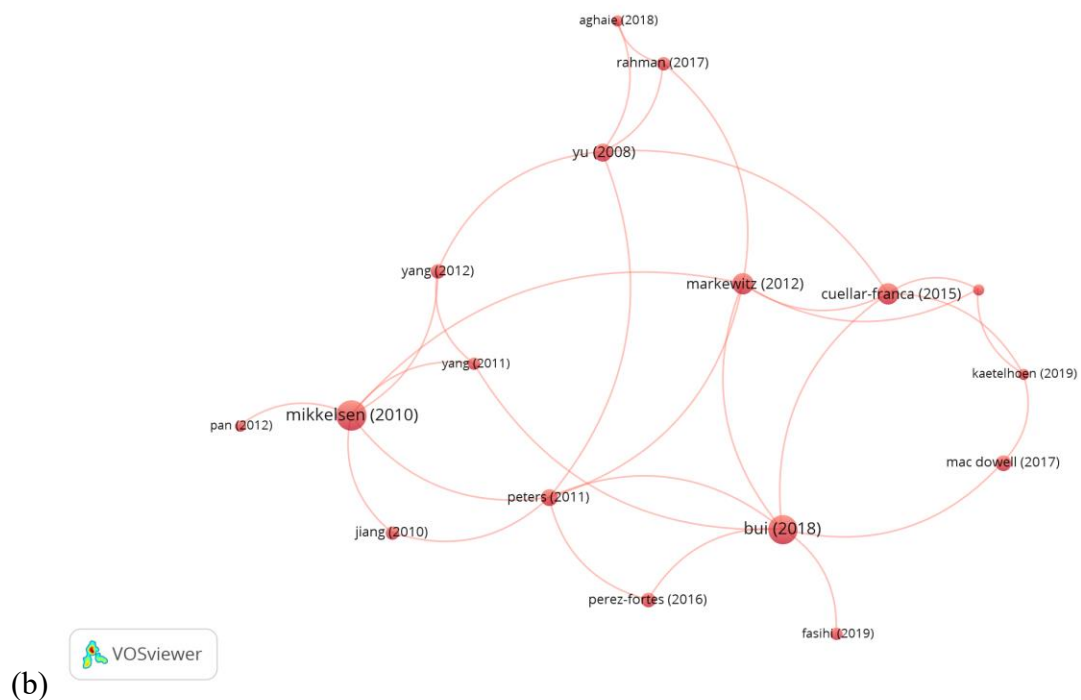
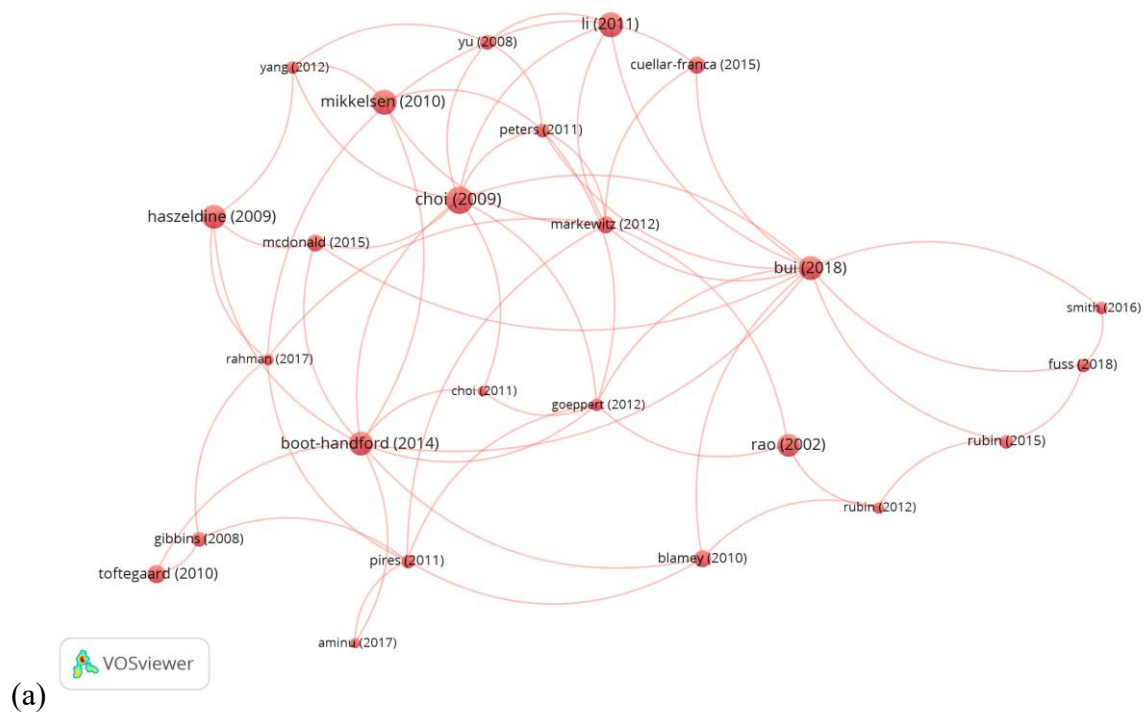


Figure 3.6 Network of top 25 (a) highly linked and cited documents for CCS with a minimum of 300 citations and (b) top 17 highly linked and cited documents for CCU with a minimum of 200 citations, using VOSviewer version 1.6.19.

Table 3.5 Year, cluster and key contribution of literature for the CCS and CCU fields from Figure 3.6, showing the most cited references in the CCS and CCU fields.

Field	Reference	Citations	Key Areas of Contribution	Country	Year
CCS	(Choi, et al., 2009)	1909	Review of behaviour of solid CO <sub>2</sub> adsorbent for large-scale CO <sub>2</sub> capture	USA	2009
CCS	(Li, et al., 2011)	1615	Review of metal-organic frameworks for CO <sub>2</sub> adsorption, storage and separation	USA	2011
CCUS	(Mikkelsen, et al., 2010)	1569	Six CCU transformations of CO <sub>2</sub> (chemical, photochemical, electrochemical, biological, reforming and inorganic) and brief CCS review	Denmark	2010
CCUS	(Bui, et al., 2018)	1536	Current state of art of CO <sub>2</sub> capture, transport, utilization and storage, including negative emissions technologies	UK, Germany, Switzerland, USA, Australia	2018
CCS	(Boot-Handford, et al., 2014)	1442	CO <sub>2</sub> capture technology for power plants, CCS, economics and policy	UK, Spain, Sweden, USA	2014
CCS	(Haszeldine, 2009)	1424	Technological, political and commercial challenges of point source CO <sub>2</sub> capture and storage	UK	2009
CCS	(Rao & Rubin, 2002)	1377	Technical, economic and environmental aspects of amine CO <sub>2</sub> absorption in power plants	USA	2002
CCS	(Toftegaard, et al., 2010)	846	Review of oxy-fuel process focusing on combustion fundamentals and highlighting areas of more research	Denmark	2010
CCS	(McDonald, et al., 2015)	816	Diamine-appended metal-organic framework adsorbents for removing CO <sub>2</sub> from gas mixtures	USA, China, Italy, France, Norway, Switzerland	2015
CCUS	(Cuéllar-Franca & Azapagic, 2015)	805	Comparison of life cycle assessment environmental impacts of CCS and CCU technologies	UK	2015

Table 3.5 (continued)

Field	Reference	Citations	Key Areas of Contribution	Country	Year
CCUS	(Markewitz, et al., 2012)	803	Status of research of three main CO <sub>2</sub> capture routes (post-combustion, oxyfuel, pre-combustion), transport and utilization	Germany	2012
CCS	(Blamey, et al., 2010)	748	Calcium looping technology to capture CO <sub>2</sub> from large power plants and integration with cement manufacturing	UK, Canada	2010
CCUS	(Yu, et al., 2008)	610	Promising research in CCS and chemical fixation of CO <sub>2</sub> into fuels, material (polymers) and chemicals	UK	2008
CCS	(Gibbins & Chalmers, 2008)	549	Investigating widespread commercial development of CCS and key advances for CO <sub>2</sub> capture technologies	UK	2008
CCUS	(Peters, et al., 2011)	540	Using multi-evaluation criteria to assess the impacts of CO <sub>2</sub> utilization for synthetic applications of fuel, chemical and material production	Germany	2011
CCS	(Rubin, et al., 2015)	482	Cost analysis of post-combustion CO <sub>2</sub> capture with supercritical coal and natural gas combined cycle, and pre-combustion capture with integrated gasification combined cycle	USA	2015
CCS	(Pires, et al., 2011)	480	Technical, economic, environmental and safety issues associated with CCS methodologies: CO <sub>2</sub> capture, transportation and storage	Portugal	2011
CCS	(Fuss, et al., 2018)	467	Costs, potential and effects of negative emission technologies: bioenergy-CCS, direct air capture, weathering, biochar, afforestation/deforestation and soil sequestration	Germany	2018
CCU	(Mac Dowell, et al., 2017)	465	Potential contributions of CCU options towards mitigation challenge and comparison with CCS and EOR	UK	2017
CCS	(Goeppert, et al., 2012)	430	Review of advantages and drawbacks of CO <sub>2</sub> capture from atmosphere	USA	2012

Table 3.5 (continued)

Field	Reference	Citations	Key Areas of Contribution	Country	Year
CCUS	(Yang, et al., 2012)	404	CCU mechanisms for CO <sub>2</sub> capture and chemical transformation with C-N bond formation pathway and CCS barriers	China	2012
CCU	(Pérez-Fortes, et al., 2016)	392	Techno-economic and environmental assessment of methanol synthesis using H <sub>2</sub> and captured CO <sub>2</sub> and conventional methanol synthesis	Netherlands, Germany	2016
CCS	(Smith, 2016)	399	Negative emission technologies potential of soil carbon sequestration and biochar addition to soil	UK	2016
CCS	(Rubin, et al., 2012)	338	Outlook for improved lower cost capture systems for power plants and industrial facilities and review of R&D programs	USA	2012
CCUS	(Rahman, et al., 2017)	327	Combining CCS and biofuel production using CO <sub>2</sub> as a feedstock	Malaysia, Saudi Arabia,	2017
CCU	(Jiang, et al., 2010)	324	Three approaches for synthetic fuel production: synthetic methanol, syngas and photochemical	UK	2010
CCS	(Aminu, et al., 2017)	310	CO <sub>2</sub> storage challenges, site selection criteria, CO <sub>2</sub> reservoir behaviour, techno-economic and public acceptance	UK	2017
CCS	(Choi, et al., 2011)	305	Solid amine-based absorbents for direct CO <sub>2</sub> capture from ambient air	USA	2011
CCU	(Yang, et al., 2011)	297	Ionic liquids for chemical capture of CO <sub>2</sub> for catalytic transformation into value-added organic compounds (solvents, fuels, chemicals, polymers)	China	2011
CCU	(Fasihi, et al., 2019)	283	Techno-economic analysis of high temperature aqueous solution direct air capture and low temperature solid sorbent direct air capture	Finland	2019
CCU	(Al-Mamoori, et al., 2017)	258	CCU challenges, opportunities, efficiency, economy and overview	USA	2017
CCU	(Pan, et al., 2012)	248	Accelerated carbonation of alkaline industrial wastes with captured CO <sub>2</sub>	Taiwan	2012

Table 3.5 (continued)

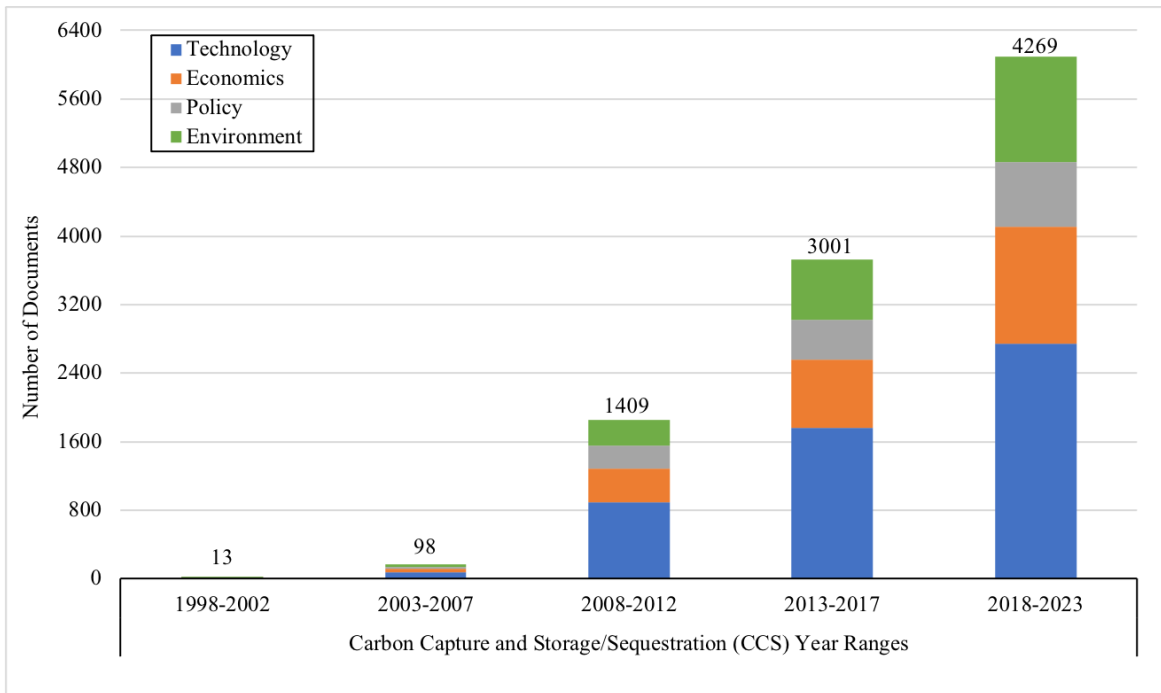
Field	Reference	Citations	Key Areas of Contribution	Country	Year
CCU	(Aghaie, et al., 2018)	233	Technical and economic aspects of CO <sub>2</sub> capture with ionic liquids for CCUS	Canada	2018
CCU	(Kätelhön, et al., 2019)	216	Modelling demands of CCU in chemical industry	Germany	2019



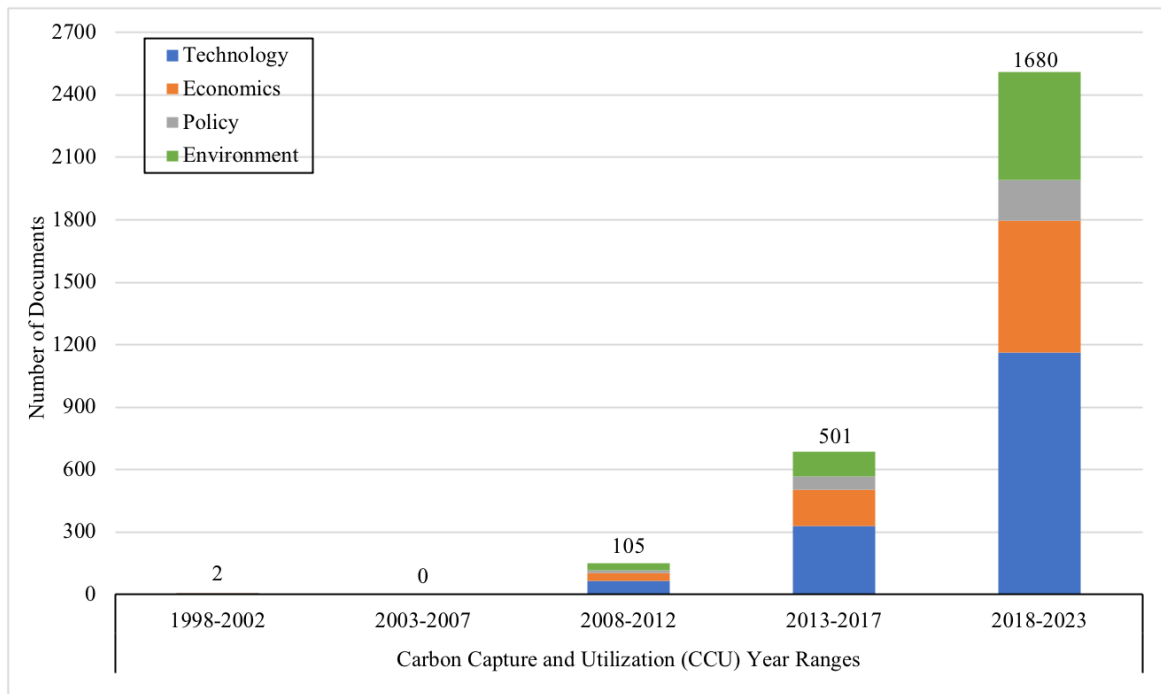
leakage into the surrounding environment (Cuéllar-Franca & Azapagic, 2015) (Aminu, et al., 2017). As evidenced here, CCS and CCU discuss many concepts within disciplines. However, several most cited studies presented in Table 3.5 are focused on carbon capture.

### ***3.3.5 Records by Four Topical Areas***

Four topical areas were examined in the CCS and CCU fields: technology, economics, policy and environment. Results show a drastic rise in the CCS research topics in 2008-2012 period compared to the previous periods (Figure 3.7a). The total CCS publications during this time period is 1,409, compared to 98 in 2003-2007 (Figure 3.2). Out of these four terms, “technology” had the most numerous records for the CCS search term (40-48%) compared to the other terms for all time periods considered (Figure 3.8). After that, economics is the second most abundant field (21-30%) (Figure 3.8). The economic records for 2018-2023 are half as abundant as technology. CCS policy records were found to be the lowest for 2008-2012, 2013-2017 and 2018-2023 time periods, that being 268, 463 and 747 records, respectively (Figure 3.7a). Similarly, compared to the other 3 fields, the largest CCU records are observed for the technology area (45-50%) (Figure 3.8). The second largest records are observed for the economic area (25-26%) (Figure 3.8). For the 2008-2012, 2013-2017 and 2018-2023 time periods the lowest records are for the CCU policy area, that is 12, 63 and 198 records, respectively (Figure 3.7b). The environment area accounts 17-21% of the CCU field and 15-20% of the CCS field. Records on policy are the least numerous of the 4 fields, that being 12-14% of the CCS field and 8-9% of the CCU field in the 3 most recent time periods (Figure 3.8). It is important to note that the prevalence of each research area doesn’t necessitate its importance or lack thereof in Figures 3.7 and 3.8. It is meant to show a comparison between the four topical areas examined in this study. It is also interesting that the relative ratios of research publications



(a)



(b)

Figure 3.7 Total number of records from 1998-2023 obtained from Web of Science for (a) CCS and (b) CCU for different time periods shown by 4 topical areas (technology – blue, economics – orange, policy – gray and environment – green). Labels show total number of publications for respective time periods.

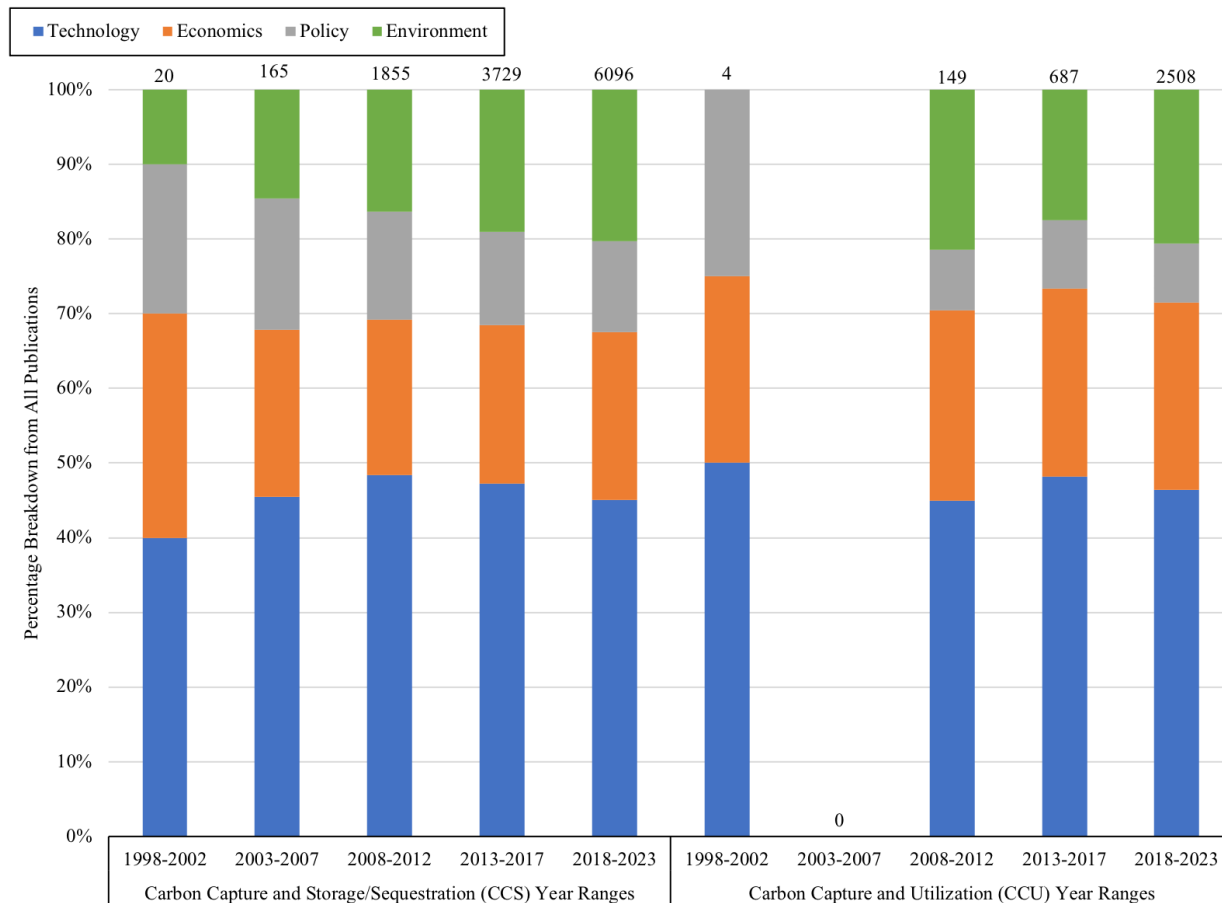


Figure 3.8 Total percentage (%) breakdown and number of documents of CCS and CCU publications from 1998-2023 obtained from Web of Science for different time periods shown by 4 topical areas (technology – blue, economics – orange, policy – gray and environment – green). Labels show sum of all topical areas for respective time periods.

amongst the four topics has remained relatively consistent over time (excluding the early periods for CCU). This observation indicates that all the research topic areas are important and are continually being advanced in an incremental fashion.

To study these 4 topical areas further, a total of 966 review papers in CCS and CCU fields are organized by most cited for 5 time periods: 1998-2002, 2003-2007, 2008-2012, 2013-2017 and 2018-2023. Once a maximum of 5 review papers that highly represent the topical areas are selected from each time period for each field (Section 3.2.3 & Figure 3.1), a total of 101 review papers are obtained. This accounts for review papers from 2007-2022 and 2008-2022 for CCS

and CCU, respectively. After accounting for duplicates within CCS, CCU and the 4 topical areas, a total of 89 review papers were read and analyzed in this paper. The results and discussion of the analysis are presented below and in Tables 3.6-3.9.

The analyzed policy research area literature shows 5 themes, these include strategies, funding (i.e., investments), cap and trade, carbon tax/price, and incentives/subsidies (Table 3.6). These papers shows high capital costs for CCS and CCU development and operations to be common challenge across the time periods examined (Jiang, et al., 2020) (Bui, et al., 2018) (Li, et al., 2019a) (Yuan & Lyon, 2012). Table 3.6 shows initially fewer specific categories exist in CCS (2007 to 2012), and none for CCU for the same time period. This is followed by appearance of several policy categories for CCS (2017 to 2021) and CCU (2018 to 2022) (Table 3.6). Many of the identified papers later on mention the need for more CCS and CCU policy supports, such as direct incentives, carbon tax and subsidies (Lee & Haites, 2012) (Babatunde, et al., 2017) (Haszeldine, et al., 2018) (Waller, et al., 2020) (Akerboom, et al., 2021) (Babin, et al., 2021) (Busch, et al., 2022). Although both CCS and CCU require policy supports, CCU policy supports for small-scale development are identified as almost non-existent, as such lowering eligibility for the 45Q tax credit is recommended (Warsi, et al., 2020).

The technology research area findings are organized in 7 categories. These include CCS geologic/oceanic storage, EOR/enhanced gas recovery (EGR)/enhanced coal bed methane recovery (ECBM), chemicals, fuels, materials, biogenic, and others (i.e., food) (Table 3.7). Results show CCU technical issues, such as efficiency, catalysts, reaction rates and energy consumption to be consistent across the time frames studied (Yang, et al., 2012) (Liew, et al., 2016) (Al-Mamoori, et al., 2017) (Norhasyima & Mahlia, 2018) (Zhang, et al., 2020). CCS and CCU topical areas discuss EOR in the early years, starting in 2007 for CCS and 2008 for CCU

Table 3.6 Selected review papers for CCS and CCU policy research area from Figures 3.7 & 3.8, their sources, publication year and number of citations organized by policy research area categories

Field	Source	Year	Citations	Strategies	Funding	Cap and Trade	Carbon Tax/Price	Incentives/ Subsidies
CCS	(Martinot, et al., 2007)	2007	86	✓				
	(Valkila & Saari, 2010)	2010	29	✓				
	(Rahman & Khondaker, 2012)	2012	49	✓				
	(Yuan & Lyon, 2012)	2012	10	✓	✓			
	(Lee & Haites, 2012)	2012	2	✓	✓		✓	✓
	(Zeng, et al., 2014)	2014	49	✓				✓
	(Babatunde, et al., 2017)	2017	112			✓	✓	✓
	(Haszeldine, et al., 2018)	2018	143				✓	✓
	(Li, et al., 2019a)	2019	30				✓	✓
	(Waller, et al., 2020)	2020	24			✓	✓	
	(Babin, et al., 2021)	2021	34	✓	✓	✓		✓
CCU	(Jones, et al., 2017)	2017	53		✓			
	(Bui, et al., 2018)	2018	1641	✓			✓	✓
	(Warsi, et al., 2020)	2020	8	✓	✓		✓	✓
	(Akerboom, et al., 2021)	2021	14	✓	✓			✓
	(Busch, et al., 2022)	2022	18	✓	✓	✓	✓	✓
CCUS	(Jiang, et al., 2020)	2020	90	✓				✓

for the literature examined in this study (Race, et al., 2007) (Radosz, et al., 2008). Additionally, CCU sources discuss chemicals and fuels early on (2010 to 2016) followed by materials and biogenic pathways in more recent years (2015 to 2021) (Table 3.7). There are also limited studies on the technological mechanisms of chemical transformations, such as a few types of carbamates from CO<sub>2</sub>, as well as environmental impacts of wastewater treatment with CCU (Baena-Moreno, et al., 2019) (Gude, 2016) (Lu Lu, et al., 2018). Currently, short term focus is suggested for building materials, chemicals, fuels and polymers, and long term for other CCU technologies, as they improve market, cost and technical barriers (Baena-Moreno, et al., 2019) (Norhasyima & Mahlia, 2018) (Zhang, et al., 2020).

The environmental research area is organized into 4 categories: emissions to air (e.g., CO<sub>2</sub> emissions and ozone depletion), emissions to water (e.g., causing acidification and eutrophication), other effects (e.g., human toxicity and contamination) and risks of storage/transport (e.g., due to leakage) (Table 3.8). Life cycle assessments are commonly seen from 2007 onwards in CCS and 2010 for the CCU literature examined in this study (Viebahn, et al., 2007) (Collins, 2010). Overall, both CCS and CCU reduce CO<sub>2</sub> (global warming potential-GWP) emissions compared to no mitigation options (Markewitz, et al., 2012) (Bobicki, et al., 2012) (Salas, et al., 2016) (Treyer & Bauer, 2016) (Matustik, et al., 2020) (Yu, et al., 2008) (Lam, et al., 2012) (Olajire, 2013) (Zhu, 2019) (Garcia-Garcia, et al., 2021). Although CCU's GHG reduction potential is realized early on (2008), other CCU impacts such as acidification and eutrophication are discussed being higher compared to incumbents mainly in later years from 2015 to 2022 (Table 3.8) (Cuéllar-Franca & Azapagic, 2015) (Thonemann, 2020) (Li, et al., 2022). The environmental impacts for CCU technologies, however, can be lowered when combined with renewable energy and hydrogen from water electrolysis (Jarvis & Samsatli, 2018)

Table 3.7 Selected review papers for CCS and CCU technology research area from Figures 3.7 & 3.8, their sources, publication year and number of citations organized by technology research area categories

Field	Source	Year	Citations	CCS Geologic/ Oceanic	EOR/ EGR/ ECBM	Chemical	Fuels	Materials	Biogenic	Others
CCS	(Balat & Öz, 2007)	2007	37	✓	✓			✓		
	(Haszeldine, 2009)	2009	1440	✓	✓					
	(Shukla, et al., 2010)	2010	321	✓	✓					
	(Jones, 2011)	2011	194	✓						
	(Rubin, et al., 2012)	2012	343	✓						
	(Boot-Handford, et al., 2014)	2014	1472		✓	✓	✓			✓
	(Bachu, 2015)	2015	218	✓						
	(Aminu, et al., 2017)	2017	335	✓	✓					
	(Minx, et al., 2018)	2018	340						✓	✓
	(Ajayi, et al., 2019)	2019	115	✓	✓					
(Snæbjörnsdóttir, et al., 2020)	2020	147	✓				✓			
CCU	(Jiang, et al., 2010)	2010	329				✓			
	(Peters, et al., 2011)	2011	543		✓	✓	✓			✓
	(Yang, et al., 2012)	2012	410			✓				
	(Liew, et al., 2016)	2016	238				✓		✓	
	(Li, et al., 2016)	2016	46					✓	✓	
	(Al-Mamoori, et al., 2017)	2017	277		✓	✓	✓	✓		
	(Norhasyima & Mahlia, 2018)	2018	155		✓	✓	✓	✓	✓	✓
	(Baena-Moreno, et al., 2019)	2019	163		✓	✓	✓	✓	✓	✓
	(Zhang, et al., 2020)	2020	218		✓	✓	✓	✓	✓	✓
CCUS	(Markewitz, et al., 2012)	2012	819	✓	✓					
	(Cuéllar-Franca & Azapagic, 2015)	2015	827	✓	✓	✓	✓	✓	✓	
	(Rahman, et al., 2017)	2017	344	✓		✓	✓			
	(Bui, et al., 2018)	2018	1641	✓	✓	✓	✓	✓	✓	✓
	(Osman, et al., 2021)	2021	191	✓	✓	✓	✓	✓	✓	✓

Table 3.8 Selected review papers for CCS and CCU environmental research area from Figures 3.7 & 3.8, their sources, publication year and number of citations organized by environmental research area categories

Field	Source	Year	Citations	Emissions to Air	Emissions to Water	Other effects	Risks of storage/transport
CCS	(Markewitz, et al., 2012)	2012	820	✓	✓	✓	
	(Bobicki, et al., 2012)	2012	367	✓			
	(Koorneef, et al., 2012)	2012	120	✓	✓	✓	✓
	(Salas, et al., 2016)	2016	123	✓	✓	✓	✓
	(Treyer & Bauer, 2016)	2016	32	✓	✓	✓	
	(Ko, et al., 2016)	2016	25		✓	✓	✓
	(Liu & Ramirez, 2017)	2017	19	✓	✓	✓	✓
	(Tcvetkov, et al., 2020)	2019	54				✓
	(Li, et al., 2019b)	2019	38				✓
	(Matustik, et al., 2020)	2020	58	✓	✓	✓	
	(Terlouw, et al., 2021)	2021	59	✓	✓	✓	
(Chen, et al., 2022)	2022	29				✓	
CCU	(Yu, et al., 2008)	2008	622	✓			
	(Lam, et al., 2012)	2012	208	✓			
	(Olajire, 2013)	2013	264	✓	✓	✓	
	(Gressel, et al., 2013)	2013	39			✓	
	(Deleebeeck & Hansen, 2014)	2014	52	✓			
	(Jang, et al., 2016)	2016	129	✓			
	(Jarvis & Samsatli, 2018)	2018	108	✓		✓	
	(Zhu, 2019)	2019	96	✓			
	(Thonemann, 2020)	2020	42	✓	✓	✓	
	(Garcia-Garcia, et al., 2021)	2021	26	✓	✓	✓	
	(Li, et al., 2022)	2022	39	✓	✓	✓	
CCUS	(Cuéllar-Franca & Azapagic, 2015)	2015	827	✓	✓	✓	✓



(Garcia-Garcia, et al., 2021). In contrast, CCS is still discussed with the same risks that it did early on (from 2012 to present), that being contamination due to release of CO<sub>2</sub> and seismicity (Koornneef, et al., 2012) (Tcvetkov, et al., 2020) (Chen, et al., 2022) (Table 3.8).

The economic research area is found to have 5 categories: industry integration (i.e., with power plants, renewables or cement plants), carbon capture costs, storage costs, transportation costs and economic benefits (Table 3.9). Economic and cost analysis are commonly seen from 1999 and 2002 in CCS and CCU literature examined in this study, respectively (Dooley, et al., 1999) (Read, 2002). Table 3.9 shows more CCS sources mention CO<sub>2</sub> capture cost as the predominant cost for CCS operations (since 2007) (Balat & Öz, 2007) (Han, et al., 2012) (Tang, et al., 2014) (Singh & Dhar, 2019), while CCU sources discuss added costs with conversion of CO<sub>2</sub> (Gude, 2016) (Mondal, et al., 2017) (Bataille, et al., 2018) (Bhatia, et al., 2019) (Chauvy & De Weireld, 2020). Early and late results, specifically 2007 to 2022 and 2008 to 2020, for CCS and CCU, respectively show mitigating high CCU and CCS costs by integration with power plants, renewable energy strategies, and industrial plants (Table 3.9) (Beer, 2007) (Wu, et al., 2016) (Ma, et al., 2014) (Sreenivasulu & Sreedhar, 2015) (Mondal, et al., 2017) (Lin, et al., 2020). Currently, some C1 chemicals (formic acid) are cost competitive with conventional fossil processes (Overa, et al., 2022) (Lin, et al., 2020). Other CCU technologies need incentives and carbon taxes to gain economic advantage and competitiveness (Overa, et al., 2022) (Bhatia, et al., 2019). EOR is identified to offset costs and appears early in the evolution of both fields, that being 2007 and 2008 for CCS and CCU, respectively (Table 3.9) (Balat & Öz, 2007) (Yu, et al., 2008). Additionally, building plants close to CO<sub>2</sub> sources decreases transportation costs for CCS options (Onyebuchi, et al., 2018).

Table 3.9 Selected review papers for CCS and CCU economic research area from Figures 3.7 & 3.8, their sources, publication year and number of citations organized by economic research area categories

Field	Source	Year	Citations	Industry Integration	Carbon Capture Costs	Storage Costs	Transportation Costs	Economic Benefit
CCS	(Balat & Öz, 2007)	2007	37	✓	✓	✓	✓	✓
	(Beer, 2007)	2007	390	✓	✓			
	(Hadjipaschalis, et al., 2009)	2009	44	✓	✓		✓	
	(Lenzen, 2010)	2010	74	✓		✓		
	(Williams, et al., 2011)	2011	13	✓	✓	✓		✓
	(Han, et al., 2012)	2012	13	✓	✓	✓	✓	
	(Streimikiene, 2012)	2012	5			✓		
	(Cusack, et al., 2014)	2014	21	✓		✓		
	(Tang, et al., 2014)	2014	12			✓	✓	
	(Wu, et al., 2016)	2016	52	✓	✓			✓
	(Leeson, et al., 2017)	2017	243	✓	✓			
	(Anderson, 2017)	2017	23		✓	✓	✓	
	(Aghaie, et al., 2018)	2018	248		✓			
	(Onyebuchi, et al., 2018)	2018	62	✓			✓	
	(Singh & Dhar, 2019)	2019	100			✓	✓	
	(Kárászová, et al., 2020)	2020	73			✓		
(Gür, 2022)	2022	67	✓	✓	✓	✓		
CCU	(Yu, et al., 2008)	2008	622	✓	✓			
	(Xie, et al., 2014)	2014	70					✓
	(Ma, et al., 2014)	2014	24	✓				✓
	(Sreenivasulu & Sreedhar, 2015)	2015	75	✓	✓			
	(Gude, 2016)	2016	275					
	(Mondal, et al., 2017)	2017	120	✓				
	(Bataille, et al., 2018)	2018	193					
	(Bhatia, et al., 2019)	2019	93		✓			
	(Lin, et al., 2020)	2020	43	✓				✓
	(Chauvy & De Weireld, 2020)	2020	40					
(Overa, et al., 2022)	2022	30					✓	

### ***3.3.6 Landscape of CCS and CCU Projects and Research***

Commercial CCS facilities, and short-lived pilot and demonstration facilities as of February 2023 are depicted in Figure 3.9. The top countries with the highest number of completed and in development CCS projects (and their total projects) are USA (116), United Kingdom (50), Canada (22), China (21), Australia (17), Norway (15) and Netherlands (14) (Figure 3.9). Germany and South Korea also have 3 projects. The USA leads in number of operational CCS facilities with 37. Followed by China (15), Canada (11), Australia (6), Norway (5) and United Kingdom (5). The total number of completed and in development CCS facilities show similar patterns with the research documents. The top 3 countries in abundance of CCS documents are USA, UK and China (Figure 3.9). Canada has the third highest number of completed and in development CCS projects, and 6<sup>th</sup> highest number of documents (Figure 3.9).

The total number of CCU projects are higher than completed and in development CCS projects. Figure 3.9 shows the number of CCU start-up companies. The top countries in abundance of CCU projects are USA (110), Canada (21), Germany (13), United Kingdom (13), Netherlands (11), Belgium (7), Norway (7) and Australia (6) (Figure 3.9). The most abundant CCU documents belong to China, USA, UK and Germany (Figure 3.9). USA leads in the number of CCU start-up companies, while China only has 2 CCU projects but the highest number of documents (601 documents) (Figure 3.9). Canada has the second highest CCU projects (21), and seventh highest CCU documents (Figure 3.9). The UK and Germany have the third highest CCU projects, and third and fourth highest documents, respectively (Figure 3.9).

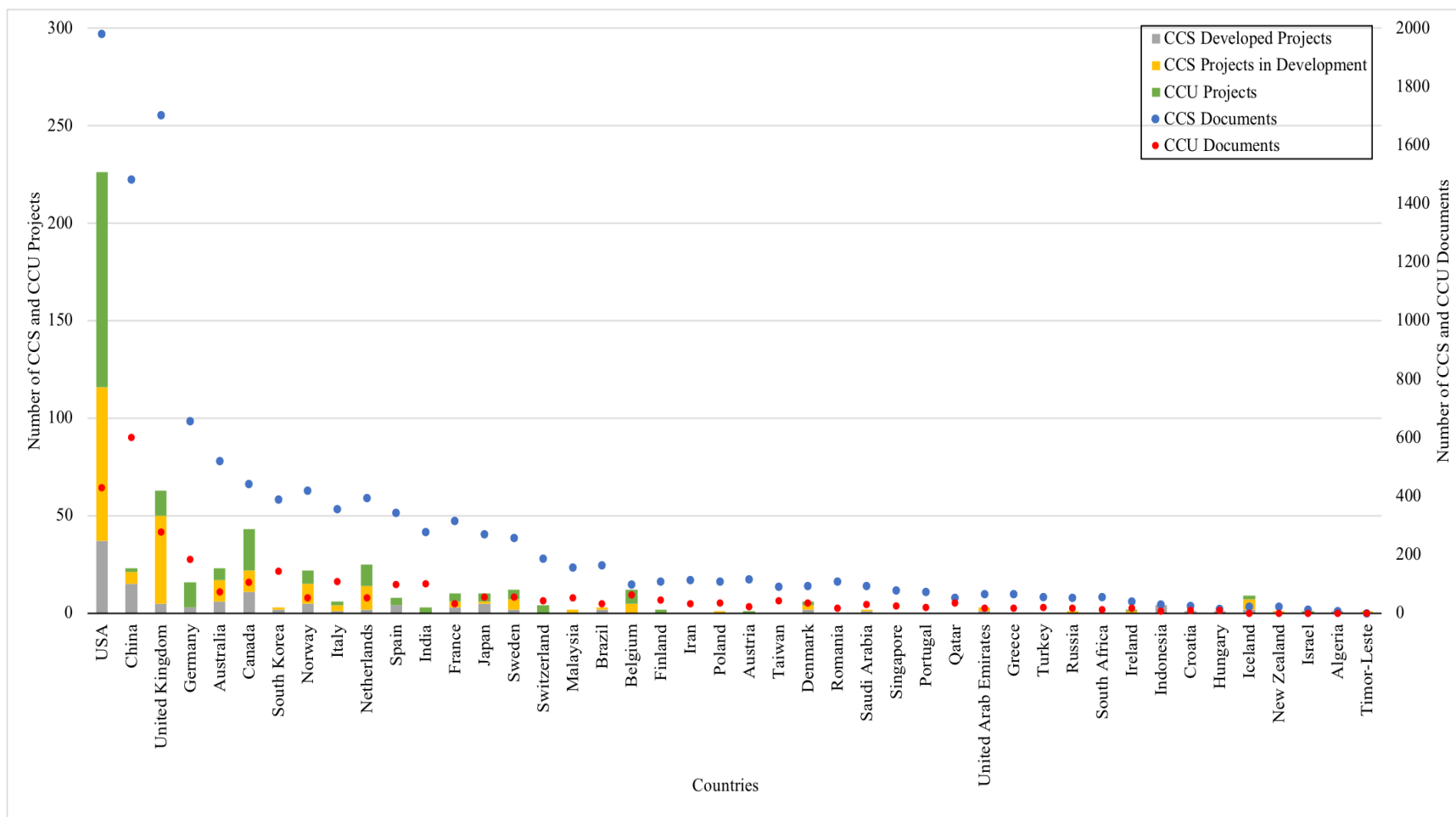


Figure 3.9 Countries showing number of CCS documents, CCU documents, CCS facilities currently completed/operational and under construction/development and CCU start-up companies. Data is organized by number of total documents. Number of commercial, pilot and demonstration CCS facilities currently completed/operational and under construction/development was gathered from Global CCS Institute (Global CCS Institute, 2022). Number of start-up carbon dioxide utilization companies' data was obtained from the Global CO<sub>2</sub> Initiative-University of Michigan (Global CO<sub>2</sub> Initiative, 2020)

Table 3.10 Annual carbon dioxide emissions released by countries in Gigatons of CO<sub>2</sub> equivalents (GtCO<sub>2</sub>) (Ritchie & Roser, 2021) and gross domestic product (USD) (The World Bank, 2021), number of CCS documents, CCU documents, CCS facilities currently completed/operational and under construction/development and CCU start-up companies are shown

Country	CCS Documents	CCU Documents	CCS Developed Projects	CCS Projects in Development	CCU Projects	Emissions (Gt CO <sub>2</sub> )	GDP (Trillion US\$)
USA	1980	428	37	79	110	5.01	23.32
China	1483	601	15	6	2	11.47	17.73
United Kingdom	1702	277	5	45	13	0.35	3.13
Germany	657	185	3	0	13	0.67	4.26
Australia	519	72	6	11	6	0.39	1.55
Canada	441	106	11	11	21	0.55	1.99
South Korea	388	144	2	1	0	0.62	1.81
Norway	420	52	5	10	7	0.041	0.48
Italy	355	108	1	3	2	0.33	2.11
Netherlands	393	52	2	12	11	0.14	1.01
Spain	344	98	4	0	4	0.23	1.43
India	278	101	0	0	3	2.71	3.18
France	316	32	3	3	4	0.31	2.96
Japan	271	56	5	1	4	1.07	4.94
Sweden	258	54	2	5	5	0.036	0.64
Switzerland	186	42	0	0	4	0.034	0.80
Malaysia	155	53	0	2	0	0.26	0.37
Brazil	164	32	2	1	0	0.49	1.61
Belgium	99	64	0	5	7	0.096	0.59
Finland	109	46	0	0	2	0.038	0.30

Many barriers presently impede the development of these projects. High cost of development and scaling are discussed as major ones (Olfe-Kräutlein, 2020) (Kant, 2017).

The country specific emissions are shown in Table 3.10. Results show China is the largest emitter at 11.47 Gigatons (Gt) CO<sub>2</sub> in 2021, followed by the USA at 5.01 Gt of CO<sub>2</sub>. These two countries also have the highest gross domestic product (GDP), specifically, 23.32 Trillion USD and 17.73 Trillion USD for USA and China, respectively. India is the third highest emitter of CO<sub>2</sub> at 2.71 Gt as well as the fifth highest GDP at 3.18 Trillion USD. As observed previously, USA leads CCS facilities and CCU projects. Although China is the highest emitter, it has 21 CCS projects and 2 CCU projects, compared to the USA's 116 CCS and 110 CCU projects. Likewise, India emits 2.71 Gt CO<sub>2</sub> and a GDP of 3.18 Trillion USD and does not appear in the top 10 countries for CCS and CCU facilities nor total documents (Table 3.10 & Figure 3.9). Similarly, Japan's emissions are at 1.07 Gt CO<sub>2</sub>, with a GDP of 4.94 Trillion USD (third highest) and does not appear in the top 10 countries for CCS and CCU facilities nor total documents (Table 3.10). Germany appears in the top 5 total CCS and CCU documents area, has the fourth highest GDP (4.24 Trillion USD), however it has the eight highest total CCS and CCU projects (Table 3.10). On the other hand, even though Canada is the 7<sup>th</sup> highest emitter of CO<sub>2</sub> emissions (0.55 Gt of CO<sub>2</sub>), it has the third highest CCS projects (22), and second highest CCU projects (21) (Table 3.10, Figure 3.9). Similarly, the United Kingdom has the 10<sup>th</sup> highest emissions (0.35 Gt of CO<sub>2</sub>), the second most abundant CCS facilities (50) and third most numerous CCU projects (13). This shows a trend that some low emissions ranked profile countries are leading in CCS and CCU projects while some high emission countries are not leading project development. Given that CCS is expected to mitigate 14-20% of global anthropogenic CO<sub>2</sub> emissions by 2050 (Mac Dowell, et al., 2017), and CCU is expected to utilize 20% of global CO<sub>2</sub> emissions by 2050

(Sapart, et al., 2022), countries should address major barriers of CCS and CCU cost and marketability to increase development.

The size of nodes of the co-authorship map of countries reflects the number of documents. The level of collaboration between authors of different countries is shown by lines. Out of the 112 countries with CCS co-authorships to other countries in the WoS records, 73 met the threshold criteria in Section 3.2.2.4, and are shown in Figure 3.10a. Out of the 88 countries relevant to the CCU field, 53 met the threshold criteria in Section 3.2.2.4, and are shown in Figure 3.10b. The co-authorship relationship for countries in the fields are from 1998-2023. The smaller the distance between two nodes, the more related their co-authorship relationship.

The 5 countries with the highest number of CCS documents are: USA (1,980), China (1,483), England (1,287), Germany (657) and Australia (519) (Figure 3.9). The total link strength of these countries is 799, 606, 727, 331 and 268, respectively. This indicates the total strength of the co-authorship links of researchers of one country to other countries. These countries also have the highest total citations, specifically, USA (74,854), England (41,508), China (30,110), Germany (20,942) and Australia (19,411). The top 5 country co-authorship relationships (and respective link strengths) are USA and China (167.91), England and China (102.28), England and Scotland (96.53), USA and England (65.21), and USA and Canada (65.21). Thus, most of the top co-authorship links and citations are in countries that also have the highest number of CCS documents.

The top 5 countries with the highest number of CCU documents are China (601), USA (428), England (207), Germany (185) and South Korea (144) (Figure 3.9). The total link strengths of these countries are 207, 164, 134, 77 and 38, respectively. Additionally, the largest citations are for USA (12,187), China (10,641), England (8,453), Germany (7,675) and Australia (4,728).

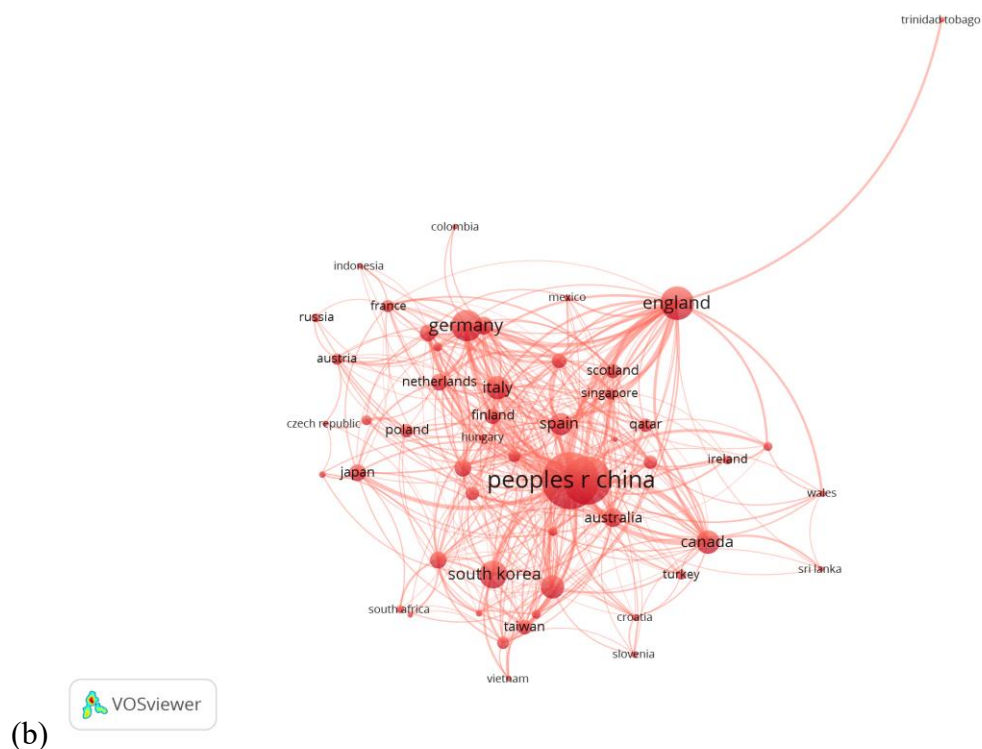
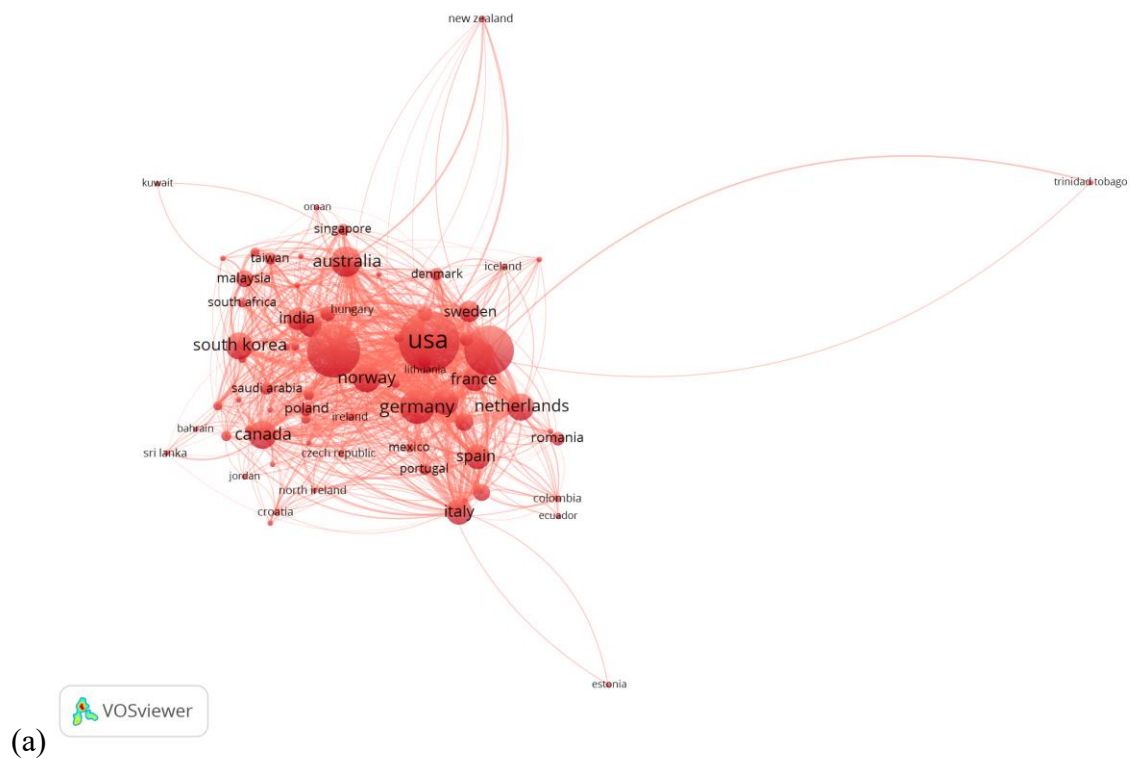


Figure 3.10 Visualization of the co-authorship network of countries for the (a) CCS and (b) CCU fields using VOSviewer version 1.6.19. Minimum number of documents for a country were selected as 5. Fractional counting method was used



Although USA is highly cited in CCS and CCU, China leads document abundance in the CCU field. South Korea has a higher ranking (5<sup>th</sup>) for abundance of CCU documents compared to CCS (9<sup>th</sup>) (Figure 3.9). The 5 strongest country co-authorship CCU relationships (and respective link strengths) are USA and China (50.65), England and China (24.87), Canada and USA (16.34), England and Spain (14.83), and Canada and China (14.4). The countries with the top two link strengths are the same for both CCS and CCU, that being (1) USA and China and (2) England and China. China appears in 3 of the top 5 CCU linkages, whereas USA appears in 3 of the top 5 CCS linkages. Canada has the sixth highest CCS documents (441) with 10,983 citations and seventh highest CCU documents (106) with 2,494 citations. It also appears in the top 5 CCU linkages in both fields. The countries with the highest number of co-authorship occurrences, citations and link strengths in CCS and CCU are USA, China and England (not organized by citations or link strengths).

### **3.4 Conclusions and Limitations**

An in-depth bibliometric analysis and meta-analysis carbon capture storage and carbon capture utilization was carried out using Web of Science data. Relationships were determined for the co-occurrence of terms, temporal distribution of publications, co-citation network of journals, network of highly linked and cited publications, findings from four topical areas- policy, technology, environment and economy, co-authorship of countries, and landscape of CCS and CCU projects. Topical areas and future direction for research and development are identified.

The CCS co-occurrence network of terms data identifies clusters on policy and perception, CO<sub>2</sub> injection and storage, CO<sub>2</sub> capture and power requirements, and CO<sub>2</sub> chemistry. The CCU co-occurrence network of terms data has clusters associated with carbon capture and carbon storage in general, and carbon conversion of CCU. Terms associated with CCS policy show

fewer lines to the other clusters. The CCU field developed after and has less publications than CCS. Trends show a sustained interest in CCU research, and more growth in CCS research. The highest CCS documents were found for USA, United Kingdom and China. The highest CCU documents were China, USA and United Kingdom. Results show the highest completed and in-development CCS projects in USA, United Kingdom and Canada (Figure 3.9). The most numerous CCU start-up companies are in USA, Canada, Germany and United Kingdom (Figure 3.9).

Clustering data for the co-citation network of journals has identified the following CCS clusters: energy & fuels, chemical engineering, multidisciplinary sciences, and geochemistry & geophysics. The following clusters have been identified for the CCU field: energy & fuels, chemical engineering, and biotechnology & applied microbiology. The highest citations for CCS and CCU were observed for the *International Journal of Greenhouse Gas Control*, which belongs to the energy and fuels cluster. This cluster also has the highest total link strengths (Tables 3.1 and 3.2).

The research findings from the most cited and research area meta-analysis show the multidisciplinary nature of CCU and CCS research. The policy research area is the least contributing field to CCS and CCU when compared to other areas such as technology, economics and environment (Figures 3.7a and 3.7b). The economic topical research area shows that both CCS and CCU have high capital costs. The policy research area also discusses that more CCS and CCU policy supports, such as direct incentives, cap and trade, carbon tax and subsidies are needed. The technology research area shows that CCU technologies face technological issues due to efficiency, catalysts, reaction rates and energy consumption. Environmental issues of CCS discuss risks of storage and leakage, whereas CCU having higher

non-GWP impacts (such as acidification and eutrophication) are discussed. In the future, increasing lines between clusters can enhance interdisciplinarity.

Finally, the study's limitation should be discussed. The research database is limited to Web of Science. Even though it is comprehensive, global and contains articles from the 1900s, those articles that are not in Web of Science may be overlooked. Additionally, the search string mentioned in the methodology may miss literature that discuss carbon capture and utilization and carbon capture and storage, which do not state the terms in the search string. For example, phrases such as "carbon capture and conversion" may be missed if they don't have utilization or storage terms. Specifically, this phrase accounts for 40 documents from the Web of Science database. Moreover, some authors may use different names or common names, which can lead to inaccurate tallying for the authorship maps. These limitations can be improved in future work by increasing the scope of research to include other databases, while removing and accounting for duplicates. Finally, it is important to note that the relevant research area documents presented don't allow for assumptions to be made about relevance and/or need of a research area compared to others; the purpose of this research is not to assess if each of these research areas are adequate.

The overall goal of this research is to study the evolution of research in CCS and CCU using bibliometric techniques and meta-analysis. The bibliometric methods show temporal results for CCS having higher maturity compared to CCU. Parallels are also made between CCS and CCU depicting similar areas for co-citation network of journals (e.g. energy & fuels, and chemical engineering). When compared with industry data, parallels can be drawn with the prevalence of research documents, revealing geographical trends in data. Additionally, the topical areas and highly cited literature investigation reveals the evolution of CCS and CCU, as

well as gaps in CCU which show a lack in carbamate synthesis and waste-water treatment pathways, this evidently suggests directions for future research.

It is hoped that this bibliometric study and meta-review will provide a good introduction and summary of the research and development landscape for both CCS and CCU for a number of relevant stakeholder groups including researchers, government, industry (e.g. technology developers, end users, industry associations), funding agencies, and the greater community. This work also advances the academic understanding of research evolution in this space, and provides a resource for those studying knowledge generation and transfer.

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## **Chapter Four: Preliminary Study of Life Cycle Greenhouse Gas Emissions and a Novel Characterization System for Carbon Conversion and Utilization Technologies**

### **4.1 Introduction**

Over the past three decades, there has been a growing concern for anthropogenic greenhouse gas (GHG) emissions due to their effect on global warming and climate change. These man-made emissions can be attributed to the burning of fossil fuels, emissions from production processes and unintentional releases to the atmosphere, and include a variety of gases including carbon dioxide, carbon monoxide, methane, nitrous oxide, fluorinated gases, water vapor and other chemistries (Folland, et al., 2001). Specific sources of these emissions by sector include electricity and heat production (25%), agriculture, forestry and other land use (24%), industry (21%), transportation (14%), buildings (6%) and other energy sources (10%) (EPA, 2023).

A recent report from the United Nations International Panel on Climate Change (IPCC) indicates that these anthropogenic GHG sources are responsible for an increase in the average global temperature by approximately 1°C since the mid-18<sup>th</sup> century (IPCC, 2018). At the current rate of warming, this average global temperature is projected to increase by 1.5°C between 2030 and 2052 and 2.2°C to 3.5°C by the end of the 21<sup>st</sup> century (IPCC, 2021) (IPCC, 2022). This rise in global temperatures is anticipated to have a profound effect of global climate resulting in a number of potential impacts including sea level rise (due to melting ice caps), extreme weather events due to climate change (e.g. heat waves, flooding from heavy precipitation), melting of glaciers, and significant changes to regional eco-systems (IPCC, 2018). In order to mitigate this human impact on the environment, 195 nations participated in drafting the Paris Agreement

(United Nations Framework Convention on Climate Change, 2015) in 2015 with the aim to keep future global temperature rises below 2°C from pre-industrial times.

In order to accomplish this task, there have been a number of approaches proposed to mitigate rising GHG emissions by either reducing or eliminating the combustion of fossil fuels (e.g. using renewable energy sources; improving efficiencies and/or losses) or by preventing the release of GHG emissions to the atmosphere (e.g. using carbon capture). In the latter approach, there has been extensive work over the past two decades on the development and implementation of Carbon Capture and Sequestration (CCS) technologies. CCS is process by which GHG emissions are captured from industrial processes or directly from the atmosphere, and then stored underground in stable geological formations (Cuellar-Franca & Azapagic, 2015). CCS technologies offer an effective solution to mitigate GHGs from processes that generate concentrated sources of emissions such as fossil fuel based electricity generation, petroleum/chemical processing, and cement manufacturing, for example. The main concerns regarding CCS is the significant infrastructure required to handle, transport and inject the produced emissions, and the potential risk of future releases due to geological damage or degradation of injection wells (Leung, et al., 2014) (Arning, et al., 2019) (Davies, et al., 2013).

An alternative to storing CO<sub>2</sub> underground using CCS is utilizing the captured CO<sub>2</sub> to manufacture valuable end products. These technologies are collectively known as Carbon Capture and Utilization (CCU). Compared to CCS, CCU technologies offer the potential to offset the cost of carbon capture and storage by creating value added product streams that can be directly sold to existing or new markets for profit. While the elimination of fossil fuel use is the best solution for reducing global warming, proponents of both CCS and CCU see these

technologies as a potential short-term or transitional solution to GHG mitigation (Cuellar-Franca & Azapagic, 2015) (Bruhn, et al., 2016) (Kätelhön, et al., 2019).

Over the past decade, there has been an exponential growth in research and development in CCU technologies spanning various production pathways (chemical and biological) and various end products including fuels, chemicals and materials (e.g. plastics or cement replacements/additives). In terms of fuel production from CO<sub>2</sub>, there have been a number of pathways proposed in the literature to produce a variety of fuels including methanol (Perez Fortes, et al., 2016) (Meunier, et al., 2020) (Kiss, et al., 2016), methane (Guilera, et al., 2018) (Reiter & Lindorfer, 2015) (Chauvy, et al., 2020), Fischer-Tropsch fuels (Choi, et al., 2017) (Zang, et al., 2021), dimethyl ether (DME) (Aguayo, et al., 2005) (Catizzone, et al., 2018) and bio-fuels from microalgae (Chen, et al., 2020) (Cheah, et al., 2015) (Khoo, et al., 2011). Fu *et al.* (Fu, et al., 2010) discussed the production of syngas from co-electrolysis of steam and CO<sub>2</sub>, which can be converted into methane, methanol or diesel using the Fischer-Tropsch process. Methane can be directly produced from CO<sub>2</sub> using a process called methanation which combines CO<sub>2</sub> with hydrogen using a fixed bed reactor or fluidized bed reactor (Schaaf, et al., 2014). Methanol can be produced from CO<sub>2</sub> and syngas feedstocks using methanol synthesis (Hoppe, et al., 2017), and dry methane reforming (Luu, et al., 2015), respectively. Additionally, the methanol produced from hydrogenation of CO<sub>2</sub> can be converted into dimethyl ether (DME) via methanol dehydration (Catizzone, et al., 2018). In terms of biological pathways, the fixation of CO<sub>2</sub> using microalgae with open and closed pond systems provides significant mitigation potential. A review by Klinthong *et al.* (Klinthong, et al., 2015) suggests cultivation using closed photobioreactors is promising for both the biofuel industry and for the commercialization of co-products (such as biomass) for additional profit. However, the cultivation stage emissions

contribute most toward global warming potential (GWP) of biodiesel derived from microalgae (Cuellar-Franca & Azapagic, 2015).

Various chemicals and their intermediates can also be produced from CO<sub>2</sub> using a number of distinct conversion pathways such as thermocatalytic, electrochemical, biochemical and photochemical (Bobeck, et al., 2019). Common chemicals and intermediates that have been produced from CO<sub>2</sub> include urea (Koohestanian, et al., 2018) (Huang & Tan, 2014), formic acid (Hao, et al., 2011) (Nakata, et al., 2014), formaldehyde (Nakata, et al., 2014), and salicylic acid (Aresta & Dibenedetto, 2007). In particular, some technologies that use CO<sub>2</sub> for the synthesis of urea, salicylic acid and polycarbonates has been commercialized with approximately 130MT of CO<sub>2</sub> used per year to manufacture these products (Chauvy, et al., 2019). Of these three products, urea consumes the most CO<sub>2</sub> industrially, and is produced by the reaction of CO<sub>2</sub> and ammonia at high temperatures and pressure (Huang & Tan, 2014). Based on preliminary studies, CO<sub>2</sub> can also be reduced to formic acid and to formaldehyde, however, the production and technology readiness levels for these technologies are lower than for the other chemicals mentioned (Chauvy, et al., 2019).

Several materials can also be produced from CO<sub>2</sub> including the production of novel polycarbonates (Wang & Darensbourg, 2018) (Liu & Wang, 2017), cement/concrete (Monkman & MacDonald, 2017) (Wang, et al., 2017) (Huang, et al., 2019), cement replacements (Benhelal, et al., 2018) and aggregates (Kirchofer, et al., 2013) (The National Academies Press, 2019). The co-polymerization of CO<sub>2</sub> and epoxides to form polycarbonates is commercialized but the market size is much lower (5MT per year) compared to mineral carbonation products, such as calcium carbonate (113.9MT per year) (Chauvy, et al., 2019). Direct and indirect carbonation are the two main approaches for combining CO<sub>2</sub> with magnesium and calcium silicate minerals to form

stable carbonates (Sanna, et al., 2014) (Giannoulakis, et al., 2013). The direct carbonation process developed at the National Energy Technology Laboratory (NETL) uses one self-contained unit to create carbonates by dissolution and precipitation reactions (Nduagu, et al., 2012). In contrast, indirect mineralization such as the Åbo Akademi University (AAU) process uses a multi-step extraction process (Olajire, 2013). Sanna *et al.*, (Sanna, et al., 2014) examined the feasibility of both of these mineral carbonation processes but determined that the current technologies are not economically viable at this time, and that further research and development is required to scale this potential mitigation pathway.

In order to determine the effectiveness of GHG mitigation for the multitude of CCU technology options available, the life cycle assessment (LCA) methodology can be used. This methodology allows for a fair comparison between different process pathways and products by using clearly defined units of comparison (e.g. functional units). While there has been a number of LCA studies performed on individual CCU pathways, there has been limited work in assessing and directly comparing a variety of CCU technologies in a single study. One of the challenges in performing such a meta-analysis from literature is the variation of LCA methodologies and assumptions (e.g. functional unit, system boundaries, emission intensity factors, etc.) used by each specific study. If properly reconciled, a comparison of LCA results can offer a fair methodology to compare the net emissions of various CCU technologies both amongst themselves and with current incumbent production pathways and products. This comparison may provide an initial method to screen promising CCU technologies, however, other factors such as economics, technology readiness, scalability, government policy and industry acceptance will all play a significant role in determining which technologies ultimately become adopted (Bobeck, et al., 2019).

In order to ensure a fair comparison when conducting comparative LCAs for CCU, a number of guidelines have been proposed in the literature (Table 4.1). In general, these guidelines provide a variety of recommendations including entire cradle-to-grave LCAs to ensure holistic comparisons (Assen, et al., 2014) (Langhorst, et al., 2022) (Müller, et al., 2020b) (Zimmermann, et al., 2022), although gate-to-gate or cradle-to-gate boundaries have also been discussed as being sufficient for comparisons if life cycle inventories are missing or products are identical, respectively (Müller, et al., 2020a) (Langhorst, et al., 2022) (Zimmermann, et al., 2022). The use of energy based functional units for fuels and mass based for non-fuels is necessary to ensure that comparisons are equitable (Assen, et al., 2014) (Assen, et al., 2013) (Müller, et al., 2020b) (Langhorst, et al., 2022) (Müller, et al., 2020a) (Zimmermann, et al., 2022). Issues of multifunctionality are commonly mentioned in guidelines where there are multiple products. As such, allocation and system expansion are discussed (Müller, et al., 2020a) (Langhorst, et al., 2022) (Müller, et al., 2020b). Additionally, including CO<sub>2</sub> capture source and upstream emissions from capture are also important (Assen, et al., 2013) (Assen, et al., 2014) (Müller, et al., 2020a) (Müller, et al., 2020b) (Langhorst, et al., 2022). Data quality aspects are addressed using sensitivity, uncertainty and Monte Carlo analysis in the guidelines (Assen, et al., 2014) (Müller, et al., 2020a) (Langhorst, et al., 2022) (Zimmermann, et al., 2022). The work presented in this research uses a gate-to-grave boundary, energy and mass-specific metrics of assessments, and applies allocation where needed. In addition, this work uses consistent emissions and conversion factors.

In terms of previous LCAs of CCU technology categories, there have only been four comparative studies published to the authors' knowledge. Cuellar-Franca and Azapagic (Cuellar-Franca & Azapagic, 2015) compared 4 different CCU technologies from the literature including

Table 4.1 Summary of recommendations from literature on guidelines for conducting life cycle assessments of carbon capture utilization technologies

Recommendations	(Assen, et al., 2014)	(Assen, et al., 2013)	(Müller, et al., 2020a)	(Müller, et al., 2020b)	(Langhorst, et al., 2022)	(Zimmermann, et al., 2022)	This study
<b>Goal and scope</b>							
Define central question and audience (goal)			✓		✓	✓	✓
Compare products with same function or functional unit (mass or energy basis)	✓	✓	✓	✓	✓	✓	✓
Ideally include system boundary with entire lifecycles	✓		✓		✓	✓	
Use gate-to-gate boundary if LCI missing			✓				
Use cradle-to-gate boundary if compared products are identical					✓	✓	
Comparison across technologies using similar functional units						✓	✓
Present all assumptions					✓	✓	✓
Get stakeholder input for benchmarks						✓	✓
<b>Life cycle inventory</b>							
Include all mass and energy flows	✓		✓		✓	✓	✓
Use LCA software and databases for LCI	✓		✓				✓
Use stoichiometry, energy, exergy and entropy balance if other LCI missing			✓		✓	✓	✓
Deal with multifunctionality using allocation, system expansion or substitution	✓	✓	✓	✓			✓
Select market competitive reference process for comparison			✓			✓	✓
Do not account for temporary storage			✓		✓		✓
Verify and compare data with industry partners						✓	✓
Use consistent emission factors, conversion factors and market sizes for respective products							✓



Table 4.1 (continued)

Recommendations	(Assen, et al., 2014)	(Assen, et al., 2013)	(Müller, et al., 2020a)	(Müller, et al., 2020b)	(Langhorst, et al., 2022)	(Zimmermann, et al., 2022)	This study
Include CO <sub>2</sub> source and upstream emissions from capture	✓	✓	✓	✓	✓		✓
Develop an LCI template and model to assess carbon conversion technologies consistently							✓
<b>Life cycle impact assessment</b>							
Include all impacts for midpoint indicators	✓	✓	✓		✓		
Include time dependent global warming potential (GWP) if profiles available		✓					
Present results for CO <sub>2</sub> sources with reduced environmental impact				✓			✓
Present worst, best and neutral scenarios						✓	✓
Estimate missing data if missing and use forecasting if needed						✓	
Market relevant metrics to show mitigation potentials shown							✓
<b>Life cycle interpretation</b>							
Include uncertainty, sensitivity, or monte-carlo analysis	✓		✓		✓	✓	✓
Incumbent and LCA result variability presented						✓	✓
Follow ISO 14044 for all reporting			✓				✓
<b>Others</b>							
Plan for significant time to conduct the LCA	✓					✓	✓
Collaborate with practioners to save time						✓	
Develop specific nomenclature for carbon conversion technologies							✓

enhanced oil recovery, mineralization, biodiesel from microalgae and dimethyl carbonate. As noted by the authors, the assessment had many limitations including noted differences in system boundaries, assumptions and level of uncertainty between studies. Another study presents a comparison between mineralization and catalytic conversion processes. However, the results are aggregated, and data variability is low (Sleep, et al., 2021). Other studies, such as the “CO<sub>2</sub> Conversion Technologies for Oil Sand Activities” report prepared by ENEA and COSIA proposed eight CCU classes and sub-classes based on products (ENEA & COSIA, 2015). However, they do not further differentiate different pathways within a sub-class which form the same product. Additionally, the Global Roadmap for Implementing CO<sub>2</sub> Utilization Report (ICEF, 2016) distinguishes four market categories and eight sub-categories based on maturity, market promise and potential to mitigate emissions (ICEF, 2016). However, this report doesn't separate markets based on different CCU technology modes which is important consideration due to the fact that a particular product may be produced by different CCU pathways. Distinguishing pathways is important to determine the exact method by which products mitigate CO<sub>2</sub> emissions. This shows inherent challenges in characterizing CCU technologies and pathways, as well as in utilizing consistent methodologies to assess various CCU technologies.

Based on the lack of comparative LCAs of CCU technologies, the goal of this current study is two-fold: (1) to develop a characterization system for categorizing CCU technologies, and (2) to compare the net carbon footprint of a number of CCU technologies using a consistent LCA approach. In this research, a framework for categorizing different and diverse CCU technologies based on the CO<sub>2</sub> reaction mechanism is proposed with sub-categorization to differentiate unique pathways within the main categories. Furthermore, life cycle assessment methodology is used to characterize the greenhouse gases mitigation potential based on pathway

and product type for a number of CCU technologies found in the literature. A standardized modeling framework was developed to ensure a fair comparison between CCU pathways including consistent system boundaries, and consistent emission and conversion factors. Additionally, comparisons between product types and the potential market impact of these CCU technologies were examined using four assessment metrics. Overall, this research demonstrates a robust categorization scheme and comparative life cycle approach which can be expanded as further CCU technologies are developed and/or refined.

## **4.2 Methodology**

### ***4.2.1 Overview and Data Sources***

The goal of this study is to develop a characterization system for CCU technologies. The technologies are first assigned to 1 of 8 categories. These include CO<sub>2</sub> reduction by a hydrocarbon, CO<sub>2</sub> reduction by hydrogen, CO<sub>2</sub> reduction by other reagents, CO<sub>2</sub> reduction involving electricity, CO<sub>2</sub> reduction involving light, CO<sub>2</sub> mineralization, CO<sub>2</sub> bioconversion and other CO<sub>2</sub> conversions. Then, a methodology was developed for the comparison of the GHG emissions of these technologies using a consistent LCA approach. The technologies are evaluated using 4 assessment metrics. To compare the technologies among each other, the kg CO<sub>2</sub> equivalents per kg CO<sub>2</sub> converted, and kg CO<sub>2</sub> equivalents per kg or megajoule (MJ) of product were developed. Two additional metrics (CO<sub>2</sub> avoided emissions and global emissions reduction potential) were also studied to compare technologies to their incumbents and markets (Figure 4.1).

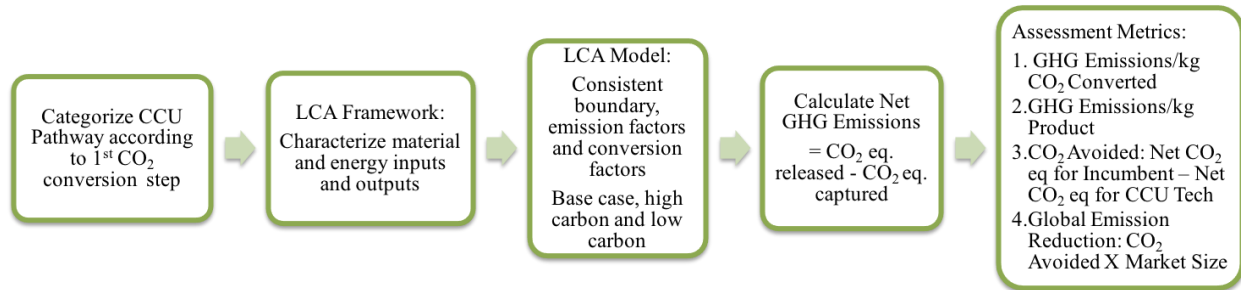


Figure 4.1 Methodology for life cycle assessment of carbon capture utilization technologies

#### 4.2.2 Categorization Scheme

A categorization scheme was developed to identify and assess the life cycle greenhouse gas emissions from these technologies on a consistent basis. The CO<sub>2</sub> reduction step was selected as the defining process by which the CCU categories were selected. After which, subcategories were selected based on pathway specific factors, such as reactants or processes. In the case of mineralization, the reactants defined the subcategories. Once a technology was identified, it was first assigned to a category and subsequently to a subcategory, based on this methodology. This subcategorization method improves previously discussed methods of CCU categorizations and allows for better comparison using the assessment metrics, as described in the results section.

#### 4.2.3 Determination of Life Cycle Greenhouse Gas Emissions

##### 4.2.3.1 Goal and Scope

The primary goal of this study is to develop a life cycle framework and to evaluate the results of this framework for consistent comparison of the energy requirements and greenhouse gas emissions associated with CCU technologies. The specific outcomes of this study are to develop a standard set of evaluation criteria and metrics, boundaries and generalized CCU life

cycle stages, a data collection template for CCU technologies and sensitivity analysis to assess hot-spots and robustness of the results from the pathways.

#### 4.2.3.2 System Boundary

The system boundary for this assessment will be gate-to-grave. The generic life cycle stages used in this study were CO<sub>2</sub> capture process, CO<sub>2</sub> conversion, hydrogen production and end use. The individual steps involved in the aforementioned stages were represented in the process diagram for each category/sub-category. Net emissions from literature for each of the identified pathways were shown as variability in the results section along with net emission variability from incumbent processes for each category.

#### 4.2.3.3 Assessment Metrics

Four assessment metrics will be used in this study to assess the different CCU technologies consistently:

- kg CO<sub>2</sub> equivalents per kg CO<sub>2</sub> converted: net emissions for each pathway divided by the amount of CO<sub>2</sub> converted
- kg CO<sub>2</sub> equivalents per kg or megajoule (MJ) of product: net emissions for each pathway divided by the amount of product formed

Two additional metrics were used to assess the market potential of CCU technologies. These metrics will be used to assess the benefits (if any) of CCU technologies compared to incumbent processes. It will also serve to identify the potential market capacity and GHG mitigation potential of CCU technologies.

These metrics are:

- Avoided emissions (AE): amount of CO<sub>2</sub> avoided by each pathway compared to the incumbent pathways

$$AE = IC - CE \quad (\text{Equation 1})$$

Where IC refers to the net incumbent emissions per kg of product, and CE refers to the net CCU emissions per kg of product. In this case, incumbent refers to a commercially available technology that produces the same product as the CCU technology without using CO<sub>2</sub> as an input.

- Global emission reduction potential (GE): amount of avoided emissions from a pathway multiplied by the market size (kg) which the product is assumed to replace

$$GE = AE \times MS \quad (\text{Equation 2})$$

Where AE refers to avoided emissions and MS refers to market size in kg

#### 4.2.3.4 Assumptions

Various assumptions were made for the energy sources.

- The base case was selected as a natural gas source. The electricity for this was assumed to be provided by natural gas, whereas the heat was assumed to be provided by a natural gas industrial furnace. The CO<sub>2</sub> capture method for the base case was assumed to be via monoethanolamine (MEA) from a natural gas power plant. The hydrogen was assumed to be provided by steam methane reforming.
- For the high carbon scenario, coal was chosen as the electricity source and combined heat and power for the heat source. The CO<sub>2</sub> capture method for this was chosen as MEA from a coal power plant. The hydrogen was assumed to be provided by coal gasification.

- The electricity for the low carbon scenario was assumed to be provided by renewable energy sources (an averaged emission factor using hydropower, solar-photovoltaics, wind and nuclear) and the heat to be from geothermal energy. The CO<sub>2</sub> capture method for the low carbon scenario was assumed to be via direct air capture from ambient air. The hydrogen for the low carbon scenario was assumed to be provided by electrolysis using renewable energy.
- The global emission reduction potentials were calculated using 50% market penetration potential. One market was selected for each product and the size was assumed to be 50% of the given market for the selected year, 2015.
- In all cases where different emission and/or conversion factors were used, the literature applied conversion factors were removed and the emission and/or conversion factors mentioned in this section were applied to ensure consistency.
- The negative values result from a reduction in emissions compared to the amount of CO<sub>2</sub> converted and incumbents for all scenarios other than the low carbon scenario. In the low carbon scenario CO<sub>2</sub> removal occurs from the atmosphere, as determined by (Langhorst, et al., 2022). Combustion pathways are not considered to be net negative in all scenarios.

Table 4.2 summarizes the key emission factors used when calculating GHG emissions released for each of the scenarios discussed above.

#### 4.2.3.5 Inventory Analysis & Data Type

A total of 86 CCU pathways from various literature sources were examined in this study (Table 4.3). A data collection template was used to obtain the material inputs/outputs and energy inputs/outputs for each unit process of the life cycle of a pathway. This data was input into a

Table 4.2 Emission factors for energy and material inputs for base case, high carbon and low carbon scenarios

Scenario	Parameter	Value	Reference
Base case	Natural gas electricity source	0.49 kg CO <sub>2</sub> eq/kWh	(Schlömer, et al., 2014)
Base case	Natural gas industrial furnace	0.25 kg CO <sub>2</sub> eq/kWh	(ISCC, 2017)
Base case	Natural gas industrial boiler steam	0.185 kg CO <sub>2</sub> eq/kWh	(EPA, 1996)
Base case	Steam methane reforming for hydrogen production	10.62 kg CO <sub>2</sub> eq/kg H <sub>2</sub>	(Spath & Mann, 2001)
Base case	CO <sub>2</sub> capture from natural gas power plant	0.18 kg CO <sub>2</sub> eq/kg CO <sub>2</sub> Captured	(Helmeth Consortium, 2017)
High carbon	Coal fired electricity	0.80 kg CO <sub>2</sub> eq/kWh	(GREET, 2015)
High carbon	Combined heat & power	0.55 kg CO <sub>2</sub> eq/kWh	(Doluweera, et al., 2011)
High carbon	Coal gasification hydrogen production	24.2 kgCO <sub>2</sub> eq/kg H <sub>2</sub>	(Mehmeti, et al., 2018)
High carbon	CO <sub>2</sub> capture from coal power plant	0.82 kg CO <sub>2</sub> eq/kg CO <sub>2</sub> Captured	(Helmeth Consortium, 2017)
Low carbon	Renewable electricity (hydropower, wind, solar-pv and nuclear)	0.024 kg CO <sub>2</sub> eq/kWh	(Schlömer, et al., 2014)
Low carbon	Electrical heater using renewable electricity (100% efficiency)	0.024 kg CO <sub>2</sub> eq/kWh	(Schlömer, et al., 2014)
Low carbon	Geothermal steam	0.006 kg CO <sub>2</sub> eq/kWh	(Schlömer, et al., 2014)
Low carbon	Electrolysis with low carbon electricity for hydrogen production	0.984 kg CO <sub>2</sub> eq/kg H <sub>2</sub>	(Simbeck & Chang, 2002)
Low carbon	CO <sub>2</sub> capture from direct air capture	0.007 kg CO <sub>2</sub> eq/kg CO <sub>2</sub> Captured	(Helmeth Consortium, 2017)



Table 4.3 CCU category classification, definitions, subcategories, main products, number of cases examined per process and literature examined for application of methodology

Category Name	Category Definition	Subcategories and Abbreviations	Main Products	Number of cases examined	Literature
CO <sub>2</sub> Reduction by a Hydrocarbon	CO <sub>2</sub> is first reduced by a chemical reaction with a hydrocarbon A hydrocarbon is a substance containing only hydrogen and carbon atoms, examples include methane and ethane but not methanol, glycerol or coke	Dry methane reforming to produce fuels (DMR)	Dimethyl Ether	1	(Schakel, et al., 2016)
		Dry methane reforming with CO purging (DMR-CO)	Methanol	1	(Luu, et al., 2015)
		Dry methane reforming with H <sub>2</sub> (DMR-H <sub>2</sub> )	Methanol	1	(Luu, et al., 2015)
CO <sub>2</sub> Reduction by Hydrogen	CO <sub>2</sub> is first reduced by a chemical reaction with hydrogen These reactions typically use a catalyst	Direct hydrogenation to produce fuels (D-H <sub>2</sub> )	Diesel	1	(Giesen, et al., 2014)
			Methanol	6	(Matzen, et al., 2015) (Van-Dal & Bouallou, 2013) (Kiss, et al., 2016) (Rihko-Struckmann, et al., 2010) (Perez Fortes, et al., 2016) (Hoppe, et al., 2017)
			Methane	4	(Reiter & Lindorfer, 2015) (Hoppe, et al., 2017) (Müller, et al., 2011) (Jean, et al., 2014)
CO <sub>2</sub> Reduction by Other Reagents	CO <sub>2</sub> is first reduced by a chemical reaction with a reagent other than hydrogen or a hydrocarbon Includes some 'CO <sub>2</sub> polymerization' and	Electrochemical-uses water for extractive distillation (EL-CHEM.I)	Dimethyl Carbonate	1	(Garcia-Herrero, et al., 2016)

Table 4.3 (continued)

Category Name	Category Definition	Subcategories and Abbreviations	Main Products	Number of cases examined	Literature
	ethylene synthesis pathways Does not include bioconversion or those involving electricity or light	Electrochemical-uses aniline for extractive distillation (EL-CHEM.II)	Dimethyl Carbonate	1	(Garcia-Herrero, et al., 2016)
		Chemical synthesis-ethylene route (CHEM.I)	Dimethyl Carbonate	1	(Kongpanna, et al., 2015)
		Chemical synthesis- urea route (CHEM.II)	Dimethyl Carbonate	1	(Kongpanna, et al., 2015)
CO <sub>2</sub> Reduction Involving Electricity	CO <sub>2</sub> is first reduced by a reaction involving externally applied electricity but not light Other forms of energy, like heat or chemical energy may also be involved	Solid oxide electrolyser cell (SOEC) co-electrolysis (SOEC-CO)	Methane	1	(Fu, et al., 2010)
		Polymer electrolyte membrane (PEM)	Diesel	1	(Giesen, et al., 2014)
			Methane	4	(Reiter & Lindorfer, 2015) (Hoppe, et al., 2017) (Müller, et al., 2011) (Jean, et al., 2014)
			Methanol	5	(Matzen, et al., 2015) (Kiss, et al., 2016) (Van-Dal & Bouallou, 2013) (Hoppe, et al., 2017) (Rihko-Struckmann, et al., 2010)
		Solid oxide electrolyser cell electrolysis (SOEC-EL)	Diesel	1	(Giesen, et al., 2014)
			Methane	4	(Reiter & Lindorfer, 2015) (Hoppe, et al., 2017) (Müller, et al., 2011) (Jean, et al., 2014)

Table 4.3 (continued)

Category Name	Category Definition	Subcategories and Abbreviations	Main Products	Number of cases examined	Literature
			Methanol	5	(Matzen, et al., 2015) (Kiss, et al., 2016) (Van-Dal & Bouallou, 2013) (Hoppe, et al., 2017) (Rihko-Struckmann, et al., 2010)
CO <sub>2</sub> Reduction Involving Light	CO <sub>2</sub> is first reduced by a reaction that involves light energy Other forms of energy may also be involved e.g. heat, chemical or electrical energy Does not include biological photosynthesis	Photocatalytic reduction of CO <sub>2</sub> using semiconductors (P.CAT)	Methane	1	(Trudewind, et al., 2014)
			Methanol	1	(Trudewind, et al., 2014)
CO <sub>2</sub> Mineralization	CO <sub>2</sub> is not reduced but instead reacts with a cation such as magnesium, calcium, or iron to form a carbonate mineral This category only includes ex-situ conversion	Direct mineral carbonation using serpentine (DMC-S)	Magnesite	3	(Khoo & Reginald, 2006) (Giannoulakis, et al., 2013)
		Direct mineral carbonation using olivine (DMC-O)	Magnesite	4	(Khoo & Reginald, 2006) (Giannoulakis, et al., 2013) (Kirchofer, et al., 2012)
		Direct mineral carbonation using wollastonite (DMC-W)	Calcite	2	(Khoo & Reginald, 2006) (Giannoulakis, et al., 2013)
		Indirect mineral carbonation using serpentine (IMC-S)	Magnesite	7	(Khoo, et al., 2011b) (Giannoulakis, et al., 2013) (Nduagu, et al., 2012)

Table 4.3 (continued)

Category Name	Category Definition	Subcategories and Abbreviations	Main Products	Number of cases examined	Literature
		Waste mineral carbonation (WMC)	Carbonated waste material (CWM)	5	(Pan, et al., 2016)
CO <sub>2</sub> Bioconversion	Microbial: CO <sub>2</sub> is first converted by unicellular biological organisms that derive their energy from light (e.g. by photosynthesis) or use chemical energy (i.e. chemotrophs) Plant-based: CO <sub>2</sub> is captured and converted by photosynthetic plants into biomass	Pond-wet extraction (P-WE)	Jet fuel	1	(Ou, et al., 2013)
			Diesel	14	(Stephenson, et al., 2010) (Frank, et al., 2012) (Lardon, et al., 2009) (Campbell, et al., 2011) (Ponnusamy, et al., 2014) (Liu, et al., 2013) (Woertz, et al., 2014) (Yuan, et al., 2015) (Gao, et al., 2013) (Torres, et al., 2013)
		Pond-dry extraction (P-DE)	Diesel	8	(Torres, et al., 2013) (Lardon, et al., 2009) (Manganaro & Lawal, 2016) (Yuan, et al., 2015) (Collet, et al., 2014)
		Photobioreactor-dry extraction (PBR-DE)	Diesel	1	(Khoo, et al., 2011a)
Other CO <sub>2</sub> Conversions	All other methods of CO <sub>2</sub> utilization. Includes CO <sub>2</sub> absorption and thermal splitting/plasma processes.				

life cycle framework for CCU (Figure 4.2) that includes common process groups which can be added/removed to model various pathways, and can be adapted for any unique CCU pathway being examined. Following this, the data was input into the model, which refers to the data and calculations carried out for a particular pathway. The model was created using Microsoft Excel<sup>®</sup> and as such all calculations were performed in this software. Expert and industry elicitation were also carried out to confirm the findings from the literature sources.

#### 4.2.3.6 Greenhouse Gas Emissions

Once all the material and energy flows were identified, the base case assumptions were applied and the GHG emissions were determined in kilograms of CO<sub>2</sub> equivalents. All material

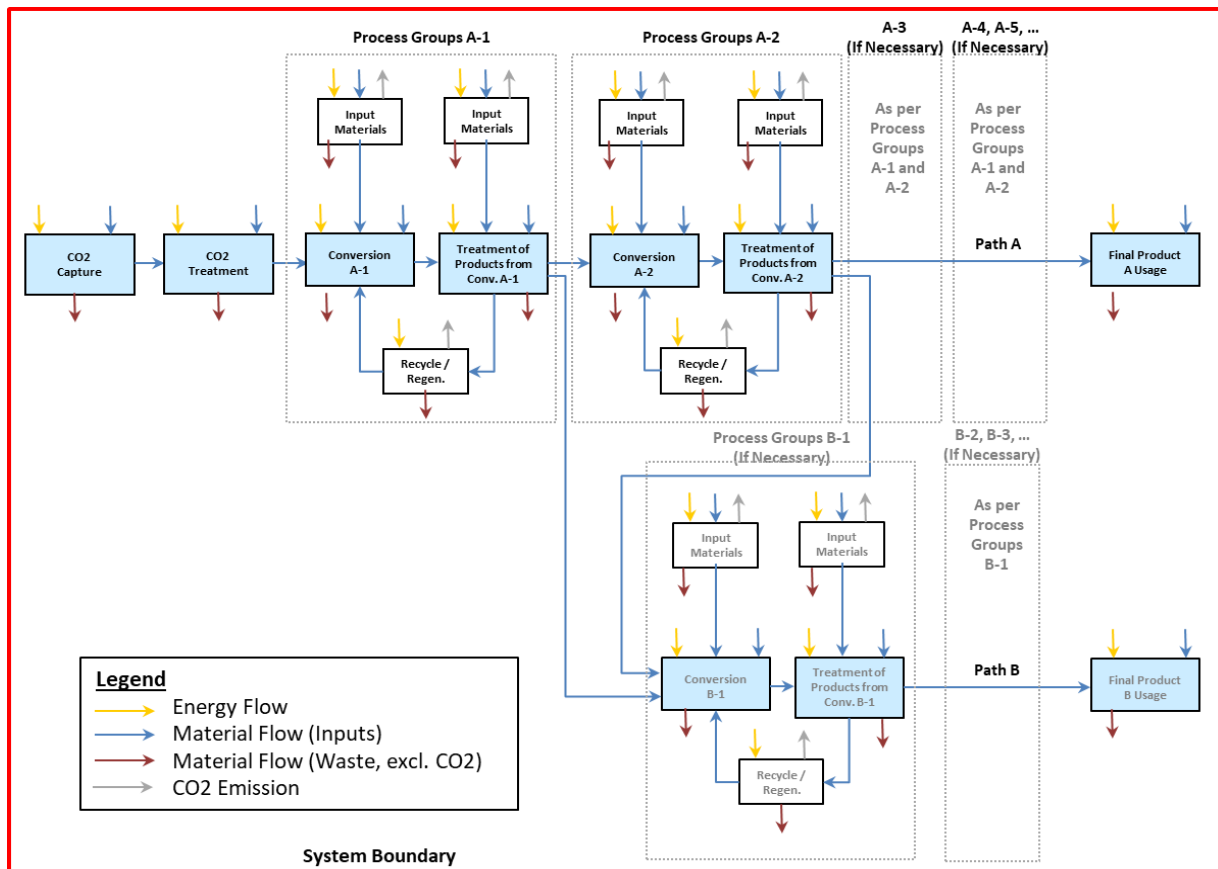


Figure 4.2: System level CCU framework showing process groups that can be added or removed based on unique CCU technology pathways.

and energy balances were included, this includes the amount of CO<sub>2</sub> converted and any heat or material recycling (if applicable). The two assessment metrics were applied to the net emissions. After which, the net emissions (NE) were calculated for each pathway using Equation 3. Similarly, the other assessment metrics were also calculated for each of the pathways using equations (1) and (2). Only the positive values for global CO<sub>2</sub> emissions reduction potential are shown in the figures. Outliers were also removed, as discussed further in the results and discussion section.

$$NE = CR - CC \quad (\text{Equation 3})$$

Where net emission (NE) is the difference between the CO<sub>2</sub> equivalents released (CR) and the CO<sub>2</sub> equivalents captured (CC).

Representative cases were then selected for each subcategory which is depicted in the results section. The other pathways within each subcategory were shown as literature variability. Representative cases were selected from literature based on those that are either already commercialized or are close to being commercialized. However, for the subcategories in which only early stage developmental cases were found (lab-scale or basic observed cases), the primary selection criterion was the greater level of detail breakdown of life cycle stages. Some other common parameters for the selection of representative cases were availability of resources, optimal heat integration, carbonation efficiency, operating conditions and prevalence in industrial installations. Details of the mineralization selection process is provided in Appendix A and Figure A1.

#### 4.2.3.7 Sensitivity Analysis

Sensitivity analysis was carried out on the parameters for the representative cases. All available operational and life cycle parameters were varied based on upper and lower bounds

found in literature. If this data wasn't available, the parameters were varied with  $\pm 50\%$ , or based on most probable percentage change. One parameter was varied at a time and the net emission results were shown using the base case scenario for the respective pathway. In the case where multiple parameters were varied, different scenarios were also expressed in the sensitivity figure.

## 4.3 Results & Discussion

### 4.3.1 CCU Categorization Scheme

The guidance table (Table 4.3) provides clear category definitions, processes, possible end products, and literature sources used in this study for the categorization of CCU technologies and the literature pathways examined in this study. The categorization scheme is used to define pathways on their first CO<sub>2</sub> conversion step (Table 4.3). A total of 8 unique CCU categories were established based on these criteria: CO<sub>2</sub> reduction by a hydrocarbon, CO<sub>2</sub> reduction by hydrogen, CO<sub>2</sub> reduction by other reagents, CO<sub>2</sub> reduction involving electricity, CO<sub>2</sub> reduction involving light, CO<sub>2</sub> mineralization, CO<sub>2</sub> bioconversion and other CO<sub>2</sub> conversions. The CO<sub>2</sub> reduction by other reagents captures all reactions other than those involving hydrocarbon or hydrogen, such as with methanol to produce dimethyl carbonate. Based on the findings, subcategories were developed to better distinguish and group specific CCU processes depending on differences in the intermediate products they produce or based on variation in the raw materials they utilize. As shown by Table 4.3, a total of 20 subcategories are defined across the categories: dry methane reforming to produce fuels (DMR), dry methane reforming with CO purging (DMR-CO), dry methane reforming with H<sub>2</sub> (DMR-H<sub>2</sub>), direct hydrogenation to produce fuels (D-H<sub>2</sub>), electrochemical- uses water for extractive distillation (El-Chem. I), electrochemical- uses aniline for extractive distillation (El-Chem. II), chemical synthesis- ethylene route (Chem. I), chemical synthesis- urea route (Chem. II), solid oxide electrolyser cell (SOEC) co-electrolysis (SOEC-

CO), polymer electrolyte membrane (PEM), solid oxide electrolyser cell electrolysis (SOEC-EL), photocatalytic reduction of CO<sub>2</sub> using semiconductors (P.CAT), direct mineral carbonation using serpentine (DMC-S), direct mineral carbonation using olivine (DMC-O), direct mineral carbonation using wollastonite (DMC-W), indirect mineral carbonation using serpentine (IMC-S), waste mineral carbonation (WMC), pond-wet extraction (P-WE), pond-dry extraction (P-DE) and photobioreactor-dry extraction (PBR-DE). The following nine end products are investigated as a result of the above categorization: diesel, methanol, methane, dimethyl ether (DME), jet fuel, dimethyl carbonate (DMC), magnesite, calcite and carbonated waste material (CWM). The combination of products and subcategories reveals twenty-eight CCU pathways.

#### ***4.3.2 Comparison of Life cycle GHG Emissions of Different CCU Technologies on a Per kg CO<sub>2</sub> Converted Basis***

Twenty-six unique CCU pathways from 8 different CCU categories and 20 subcategories are depicted in Figure 4.3. This figure also shows a total of nine unique products. The results depicted in this figure use the base case (natural gas) (Figure 4.3a) and low-carbon (Figure 4.3b) assumptions described in the methodology. The negative bars indicate the amount of CO<sub>2</sub> captured by the process/pathway. Whereas the bars on the positive axis indicate the contribution of the life cycle stages towards GHG emissions. Emissions from four stages were examined for each process: CO<sub>2</sub> capture process, CO<sub>2</sub> conversion, H<sub>2</sub> production and end-use. This allows for consistent comparison across different CCU categories. Outliers, for the following two subcategories from the CO<sub>2</sub> reduction involving other reagents category are not depicted: electrochemical-uses water for extractive distillation and electrochemical-uses aniline for extractive distillation. This was due to the relatively high contribution of the CO<sub>2</sub> conversion stage for these pathways. Analysis using the per kg CO<sub>2</sub> converted assessment metric allows for

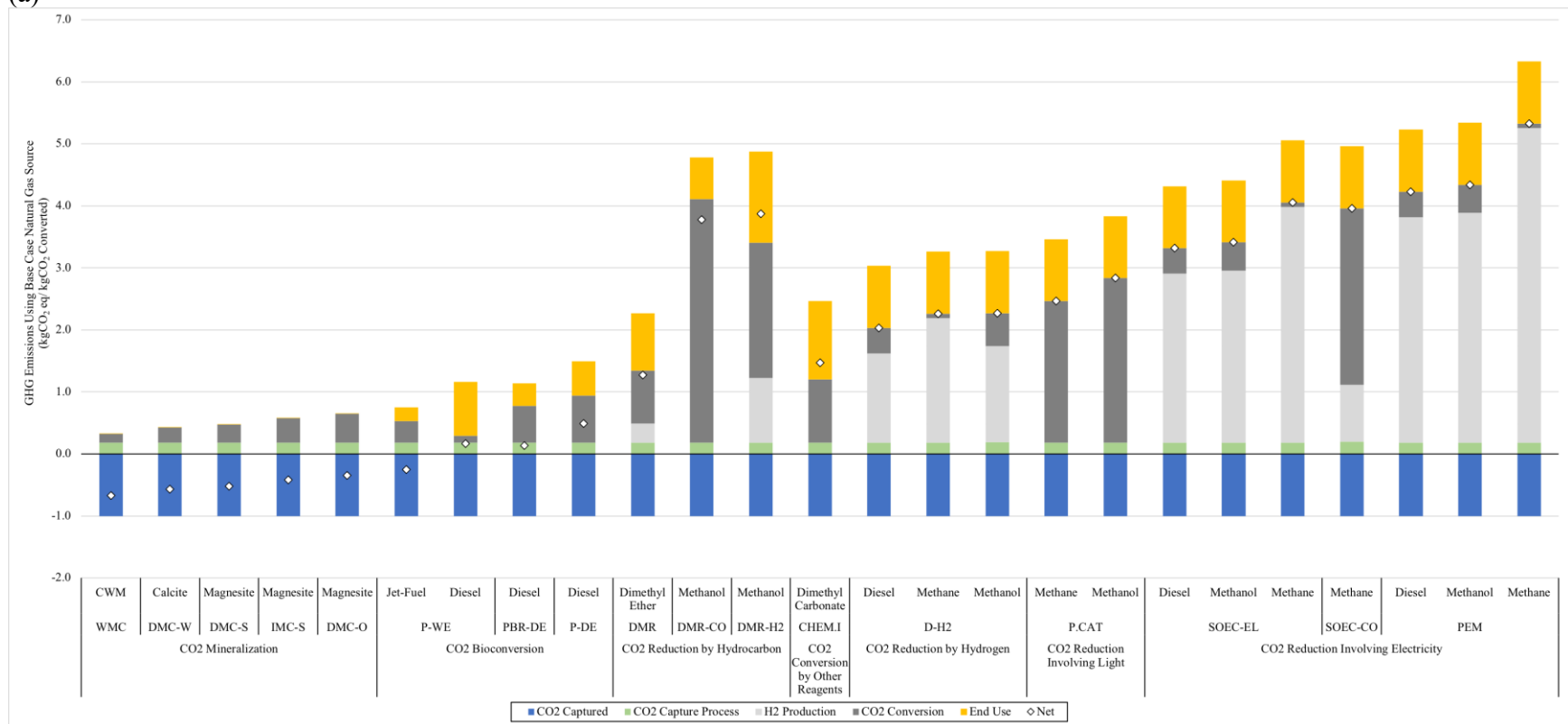


equal comparison across different CCU categories due to the fact that the same amount of CO<sub>2</sub> is converted in all CCU categories in Figure 4.3. This assessment metric allows for comparing different CCU pathways and products to identify low GHG intensity pathways for producing a product or selecting a particular preferred configuration within a CCU category.

It is important to note that the negative values shown in Figure 4.3 represent the conversion of CO<sub>2</sub> compared to processes that do not convert CO<sub>2</sub>, such as incumbent or conventional processes. They do not represent negative emissions, nor do they imply CO<sub>2</sub> capture from the atmosphere and environment. Additionally, base case pathways from fuel combustion depict the release of CO<sub>2</sub>, not sequestration of negative amounts of CO<sub>2</sub> converted. The technologies that reflect environmental advantage as a consequence of negative emissions would be those that utilize direct air capture using renewable energy sources and sequester the CO<sub>2</sub> during the end-use stage of the pathway.

For the base case (natural gas) assumption, CO<sub>2</sub> mineralization, bioconversion and reduction by hydrogen are shown to emit less GHGs than other categories like CO<sub>2</sub> reduction involving light and CO<sub>2</sub> reduction involving electricity, as well as some of the CO<sub>2</sub> reduction by hydrocarbon and reduction by other reagents pathways. CO<sub>2</sub> mineralization shows the lowest (most net-negative) GHG emissions compared with any other CCU technology category. This is primarily due to the lower CO<sub>2</sub> conversion stage emissions and the fact that there are much lower end-use emissions arising from mineralization compared to the other CCU pathways. The end-use emissions in this case were allocated to the amount of cement that was replaced by mineral carbonation products in the production of 1m<sup>3</sup> of concrete. Details of this are described in the sensitivity analysis (Section 4.3.8). In contrast, most other categories produce fuels and due to their combustion, add additional emissions to the life cycle. Additionally, the base case (natural

(a)



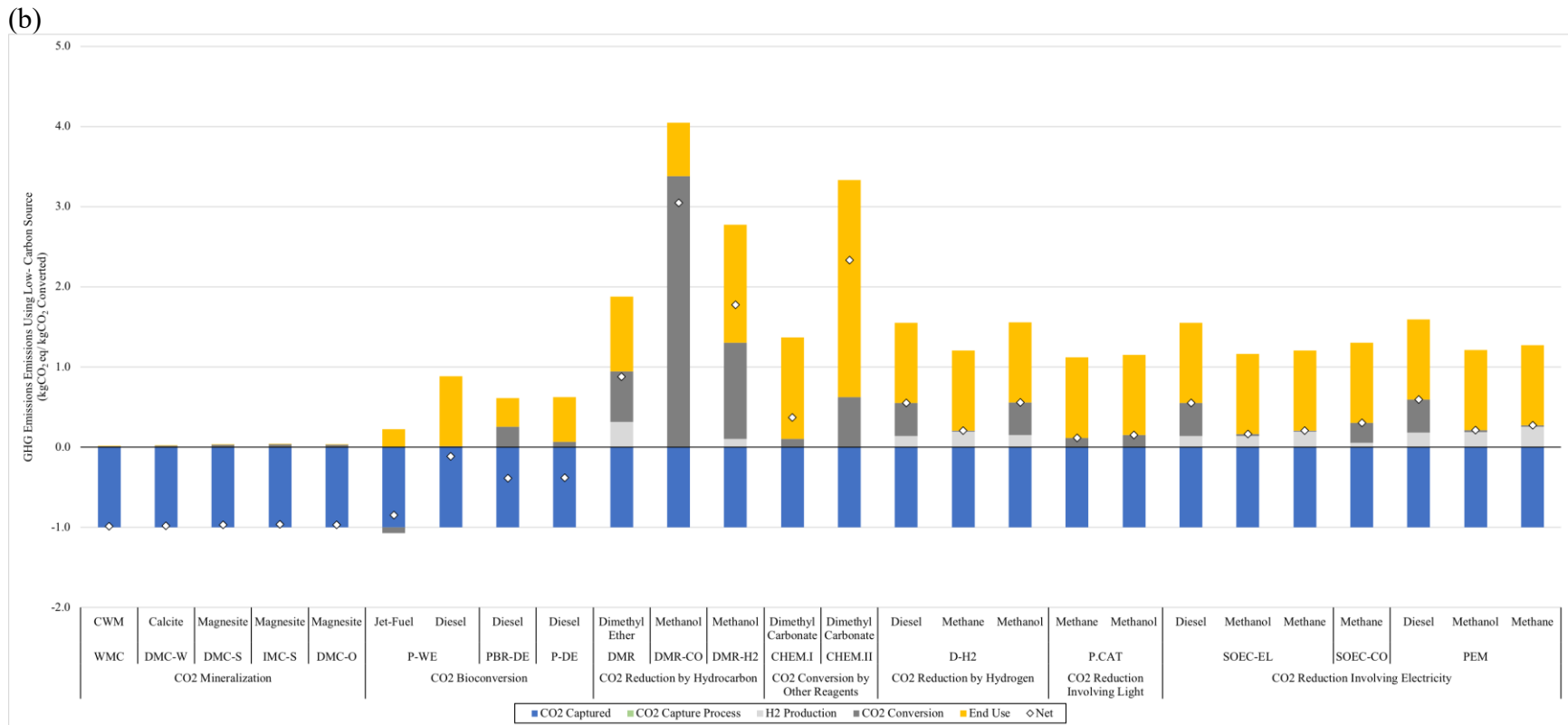


Figure 4.3 (a) Base case (natural gas) greenhouse gas emissions from different life cycle stages in kgCO<sub>2</sub>eq/CO<sub>2</sub>converted. (b) Low-carbon source greenhouse gas emissions from different life cycle stages represented in kgCO<sub>2</sub>eq/CO<sub>2</sub>Converted. Life cycle stages organized by category, subcategory and product.

gas) CO<sub>2</sub> reduction by hydrogen, hydrocarbon and electricity pathways utilize hydrogen in their processes. Hence, the energy cost of hydrogen production adds additional GHG emissions to their life cycles; the contribution of hydrogen production is up to 80% in some CO<sub>2</sub> reduction involving electricity pathways (Figure 4.3a). Replacing the hydrogen use stage provided from steam methane reforming in the base case, by electricity from renewable sources in the low-carbon scenario reduces GHG emissions for the life cycle of various pathways (Figure 4.3b). The reduction in GHG emissions for CO<sub>2</sub> reduction involving electricity and CO<sub>2</sub> reduction involving hydrogen using low carbon sources is noticeable compared to the base case pathways (Figures 4.3a and 4.3b). The PEM-Methane pathway GHG emissions are 5-fold lower (Figure 4.3b). Alternatively, in Figure 4.3b the CO<sub>2</sub> reduction by hydrocarbon show high CO<sub>2</sub> conversion emissions due to emissions arising from the methane input and carbon monoxide evolution stages. The other low-carbon source net negative emissions such as CO<sub>2</sub> bioconversion is as a result of decreased CO<sub>2</sub> conversion process emissions. This is due to the fact that electrical and heat requirements from the natural gas source, that being a natural gas power plant and an industrial furnace, respectively, are replaced by renewable electricity (hydropower, solar-photovoltaics, wind and nuclear) and geothermal heat source, respectively. Moreover, in another study, CO<sub>2</sub> mineralization has been shown to have lower emissions than other CCU options, such as, biodiesel from microalgae (Cuellar-Franca & Azapagic, 2015). However, the novelty of the work presented here is that it accounts for the amount of CO<sub>2</sub> captured which makes some technologies, notably CO<sub>2</sub> mineralization, net negative. Additionally, in the low-carbon scenario, the CO<sub>2</sub> is considered to be a CO<sub>2</sub> removal technology, as the process uses DAC.

CO<sub>2</sub> mineralization is shown to have the lowest GHG emissions using low-carbon energy sources: -0.98 to -0.96 kgCO<sub>2</sub>/CO<sub>2</sub>Converted for waste mineral carbonation and indirect mineral

carbonation using serpentine, respectively. Additionally, mineralization is also depicted as clearly the lowest GHG intense category for the base case (natural gas) source: -0.68 to -0.35 kgCO<sub>2</sub>/CO<sub>2</sub>Converted for waste mineral carbonation and direct mineral carbonation using olivine, respectively. The lowest net emissions are shown for the waste mineral carbonation subcategory that uses basic oxygen furnace slag (BOFS) to produce carbonated waste materials and the direct mineral carbonation using wollastonite subcategory that produces calcite (Figure 4.3a and Figure A2). Literature confirms these findings as wollastonite-ordinary Portland cement (OPC) blocks and slag-OPC blocks have the lowest GWPs from all of the different mix designs examined in the study, the boundary of this study is cradle-to-gate (Huang, et al., 2019). Direct aqueous carbonation, carbonation mixing and carbonation curing routes show negative values in the scenarios assessed (Thonemann, et al., 2022), as well as aqueous carbonation of BOFS generating lower carbon emissions than other types of slag (Shao, et al., 2022). However, additional factors must also be considered in order to determine the viability of CO<sub>2</sub> mineralization, these include techno-economics, such as developing economies of scale to compete with conventional cement production, the durability of the products and associated quality control measures for their replacement, chemical barriers to reactions such as competitive reactions, as well as additional equipment requirements for mineral carbonation facilities (The National Academies Press, 2019). Challenges also exist for other CCU technologies, such as economic challenges in production chains from cultivation to biodiesel production that hinder the commercialization of biodiesel from microalgae (Slade & Bauen, 2013).

#### ***4.3.3 Global CO<sub>2</sub> Emissions Reduction Potential using the Base Case for CCU Technologies***

The global CO<sub>2</sub> reduction potential is a function of the difference between the net emissions of the CCU technology and the incumbent processes, multiplied by the market size of the

product. This ratio allows to distinguish and compare benefits provided by large market sizes. It is also product specific, in that it compares products on a kilogram of product converted basis to the incumbent product. The year 2015 was selected as the market year and 50% market penetration was assumed, based on available market information (Section 4.2.3.4). Positive global CO<sub>2</sub> emissions reduction potential represents pathways which offer CO<sub>2</sub> mitigation potential compared to incumbent processes. These positive global CO<sub>2</sub> emission reduction potential for the base case, which assumes all inputs to be from natural gas sources are shown by category and subcategory combinations with their respective products in Figure 4.4.

The CO<sub>2</sub> mineralization category has the highest global CO<sub>2</sub> reduction potential. This is an order of magnitude larger than several other CCU pathways. CO<sub>2</sub> mineralization is considered to replace the cement market due to its high-volume application, as supported by a vast number of literature sources (Pan, et al., 2016) (Sanna, et al., 2012) (Monkman & MacDonald, 2017) (CEMCAP, 2017). The higher global CO<sub>2</sub> reduction potential is attributed to a large market size for cement replacement (4.4 Gt/year) (Kurad, et al., 2017), compared to lower market sizes for other products such as methanol (0.07 Gt/year) and methane (2.33 Gt/year) (ICEF, 2016). It is also attributed to the large avoided emission potential of CO<sub>2</sub> mineralization, as will be discussed in Section 4.3.7. Similar gigaton level potentials have also been projected for mineralization products in (Hepburn, et al., 2019).

The second factor affecting the annual global CO<sub>2</sub> emission reduction potential is the difference of the CCU technology from the incumbent emissions (avoided emissions). Compared to CO<sub>2</sub> mineralization, all other categories except some CO<sub>2</sub> bioconversion pathways and CO<sub>2</sub> conversion by other reagents, have lower CO<sub>2</sub> mitigation potential. For example, CO<sub>2</sub> bioconversion for pond-wet extraction has lower emissions (0.547 kgCO<sub>2</sub>eq/kg Diesel)

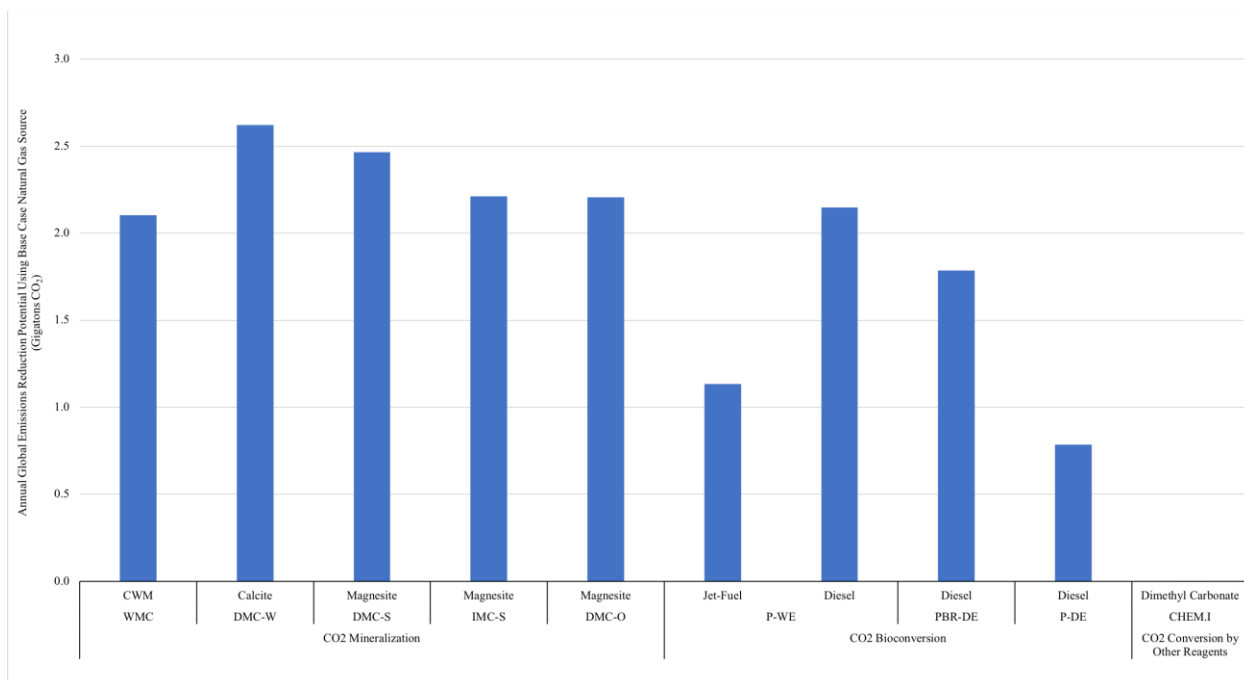


Figure 4.4 Positive values for annual global CO<sub>2</sub> emission reduction potential using base case (natural gas) source assuming 50% market penetration, represented by category, subcategory and product in Gigatons (Gt) of CO<sub>2</sub>.

compared to incumbent processes (3.8 kgCO<sub>2</sub>eq/kg Diesel) (Argonne National Laboratory, 2019). The market size is also quite small for diesel (1.32 Gt/year) (BP P.L.C, 2020) compared to CO<sub>2</sub> mineralization (4.4Gt/year) (Kurad, et al., 2017). CCU categories such as CO<sub>2</sub> reduction involving electricity, light, CO<sub>2</sub> reduction by hydrocarbon, hydrogen, as well as some of the CO<sub>2</sub> conversion by other reagents pathways have adverse CO<sub>2</sub> mitigation potential. For example, CO<sub>2</sub> reduction involving electricity with polymer electrolyte membrane (PEM) does not contribute to reducing emissions because the products emit higher CO<sub>2</sub> emissions (0.29 kgCO<sub>2</sub>eq/MJ Methane) compared to the incumbent process (0.06 kgCO<sub>2</sub>eq/MJ Methane) (Argonne National Laboratory, 2019). These findings are only applicable for the base case energy source (natural gas), findings for low-carbon emitting sources are also shown in the next section. For reference, the annual global anthropogenic CO<sub>2</sub> emissions were 36.2 Gt for the year 2015 (Jos, et al., 2016).

Based on these findings, CO<sub>2</sub> mineralization depicts some of pathways with the highest potential to mitigate emissions, using base case natural gas.

#### ***4.3.4 Global CO<sub>2</sub> Emissions Reduction Potential using the Low Carbon Source for CCU Technologies***

Figure 4.5 shows the global CO<sub>2</sub> emission reduction potential for CCU categories based on a low-carbon energy source. Only positive global CO<sub>2</sub> emission reduction potentials are shown in the figure. Figure 4.5 shows more categories, subcategories and products compared to Figure 4.4, due to the fact that utilizing a low-carbon energy source confers GHG emission reductions for CCU pathways, thereby making the emissions from these pathways lower than their incumbents hence, increasing the potential to mitigate CO<sub>2</sub>. Moreover, base case positive global CO<sub>2</sub> emission reduction potential categories have increased potential when a low-carbon source was used. The highest annual global emission reduction potential is observed for the photobioreactor-dry extraction subcategory (4.57Gt), with a market share of 1.32Gt (BP P.L.C, 2020). This is as a result of the relative amount of CO<sub>2</sub> converted per kg product.

The greatest number of subcategories with higher annual global emission reduction potential is observed for the CO<sub>2</sub> mineralization category. Specifically, direct mineral carbonation using wollastonite has a global CO<sub>2</sub> emission reduction potential of 3.21 Gt. Compared to Figure 4.4, it's obvious that in Figure 4.5, the direct mineral carbonation using olivine has a higher CO<sub>2</sub> emission reduction potential than indirect mineral carbonation using serpentine. This difference is attributed to the fact that the emissions from energy are similar for both pathways using the low-carbon source. But, the ratio of CO<sub>2</sub> converted to product formed is higher for the direct mineral carbonation using olivine pathway compared to the indirect mineral carbonation using serpentine pathway. Other literature findings indicate large variability in GWP with the biodiesel



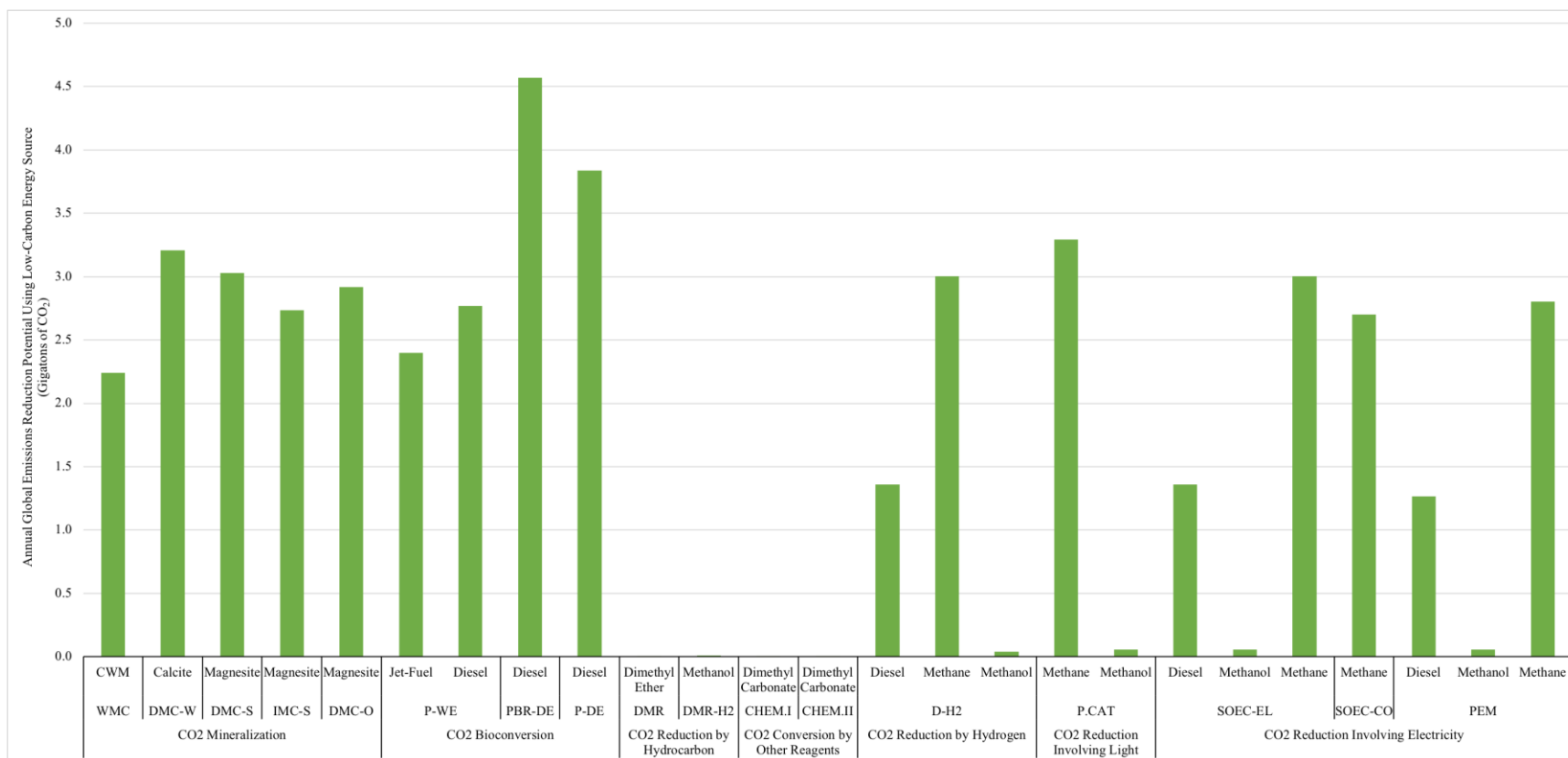


Figure 4.5 Positive values for annual global CO<sub>2</sub> emission reduction potential using low-carbon source assuming 50% market penetration, represented by category, subcategory and product in gigatons (Gt) of CO<sub>2</sub>.

from microalgae production (Cuellar-Franca & Azapagic, 2015). Hence, depicting scenarios when this pathway has the lowest GWP, even so, much lower than mineralization. However, boundary and conversion factor consistency are not maintained in this literature.

Drastic increases in global CO<sub>2</sub> emission reduction potential using low-carbon versus base case (natural gas source) is observed for the CO<sub>2</sub> reduction involving electricity pathways. Methane from the solid oxide electrolyser cell electrolysis subcategory was observed to be 3.0Gt with the low-carbon source, whereas it was -9.3Gt using the base case energy source. This is primarily because as shown in Figure 4.3, this pathway depends heavily on hydrogen requirements. Thus, substituting electrical needs with a low-carbon source for the hydrogen production reduces the GHGs significantly and increases the global CO<sub>2</sub> emission reduction potential drastically. Products such as methanol and jet-fuel do not have a pronounced market potential because their market sizes are relatively small, 0.07 Gt/year (ICEF, 2016) and 0.29 Gt/year (U.S. Energy Information Administration, 2020) respectively, for 2015.

#### ***4.3.5 Comparison of Life Cycle GHG Emissions on per kg Product Basis for Fuels and Chemicals***

As shown in Figure 4.6a, GHG emission from diesel production via the bioconversion pathway is below the incumbent (0.088 kg CO<sub>2</sub> eq/MJ Diesel). Compared to wet extraction (0.013 kg CO<sub>2</sub> eq/MJ Diesel), a higher GHG emission was observed from dry extraction (0.025 kg CO<sub>2</sub> eq/MJ Diesel). These emissions were contributed primarily from the drying and lipid extraction processes, as reflected from the CO<sub>2</sub> conversion process stage (Lardon, et al., 2009). CO<sub>2</sub> reduction involving electricity has more emissions than bioconversion. PEM has more emissions because it has higher electrical energy demand (54 kWh/kg H<sub>2</sub>) (Hoppe, et al., 2017) for electrolysis than SOEC-EL (41 kWh/kg H<sub>2</sub>) (Sunfire, 2018). The net CO<sub>2</sub> emissions from the

CO<sub>2</sub> reduction involving electricity category is higher than the incumbent process (crude oil upgrading and refining) (0.088 kg CO<sub>2</sub> eq/MJ Diesel) (Argonne National Laboratory, 2019). The performance of diesel production via hydrogenation is better than the involving electricity pathways.

Among all the categories in Figure 4.6b, CO<sub>2</sub> reduction by hydrogen results in the lowest net CO<sub>2</sub> emissions (0.16 kg CO<sub>2</sub>eq/MJ Methanol) compared to all other categories and incumbent technology (0.093 kg CO<sub>2</sub> eq/MJ Methanol) (Argonne National Laboratory, 2019). The production of methanol with syngas from natural gas via steam methane reforming is an established commercial process and was considered as the incumbent technology for comparison across the categories and sub-categories in Figure 4.6b. The CO<sub>2</sub> reduction by hydrogen category does not have significant energy demand compared to CO<sub>2</sub> reduction by hydrocarbon and involving electricity, which have high electricity requirement and hydrogen demands, respectively. Comparing dry methane reforming with CO purging and external hydrogen show a CO<sub>2</sub> to methanol ratio of approximately 2:1 and 1:1, respectively. Thus, external hydrogen has better CO<sub>2</sub> utilization (Luu, et al., 2015). Additionally, it is noted regarding the carbon footprint, that the combination of syngas and conventional synthesis and direct hydrogenation are equally suitable processes for methanol production (Artz, et al., 2017).

Similar to methanol, the production of methane resulted in the lowest CO<sub>2</sub> emissions when CO<sub>2</sub> was reduced by hydrogen compared to other categories (light, electricity) (Figure 4.6c). The CO<sub>2</sub> reduction involving electricity requires electricity and in the process, it creates significant CO<sub>2</sub> emissions. Comparatively, the hydrogen produced from steam methane reforming in the reduction by hydrogen pathway releases less CO<sub>2</sub> emissions. In the reduction involving electricity subcategory, the main challenge is coming from the energy and material demand of

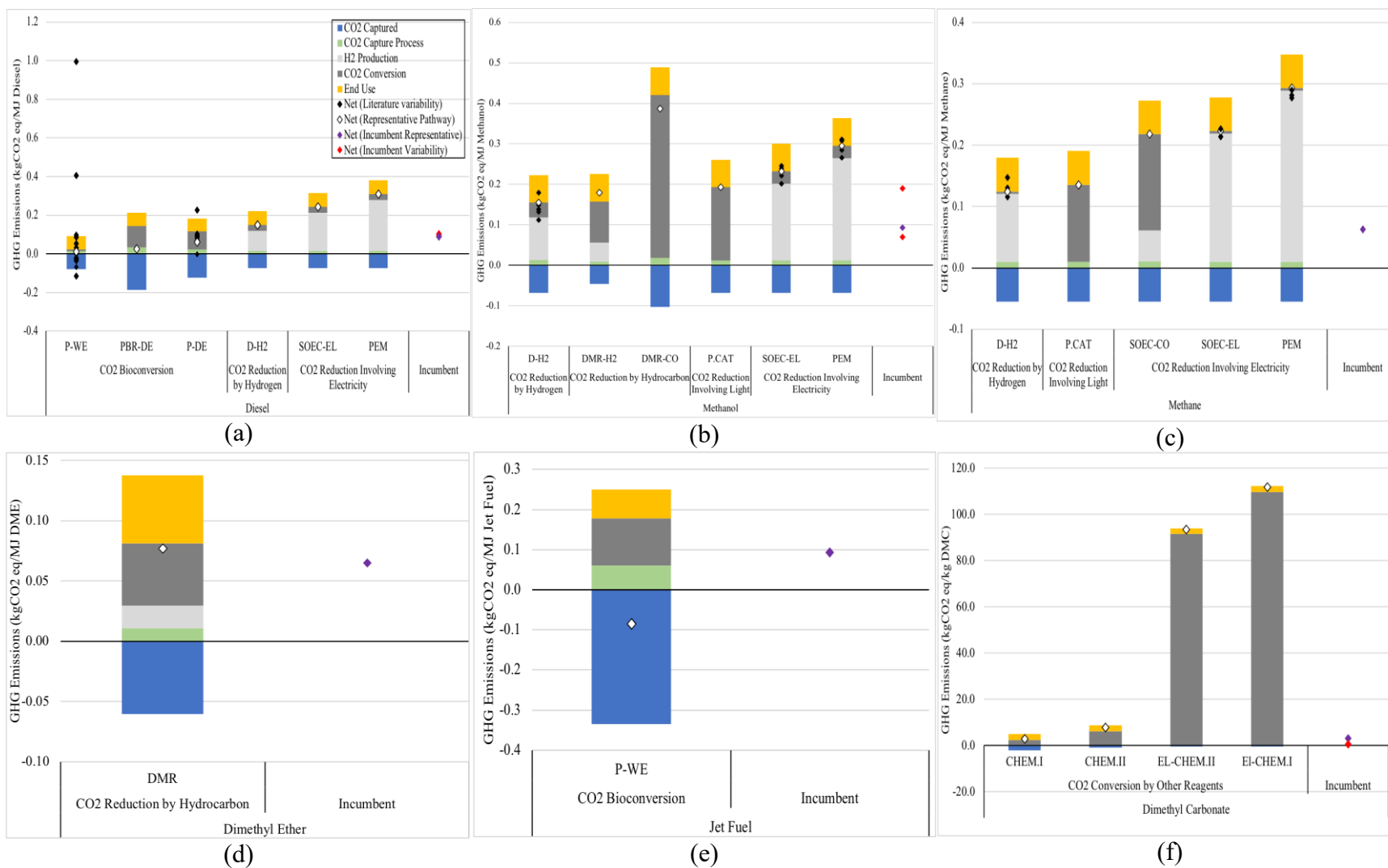


Figure 4.6 Base case GHG emissions (kgCO<sub>2</sub>eq/MJ) for diesel (a), methanol (b), methane (c), dimethyl ether (DME) (d) and jet-fuel (e) and GHG emissions (kgCO<sub>2</sub>eq/kg) of dimethyl carbonate (DMC) (f). White diamonds represent net GHG emissions, black diamonds represent net literature variability, purple diamonds represent net incumbent representative and red diamond represents net incumbent variability.

the electrolysis process (Hoppe, et al., 2017). Use of renewable sources of energy lessens this challenge and thus lowers overall GHG emission. As can be seen from Figure 4.6c, the largest contributor to GHG emission is hydrogen production stage. This is noted in another source which mentions methane via CCU reduces emissions only if water electrolysis is powered by renewable energy to produce the hydrogen (Garcia-Garcia, et al., 2020). The net CO<sub>2</sub> emissions from these categories are compared to an incumbent technology, which is natural gas from conventional recovery (0.063 kg CO<sub>2</sub> eq/MJ Methane) (Argonne National Laboratory, 2019).

Figure 4.6d compares GHG emission from dimethyl ether (DME) production via CO<sub>2</sub> reduction by hydrocarbon to GHG emission from incumbent DME production. GHG emission during DME production via dry methane reforming reduction reaction was higher (0.080 kg CO<sub>2</sub> eq/MJ DME) by 18% compared to GHG emissions from DME production via incumbent processes (0.065 kg CO<sub>2</sub> eq/MJ DME) (Schakel, et al., 2016). DME production stage contributed over 68% of the net GHG emission followed by H<sub>2</sub> production (25%). However, with a single LCA result it is difficult to fully understand the performance of DME production via CO<sub>2</sub> utilization.

Figure 4.6e shows GHG emission from one case for jet fuel production via photosynthetic carbon dioxide conversion. The difference in net emission from the wet extraction subcategory (-0.084 kg CO<sub>2</sub> eq/MJ Jet-fuel) was 189% lower compared to the incumbent (0.094 kg CO<sub>2</sub> eq/MJ Jet-fuel) (Ou, et al., 2013). While this result gives some insights, it is difficult to fully understand the performance of jet-fuel production from microalgae using a single LCA result.

Figure 4.6f compares GHG emissions from chemical synthesis of dimethyl carbonate (DMC) from CO<sub>2</sub> via ethylene (2.93 kg CO<sub>2</sub> eq/kg DMC) and urea routes (7.78 kg CO<sub>2</sub> eq/kg

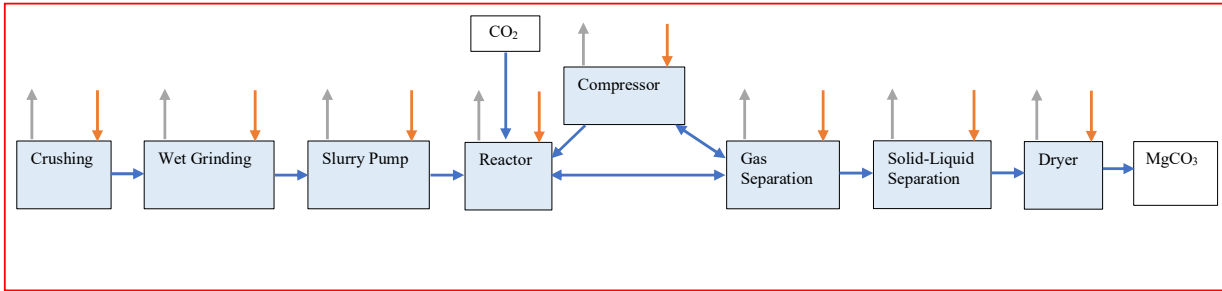
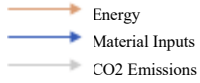
DMC), as well as electrochemical synthesis processes (93.5-111.8 kg CO<sub>2</sub> eq/kg DMC) to incumbent process (3.18 kg CO<sub>2</sub> eq/kg DMC) (Garcia-Herrero, et al., 2016). El-Chem.I and El-Chem.II are two electrochemical CO<sub>2</sub> reduction processes that use water and aniline for extractive distillation, respectively. Both of the latter processes showed higher GHG, which was mainly contributed by the energy consumption in the DMC separation process and burning of natural gas for steam generation (Garcia-Herrero, et al., 2016). This is supported by other meta-analysis which shows CO<sub>2</sub> based DMC have higher impacts compared to conventional processes (Nils, 2020). Comparatively, the chemical synthesis processes of DMC production via ethylene route reduced GHG emissions by 8% compared to the incumbent. Compared to the incumbent DMC production processes, it is claimed that the net CO<sub>2</sub> emission and global warming potential via ethylene route processes have improved by 11.4% and 58.6% respectively (Kongpanna, et al., 2015).

#### ***4.3.6 Comparison of Life Cycle GHG Emissions on a Per kg Product Basis for Materials***

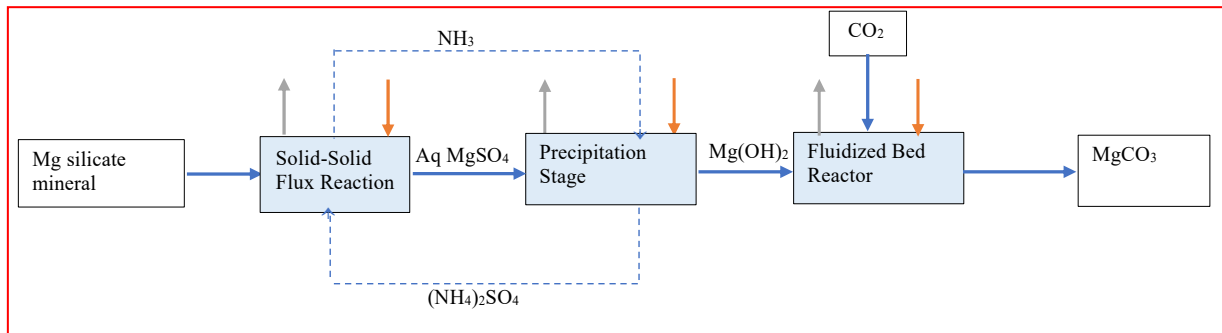
Three forms of ex-situ mineralization processes are examined in this paper: direct (Figure 4.7a), indirect (Figure 4.7b) and waste mineral carbonation (Figure 4.7c). Direct carbonation refers to a single step carbonation reaction of the magnesium and calcium ions. The process from the National Energy Technology Laboratory (NETL) (Gerdemann, et al., 2007) is categorized as such, which has been developed for serpentine, olivine and wallastonite. Indirect carbonation is composed of two steps, where the active compounds are first precipitated, followed by their carbonation. The process was developed at Åbo Akademi University (AAU) (Highfield, et al., 2012). This two-step process is only currently viable for serpentine, due to achievable reaction efficiencies (Giannoulakis, et al., 2013). Waste carbonation refers to the carbonation of waste materials such as basic oxygen furnace slag (BOFS) (Sanna, et al., 2014).

Results for the comparison of GHG life cycle emissions using base case (natural gas source) for five unique CO<sub>2</sub> mineralization subcategories examined in this study and their incumbent process emissions are shown on a per kilogram product basis in Figure 4.8. Products of CO<sub>2</sub> mineralization can replace three main markets: cements, aggregates and novel concretes (Pasquier, et al., 2018). In this study, cement is selected to be the incumbent, because all products examined in this study can replace cement directly and because cement is a high-volume market for these products (CEMCAP, 2017) (Iizuka, et al., 2004) (Sanna, et al., 2014). Additionally, adding limestone or curing cement with CO<sub>2</sub> has been noted to provide significant strength advantage to the cement mixture (Monkman & MacDonald, 2017). But it is important to note that different formulations will confer differences in important properties such as compressive strengths ( Walling & Provis, 2016). Future work should examine these potential differences in order to reconcile the true market replacement potential of these products, which is assumed to be 50% in Figures 4.4 and 4.5.

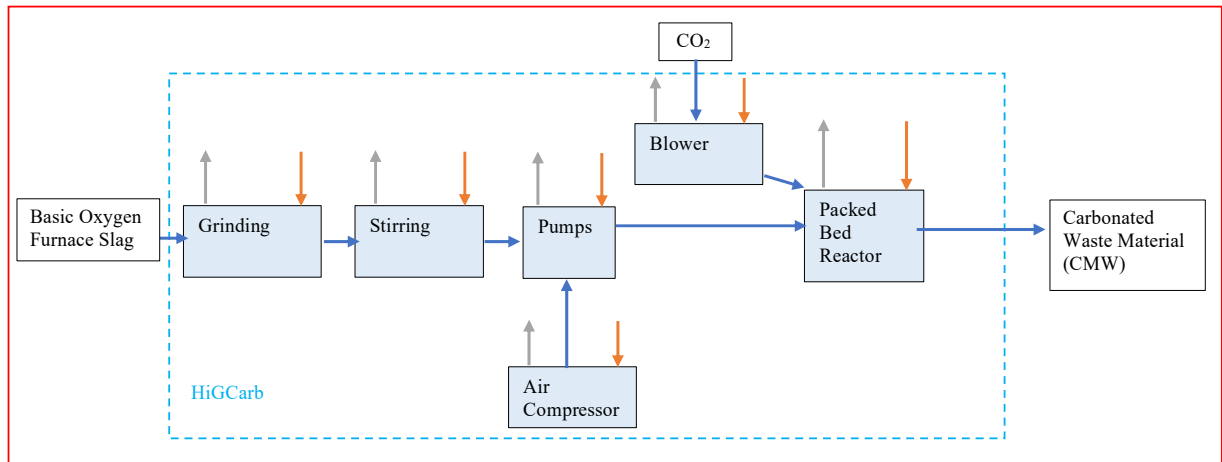
All subcategories show net negative GHG emissions on a per product basis (Figure 4.8). The lowest net GHG emissions are observed for calcite with direct mineral carbonation (-0.37kgCO<sub>2</sub>eq/kg Calcite), which is due to the high reactivity of the mineral (Sanna, et al., 2014). The serpentine minerals also show more net negative emissions than olivine. This is attributed to the fact that olivine requires mechanical activation and serpentine requires thermal activation. Thus, olivine releases more GHG emissions than serpentine (Figure 4.8), due to the fact that the heat source for the base case in this study uses a natural gas industrial furnace with a lower emission factor than natural gas electricity, that being 0.25kgCO<sub>2</sub>eq/kWh and 0.49kgCO<sub>2</sub>eq/kWh, respectively (Table 4.2). It was also observed that the emissions from the indirect mineral carbonation using serpentine were lower than those from direct mineral



(a)



(b)



(c)

Figure 4.7 System boundary for (a) direct mineral carbonation process (DMC), adapted from (Sanna, et al., 2014) (b) for indirect mineral carbonation process (IMC), adapted from (Sanna, et al., 2014) (c) for waste mineral carbonation process (WMC), adapted from (Pan, et al., 2016).



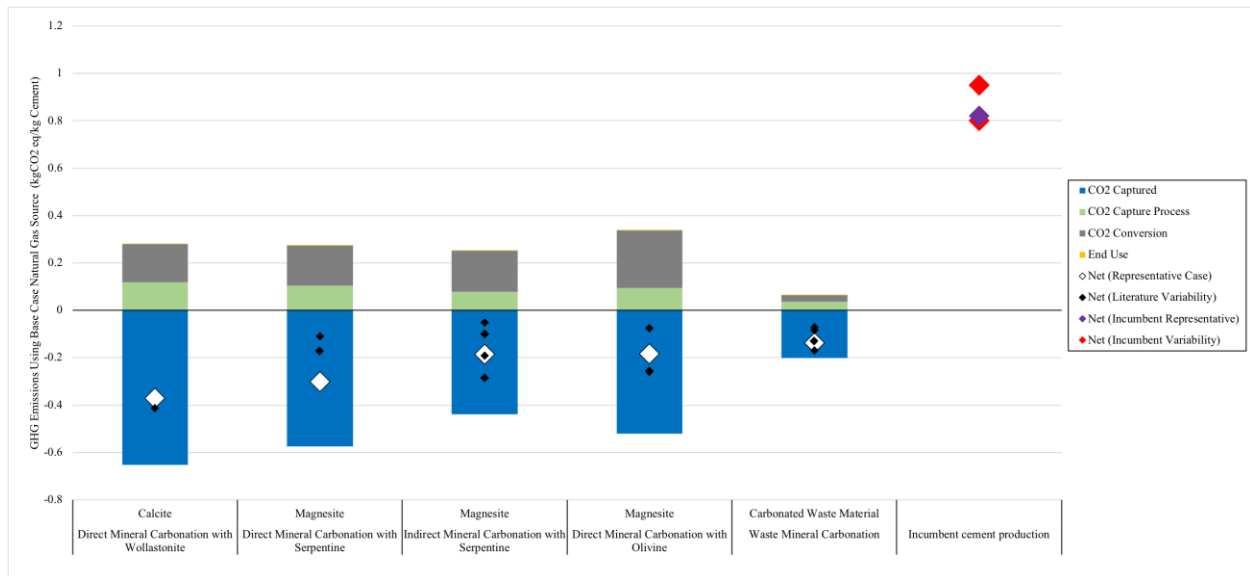


Figure 4.8 Base case (natural gas source) greenhouse gas emissions from different life cycle stages represented by category, subcategory and product in kgCO<sub>2</sub>eq/kgCement. Literature variability is depicted for each subcategory. Incumbent cement production (Josa, et al., 2004) (Smith & Durham, 2016) and variability associated with incumbents are also shown.

carbonation, that being 0.25kgCO<sub>2</sub>eq/kg Cement and 0.27kgCO<sub>2</sub>eq/kg Cement, respectively.

However, the net emissions were lower for direct mineral carbonation compared to indirect carbonation due to the larger ratio of kgCO<sub>2</sub>eqCaptured/kg Cement. Lastly, although carbonated waste materials had the lowest process emissions compared to any other observed pathway, i.e. 0.066kgCO<sub>2</sub>eq/kg WMC, it appears to have the highest net emissions (-0.14kgCO<sub>2</sub>eq/kg Carbonated Waste Material). This is attributed to a lower ratio of CO<sub>2</sub> converted to the amount of product formed.

The end use emissions for all mineral carbonate products is at least two orders of magnitude lower than the CO<sub>2</sub> converted emissions. The lowest end use emissions per kilogram of product was for direct mineral carbonation using wollastonite. It was assumed that these products replaced 5% of the cement in concrete (ASTM International, 2019) (Monkman & MacDonald, 2017). Allocation was used to determine the emissions attributed to the replacement of cement with mineral carbonation products in 1 m<sup>3</sup> of concrete.

### 4.3.7 CO<sub>2</sub> Avoided Emissions for Mineralization

All CO<sub>2</sub> avoided emissions for CO<sub>2</sub> mineralization subcategories using base case (natural gas source) are on the positive axis as shown in Figure 4.9. This indicates that all of the categories have CO<sub>2</sub> mitigation potential if used as a replacement for the cement market. The highest CO<sub>2</sub> avoided emissions are observed for wollastonite (1.19kgCO<sub>2</sub>eq/kgCement). Indicating that it has the largest difference in net emissions compared to the incumbent, cement. Serpentine pathways use heat for the activation of the minerals, thus using heat recovery will further minimize emissions and increase the CO<sub>2</sub> avoided emissions further. Additionally, using low carbon sources will further enhance the potential of these subcategories and pathways, as discussed in the next section.

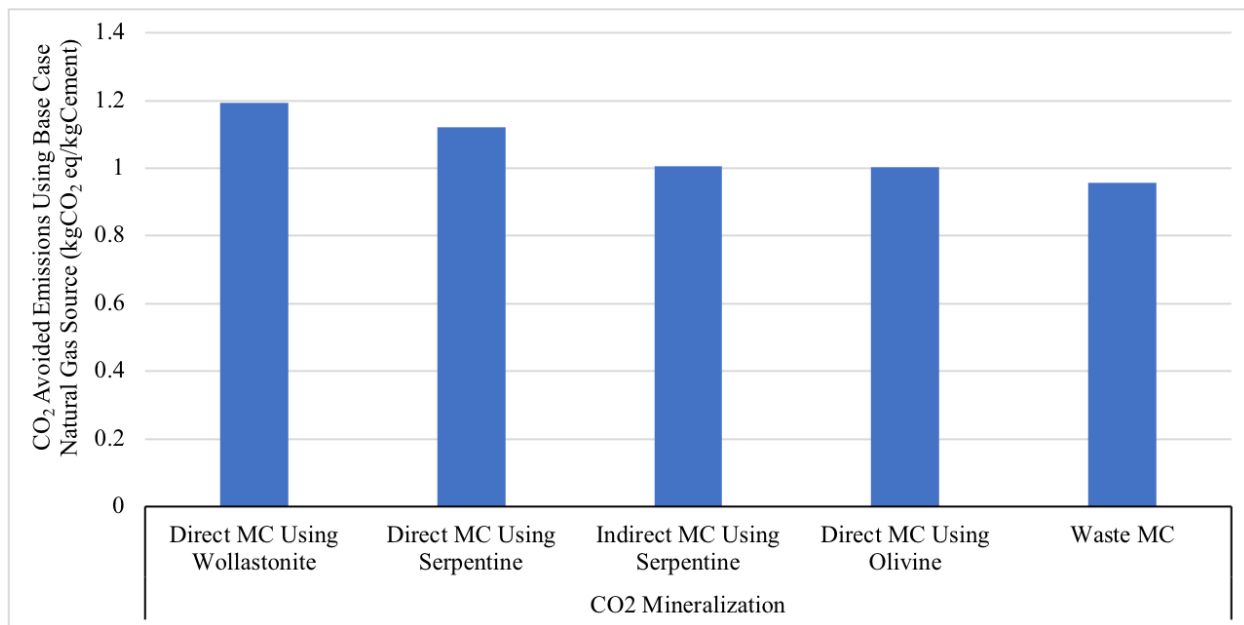


Figure 4.9 Mineralization CO<sub>2</sub> avoided emissions using base case (natural gas source) greenhouse gas emissions from different life cycle stages represented by category, subcategory and product. The selected incumbent is cement

#### ***4.3.8 Sensitivity Analysis for CO<sub>2</sub> Mineralization***

Indirect mineral carbonation using serpentine has many variables that can be varied in the sensitivity analysis, including heat requirements, electricity source and end-use applications. The sensitivity parameters that were examined in this study and their effects on the net emissions are depicted for indirect mineral carbonation using serpentine in Figure 4.10. These parameters are organized from the largest variability at the top of the figure to the smallest at the bottom.

Twenty-two sensitivity parameters were varied for this analysis, showing that electricity source differences confer the highest variability in net emissions. The electrical source shows a range between -0.36 kgCO<sub>2</sub> eq/kg Cement and 0.16 kgCO<sub>2</sub> eq/kg Cement for low-carbon and coal, respectively. Suggesting that using low-carbon source has the potential to mitigate CO<sub>2</sub> emissions most drastically. As shown, the next highest variability parameter is the CO<sub>2</sub> capture source. Direct air capture using low-carbon energy source provides significant GHG savings compared to capture from coal power plant flue using energy from the power plant.

Interestingly, the scenario with a reduction in ordinary Portland cement by 25% and absorption of 16.2% CO<sub>2</sub> during curing (Lim, et al., 2019) results in 0.05kgCO<sub>2</sub>eq/kg Concrete. The emissions are for concrete and do not use allocation because cement is not being replaced in this scenario. This shows the distinction between cement and concrete products as replacement markets. Allocation also affects the net emissions. Five percent cement replacement without allocation results in -0.12kgCO<sub>2</sub>eq/kg Concrete, which is higher than the base case shown in Figure 4.10. In contrast, when 50% and 0% cement replacement and allocation is applied to the concrete production using natural gas energy source, the variation was not large as shown in the figure, and the emissions for the 50% scenario is higher than the base case scenario. Indicating that larger cement replacement with mineral carbonate products via allocation doesn't always lead to greatly lower emissions. This is due to the fact that the emissions represent the percentage

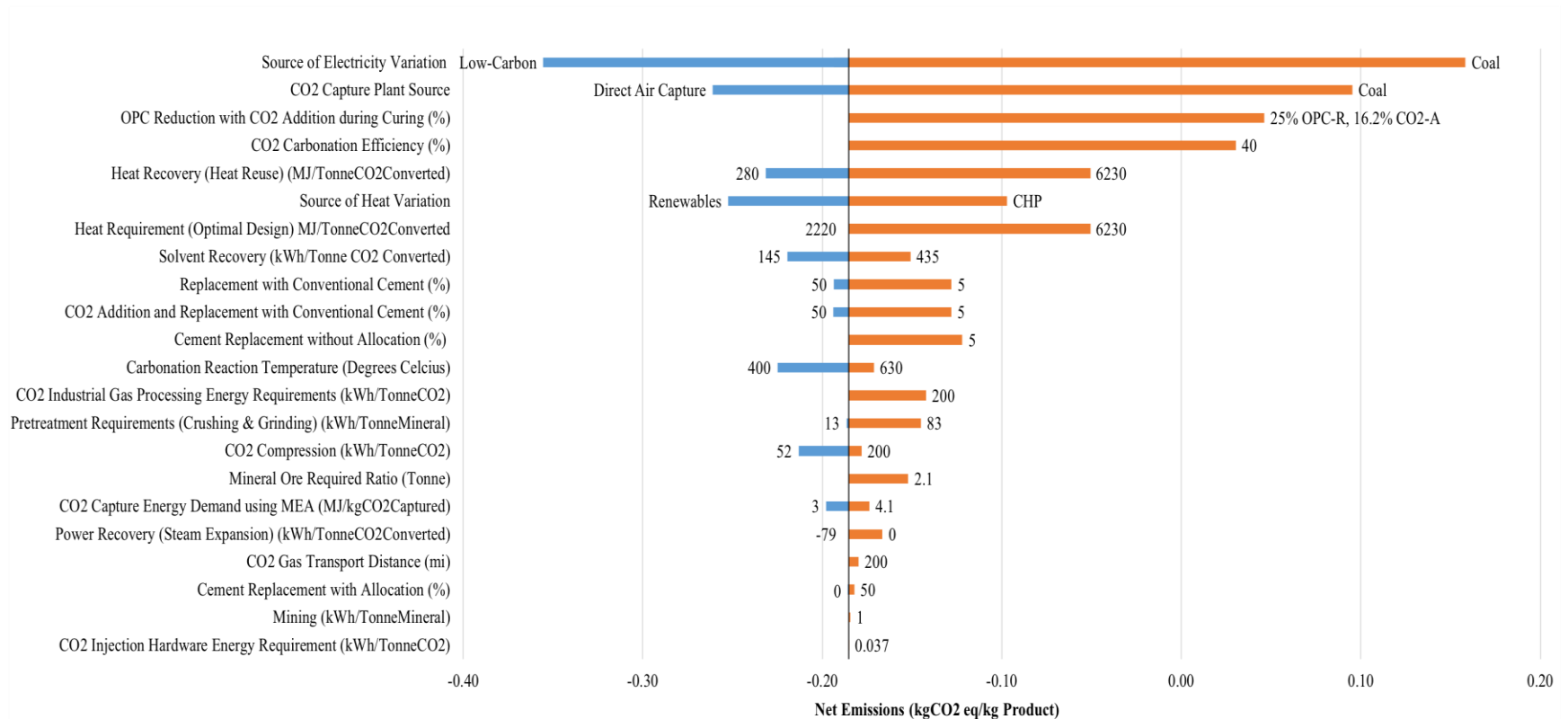


Figure 4.10 Indirect mineral carbonation using serpentine sensitivity results showing effects on net emissions of varying each parameter indicated on the left. If energy source is not varied, assume base case natural gas emission factors were utilized

of concrete emissions allocated to the amount of cement that is replaced in the concrete. Replacing 5% cement with conventional cement production (1.04kgCO<sub>2</sub>eq/kg Cement) (Monkman & MacDonald, 2017) has very similar GHG emissions as 5% cement replacement without allocation. But, increasing the replacement ratio to 50% causes a further decrease in emissions using the replacement with conventional cement to -0.19kgCO<sub>2</sub>eq/kg concrete, contrary to the without allocation and 50% allocation scenario. The raw material data for concrete production modelling in this study was retrieved from (Nisbet, et al., 2002). A cement replacement value of 5% was selected because it doesn't compromise the strength of the cement mixture, as discussed in literature and international standards organizations (Matschei, et al., 2007) (Monkman & MacDonald, 2017) (ASTM International, 2019).

A decrease in carbonation reaction efficiency to 40% (Sanna, et al., 2014) is also shown to adversely affect GHG emissions. Heat recovery can also assist in further reducing emissions by 1940MJ/Ton CO<sub>2</sub> (Nduagu, et al., 2012), leading to net emissions of -0.23 kgCO<sub>2</sub>eq/kg Cement. Moreover, altering heat sources by using electrical heaters with 100% efficiency with low-carbon sources can also reduce net emissions further, whereas combined heat and power plants, which accounts for allocation of GHG emissions, leads to an increase in emissions. The use of optimally designed heat exchangers also contributes to reducing energy consumption by over three-fold (Giannoulakis, et al., 2013) as indicated in Figure 4.10.

The industrial gas processing energy requirements (200 kWh/Tonne CO<sub>2</sub>) to create liquid CO<sub>2</sub> affects the GHG emissions more than the other gas related factors, such as the energy required for the injection hardware for the gas during concrete batching and mixing. The gas transport using heavy duty trucks was also determined to be low for distances up to 200 miles (Monkman & MacDonald, 2017). Similar results were observed for the mineral mining

emissions, whereby 0.25 kWh/Tonne (Bleiwas, 2011) was increased by 4-fold using the sensitivity shown. This had relatively negligible effects on the base case emissions.

The parameter, solvent recovery/consumption, was varied by plus and minus 50%, this stage can further reduce GHGs by 0.03 kgCO<sub>2</sub>eq/kg Cement. The operational temperature for heat treatment of the minerals was 550°C, which can vary between 630°C and 400°C, with lower temperatures compromising mineral reactivity (Penner, et al., 2004). Pre-treatment requirements, specifically crushing and grinding increases the base case emissions as shown in the figure. Decreasing the mineral ore requirements to 2.1Tonnes (Sanna, et al., 2014) also increases the emissions to -0.15kgCO<sub>2</sub> eq/kg Cement. Changing the base case CO<sub>2</sub> compression energy requirements to 52 kWh/t-CO<sub>2</sub> (Nduagu, et al., 2012) also contributed to decreasing the net emissions of the process. The CO<sub>2</sub> capture energy demand was also varied from 4.1MJ to 3MJ, due to variability in energy requirements (Helmeth Consortium, 2017). Lastly, power recovery from the steam expansion process derived from the fluidized bed reactor contributes to a 79kWh/Tonne CO<sub>2</sub>Converted (Giannoulakis, et al., 2013) reduction in energy consumption. As expected, if this is not incorporated it can also increase the base case emissions of the process.

#### **4.4 Conclusion**

This study attempts to characterize, assess and compare different carbon conversion utilization technologies among and within each other. Due to the vastness of the field, a categorization scheme is presented to categorize eight relevant CCU technologies. These include CO<sub>2</sub> reduction by a hydrocarbon, CO<sub>2</sub> reduction by hydrogen, CO<sub>2</sub> reduction by other reagents, CO<sub>2</sub> reduction involving electricity, CO<sub>2</sub> reduction involving light, CO<sub>2</sub> mineralization, CO<sub>2</sub> bioconversion and other CO<sub>2</sub> conversions. The categories were filtered into subcategories with

respect to products and starting materials. The life cycle GHG emissions of these CCU technologies were assessed using consistent system boundaries, emission factors and conversion calculations. Two assessment metrics were examined: kg CO<sub>2</sub> equivalents per kg CO<sub>2</sub> converted, and kg CO<sub>2</sub> equivalents per kg or megajoule (MJ) of product. Two additional metrics were also studied to compare technologies to their incumbent processes and markets using CO<sub>2</sub> avoided emissions and global emissions reduction potential, respectively. A full sensitivity analysis was conducted. Results showed base case net negative GHG emissions for CO<sub>2</sub> mineralization due in part to the fact that there are lower end-use emissions when used as a replacement for cement in concrete applications. This is also advantageous due to the large cement market size compared to the other markets presented in this study. CO<sub>2</sub> mineralization also doesn't utilize hydrogen contributing further to its competitive advantage over other categories presented in this study. Net negative GHG emissions for the base case were also observed for CO<sub>2</sub> bioconversion.

The quality of data presented in this study has limitations, given that early stage TRL technologies are examined. As such, the impact results may improve as these technologies are scaled and advance to full commercialization as a result of improved design and efficiencies. Additionally, a limitation to this study is characterizing the exact level of market penetration by the replacement of the CCU incumbent processes.

Future work should expand findings to these and other CCU categories. The purity of the CO<sub>2</sub> captured and its effect on the different pathways needs further investigation. Additionally, the life cycle emissions of other impacts, besides GHG emissions may also be carried out using a similar consistent assessment method. Finally, a more sophisticated market and techno-economic assessment would assist in getting a holistic view. Collaboration with industry and companies currently involved in this field would also assist in achieving these goals. This work informs part

of an open source, accessible tool that estimates GHG emissions from representative CCU pathways (Nishikawa, et al., 2020)<sup>1</sup>. This can assist in structuring CCU technologies and act as an investment tool for decision makers.

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<sup>1</sup> The author Shah Ahmad's contribution are concept formation, methodology development, incumbent selection, data collection, analysis, and technical documentation writeup for the mineralization, bioconversion and CO<sub>2</sub> reduction involving light pathways.



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## **Chapter Five: Barriers and Opportunities of Carbon Utilizing Mineralization Technologies in Canada**

### **5.1 Introduction**

In response to rising atmospheric greenhouse gas (GHG) emissions successive international mitigation agreements increasingly force governments and carbon intensive industry alike to shift from policy pledges to investing in demonstratable mitigation technologies (Victor, et al., 2017). Among several adaptation strategies the deployment of carbon mitigating clean technology solutions at scale is being viewed as essential to meeting the climate change targets laid out by the IPCC (IPCC, 2018).

One such technology pathway is carbon capture utilization (CCU) technologies that convert carbon dioxide from industrial emission sources into value-added products (e.g., feedstock materials) (Wang, et al., 2021). Relative to more widely known carbon capture and storage (CCS), the environmental benefits of CCU lie in its ability to replace fossil and other feedstock resources as opposed to mitigating climate change by reducing atmospheric carbon dioxide (CO<sub>2</sub>). While CCU yields vary by application, the technology's potential for reducing global GHG emissions is estimated at 10-15% of annual global CO<sub>2</sub> output, especially if appropriate technical, economic and policy framework conditions are put in place (ICEF, 2016); (NAP, 2019). Among many CCU technologies discussed by experts for their mitigative potential and innovative properties (Waxman, et al., 2021) mineralization has emerged as the most promising application of CCU (Cuéllar-Franca & Azapagic, 2015); (Sanna, et al., 2014). Mineralization, the process where carbon dioxide reacts with mineral substrates to form mineral carbonates permanently sequesters CO<sub>2</sub> and provides cost-effective mitigation strategies especially for carbon-intensive industrial sectors such as cement and concrete where other means

of GHG mitigation (e.g., electrification) are currently not viable (Cuéllar-Franca & Azapagic, 2015); (Hepburn, et al., 2019); (Perez Fortes, et al., 2016). In the literature mineralization is well recognized for its ability to sequester CO<sub>2</sub> (Sanna, et al., 2014); (Giannoulakis, et al., 2013) in a process known to enhance the compressive strength of resulting concrete products (Guo, et al., 2019); (Li, et al., 2019c). However, while other climate-mitigative technologies, such as solar PV, have made significant advances in both technology capacity and cost-effectiveness towards widespread commercial adoption, CCU remains at early stages in the technology development process, with mineralization currently being considered at the demonstration stage (Jones, et al., 2017b); (Zimmermann & Schomacker, 2017); (Renssen, 2011).

A major known barrier to the commercial adoption of CCU technology pathways are economic hurdles stemming from high breakeven costs, limited mitigation potentials, and regulatory uncertainty (Al-Mamoori, et al., 2017); (Sanna, et al., 2012). While mineralization pathways show comparably low breakeven costs and significant potential to mitigate CO<sub>2</sub> through permanent storage their high degrees of uncertainty due to variable, early-stage technology readiness levels TRLs are found to inhibit market penetration (Hepburn, et al., 2019).

Beyond technology cost factors, few studies have conducted detailed sector-level analysis of framework conditions that govern CCU technology acceptance and adoption (Arning, et al., 2020). Accordingly, evidence of technology benefits, social, economic, regulatory and policy framework conditions needed to advance CCU development as a climate-mitigative strategy is limited. Studies by Rafiaani et al. (2020), Heek et al. (2017a), and Arning et al. (2017) are among a small literature that investigate individuals' CCU technology risk and benefit-cost perceptions as broad indicators of public acceptance using qualitative and survey research methods. These studies identify health and environmental risks and safety of CCU applications

and individuals' sustainability concern as important factors in CCU acceptance. However, this literature also indicates that the public tends to have limited awareness and knowledge of CCU, indicating a need for a regulation of product labelling and future life-cycle analyses (Arning, et al., 2019); (Heek, et al., 2017a). While public technology acceptance matters, the success of CCU as a CO<sub>2</sub> mitigating strategy rests on the critical nexus of technology adoption and market acceptance and thus the views of stakeholders along the CCU technology and mineralization supply chain. A second stream of previous studies on the topic of CCU adoption employed qualitative research methods to elicit barriers to CCU adoption among technology and industry experts (Arning, et al., 2020); (Muslemanni, et al., 2020); (Kant, 2017). A common conclusion drawn in this literature is that a lack of viable business models and a lack of government support for CCU technology have held back companies from commercialization and investment (Muslemanni, et al., 2020).

The objective of this study is to fill the gap in the CCU literature regarding socio-economic and regulatory factors that influence technology acceptance and views of adoption from knowledge to persuasion among industry stakeholders along the CCU supply chain. We conduct a series of semi-structured interviews with industry stakeholders ranging from CCU technology developers to potential users in the cement and concrete industry to assess their perceptions about the future adoption of mineralization technologies, perceived stakeholder benefits, and perceived risk factors and uncertainties regarding CCU-derived product, their marketability, as well as issues concerning regulatory policy conditions.

We conduct this study in the province of Alberta, Canada. As a major global producer of fossil fuels and host to a large petrochemical industry Alberta account for 38% of Canada's GHG emissions (Government of Canada, 2020). Alberta is recognized internationally as a leader in

CCS as host to the Alberta Carbon Trunk Line – one of the world’s largest CCS projects (ACTL, 2020). As such stakeholders in the Alberta CCUS industry stand exemplary for a carbon intensive economy destined to benefit from the development of commercial CCU solutions to address climate change by reducing its GHG footprint.

Given CCU’s current low technology readiness levels (TRL), we believe that a detailed case-study assessment of stakeholders’ viewpoints is critical to identifying challenges in the further technology development process. This paper contributes to this gap in knowledge by identifying barriers to the future development of CCU mineralization technologies and generating information as inputs into the development of important policy and regulatory frameworks.

## **5.2 Conceptual Framework for CCU Technology Development**

To enable the identification of challenges to mineralization technologies across different stakeholder groups, this analysis uses the concept of technology readiness levels (TRLs) as a set of metrics that assess and compare technology maturity. Ranging from basic technology research in TRL 1 through demonstrating successful implementation in operational environments in TRL 9, TRLs present a standardized assessment of mineralization’s technology maturity that facilitates comparisons of often complex techno-economic properties and communication between technology developers and diverse stakeholder groups (Zimmermann & Schomacker, 2017).

Yet, literature on socio-economic models assessing TRLs in the context of CCU technologies remains scarce. Brillhuis-Meijer et al. (2016) propose a dual-innovations model to study the dynamic interactions of technology and product development process, whereas Hepburn et al. (2019) and Zimmermann and Schomacker (2017) strictly compare technology’s

TRL. A small number studies including Kobos et al. (2018) develop integrated assessment model of CCU technology development by combining TRL measures with regulatory readiness level (RRL) and market readiness level (MRL) to conduct joint analyses of the techno, regulatory and market dimensions of technology development. Yet, none of these previous studies attempted an integrated TRL assessment across technological, economic, environmental and particularly social indicators that have been ignored to date. Several studies note that a lack of qualified data and environmental assessments hold back comparative assessments of early-stage TRL projects (Zimmermann & Schomacker, 2017).

Our analysis of stakeholder perceptions of the CCU and mineralization technology development process builds on Rogers' diffusion of innovations theory (Rogers, 1995) that describes the technology (innovation) development process towards adoption or rejection in 5 sequential stages: knowledge, persuasion, decision, implementation and confirmation (Figure 5.1). According to Rogers technology success depends on the adoption rate as a function of the attributes of the innovation, its communication through channels, the amount of time it takes and relevant social systems. Stakeholders, such as investors and private companies progress through the diffusion model guided by their strategic motives, defined targets, and outcomes for the goals (Jones, et al., 2017b). Given CCU mineralization technologies critical early TRL stage and uncertain commercial feasibility Rogers (1995) stresses the interplay of knowledge and persuasion stages as influencers of industry uptake decisions. CCU implementation in turn will predict the degree of technology diffusion and eventual commercialization of mineralization, which ultimately determine the potential carbon mitigation benefit of CCU mineralization. Research evidence of these effects in turn will be critical in providing an understanding of the broader risks and benefits of CCU technologies to policy processes.

Prior Conditions:

1. Previous practice
2. Felt needs/problems
3. Innovativeness
4. Norms of the social system

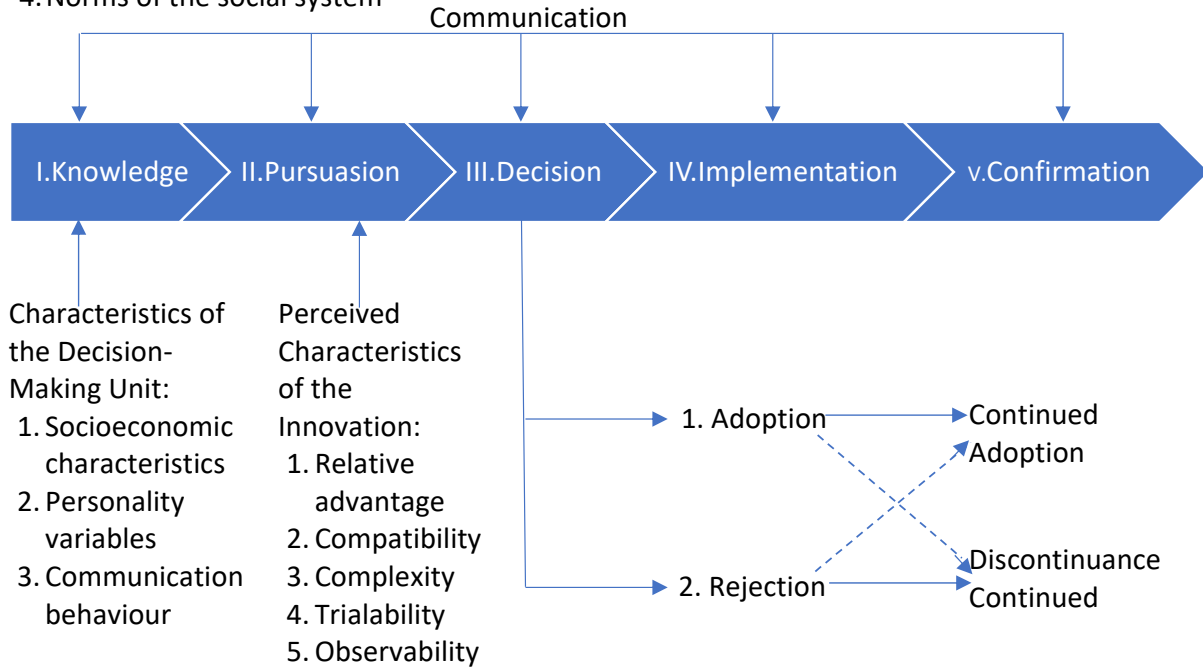


Figure 5.1 The innovation decision process through which an individual (or decision-making unit) passes through, from knowledge of an innovation, to forming an attitude, to a decision, to accepting or rejecting it, to implementation of the idea and to confirmation of the decision. (Rogers, 1995)

We investigate mineralization technology development in the context of stakeholder (company) background, areas of expertise, and perceptions of need for innovation, benefits of mineralization, as well as technology risks and challenges. Our goal is to develop a categorization of critical drivers that predict CCU mineralization adoption or rejection. Although previous modelling results for CCU (and CCS) industry investment decisions are available (e.g., (Zhang, et al., 2021); (Zhang & Liu, 2019); (Li, et al., 2019a); (Wang & Du, 2016)), few studies have used Rogers diffusion of innovation model to evaluate factors critical to supply chain and industry expert views on industry mineralization adoption decisions (Jones, et al., 2017b); (Sharp



& Miller, 2016). In doing so, we investigate five factors deemed critical for industry uptake decisions to proceed with technology adoption: perceptions of carbon emissions, technology development (Arning, et al., 2019) (Zimmermann & Schomacker, 2017), competition and collaboration, as well as mineralization risks and regulatory policy uncertainties.

Industry perceptions of carbon emission intensity and technology development have been documented as drivers of mineralization development (Arning, et al., 2019) (Zimmermann & Schomacker, 2017). Technological growth depends on the balance of competition and collaboration within an organization also referred to as coopetition (Hoffmann, et al., 2018). Coopetition may benefit firms' technology development through access to knowledge sharing networks, resource access, cost sharing, risk mitigation, whereas challenges of knowledge leakage and opportunistic behaviour can be mitigated by the creation of clear collaborative boundaries by means of clear organizational, temporal and domain separation (Audretsch, et al., 2014).

New technologies bring with them inherent risks and uncertainties which need mitigation. Firms in the CCU space assess technology uncertainties using both quantitative and qualitative methods and simulation techniques (e.g., Monte-Carlo analysis) that can help to evaluate the risks of alternative scenarios, collaborations, and real options (Zhang & Wei, 2011); (Heydari, et al., 2012). Areas of technological conflict among project partners arising from withholding of information regarding risks, risk channeling, or risk exclusion can be mitigated through transparency and uncertainty analysis that enhances trust and communication with partners and the public alike (Arning, et al., 2020); (Stankiewicz, 2009). Aside from technology- or sector specific technology risks and uncertainties, policy design and the resulting regulation of CCU mineralization can act as additional barriers to CCU commercialization and adoption at the firm

level (Xu & Dai, 2021). To date, several policy options have been proposed to enhance CCU development: tax rebates and subsidies, such as the Federal 45Q tax credit in the United States, demonstration, deployment research and infrastructure support, and market enablers (Meckling, et al., 2017); (Bobeck, et al., 2019). However, industry insight that would enable the development of policies that help companies overcome commercialization challenges via information exchange, collaboration, and cooperation are lacking to date (Yao, et al., 2018). Gaps that have inhibited this policy process include the creation of consistent standards for life-cycle analysis of GHG emissions and updated industry codes and standards to include these and other metrics, such as embodied carbon, with the expectation that they meet quality and safety standards also enhance markets for CCU mineralization-based products (Bobeck, et al., 2019). Hence, it is the goal of this study to generate empirical evidence on the above critical factors to inform CCU mineralization adoption decisions, which will in aggregate determine the pathway of diffusion and commercialization for mineralization technologies in Alberta, Canada and elsewhere.

### **5.3 Empirical approach**

This study employs a semi-structured qualitative interview and a case study approach to investigate CCU mineralization stakeholders' technology risk and benefit perceptions, acceptance, and views on adoption. Six different stakeholder groups were included: technology developers, service providers (designers, engineers, consultants), companies in the cement and concrete sector, building and construction contractors, as well as related industry associations and governmental organizations (policy development, funding). The study received human ethics research approval at the University of Alberta (Pro00100870).

Given the lack of qualified data on socio-economic factors in CCU development (Zimmermann & Schomacker, 2017) we opt for a qualitative research approach over quantitative, to investigate the experiences and perspectives of stakeholder groups in the CCU mineralization development process in more detail (Jones, et al., 2017a). Capturing unique stakeholder responses in this manner can only be carried out through conversations rather than specific surveys, which would result in a small number of fragmented responses, creating challenges in comparing and drawing meaningful conclusions and implications. Moreover, with the increased flexibility of interviews, underlying drivers and motivations of viewpoints on CCU can be explored in the context of Rogers' (1995) diffusion of innovations theory. The interview questions were generated from literature findings (Section 2). Drawing on the benefits of mineralization technologies in GHG reduction (IPCC, 2005), as well as risks and challenges of technology development with respect to technical, economic and policy aspects (ICEF, 2016) (NAP, 2019). Specifically, captured risks include health and safety (Rafiaani, et al., 2020) (Heek, et al., 2017b), perceived environmental sustainability (Arning, et al., 2018) (Arning, et al., 2020) and low knowledge of CCU products (Arning, et al., 2017) (Heek, et al., 2017b) (Arning, et al., 2018) (Arning, et al., 2019). The interview questions cover interviewee position and background, organizational position along the CCU mineralization supply chain and knowledge of expertise in CCU. The core portion of the interview was dedicated to exploring participants' perceptions of the benefits, risks, and barriers with regards to the safety, economics, design and facility challenges, market penetration/access, and regulatory/policy hurdles of CCU mineralization technology development in their market environment (Alberta and Canada). Probing and follow up questions were used to clarify and facilitate results interpretation (Roulston & Choi, 2018). The detailed interview guide is provided in Appendix B.

### ***5.3.1 Interview Process***

Relevant organizations were identified from membership databases provided by industry bodies and specific internet searches and assigned to the stakeholder groups accordingly. Potential participants in each stakeholder group were then selected randomly under the condition the organization was associated with the mineral carbonation market and has an office and/or operates in the province of Alberta. Study invitations were made via emailed letters and follow-up phone calls after two weeks from the date of initial contact. Interviews were conducted from August/2020 to December/2020, over Zoom (Zoom Video Communications, Inc, 2021). Respondents did not receive any remuneration or other form of incentive for participation in the study.

Prior to commencing the interview, participants were informed about the interview process. Consent was obtained from interviewees for audio recording. The maximum time allotted per interview was set at 45 minutes.

Before entering into the structured interview, participants were asked four pre-interview questions to gauge their knowledge of and experience with CCU and mineral carbonation in specific areas of the discipline (Appendix B). Private companies operating in the CCU field are thought to have high operating and scientific knowledge, which has been shown to help form favourable attitudes towards a technology adoption, a critical step to the persuasion stage of Rogers' innovation-decision process and a subsequent adoption decision (Jones, et al., 2017b); (Rogers, 1995). The pre-interview stage in this study also functioned as self-selection criterion for potential participants of whom some chose not to participate in the study due to their inadequate knowledge and experience of mineralization. Moreover, in the cases of study

participants holding junior positions within their respective organization, interviews were conducted to elicit senior management views and knowledge of mineralization technology.

After completion of the pre-interview, respondents answered 5 open-ended interview questions focused on interviewee and organizational background, work and expertise within the context of mineral carbonation, understanding of the process, perceived and experienced technology benefits, risks and challenges (Appendix B). To help facilitate the conversation the interviewer could make use of several probing questions for each of the five interview questions. Transcripts were then generated using the Zoom Captioning (Zoom Video Communications, Inc, 2021), the software's automatic transcription service. Interview transcripts were manually examined and corrected to rectify mistakes between the audio and written forms prior to data analysis.

### **5.3.2 Data Analysis**

To identify the critical factors at the adoption decision stage of mineralization technology as a viable CO<sub>2</sub> mitigation technology a total of 64 CCU technology stakeholders were invited to participate in this study, representing 49 individual organizations. A total of 9 interviews across each of the six stakeholder groups with between one (CCU technology developers, service providers, building and construction contractors) to two participants (governmental organizations, cement and concrete companies, industry associations) were conducted. The overall response rate was 18%. The average interview time was 25 minutes, with the longest interview lasting 39 minutes. None of the interviews exceeded the allotted 45-minute time limit.

In comparison to previous qualitative research studies in the CCU field (Muslemani, et al., 2020); (Heek, et al., 2017a); (Kant, 2017) with interview samples of between six and twelve participants, this study yields a small yet information rich expert sample that covers the entire

CCU supply chain deemed critical to the better understanding of how the uncertainty and risks of mineralization's early-stage technology readiness level may inhibit its future market penetration (Hepburn, et al., 2019).

Our analytical approach follows Arning et al (2020) Heek et al. (2017a), and Jones et al. (2017a) in developing inductive categories based on manual coding of interview data to generate issue categories based on their importance as expressed by CCU industry stakeholders. Following automated and manual transcription, interview transcripts were manually tagged using keywords of interest, e.g., the economics of mineralization technologies as a factor in commercialization and adoption, a prevalent topic in the CCU literature (e.g., (Hepburn, et al., 2019)). Thus, unique keywords on the economics of mineralization were assigned to distinct codes. Keywords were then tracked across transcripts of different stakeholders allowing similar codes to be binned into distinct issue clusters. For instance, incumbency barriers, regulatory acceptance, technical performance, safety, and integrity testing were all binned into the "mineral carbonation risks and challenges" cluster. Cement replacement, concrete addition, and supplementary cementitious materials were binned into the cluster "relevant area of work". This procedure resulted in the creation of five main categories and themes based on different codes: (1) interviewee and company background, (2) relevant area of work, (3) areas of opportunities/improvement, (4) benefits of mineral carbonation, and (5) mineral carbonation risks and challenges. Within these categories five themes were identified and analyzed with regards to their influence on stakeholder views on adoption or rejection of CCU mineralization technology at Roger's persuasion stage of the innovation decision process: (1) carbon emissions, (2) technology development, (3) competition/collaboration, (4) policy/lobbying, and (5) risk & uncertainty.

## 5.4 Results and Discussion

A total of 55 codes were extracted during the iterative transcription process ranging from interviewee's industry experience to their company's role in CCU advocacy and policy. Stakeholder information and group classifications were verified with participants. This ensured that the rationale for selection was purposive sampling. Participants expressed diverse views about technology concerns (e.g., cementing efficiencies), social license to operate, and risk of technology transparency within the Canadian mineralization context. For an exhaustive list of all codes organized by categories see Appendix B (Table B1).

We present the results as average occurrences for each code by participant in each respective stakeholder group. We then discuss each stakeholder groups in order of its proximity to the technology and degree of influence on commercial adoption from nearest to farthest: CCU technology developers, followed by governmental organizations recognizing their role in supporting early-stage commercialization of CCU technology development. Cement and concrete companies, industry associations and finally service providers, and building and construction contractors' who work at greater distance from the technology design locus are discussed. We present the frequency of each code (>1) as the average occurrence for all stakeholder groups (Appendix B - Figures B1-B7) and focus our discussion on the most prevalent (top 5) codes for each stakeholder group with respect to categories and themes that provide insights on factors that influence CCU mineralization technology adoption or rejection in the Alberta, Canadian context

Organizational categories were developed based on areas we wished to investigate, followed by the themes to identify critical factors at the juncture of Roger's (1995) knowledge and persuasion stages for technology uptake decisions. Five organizational categories were

developed from the coded data based on results in line with the interview guide: (1) professional and company background, including roles and responsibilities; (2) relevant area of work, including stakeholder position along the supply chain; (3) areas of opportunities or improvement, including market penetration potential and areas of technological improvement; (4) benefits of mineral carbonation, including environmental, financial and social dimensions; and (5) mineral carbonation risks and challenges, including product safety, marketability, financial, regulatory and policy aspects.

Figure 5.2 presents the sum frequency of individual codes across all stakeholders. The three most frequently occurring codes were carbon emission reduction (51), market penetration, retrofitting and scalability (48), and social license to operate (41). These results broadly reflect the significant potential of mineralization as a viable solution for climate mitigation for the cement and concrete industry, with the caveat of the need to generate scale if the technology is to accomplish market penetration (e.g., (Wang, et al., 2021); (Hepburn, et al., 2019)). Also, among the top 10 codes identified were providing technology development support (33) through funding, and challenges with policy structures (31); topics well represented in the CCU literature that underscore the need for (financial) support to overcome the barriers of rarely TRL-stage CCU development via targeted policy supports for mineralization (e.g., (Muslemani, et al., 2020); (Kant, 2017)).

Furthermore, five themes were identified of critical factors at the juncture of Roger's (1995) knowledge and persuasion stages as determinants of technology uptake decisions: (1) carbon emissions, including environmental benefits of using mineralization to reduce CO<sub>2</sub>; (2) technology development, including technology performance and its related financial, policy and



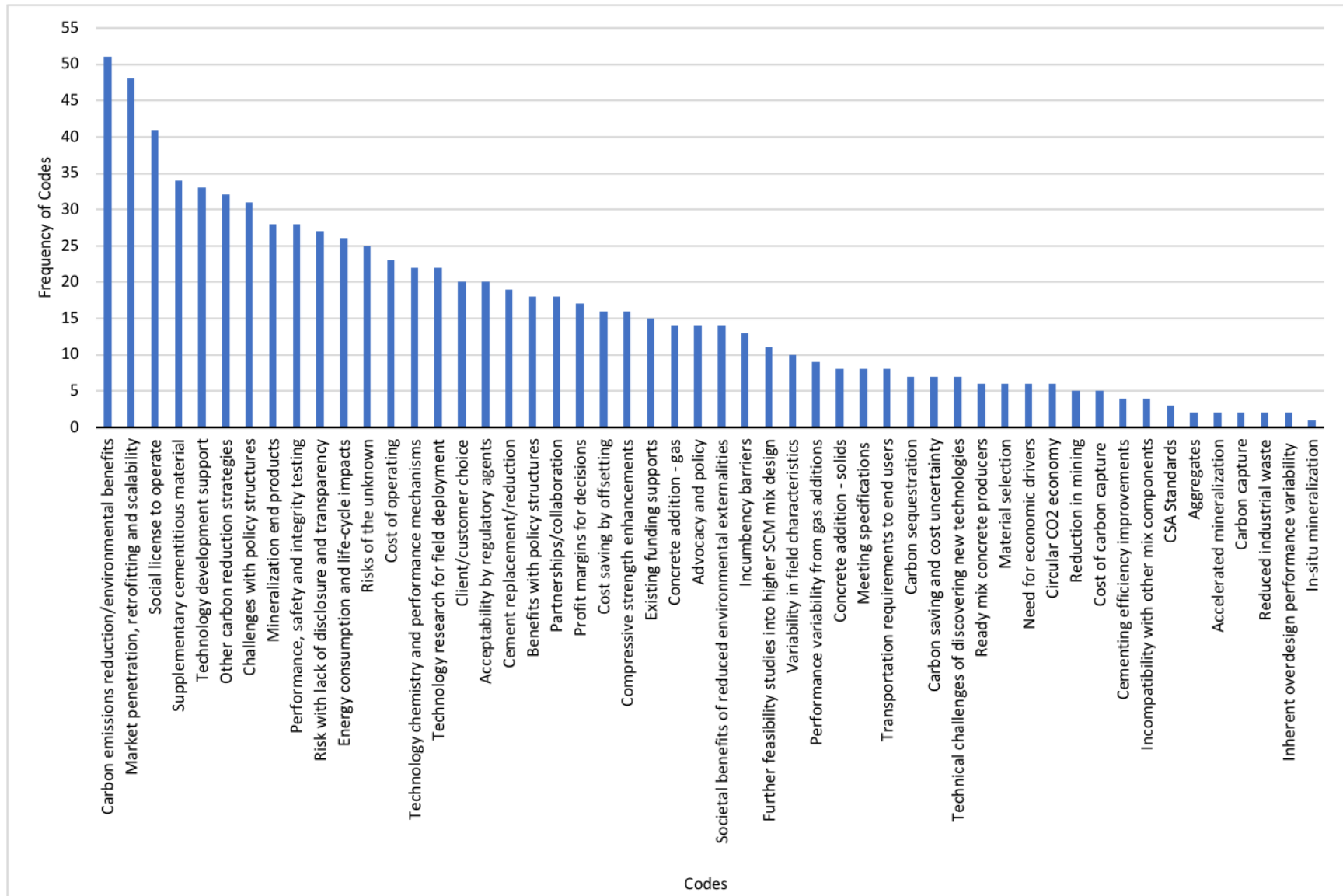


Figure 5.2 Total occurrence-sum of all individual codes represented for all stakeholder groups.

market barriers; (3) competition and collaboration, referring to the challenging process of partnerships within the mineralization supply chain; (4) policy, including challenges resulting from existing policies and the benefits of enhanced policy supports through tax credits and/or subsidies; and (5) risk and uncertainty, including issues of technology performance and transparency between mineral carbonation stakeholders and their clients.

Previous climate strategies for the cement industry have geared on reducing carbon dioxide emissions by four levers: improvements in energy efficiency, alternative fuel use, reducing clinker in cement, and technologically innovative technologies, such as CCUS (IEA, 2018). Figure 5.2 reveals that known risks and challenges of mineralization rank among the top 10 codes, namely challenges with performance, safety and integrity testing (28), and risks associated with the lack of disclosure and transparency among CCU stakeholder groups (27). This finding reiterates the role perceived risks and uncertainty play in stakeholders' adoption decision process ( (Zimmermann, et al., 2020); (Esposito, et al., 2011)). It also highlights the important role enhanced and collaborative technology development research plays in increasing market access and the transfer of technology knowledge in the development process (Audretsch, et al., 2014); (Polenske, 2004); (Harding, 2002). In line with Rogers' (1995) diffusion model, enhancing knowledge transfer can move stakeholders from the knowledge to the persuasion stage as collaboration contributes to adoption decisions through technology trialability and observability. Respondents also mentioned other carbon reduction strategies among in the context of mineralization (among the top 10 codes). Specific issues deemed important to CCU technology development in the broader literature, however, are barely mentioned (Figure 5.2). These include in-situ mineralization, carbon capture, or the production of aggregates. We ascribe this finding to fact that the majority of CCU stakeholders we interviewed identified as experts in

the ex-situ mineralization field that is primarily concerned with the direct addition or substitution of cement and/or concrete.

Rogers (1995) cites knowledge is a critical precursor to persuading agents to consider adopting technological innovations. Perceived technology risks and uncertainties due to existing knowledge gaps among stakeholders may act as barriers at the critical nexus of technology adoption and market acceptance (Arning, et al., 2020); (Muslemani, et al., 2020); (Kant, 2017). We therefore continue the discussion of results with a focus on stakeholder's self-reported knowledge scores. We then proceed with more in-depth discussion of the viewpoints of individual stakeholder groups in sequence of their proximity to mineralization technology development and the commercial adoption process.

#### ***5.4.1 Self-reported Knowledge and Experience Scores***

While the literature has documented low levels of public knowledge when it comes to CCU technologies (e.g (Arning, et al., 2017); (Arning, et al., 2018)) the involvement of CCU industry stakeholders in strategic technology use and investment decisions would suggest high knowledge scores across the board (Jones, et al., 2017b). Figure 5.3 presents participant's self-reported knowledge, understanding, and experience as it pertains to the technical, economic, policy and environmental dimension of mineralization. Our results indicate that the assumption of high knowledge does not hold across stakeholders. In fact, construction industry associations and building/construction contractors as those not directly involved in the technology supply chain show a prevalence in low knowledge scores.

Overall, Figure 5.3 shows that most participants had moderate overall knowledge of mineralization with scores of five to seven out of 10. Service providers, such as engineers and consultants stand out for their overall high knowledge of the economic, policy, and

	CCU Technology Developers	Government	Cement/Concrete Companies	Industry Associations	Service Providers	Building and Construction Contractors
Knowledge	■	■	■	□	■	□
Technical	□	■	■	□	■	□
Economic	■	■	■	□	■	■
Policy	■	■	■	■	■	■
Environmental	■	■	■	■	■	■
Applicability	Y	Y	Y	Y	Y	Y

High (>8)    
 Moderate (5-7)    
 Low (4-6)    
 Very Low (0-3)

Figure 5.3 Participant’s self-reported mineralization knowledge and experience scores. Scores are represented as averages and on a scale of 0-10, with 10 representing a high degree of knowledge/experience. ‘Y’ represents yes, for binary responses.

environmental properties of mineralization, and to lesser extent technical expertise. Cement and concrete companies, in comparison, many of whom conduct their own research of mineralization technology, stand out for their superior (>8) scores on the technical, economic, and environmental properties of mineralization. The Alberta CCU industry shows high overall awareness of mineralization as a solution to mitigate the province’s high emissions profile.

With the exception of construction industry associations and contractors, the absence of low knowledge scores indicates that members in the Alberta CCU industry possess moderate to high level of functional principles and the associated economic, policy and environmental circumstances of innovative mineralization processes. This evidence places the majority of

stakeholders at the persuasion stage of Rogers (1995) innovation diffusion model, decision makers are driven by targets, motives and technology outcomes (Jones, et al., 2017b). This argument is underscored by many stakeholders citing the importance of mineralization technologies in meeting targets for carbon emissions reduction and their role in maintaining social licences to operate. However, issues of financial barriers and performance testing also rank high (Figure 5.2) and need to be along the technology development process in order for stakeholders to be persuaded into adopting mineralization technologies.

#### ***5.4.2 CCU Technology Developers***

CCU technology developers, as the group working most directly on mineralization technology most frequently mentioned knowledge of the mineralization field during the interviews (Figure 5.4) as discussions centered on end products (17) and supplementary cementitious materials (15). Technology developers are searching for areas of opportunities in the technology and ways to increase the marketability of mineralization and other end-products. The creation of such low-carbon (green) product markets to pass technology costs along the supply chain is a supported approach for CCU technologies (Muslemani, et al., 2020); (Arning, et al., 2018). Developers frequently discussed areas of technology development, including technology chemistry and performance mechanisms (15), market penetration, retrofitting and scalability (14), and technology research for field deployment (11). These factors were considered important to the technology's experimental success or "trialability" and critical to adoption or rejection decisions along the commercialization decision process (Figure 5.1). Although, developers did identify several benefits of mineral carbonation, energy consumption and life-cycle impacts (10), and carbon emissions reduction (9), perceived risks and challenges arising from a lack of disclosure and transparency (7) and performance, safety and integrity

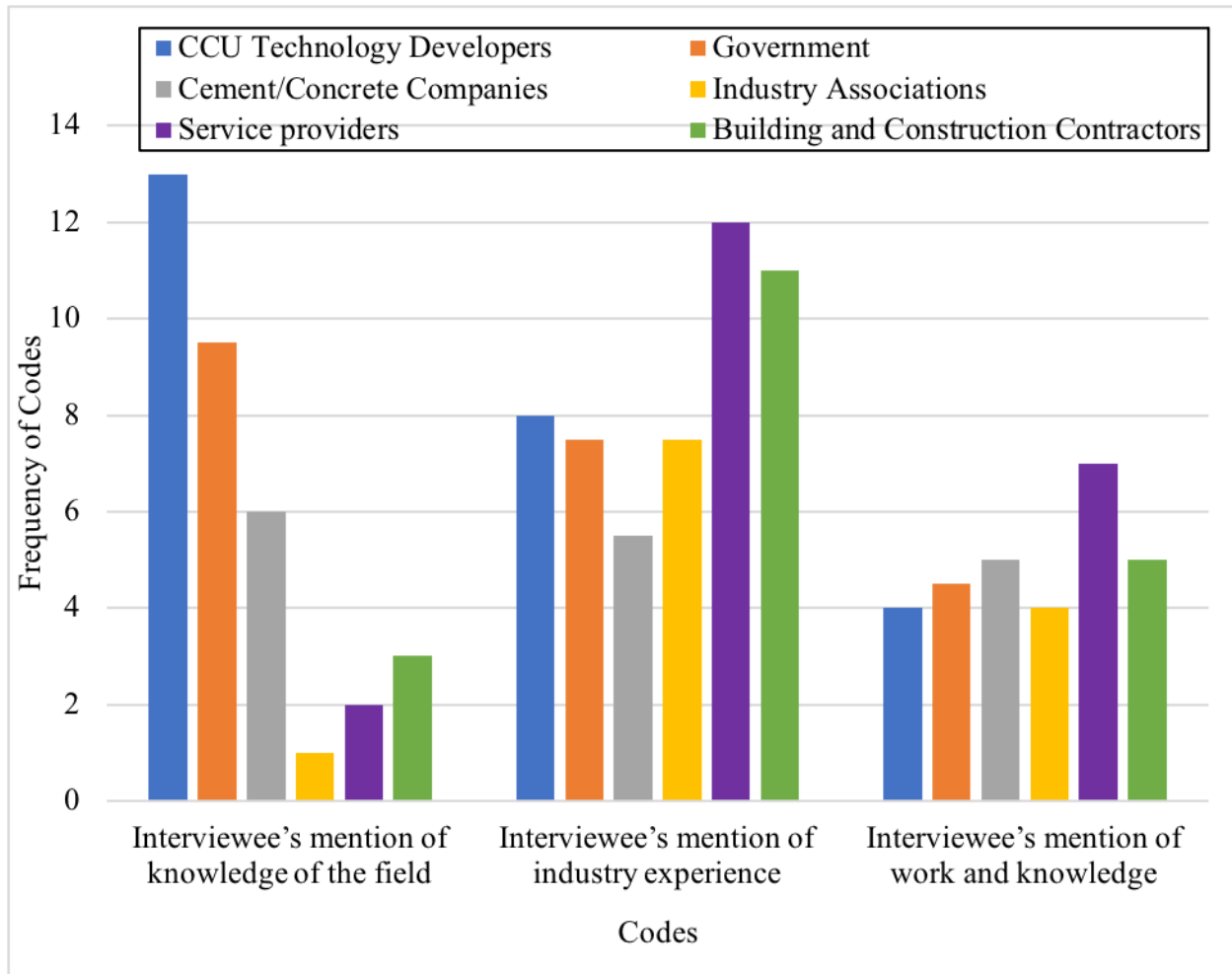


Figure 5.4 Frequency of individual and company background coded results presented for each stakeholder group. Occurrence scores are shown as averages of respondent's responses.

testing (6) were mentioned repeatedly (Figure B2). These results demonstrate that perceived risks resulting from uncertainty and a lack of institutions governing disclosure and transparency among players within the mineralization industry act as a barrier that may impede its further commercialization. An immediate possible solution to the concerns is the creation of organizational boundaries (Hoffmann, et al., 2018) that facilitate collaboration and contribute to knowledge transfer as vectors for risk mitigation.

### **5.4.3 Government Stakeholders**

Like technology developers government stakeholders were identified to possess high levels of observed technology knowledge (Figure 5.4). However, in contrast the top codes discussed by this group were led by technology development support (13), areas of opportunities/improvement as in other carbon reduction strategies (7.5), risks and challenges from performance, safety and integrity testing (6.5), and incumbency barriers (5). Benefits of mineral carbonation from carbon emissions reduction/environmental benefits (4), partnerships/collaboration (4), and energy consumption and life-cycle impacts (4) played minor roles (Figure 5.5).

A high level of technology knowledge and focus on providing technology development support, uniquely positions government agencies involved in CCU (Kant, 2017); (Mikunda, et al., 2014).

While government stakeholders acknowledge the environmental benefits of mineralization technologies in reducing GHGs, our interviews reveal that (overcoming) barriers to commercialization such as testing and integrity aspects of mineralization, and the market position of incumbents matter greatly at the persuasion stage of technology diffusion. The literature on CCU acknowledges the need for financial incentives to overcome challenges of high investment costs, for instance through sector collaboration (Tönjes, et al., 2020); (Kant, 2017). Albeit organized industry collaboration in the CCUS field is limited to date (Muslemani, et al., 2020), government stakeholders argued that that this can be done through for example, infrastructure sharing and facilitated knowledge transfer initiatives.

Besides mineralization governmental stakeholders also mentioned other carbon reduction strategies as to contributing to competition within the CCU field, and collaboration is generally viewed as a means to avoid such competition. While the issues raised by CCU technology

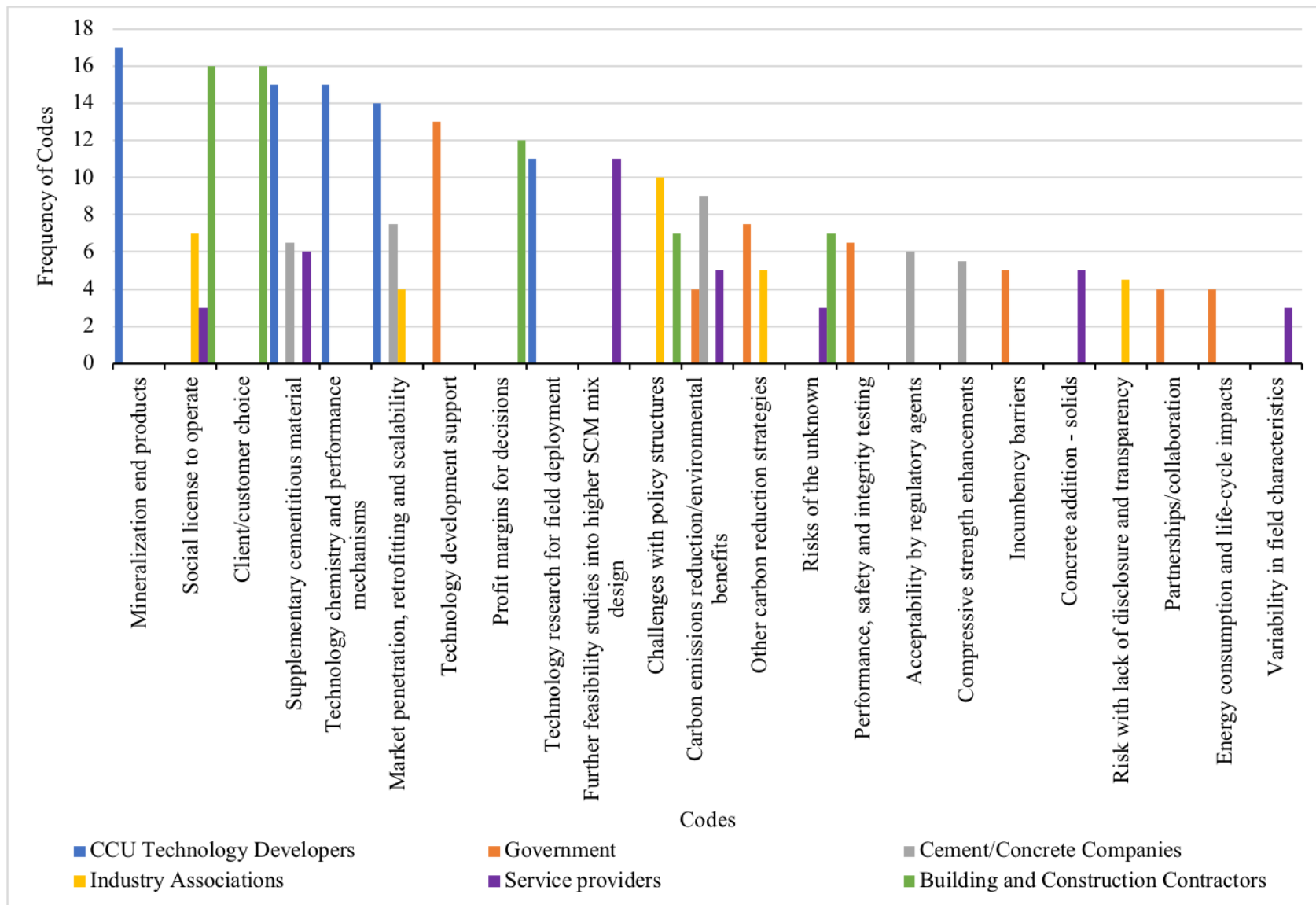


Figure 5.5 Top 5 most frequent codes represented for all stakeholders. Average occurrence scores were used for respondent's responses within each stakeholder group.



developers were located further along the TRL continuum (e.g., penetration and commercialization) government stakeholders' focus on technology development and support collaboration can be viewed as positive to advancing mineralization in the Albertan (and Canadian) context at the critical juncture.

#### ***5.4.4 Cement and Concrete Companies***

The highly technical and direct involvement with mineralization technologies stands behind cement and concrete companies' high self-reported (Figure 5.3) and observed knowledge scores (Figure 5.4). Interviewees highlight the benefits of mineralization during interviews: carbon emissions reduction/environmental benefits (9), compressive strength enhancements as an opportunity for the technology, and market penetration, retrofitting, and scalability (7.5) as aspects that matter to market access and capital investment for mineralization. Companies also discussed their focus on supplementary cementitious material (6.5) but highlight major risks and challenges in the acceptability of mineralization by regulatory agents (6), and compressive strength enhancements (5.5) (Figure 5.5). As expected, companies in the cement and concrete sector highlight issues in technology development, operating cost and market penetration. While cost uncertainties of mineralization have previously been identified as barriers (Hepburn, et al., 2019); (Sanna, et al., 2014), cost reduction strategies on both material input and output market side have been hampered by reliable data regarding their capital and operating expenditure effects (Sanna, et al., 2012).

Beyond financial investment and economic uncertainties, interviewees point to a number of concrete issues in the reliability of applying mineralization under real world conditions. For instance, applications issues with uncertainty in mix designs can lead to variability in field characteristics of concrete products from gaseous CO<sub>2</sub> additions, such as slump, freeze-thaw

cycles, viscosity, air entrainment and temperature differences; issues that raise concerns over meeting critical sector standards. Currently, the Canadian Standards Association's standard for "Concrete materials and method of concrete construction/test methods and standard practices for concrete" (CSA, A23.1:19/CSA A23.2:19, Annex S), lays out the regulatory, performance and safety specifications for CO<sub>2</sub> addition into cement (CSA, 2019). Moreover, the Cement Association of Canada has also registered an Environmental Product Declaration (EPD) on Portland-Limestone cements with the CSA (2016). Credits associated with the reduction of embodied carbon in materials using Portland limestone cement and other supplementary cementitious materials are also discussed in the Leadership in Energy and Environmental Design (LEED) (BC MECCS, 2017). Ensuring mineralization meet Canadian testing standards compared to incumbent technologies has the potential to create a (more) level the playing field for the industry and this address existing concerns over incumbency barriers also mentioned by the government stakeholder group. By addressing risk and uncertainty through increased technology trialability and modelling of real-world capital and operating expenditure effects of mineralization technology in the cement and concrete sectors should benefit future adoption decisions as perceived risks and uncertainties are mitigated.

#### ***5.4.5 Industry Associations***

Interviews with industry associations highlighted their role in communicating the various challenges of mineralization technology between private industry and existing policy structures (10) (Figure 5.5). While industry bodies invest in technology development alongside of advocacy and policy support, their main cause is the call for greater policy supports, especially from the Canadian Federal government. Increased policy supports through such vehicles as tax credits, subsidies, and infrastructure supports for decarbonation are well documented in the CCU

literature (Bobeck, et al., 2019); (Meckling, et al., 2017). We note that while industry associations point to importance of policy supports, their governmental counterparts point to the need for increased performance, safety and integrity testing, reflective of the role of industry in advocacy and government stakeholders in providing supports. Beyond their role as industry lobby, CCU association also recognize the risks and uncertainties from a lack of disclosure and transparency for the sector (4.5). In contrast to other CCU stakeholders, this expresses a concrete solution by advocating for the negotiation of contractual and non-disclosure agreements that would allow collaborating CCU parties to pre-determine the term and boundaries of cooperation (Hoffmann, et al., 2018). CCU industry associations also recognize the importance of social license to operate (7), the need for market penetration, retrofitting and scalability (4) to achieve technological implementation. Competition for funding and development for other carbon reduction strategies (5) are perceived as additional factors impeding the adoption of CCU mineralization at the critical stage of industry adoption decisions (Figure 5.5). In comparison to other groups industry associations group stand out for their lowest observed knowledge score, but level of experience and average working knowledge as foundations of their advocacy role for this innovative technology (Figure 5.4).

#### ***5.4.6 Service Providers***

CCU industry service providers interviewed in this study worked mainly in the supplementary cementitious materials (6), concrete addition, and solids (5) fields. This group stands out for its high industry experience score (Figure 5.4) due to their direct involvement with CCU technologies as designers, engineers, and consultants; a result that mirrors their self-reported knowledge scores (Figure 5.3).

Service providers acknowledged the emissions reduction benefits (5) of CCU technologies and their role in social license to operate (3). However, interviewees identified unknown risks of high percent CO<sub>2</sub> mix designs (3) due to a current lack of (applied) industry data from mix design studies as a significant risk for mineralization technology. They suggest that further feasibility studies into higher supplementary cementitious material (SCM) mix design (11) are needed to help alleviate these risks to their sector. Studies by Juenger and Siddique (2015) and Khokhar et al. (2010) indicate that high mix design studies tend to not conform to prevailing industry standards that limit the use of SCMs. Consequently, one key recommendation made service providers is to update industry standards to allow for flexible uses of mineralization implementation (Bobeck, et al., 2019). In line with other stakeholder groups, facilitating trialability is viewed to mitigate perceived risks and uncertainties, thereby enabling mineralization to overcome current barriers at a critical juncture of technology adoption.

#### ***5.4.7 Building and Construction Contractors***

Building and construction contractors are near the end of the mineralization supply chain, and the stakeholders working most closely with clients. While they are not necessarily directly involved in technology adoption, their role is to persuade clients of the benefits of CCU mineralization. Accordingly, contractors expressed most concern about client/customer choice (16). The ability to advise and influence client choice decisions on new innovations was a top priority, making contractors influential vehicles of change known as change makers in the CCU sector. In light of the technological shift present by mineralization, a primary concern for contractors was the ability to retain profit margins for decisions (12) made by clients on whether to utilize CCU product in their projects. They also cited challenges with policy structures (7) as standing in the way of mineralization adoption and indirectly influencing client choices. CCU

perceptions in the general public are perceived to be hindered by product use risks, and related fears of unknown performance issues. This includes such fears as the risk of CO<sub>2</sub> leakage (Arning, et al., 2017); (Arning, et al., 2019). Contractors also share the view that a lack of trialability and resulting industry know-how contributes to their and their clients' fear of the unknown (7), in other word uncertainty. A result reflected in contractors' high industry experience, yet low knowledge score (Figure 5.4). Contractors recognized the important benefits of mineralization for the environment and its role in supporting the industry's social license to operate (16) (Figure 5.5). They also expressed the believe that current perceptions of risk and uncertainty associated with mineralization would ease as knowledge changes perceptions. This view is indicative of Rogers (1995) innovation decision process, whereby risk decreases as innovations pass through the critical stage of adoption.

## **5.5 Conclusion and Policy Implications**

The Government of Alberta and others provide funding supports for CCUS technologies which take benchmark technologies to commercialization (Government of Alberta, 2021). This includes the Government of Alberta's support of two major CCUS projects- the Quest and Alberta Carbon Trunk Line (ACTL, 2020). Alberta Innovates' funding of CCUS projects of TRL 3-7, the NRG COSIA Carbon XPRIZE competition for \$20 Million and Emissions Reduction Alberta's (ERA) Grand Challenge for \$35 Million accelerate projects by funding demonstratable and commercial scale conversion of CO<sub>2</sub> into valuable end products (XPRIZE Foundation, 2021) (ERA, 2020) (Alberta Innovates, 2021). Although these support frameworks exist for all stakeholders in Alberta, there are many hurdles and stumbling blocks for stakeholders that prevent widespread technology adoption. This study shows critical factors beyond financial and market barriers. Competition within the mineralization supply chain, as well as with incumbents

is identified as a barrier by this research. Challenges with current policies such as lack of direct tax credits and subsidies impede technology development. Risk and uncertainty of technology performance metrics as well as friction in the transparency of results is another stumbling block for these technologies. Mitigation of these factors through sound technological and policy supports will benefit future commercial adoption and realization of the environmental and social benefits of mineralization technologies.

Few studies have used Rogers diffusion of innovation model to evaluate factors critical to adoption decisions (Jones, et al., 2017b); (Sharp & Miller, 2016). In this study, qualitative design methods using expert elicitation for the mineralization field was carried out. Semi-structured interviews were conducted with 6 stakeholder groups: CCU technology developers, governmental organizations, cement and concrete companies, industry associations, service providers, and building and construction contractors. The innovation decision process from the diffusion of innovations theory is applied for the factors that affect market acceptance of CCU technologies for stakeholders. We have identified five critical factors as enabling the adoption and eventual implementation of innovative mineralization technologies. The success or failure at this critical juncture depends on mineralization's perceived carbon reduction benefits, technology development, competition/collaboration, policy/lobbying, and risk and uncertainty. Many of the elements of the diffusion of innovations are lacking. Literature indicates the public tends to have limited awareness and knowledge of CCU (Arning, et al., 2019); (Heek, et al., 2017a). Likewise, the knowledge is low in some stakeholders and perceived characteristics of the innovations are not completely known, thus, communication channels to share information between sources need development (Figure 5.1). Mineralization, a relatively new innovation researched since the late 1990s has high degrees of uncertainty due to variable, early-stage TRLs.

(Sipilä, et al., 2008) (Hepburn, et al., 2019). This study identified that the stakeholders share a sense of risk & uncertainty with technology development and performance and testing.

Additionally, there's a lack of social systems or units unifying the technology due to elements of competition and stakeholders' requests for more policy support. Given these findings, the results presented in this study support mineralization to be in the knowledge and persuasion stages of the innovation decision process (Figure 5.1).

Most mineralization industries are in the demonstration stage (Hepburn, et al., 2019) (Jones, et al., 2017b) (Zimmermann & Schomacker, 2017). Improvements in technology development by cementing efficiency, and market penetration, retrofitting and scalability (48) discussed by cement and concrete companies are required for commercialization (Figure 5.2). Mineralization will benefit from enhanced performance, safety and integrity testing (28) to address acceptability by regulatory agents (20) and CSA standards and overcome incumbency barriers, as discussed in literature (Bobeck, et al., 2019). Having clear standards will increase policy makers' abilities to choose technologies based on clearly defined aspects of performance in a transparent manner. Literature shows lack of government support for CCU (Muslemeni, et al., 2020). This study found that challenges with policy structures is a top area of concern (31). Additional challenges include cost of operating (23) and profit margins for decisions (17). To mitigate these, technology development supports (33) through tax credits and subsidies are needed for uptake and eventual commercialization (Figure 5.2). Advocacy (14) by stakeholder groups will further increase these lobbying efforts for much needed policy support mechanisms (Meckling, et al., 2017). These policy implications which increase the level of knowledge, legislation, federal leadership and clarity in standards will create grounds for the commercial adoption of mineralization in Alberta.

Areas of technological conflict among partners as a result of withholding information can be reduced by increasing transparency, which will help decision makers and the public (Arning, et al., 2020); (Institute of Medicine, 2013). Many stakeholder groups (governmental organizations, industry associations, and building and construction contractors) view partnership/collaboration (18) as an opportunity to avoid competition. However, risk with lack of disclosure (27) is present across all stakeholder groups and signals an inherent risk with competition and collaboration. It shows competitive behaviour, a contributor to this is the fear of data leakage, as suggested by a previous source (Hoffmann, et al., 2018). Creating boundaries to address this is suggested by this source and is supported by the findings from the industry associations that mentioned usage of non-disclosure agreements. Policy implications to expand the role of the regulators to platforms for non-competitive operations will further increase collaborative efforts. Literature discusses long-term uncertainty as a major known barrier to the commercial adoption of CCU technology pathways (Al-Mamoori, et al., 2017). This research shows risks and uncertainty of the unknown (25)- which refers to potential risks to industry, specifiers and the public. These risks, if not mitigated, can negatively impact the industry's brand. Overall, as mentioned by the stakeholders, policies and policy makers must fairly and transparently select or reject technologies in order to reduce technological conflict.

Future work should deepen this research by increasing more stakeholders from different CCU pathways. Public and consumer perception of mineralization technologies for future adoption decisions is an important gap in research which should be addressed. Increasing targeted policy supports for mineralization and CCU technologies and increasing technology development research activities through enhanced testing will minimize performance risks. Enhancing collaboration across all stakeholder groups and individual companies/partners for



knowledge transfer will address the current challenges that these technologies face regarding the need for more transparency. These factors will lead to future diffusion and commercialization of mineralization technologies.

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## **Chapter Six: Overall Conclusions and Recommendations for Future Work**

### **6.1 Conclusions**

The implications of using mineral carbonation technologies and its potential was assessed in this research. This was done by carrying out three different investigations. First, the evolution of research in CCU and CCS was assessed with bibliometric analysis, topical research areas and industry data. Then, global warming GHG emissions assessment was carried out for mineralization technologies and compared to incumbents and other CCU technologies using a life cycle assessment approach. Third, industry and stakeholder expert elicitation were conducted using qualitative assessment to identify mineralization benefits and challenges.

The research problem that this work addresses is to show the differences between CCS and CCU, and to characterize GHG emissions of mineralization along with their market barriers and opportunities. One of the main long-term objectives of this, as mentioned in Chapter 1, was to compare mineralization technologies, in order to develop fair and consistent methodologies to assess them. This research was carried out using specific objectives, each of which contribute their own conclusions and recommendations. Their major conclusions will be discussed here.

The first objective was performing bibliometric analysis to compare the evolution of and parallels between carbon capture storage and carbon capture utilization. In this research CCU was discussed separately from CCS to compare and make recommendations. Few papers exist in this field and they do not compare CCS independently from CCU using bibliometric assessments (Tapia, et al., 2018) (de Cruz, et al., 2021) (Nawaz, et al., 2022). Doing so identified several clusters. This process reveals that the CCS physical sciences, represented by the CO<sub>2</sub> injection and storage (yellow), CO<sub>2</sub> chemistry (blue) and CO<sub>2</sub> capture and power requirements (green) clusters, are not highly interconnected by lines to the social sciences, represented by the policy

and perception (red) cluster. Additionally, the highest citations for the co-citation network of journals for CCS and CCU were seen for the energy and fuels cluster.

CCU records show considerably lower publications than CCS records. Later topics from 2013-2023 in the CCU map show more mature landscapes focussed on CO<sub>2</sub> capture, carbon storage and carbon conversion than earlier topics from 1998-2012, which focused on carbon utilization and capture together. Comparatively, more mature clusters were observed in both CCS maps, from 1998-2012 and 2013-2023.

Results from industrial projects show the highest completed and in-development CCS projects in USA, the United Kingdom and Canada. While the most numerous CCU start-up companies are in USA, Canada, Germany and the United Kingdom.

Additionally, 4 topical areas were examined for a meta-review showing that the policy research area is the least contributing field to CCS and CCU, compared to the other areas of technology, economics and environment. The policy research area discusses the need for more CCS and CCU policy supports, including direct incentives, cap and trade, carbon tax and subsidies. The economic area shows both to have high capital costs. The technology area shows CCU technologies facing technological issues with efficiency, catalysts, reaction rates and energy consumption. Finally, the environmental area discusses CCS risks of storage and leakage, and CCU having higher non-global warming potential impacts.

The second objective of this research was to determine process life cycle GHG emissions for mineral carbonation and compare them with other CCU pathways and incumbents. This research focusses on developing a consistent and fair life cycle methodology and nomenclature to compare across CCU pathways, which is currently lacking in other literature: (Cuellar-Franca & Azapagic, 2015) (ENEA & COSIA, 2015) (ICEF, 2016). The categorization scheme is based



on the first conversion step and identifies 8 distinct CCU categories: CO<sub>2</sub> reduction by a hydrocarbon, CO<sub>2</sub> reduction by hydrogen, CO<sub>2</sub> reduction by other reagents, CO<sub>2</sub> reduction involving electricity, CO<sub>2</sub> reduction involving light, CO<sub>2</sub> mineralization, CO<sub>2</sub> bioconversion and other CO<sub>2</sub> conversions. These were subsequently broken down into subcategories. A total of 5 subcategories were identified for mineralization: direct mineral carbonation with wollastonite, direct mineral carbonation with serpentine, indirect mineral carbonation with serpentine, direct mineral carbonation with olivine, and waste mineral carbonation. Their products are calcite, magnesite and carbonated waste materials. Four metrics of assessment were developed in this research: kgCO<sub>2</sub>equivalents per kgCO<sub>2</sub>converted, kgCO<sub>2</sub>equivalents per kg or megajoule (MJ) of product, CO<sub>2</sub> avoided emissions and global emissions reduction potential. The last two metrics are based on market sizes and incumbent emissions. This methodology also employed consistent boundaries, emission factors and conversions, which is an approach that is previously not addressed in literature when examining across different CCU pathways.

It was determined that the net emissions from the mineralization pathways were all net negative and lower compared to the incumbent of cement production. That being between -0.68kgCO<sub>2</sub>eq/kgCO<sub>2</sub>Converted and -0.35 kgCO<sub>2</sub>eq/kgCO<sub>2</sub>Converted. The large cement market size (4.4 Gt/year) (Kurad, et al., 2017) compared to other pathways also meant considerable advantages for mineralization's global emission reduction potentials compared to the other pathways examined in this research. The source of electricity was the most sensitive parameter. The lowest contributing life cycle stage was the end-use stage, as the products were used as cement replacements for concrete applications. This work also led to an open-source tool (Nishikawa, et al., 2020), in which users from industry, government and academia can model

their unique CCU pathways to determine hot spots and compare across different pathways using the metrics identified previously.

The final objective of this thesis was to investigate the benefits, risks and opportunities for mineral carbonation technologies. Studies were conducted into perceptions of CCU products in general (Arning, et al., 2017) (Heek, et al., 2017) (Rafiaani, et al., 2020). But none of these studies focused on mineralization technologies, moreover none were found to focus on the building and construction industry. Thus, a qualitative assessment research design was developed using semi-structured qualitative interviews with 6 different stakeholder groups: CCU technology developers, governmental organizations, cement and concrete companies, industry associations, service providers, and building and construction contractors. Respondents were asked 4 pre-interview questions followed by 5 open ended interview questions.

The interviews were recorded and transcribed. Data analysis was performed using open coding. The codes were organized into organizational categories, followed by the development of themes. The 5 categories were: individual and company background, relevant area of work, areas of opportunities/improvement, benefits of mineral carbonation, and mineral carbonation risks and challenges.

The analysis of stakeholder perceptions of mineralization technology builds on Rogers' diffusion of innovations theory (Rogers, 1995), which describes the technology development process for adoption/rejection in 5 sequential stages: knowledge, persuasion, decision, implementation and confirmation. In this research, five themes were identified as critical factors at the juncture of Roger's (1995) knowledge and persuasion stages as determinants of technology uptake decisions. The 5 themes were: carbon emissions, technology development, competition/collaboration, policy/lobbying, and risk & uncertainty. Among the top 10 codes that

were identified for these technologies were: challenges with policy structures, performance and integrity testing, and risk with disclosure and transparency. The findings place the majority of stakeholders at the persuasion stage of Rogers (1995) innovation diffusion model.

Recommendations were made for specific policy supports for mineralization technologies and increased collaboration across all stakeholder groups for knowledge sharing and transparency.

The results from this research can be used for academics, policy makers, industry partners and government agencies. The implications and rationale for better policy structures from the stakeholders and research examined in this study could be used by government and policy groups to design specific policy programs for mineralization and CCU technologies. This can be done by incorporating a life cycle framework to evaluate technologies for funding support. The building and construction industry partners can also evaluate GHGs of mineralization technologies for their own processes by using the tools and methodologies mentioned in this research to model scenarios for their processes. They can also identify where their respective industries place in the larger CCU and CCS fields. Lastly, academics will further benefit from this research by the contributions of the results towards understanding of the field.

## **6.2 Future Work**

This work presents many opportunities and avenues for advancement of research. First, the bibliometrics assessment could be expanded by increasing the number of databases analyzed. This may increase data in terms of the number of nodes and edges. Additionally, the life cycle assessment approach mentioned in this study should be expanded to more mineralization technologies. Having further industry feedback for individual life cycle assessments of specific technologies will also add to literature. Additional metrics can also be developed for market size replacement representations. Techno-economic indicators can also be used for CCU specific

inventories to gain a holistic understanding of the technologies. Expansion of the life cycle approach to other non-global warming potential categories, such as, ozone depletion, acidification, eutrophication, terrestrial and aquatic toxicity, human health, resource depletion, land use and water use, using consistent boundaries, methodologies, functional units and conversions would also contribute to better understanding the holistic impacts of CCU technologies.

Another major future area of expansion includes increased industry stakeholder engagement to investigate the benefits, risks and opportunities of mineralization using qualitative research methods. This can be done by increasing the number of stakeholder groups and increasing the number of participants within each group. Moreover, this study can be expanded from an Alberta case study to a Canada wide investigation. This approach should also be followed for other CCU technologies. It will act as a bridge between academics and industry to evaluate the feasibility of these technologies further, as many of them have industry applications. Additionally, conducting community perception studies would investigate the uptake of mineralization technologies by the public and identify if there are any challenges. Lastly, country specific legal, regulatory and policy metrics for CCU technologies would assist in efficiently identifying hot spots and could serve as a means to push for targeted regional policies.

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## Appendices

### APPENDIX A: Supplementary Materials for Chapter 4

#### Selection of Representative Pathway

Figure A1 shows GHG emissions for all mineral carbonation processes observed on a per kilogram carbon dioxide converted, represented by subcategory, mineral input and product.

Three possible subcategories are depicted for the direct mineral carbonation pathway, this includes three resource mineral sources and two product combinations. For direct mineral carbonation, the serpentine mineral source is typically utilized due to its high resource availability compared to the rest of the minerals (Sanna, et al., 2014).

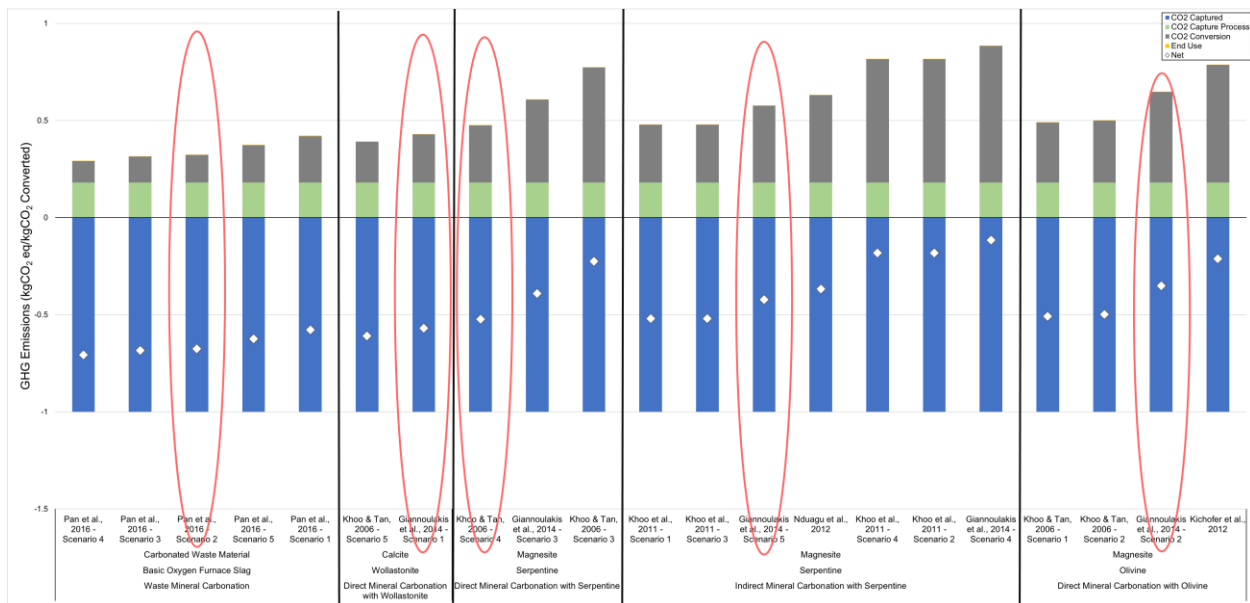


Figure A1 GHG emissions for all mineral carbonation literature cases shown on a per kilogram carbon dioxide converted, represented by subcategory, mineral input and product. Representative cases are circled in red.

Specifically, the representative case depicted for the direct mineral carbonation using serpentine producing magnesite is (Khoo & Tan, 2006, Scenario #4) (Khoo & Reginald, 2006).

This source was selected because data from this reference was obtained from other peer-

reviewed papers and from communication with scientists involved with mineral carbonation technologies and it operates with a high carbonation efficiency, which will optimize its potential for commercialization (Khoo & Tan, 2006, Scenario #4). Comparatively, the other scenario (Khoo & Tan, 2006, Scenario #3) has lower carbonation efficiency and higher energy consumption for activation, thus is less likely to have the highest potential for commercialization. The (Giannoulakis et al., 2014, Scenario #3) (Giannoulakis, et al., 2013) references the chosen representative case (Khoo & Tan, 2006, Scenario 3), thus it will be best to select the source of the data rather than a paper that references the source, which is available in this case.

The representative case depicted for the direct mineral carbonation using olivine is from (Giannoulakis et al., 2014, Scenario #2) (Giannoulakis, et al., 2013). This was selected because the (Kirchofer et al., 2012) (Kirchofer, et al., 2012) scenario uses a much lower and unrealistic weight percentage of mineral to rock. The (Khoo & Tan, 2006, Scenarios 1 and 2) (Khoo & Reginald, 2006) were not selected because they do not show a great deal of breakdown of the life cycle stages. The selected case shows a greater life cycle stage breakdown and has the typical percent weight of mineral on rock as referenced by other literature (Sanna, et al., 2014).

For the direct mineral carbonation using wollastonite subcategory, the chosen representative case was (Giannoulakis et al., 2014, Scenario #1) (Giannoulakis, et al., 2013). There is no huge discrepancy from other sources, the chosen case has more life cycle stages and hence variables which can be varied for the sensitivity/tornado plot.

For the indirect mineral carbonation using serpentine subcategory, the selected case was (Giannoulakis et al., 2014, Scenario #5) (Giannoulakis, et al., 2013), this case has optimal heat integration, compared to others such as (Giannoulakis et al., 2014, Scenario #4), which is typical

and necessary for this subcategory. The (Khoo et al., 2011, Scenarios 2 and 4) (Khoo, et al., 2011) are discussed as being worst case scenarios (as discussed in paper). The (Khoo et al., 2011, Scenarios 1 and 3) (Khoo, et al., 2011) are discussed as being best cases (as discussed in paper). The (Nduagu et al., 2012) scenario (Nduagu, et al., 2012) shows an extreme heat consumption process.

For the waste mineral carbonation using carbonated waste materials subcategory, the chosen scenario (Pan et al., 2016, Scenario 2) (Pan, et al., 2016) was under medium operating conditions with the highest carbonation efficiency. This was chosen over the others due to the fact that it presents medium carbon dioxide removal from the flue gas which is likely to not be as energy intensive as the other processes nor as costly, thereby representing the case that has the most potential to be commercial. This case and the other scenarios in this subcategory are based on industrial installations.

Figure A2 shows GHG emissions for chosen representative cases on a per kilogram carbon dioxide converted basis. The representative cases are shown by subcategory, mineral input and product.

The chosen incumbent process for mineral carbonation was selected as cement. All of the products discussed in this paper can serve as replacements for cement. They act to lower the volume of cement required, thereby reducing the GHG emission profiles for mineral carbonation-based cements. The incumbent emissions (Flower & Sanjayan, 2007) were supported as being within range when compared to two other literature sources (Smith & Durham, 2016) and (Josa, et al., 2004).

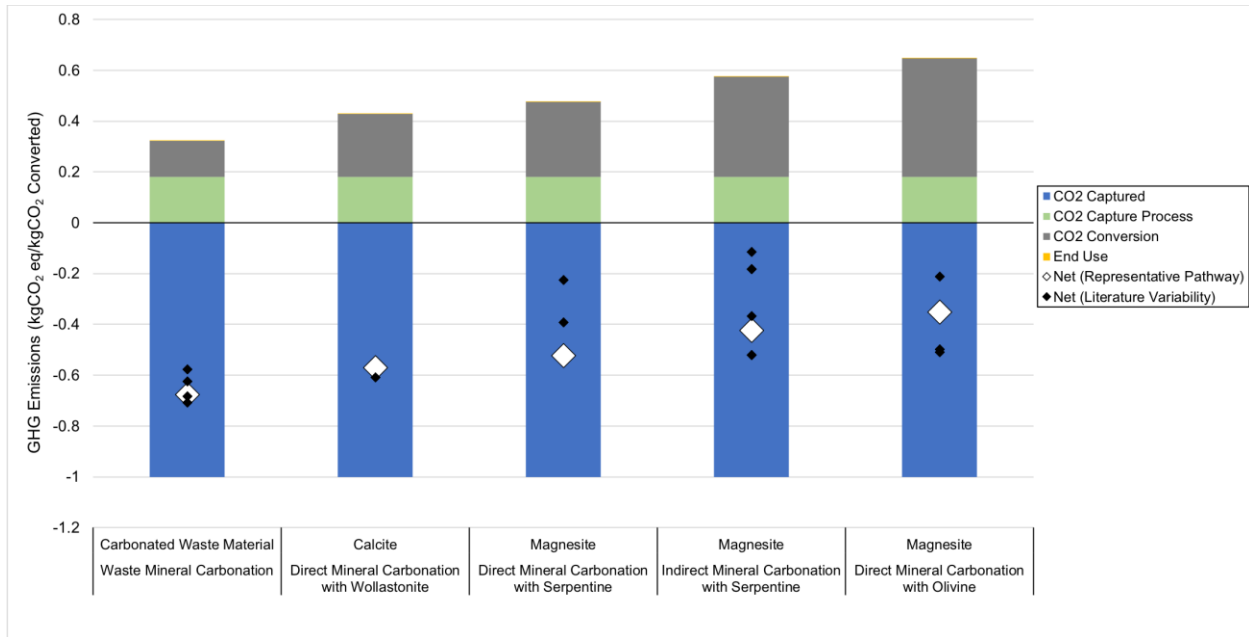


Figure A2 GHG emissions for chosen representative cases of mineralization carbonation shown on a per kilogram carbon dioxide converted basis. Cases are shown by subcategory, mineral input and product. Literature variability is indicated by the black diamond.

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## APPENDIX B: Supplementary Materials for Chapter 5

### PRE-INTERVIEW QUESTIONS

1. Please rate your knowledge with mineralization and carbon capture utilization?

1      2      3      4      5      6      7      8      9      10

2. Is mineralization applicable to your work or that of your organization?    YES    NO

3. Please rate your knowledge/experience of mineralization in the following categories?

Technical      1      2      3      4      5      6      7      8      9      10

Economic      1      2      3      4      5      6      7      8      9      10

Policy      1      2      3      4      5      6      7      8      9      10

Environmental    1      2      3      4      5      6      7      8      9      10

4. If you do not have knowledge/experience to speak about mineralization, can you direct me to someone who would know more and may be willing to provide an interview?

## INTERVIEW GUIDE

Below are the interview questions for all stakeholder groups

1. Background information
  - a) What is your position title and the length of time you have served it?
  - b) What are your roles and responsibilities as it pertains to mineral carbonation?
  - c) What decision-making capacity do you have as it pertains to mineral carbonation projects within your current position?
  
2. Describe where your work fits in CCU and mineral carbonation in the supply chain?
  - a) Cement
  - b) Concrete: ready-mix, precast, mortar, blocks
  - c) Material selection: client, project manager, contractor, design team (architect, structural engineer, health and safety advisor)
  - d) Advocacy, policy and standards
  
3. Can you describe your understanding of mineralization and CCU?
  - a) Chemical process of conversion
  - b) End-product applications
  - c) Efficiency of conversion
  
4. What benefits do you anticipate for implementing mineral carbonation/CCU technologies?



- a) Environmental
- b) Financial
- c) Social

5. What are the risks and barriers you foresee, and can you give any examples of a situation where this occurred?

- a) Safety
- b) Economic – CAPEX, OPEX
- c) Design and facility challenges
- d) Market penetration/access (new and existing)
- e) Regulatory and policy hurdles – carbon tax, etc.

Table B1 All codes obtained from qualitative interviews with 6 stakeholder groups in the mineralization field in Alberta. Codes are organized by relevant categories.

Category	Code
Individual and Company Background	Interviewee's mention of knowledge of the field
	Interviewee's mention of industry experience
	Interviewee's mention of work and knowledge
Relevant Area of Work	Ready mix concrete producers
	Concrete addition - solids
	Concrete addition - gas
	Cement replacement/reduction
	Supplementary cementitious material
	Material selection
	Mineralization end products
	Aggregates
	In-situ mineralization
	Accelerated mineralization
	Advocacy and policy
	Client/customer choice
	Technology development support
Areas of Opportunities	Technology chemistry and performance mechanisms
	Cementing efficiency improvements
	Technology research for field deployment
	Market penetration, retrofitting and scalability
	Carbon capture
	Other carbon reduction strategies
	Need for economic drivers
	Existing funding supports
Benefits of Mineralization	Cost saving by offsetting
	Carbon emissions reduction/environmental benefits
	Circular CO <sub>2</sub> economy
	Carbon sequestration
	Reduced industrial waste
	Compressive strength enhancements
	Benefits with policy structures
	Societal benefits of reduced environmental externalities
	Reduction in mining
	Social license to operate
	Partnerships/collaboration
	Energy consumption and life-cycle impacts
Mineralization Risks and Challenges	Cost of operating
	Cost of carbon capture
	Incumbency barriers
	Profit margins for decisions
	Carbon saving and cost uncertainty

	Challenges with policy structures
	Acceptability by regulatory agents
	CSA standards
	Meeting specifications
	Risk with lack of disclosure and transparency
	Risks of the unknown
	Performance, safety and integrity testing
	Inherent overdesign performance variability
	Performance variability from gas additions
	Variability in field characteristics
	Incompatibility with other mix components
	Technical challenges of discovering new technologies
	Transportation requirements to end users
	Further feasibility studies into higher SCM mix design

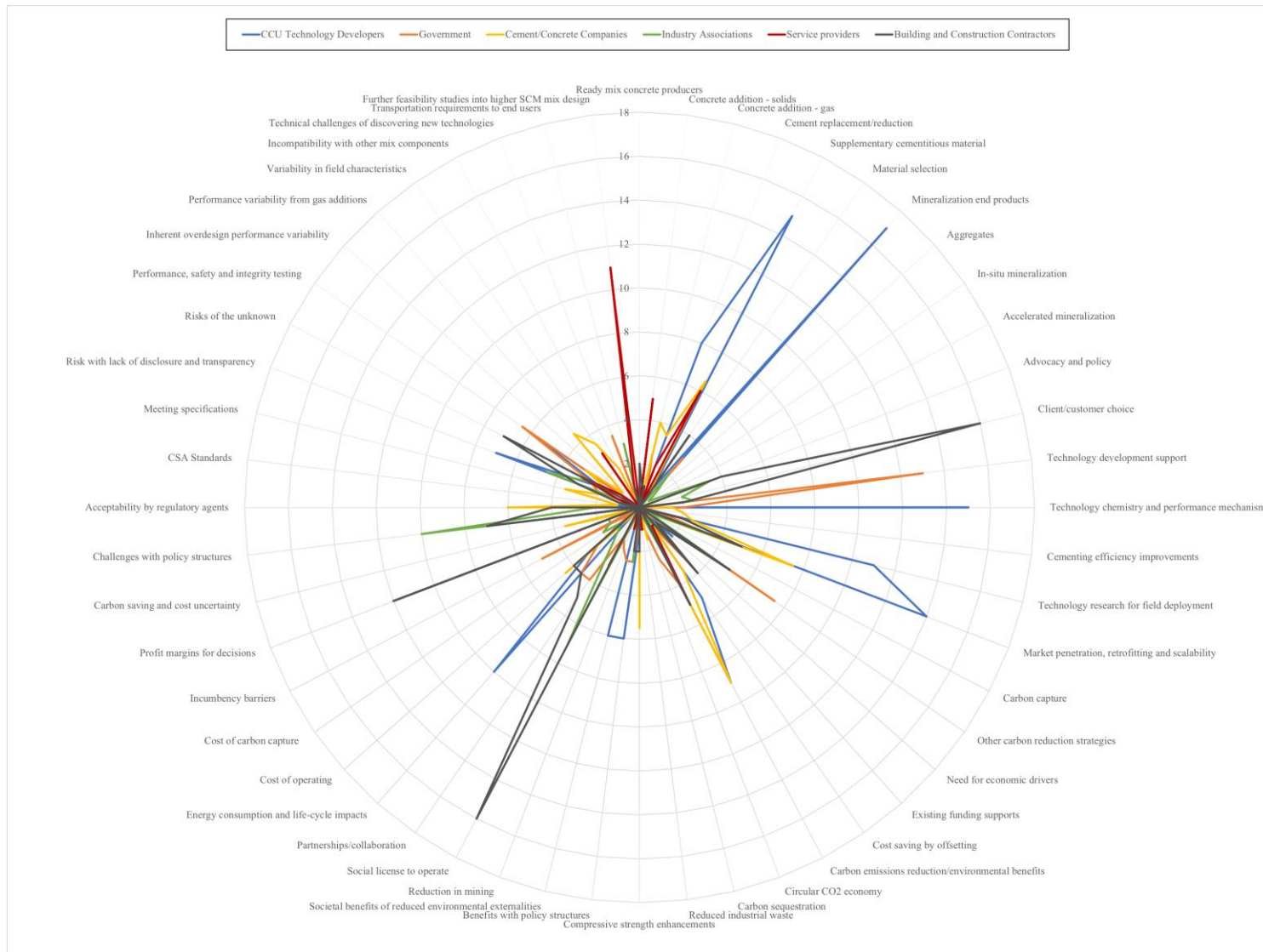


Figure B1 Occurrence scores for all individual codes represented for respective stakeholder groups. Occurrence scores are shown as averages of respondent's responses within each stakeholder group

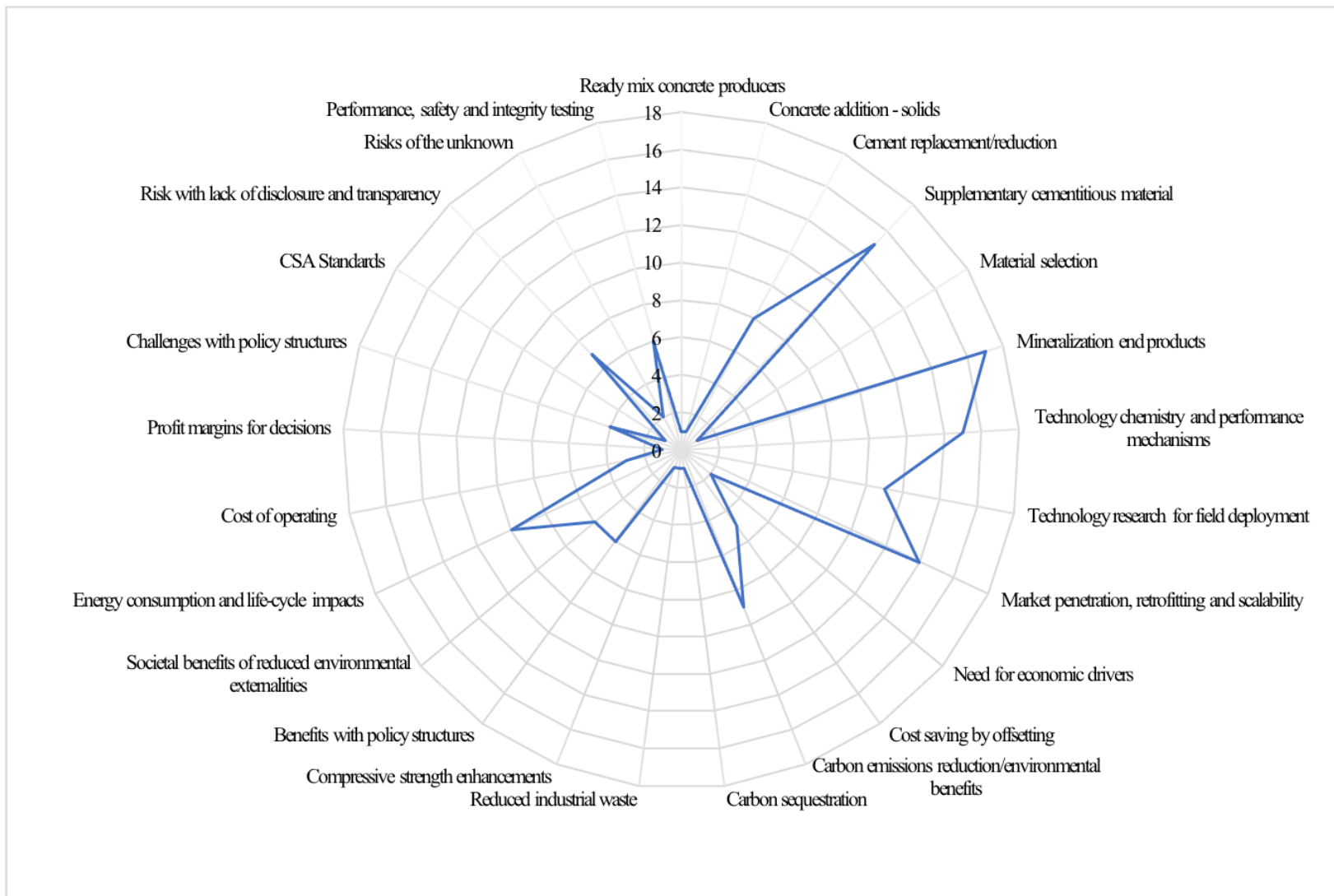


Figure B2 Occurrences of codes represented for CCU technology developers' stakeholder group. Occurrence scores are shown as averages of respondent's responses.

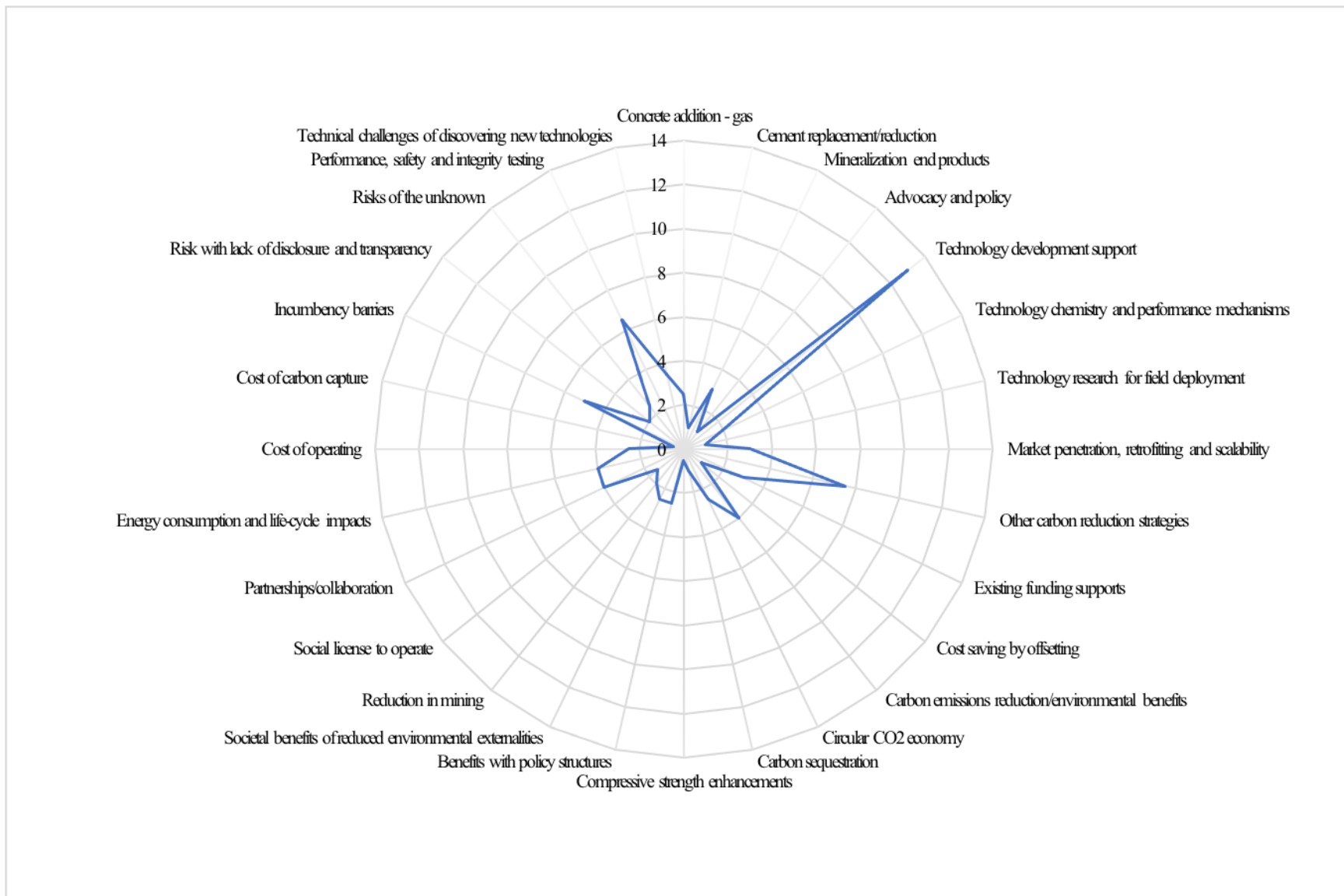


Figure B3 Occurrences of codes represented for CCU governments' stakeholder group. Occurrence scores are shown as averages of respondent's responses.

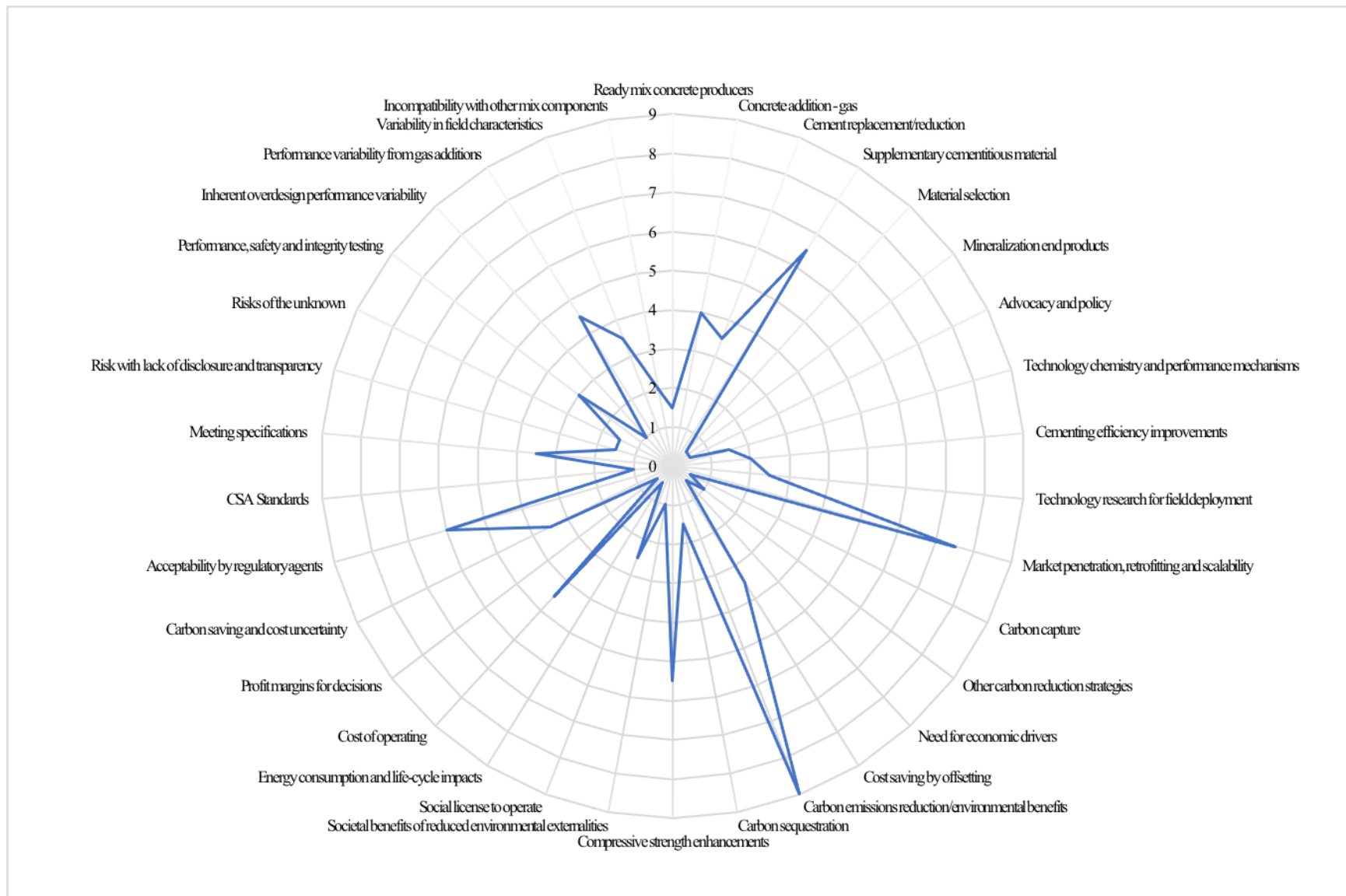


Figure B4 Occurrences of codes represented for CCU cement and concrete companies' stakeholder group. Occurrence scores are shown as averages of respondent's responses.

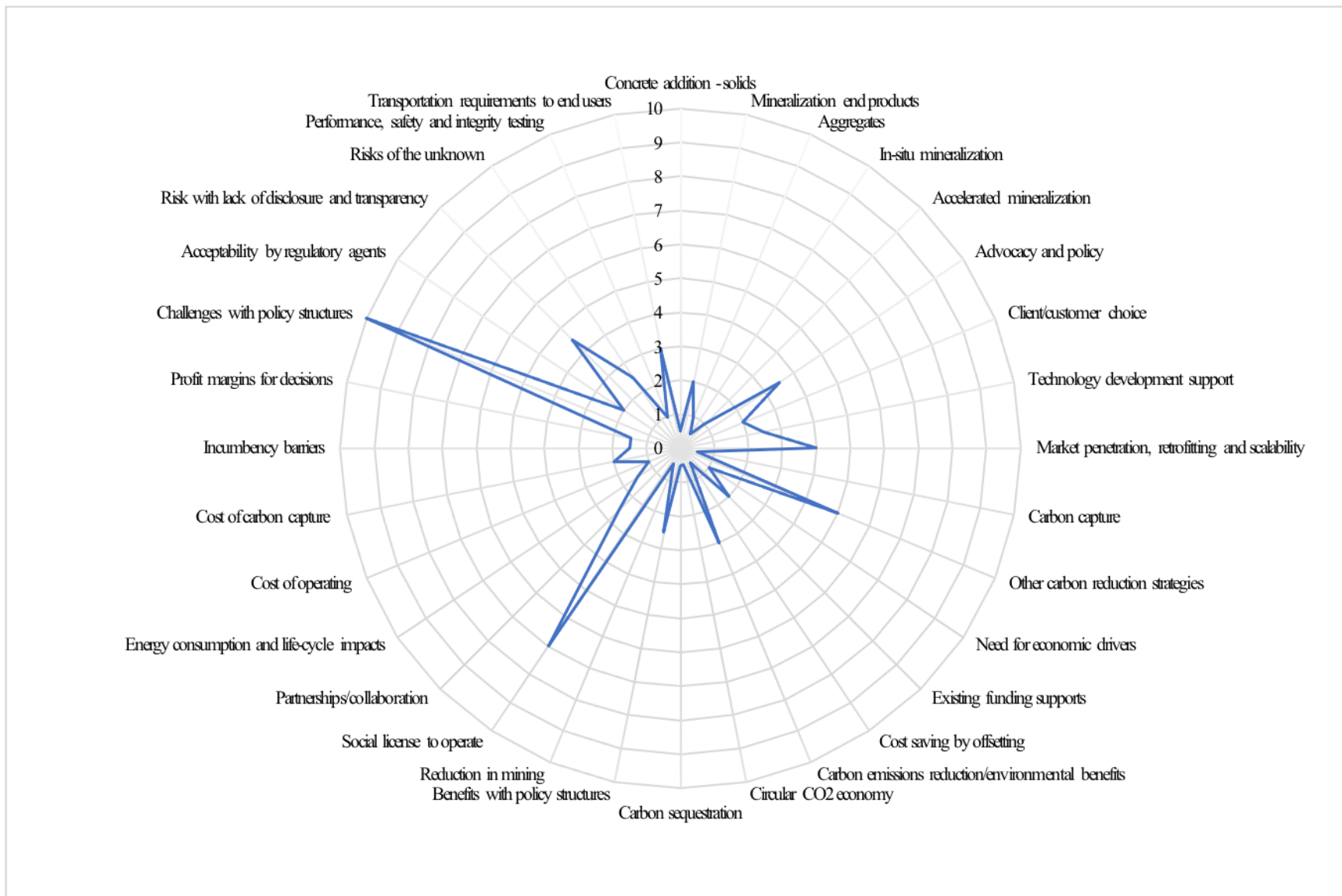


Figure B5 Occurrences of codes represented for CCU industry associations' stakeholder group. Occurrence scores are shown as averages of respondent's responses.



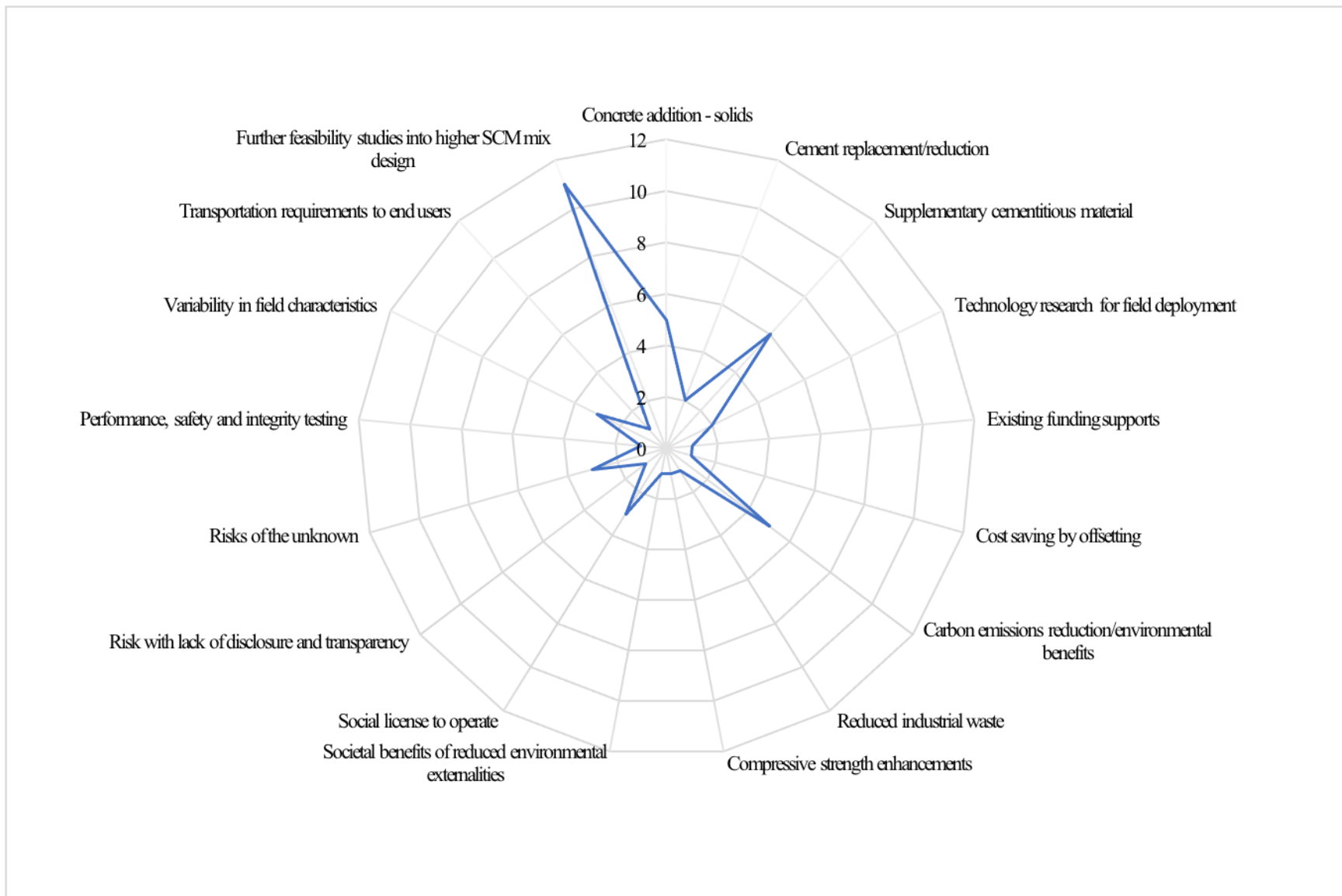


Figure B6 Occurrences of codes represented for CCU service providers' stakeholder group. Occurrence scores are shown as averages of respondent's responses.

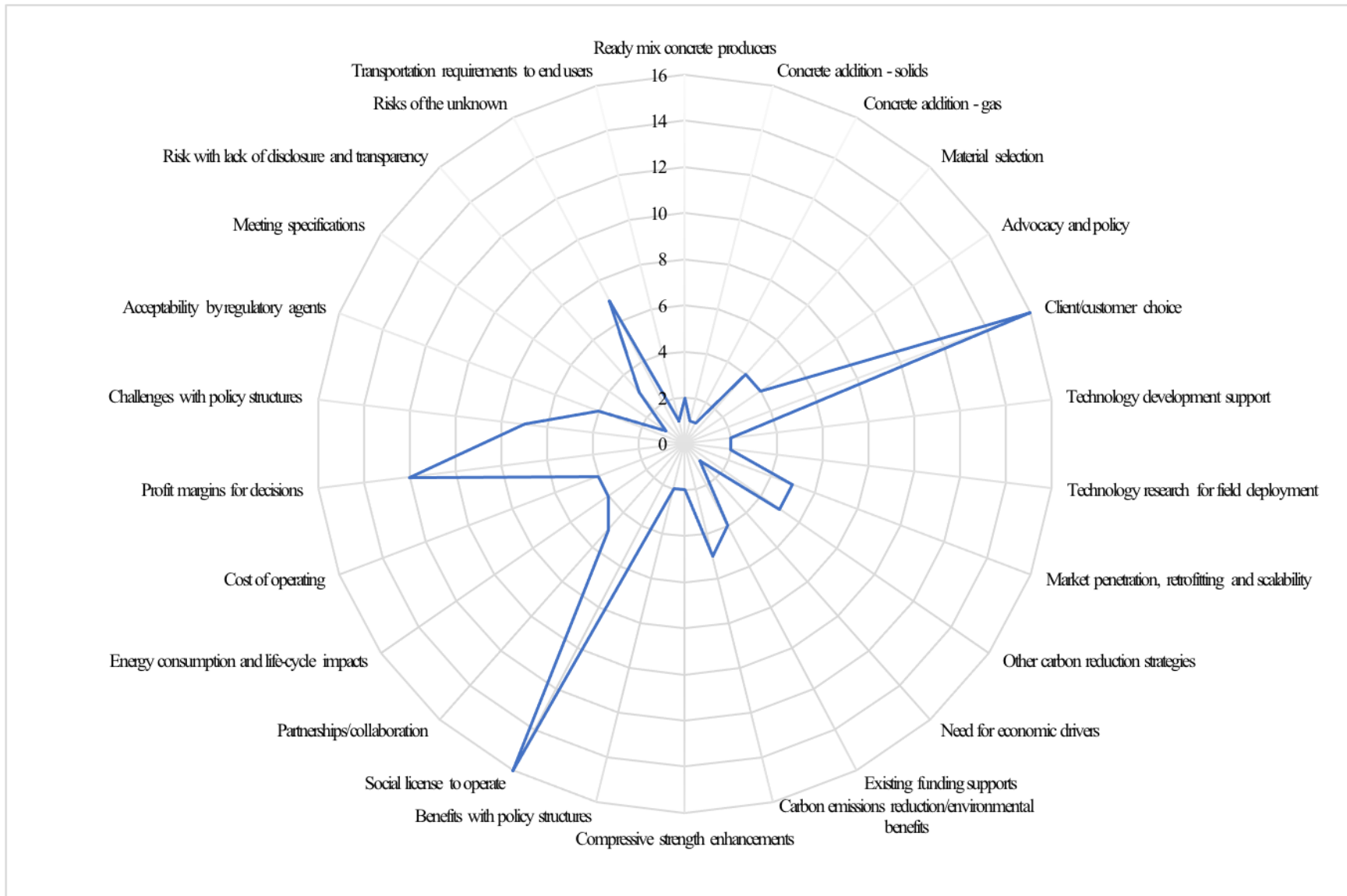


Figure B7 Occurrences of codes represented for CCU building and construction contractors' stakeholder group. Occurrence scores are shown as averages of respondent's responses