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Abstract

Carbon dioxide (CO_2) concentrations in the atmosphere are increasing and are now higher than they have ever been, mostly because of CO_2 released from burning fossil fuels for energy. Global energy demand will continue to rise in the future, resulting in increased carbon emissions. Hydrogen (H_2) could potentially decarbonize energy systems and replace fossil fuels. Climate impact from H_2 production varies but can be considered carbon-neutral if production is replaced with biomethane as feedstock instead of natural gas. Then it is referred to as biogenic hydrogen (bioH_2). If carbon capture and storage (CCS) is applied to bioH_2 production, it can result in negative emissions. Carbon mineralization is a CCS method where CO_2 is injected into underground basaltic reservoirs to rapidly convert into carbonate minerals, providing permanent CO_2 storage.

This study used a life cycle assessment (LCA) method to identify greenhouse gas (GHG) emissions and climate impact from bioH_2 production coupled with carbon storage mineralization. The chosen climate metric for the study is the global warming potential (GWP). This study used a hypothetical biogas plant where the bioH_2 is produced with biomethane as feedstock via the steam methane reforming (SMR) method. The hypothetical carbon mineralization storage site is located right outside of Uppsala where the CO_2 emissions are injected and long-term stored each day.

The results show that implementing CO_2 mineralization to bioH_2 production reduces climate impact and leads to carbon removal. The climate impact can be reduced or increased depending on different factors such as electricity source, CH_4 leakage rate from biogas, CO_2 capture efficiency from bioH_2 production, the scope of CO_2 capture, and different GWP time horizon. The biggest improvement is when the bioH_2 production is operated with renewable energy and the CO_2 capture is increased both in scope and efficiency. Mineralization of CO_2 emissions from bioH_2 production results in significantly reduced climate impact. By integrating the two technologies, it is possible to contribute, to meet the Paris Agreement targets of keeping global warming below 1.5°C to 2°C.

Keywords: Carbon capture and storage, carbon dioxide mineralization, biohydrogen production, global warming potential, greenhouse gases

Sammanfattning

Koncentrationen koldioxid (CO_2) i atmosfären ökar och är idag högre än någonsin, framför allt på grund av CO_2-utsläppen från förbränningen av fossila bränslen till energi. Den globala efterfrågan på energi kommer fortsätta stiga i framtiden vilket resulterar i ökade CO_2-utsläpp. Vätgas (H_2) kan potentiellt de-karbonisera energisystem och ersätta fossila bränslen. Vätgasproduktionens klimatpåverkan varierar men kan betraktas som CO_2-neutral om produktionen ersätter naturgas med biometan som råmaterial, då kallas det för biogent väte (bioH_2). Om kolavskiljning och lagring (CCS) appliceras på BioH_2-produktionen kan det resultera i negativa utsläpp. Kolmineralisering är en CCS-metod där CO_2 injiceras i underjordiska basaltreservoarer i syfte att snabbt omvandlas till karbonatmineraler, vilket skapar permanent CO_2-förvaring.

I den här studien har en livscykelanalysmetod (LCA) använts för att undersöka växthusgasutsläppen (GHG) och miljöpåverkan från produktion av bioH_2 i kombination med kolmineraliseringsförvaring. Som mätsystem i studien används uppvärmningspotential (GWP). Studien utgår från en hypotetisk biogasanläggning i Sverige och produktionen av bioH_2 använder biometan som råvara genom metoden ångmetanreformering (SMR). Anläggningen för den hypotetiska kolmineraliseringsförvaringen är belägen precis utanför Uppsala och injicerar samt långsiktigt förvarar CO_2- utsläpp per dag.

Resultaten visar att implementering av CO_2 mineralisering till bioH_2 produktion reducerar klimatpåverkan och leder till kolavlägsning. Klimatpåverkan kan minska eller öka beroende på faktorer som elkälla, CH_4 läckagehastighet från biogas, CO_2-avskiljningseffektivitet och omfattningen från bioH_2-produktion, och olika GWP-tidshorisonter. Den största förbättringen som kan göras för en bioH_2 - produktion är genom att driva det med förnybar energi och utöka CO_2-avskiljningen både i omfattning och effektivitet. Kolmineralisering av CO_2-utsläpp från bioH_2-produktion resulterade i betydande reducering av GHG-utsläpp. Genom att integrera de två teknologierna är det möjligt att, i en begränsad skala, bidra till att uppfylla Parisavtalets mål att hålla den globala uppvärmningen under 1.5-2°C.

Nyckelord: Koldioxidavskiljning och lagring, koldioxidmineralisering, bioH_2-produktion, uppvärmningspotential, växthusgaser

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Abbreviations

GHG	Greenhouse gases
H_2	Hydrogen
$BioH_2$	Biogenic hydrogen
O_2	Oxygen
CO_2	Carbon dioxide
CH_4	Methane
$BioCH_4$	Methane non-fossil origin
N_2O	Nitrous oxide
SMR	Steam methane reforming
WGS	Water-gas shift
PSA	Pressure swing adsorption
CCS	Carbon capture and storage
LCA	Life Cycle Assessment
LCIA	Life Cycle Impact Assessment
GWP	Global warming potential
FU	Functional unit
LHV	Lower heating value

1. Introduction

Carbon dioxide (CO_2) levels in the atmosphere are higher than ever and growing faster than they have ever been throughout human history (IEA 2021b). Every year a new record amount of global average atmospheric CO_2 is hit. Where CO_2 increases at rate 100 times faster than previous natural changes (Lindsey 2021). Owing it mostly to human activities, such as the combustion of fossil fuels and natural gas. On a global scale, fossil fuels account for 80% of energy production (Hassan et al. 2021). Meanwhile, global energy consumption will continue to rise in the future decades, with fossil fuels still dominating (IEA 2021a). Continuing to use fossil fuels will raise CO_2 concentration in the atmosphere and can lead to climate impact consequences such as more extreme temperatures, rising sea levels, and significant biodiversity losses. The Paris Agreement aims to limit global warming below 1.5 and 2°C, compared to pre-industrial levels (UN n.d).

The International Energy Agency (IEA) has long stated that achieving international energy and climate goals does not require a single or simple answer (IEA 2021c). To achieve net-zero energy systems a significant shift in how energy is used and produced must happen, which can only be accomplished with a diverse set of technologies. In addition to electrification, hydrogen- and sustainable bioenergy, carbon capture and storage (CCS) will be critical (IEA 2020a).

Hydrogen (H_2) is thought to be crucial for Europe's energy transition as a future decarbonized energy system. This shift could have a significant impact on how energy is distributed, stored, and consumed (Fuel Cells and Hydrogen 2 Joint Undertaking. 2019). Hydrogen and hydrogen-based fuels are thought to be able to fill the gaps where electricity cannot readily or economically replace fossil fuels, and also where sustainable bioenergy sources are insufficient to meet demand (IEA 2021c). It is also important in the decarbonization of transportation, buildings, and industries (Fuel Cells and Hydrogen 2 Joint Undertaking. 2019). Nonetheless, to meet the transition, H_2 production processes must become carbon-neutral (European Commission 2020). Existing biogas plants can convert their CH_4 production into biogenic hydrogen (bioH_2). This process results in biogenic CO_2 emissions which are neutral from a climate perspective, but if this CO_2 is captured and stored, then it could result in net negative emissions (Antonini et al. 2020).

Carbon capture and storage is the only technology that directly reduces CO_2 emissions in key sectors as well as removing CO_2 that cannot be avoided. It can reduce emissions from fossil fuel power plants, limit industrial processes, and remove CO_2 directly from the atmosphere and therefore deliver negative emissions (IEA 2020a). Carbon capture and storage is when CO_2 is captured and stored long-term in underground geological formations (European Commission n.d.). One CCS method is carbon mineralization, where CO_2 is injected into basaltic formations using mineralization, which provides significant advantages including permanent storage and a great potential storage volume (Gutknecht et al. 2018).

Combining $bioH_2$ production with CCS could be an effective way of carbon removal. This thesis aims to assess the climate impact of $bioH_2$ production coupled with the CCS method of carbon mineralization and to comprehend how these two technologies can be accomplished and what are their potentials. This is accomplished by the application of the life cycle assessment (LCA) methodology. The following research questions will be answered to achieve the thesis's goal:

- For a Swedish case study, what is the climate benefit of $bioH_2$ production using CO_2 mineralization for CCS technologies?

- How does a sensitivity analysis of the most relevant process steps and parameters affect the climate benefits?

- What is the maximum amount of net negative emissions that the process can achieve?

2. Background

2.1 Hydrogen production

Hydrogen as an energy source has numerous advantages; it can support energy systems in a variety of ways, and it can be used to replace carbon-containing fuels in transportation and industry (IEA 2019). Different technologies are used to produce hydrogen. The most common one is the use of natural gas as a feedstock in the steam methane reforming (SMR) process, which releases large amounts of greenhouse gas emissions (GHG) (Bhat et al. 2009; Nikolaidis et al. 2017). If natural gas were replaced by biogas the process would be declared carbon neutral. The biomass absorbs CO_2 through photosynthesis, and when biogas is used in production, about the same amount of CO_2 is emitted, resulting in carbon neutrality (Antonini et al. 2020).

2.2 Carbon capture and storage

In general, CCS methods can be categorised into two approaches: through direct air capture with CCS (DACCS), or from the exhaust system of combustion or off-gas from fermentation processes (BECCS) to produce negative emissions (carbon removals). Then the CO_2 is compressed and/or liquified and transported to a location where it will be long-term stored (European Commission n.d.). The amount of CCS operations globally is nowhere near what is required to put the world's CO_2 emissions on a sustainable course (IEA 2020a). Most CCS projects inject gaseous, liquefied, or supercritical CO_2 into sedimentary basins. That requires an impermeable cap-rock for long-term storage. Some of the CO_2 becomes trapped in pores, and some dissolves in groundwater, or reacts and forms stable carbonate minerals. This method has the risk of the CO_2 being buoyant and can migrate back to the surface (Snæbjörnsdóttir et al. 2014).

In the carbon cycle, carbon flows from one terrestrial reservoir to another and spends millions of years in rocks. Rocks are the largest reservoir of carbon on Earth. As a result of this natural evidence, mineral carbon storage appears to be the most

stable option for long-term carbon storage (Snæbjörnsdóttir et al. 2020). A new geological storage method through carbon mineralization has been developed in Iceland where captured CO_2 is dissolved in water during injection into porous basaltic rock (see figure 1), so-called in-situ mineralization (Snæbjörnsdóttir et al. 2014; Romanov et al. 2015). Through mineral carbonation the CO_2 is completely dissolved in water that is denser than the CO_2-free formation fluids, thus no cap rock is required and CO_2 is no longer at risk of it being buoyant and migrating to the surface (Snæbjörnsdóttir et al. 2014). Therefore, mineral carbonation via CO_2-fluid-rock reactions reduces the possibility of leakage, allowing for long-term and safe carbon storage (Matter et al. 2016).
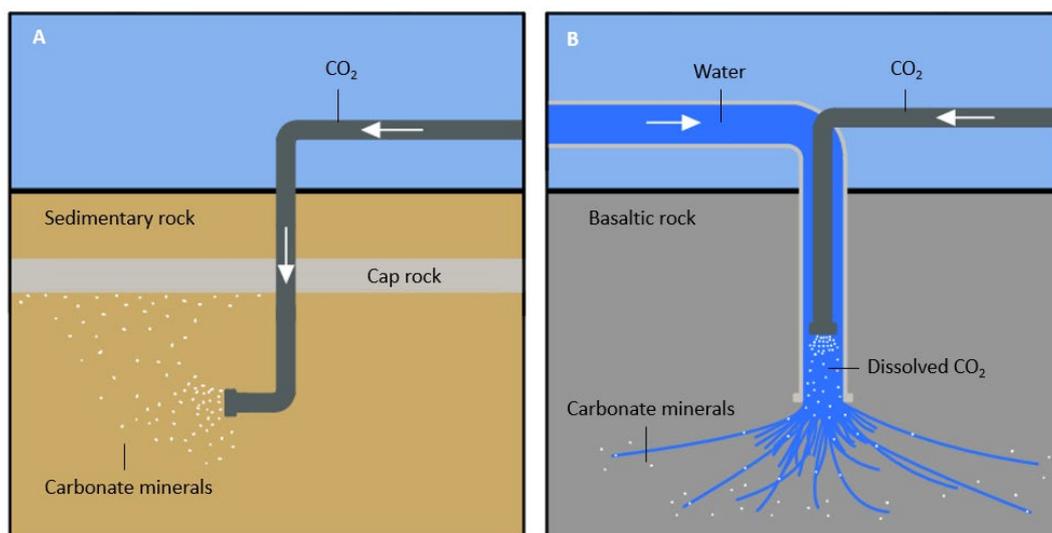
Figure 1. Two methods of carbon storage. (A) CO_2 is injected into sedimentary basins with caprock. (B) CO_2 is dissolved in water (H_2O) and injected into porous basaltic rock (CO_2 mineralization with the Carbfix method)(Gislason & Oelkers 2014).

Before CO_2 enters the rock when performing CO_2 mineralization, the CO_2 bubbles dissolve in the water. The CO_2 must be dissolved during injection at a minimum pressure of 25 bar, which is 250 m below the water table, thus making 350 meters depth ideal for injection. Water that has been charged with CO_2 is acidic, with a pH of 3–5 (Snæbjörnsdóttir et al. 2014). To form carbonate minerals the combination of CO_2 with metals is essential. There, basaltic and ultramafic rocks are most efficient, because of the presence of divalent metal cations in silicates and their strong reactivity (Snæbjörnsdóttir et al. 2020). The required metals are divalent cations such as Mg^{2+}, Fe^{2+}, and Ca^{2+} (Gislason et al. 2010). Then the CO_2 charged water interacts with reactive rock which releases these divalent cations and subsequently binds the CO_2 by forming a solid carbonate mineral such as magnesite ($MgCO_3$), siderite ($FeCO_3$), and calcite ($CaCO_3$), resulting in the permanent storage of carbon (Gutknecht et al. 2018; Ratouis et al. 2022; *How it works* n.d.). The

essential presence of divalent cations makes mineral carbonation geologically dependent. The carbon dioxide stored as a carbon mineral becomes immobilized for geological time periods and a stable carbon storage host (Snæbjörnsdóttir et al. 2014).

The alternative carbon mineralization method is called Carbfix and this study follows the CO_2 mineralization approach of the CarbFix project (Gislason et.al 2014; Matter et al. 2016; Gutknecht et al. 2018; Voigt et al. 2021; Ratouis et al. 2022; Snæbjörnsdóttir et al. 2022). Today this CO_2 mineralization method is utilized at a geothermal power plant in Iceland. There, a fraction of CO_2 is captured when producing geothermal electricity. The CO_2 is dissolved in water in a scrubbing tower and injected via injection well device into the subsurface, which reaches approximately 2000 m down into the reservoir. Roughly 12000 tonnes of CO_2 are turned into carbon mineral rock every year at the Hellisheidi geothermal plant (Snæbjörnsdottir et al. 2021).

2.3 LCA methodology

Life cycle assessment (LCA) is a method to assess the environmental impacts of products, processes, or activities throughout their life cycle (Klöpffer & Grahl 2014). LCA can assess different impact categories associated with all stages of a process from cradle to grave, preventing environmental burden shifting between life cycle stages and different environmental impact categories (*International Standard ISO 14040* 2006). LCA can identify opportunities to improve the environmental performance of products along their life cycle. There are generally four phases in an LCA study: Goal and scope, life cycle inventory analysis (LCI), life cycle impact assessment (LCIA), and interpretation of results (Figure 2). Goal and scope determine and describe the reason for conducting the study, the functional unit (FU), and the system boundary. A FU is a quantitative description of the function or service provided by the studied system, and it can be used as a basis for comparison (Klöpffer & Grahl 2014). Following the goal and scope definition phase, the core activities of LCI are to collect and compile all inputs and outputs data. LCIA assigns and combines all the inputs and outputs collected from LCI into known environmental impact categories, such as global warming, eutrophication, resource depletion, and acidification (Nieto et. al 2014).
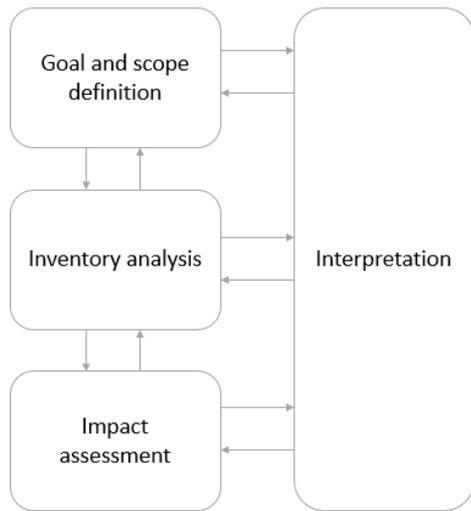
*Figure 2. Life cycle assessment framework from ISO 14040:2006. (*International Standard ISO 14040 *2006).*

Human activities have led to increased concentration of GHG, leading to changes in the earth's average surface temperature. The most common GHGs are CO_2, methane (CH_4), and nitrous oxide (N_2O). Carbon dioxide is the primary GHG, emitted through the combustion of fossil fuels. Methane is mostly emitted through livestock and agricultural practices as well as the production of coal, natural gas, and oil. Agriculture, fuel combustion, wastewater management, and industrial processes all contribute to the increase of N_2O in the atmosphere (US EPA 2015).

Global warming potential (GWP) reflects the average residence time of each GHG in the atmosphere and how strongly it absorbs heat. The gases that absorb more energy have higher GWP and therefore contribute more to warming Earth (US EPA 2015). The given global warming period is normally 100 years (GWP_{100}), but can also be given in 20 years (GWP_{20}) (Myhre et al. 2014). Global warming potential is an index used when working with the climate impact category, with CO_2 having the index value of 1, and the GWP for all other GHGs is the number of times more warming they cause compared to CO_2 (Brander 2012). Table 1 shows the metric used to transform the effects of different GHG emissions to the common scale CO_2-equivalents (CO_2-eq.) according to the 5th IPCC Assessment Report (Myhre et al. 2014).

Table 1. Global warming potential for the time horizon of 100 years and 20 years for each greenhouse gas is given in CO_2-equivalent (Myhre et al. 2014).

Greenhouse gas	GWP_{100} (CO_2-eq.)	GWP_{20} (CO_2-eq.)
Carbon dioxide (CO_2)	1	1
Carbon dioxide ($bioCO_2$) [1]	0	0
Methane (CH_4) fossil-origin	30	85
Methane ($bioCH_4$) non-fossil-origin	28	84
Nitrous oxide (N_2O)	265	264

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[1] $BioCO_2$ is CO_2 that has already been taken up by biomass and can therefore be considered carbon neutral, resulting in zero-emission (Antonini et al. 2020; IEA bioenergy 2022).

3. Method

A cradle-to-gate LCA was conducted to quantify the GHG of $bioH_2$ production. This LCA study follows the methodological framework, principles, and standards of ISO 14044:2006 and ISO 14040:2006, including four standard phases: goal and scope definition, LCI, LCIA, and interpretation.

3.1 Goal and scope

The purpose of this LCA study is to identify GHG emissions in $bioH_2$ production from biogas with the CCS method of carbon mineralization. Only the climate impact category was evaluated in this study and analysed using the GWP. The functional unit (FU) is the production of 1 kg H_2.

As shown in Figure 3, the system can be divided into two life cycle stages: the biohydrogen production and the carbon capture and storage stage. The $bioH_2$ production stage can be split into two processes: The biogas process and the $bioH_2$ process. Where the biogas process includes feedstock production, biogas production, and biogas upgrading, while the $bioH_2$ process includes SMR, water gas shift (WGS), and pressure swing absorption (PSA). The CCS stage includes the processes of CO_2 capture, CO_2 compression, CO_2 transportation, and long-term CO_2 storage.

Collection and transport of raw material were not included in the system as well as emissions from construction and upkeep of both the $bioH_2$ production plant and the CO_2 storage site. Water was used during $bioH_2$ production during the steps of the SMR and WGS, but no emission associated were considered. This contrasts with water used during CO_2 storage. Minor loss and specialized waste were excluded from the system boundaries. The excluded parts are related to the internal transport of workers and machinery, human work, and storage spaces.
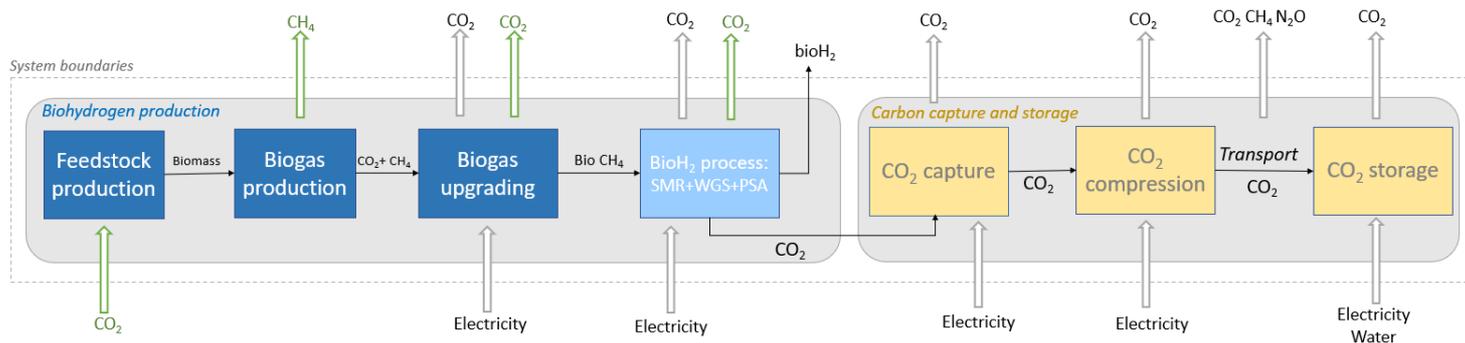
Figure 3. The process tree of the system studied. All the system's inputs are shown below the system, outputs are displayed above the system. The biogenic input and output of CH₄ and CO₂ is shown by the green color of the arrows.

3.2 Life cycle inventory and Life cycle assessment

3.2.1 Biogas process

In this study, a hypothetical biogas plant in the city of Uppsala, Sweden was considered. In this hypothetical case, Uppsala Vatten's production scale was used, where CO_2 emissions from biogas upgrading are approximately 10 tonnes of CO_2 per day (Uppsalavatten n.d.). The biogas plant is assumed to have an energy output of 30 GWh per day, and the amount of 15.4 kWh/kg CH_4 (Stolpe 2021). Electricity consumption is considered when assessing the biogas process, using the environmental impact from the Nordic residual mix 2020, which was 0.365 kg CO_2-eq./kWh. The residual mix refers to the volume in the Nordic region of the electricity that is purchased when no specific power source or origin-marking is set (EI n.d.). This was used throughout the study in all processes that need to be powered by electricity.

Feedstock production

The biogas process uses biomass as feedstock, where CO_2 was taken up during the feedstock production by photosynthesis. Potential emissions from upstream processes like land use and agriculture are not included because the feedstock used for biogas production is based on waste streams like sewage sludge and bio-waste.

Biogas production and biogas upgrading

Biogas is composed of mainly CH_4, with the rest primarily being CO_2, and a small fraction of other gases. By purifying the biogas from CO_2 contamination, bioCH_4 is produced. This method is called biogas upgrading (Andriani et al. 2014; IEA 2020b). The resultant CO_2 stream was concentrated from the biogas upgrading (e.g., amine scrubbing).

Methane leakage is an issue for many biogas plants, which might result in big negative effects on the total GHG emission performance of the whole system. To calculate the CH_4 emissions from the biogas process the amount of bioCH_4 required per day was based on a 1.4% methane leakage rate and a 97% methane concentration in biomethane (Uppsala vatten 2018; Stolpe 2021). Carbon dioxide emissions associated with biogas upgrading are mainly from two sources: the bioCO_2 separated from the biogas, and the CO_2 generated from the upstream electricity production. The latter part can be calculated using the electricity grid emission factor (in kg CO_2/kWh) multiplied by the electricity required for biogas upgrading (in kWh). No emissions of N_2O from the biogas process were considered.

3.2.2 BioH$_2$ process

It is assumed that the bioH_2 is produced by the SMR method using biomethane as feedstock. SMR involves two steps, the first step is reforming, and the second step is WGS. In the first step, CH_4 and water in the form of steam are combined and are carried out at 750-800°C to produce synthesis gas, which is H_2 – rich syngas, with carbon monoxide (CO) as a by-product. The second step (WGS) involves a catalytic reaction of CO with steam to form H_2 and CO_2. Purification of the produced H_2 is then usually accomplished using PSA (Bhat et al. 2009).

There are two sources of CO_2 emissions in an SMR plant: the first (60%) comes from the feedstock during reforming and shift, and the second (40%) comes from the combustion in the reformer furnace (Antonini et al. 2020). The electricity consumption of the whole bioH_2 process was calculated by multiplying the H_2 produced per day (in kg H_2/d) and the residual emission factor (in kg CO_2/kWh by the specific power consumption of the SMR process, which is 2.6 kWh/kg H_2 (Valente et al. 2020; Stolpe 2021). Then the total was added by the amount of CO_2 released from the processes (in kg CO_2/kg H_2). Because the process uses bioCH_4 as a fuel, there are also biogenic CO_2 emissions, but the N_2O emissions were not included.

3.2.3 Carbon capture and storage

CO₂ capture

Carbon dioxide is already separated during biogas upgrading, the CO_2 capture efficiency was assumed to be 54% (Timmerberg et al. 2020), and the total amount of CO_2 captured per day will be 7781.4 kg. To calculate the emissions from electricity consumption of CO_2 capture, the CO_2 released from the $bioH_2$ process (in kg CO_2/d) was multiplied by the capture rate efficiency and specific electricity consumption of CO_2 capture, which is 0.196 kWh/kg CO_2 (Stolpe 2021). Then the outcome was multiplied by the residual mix electricity emission factor (in kg CO_2/kWh).

CO₂ compression

The CO_2 was compressed once it had been captured. A single-state water-cooled compressor was used and CO_2 was compressed from 1 bar(a) to 13.5 bar(a), with specific electricity consumption of 0.185 kWh/kg CO_2 (Janke et al. 2022).

   To calculate the amount of GHG emissions from compressing CO_2, the specific electricity consumption to compress CO_2 was multiplied by the amount of CO_2 captured per day from the $bioH_2$ process (in kg CO_2/d). The total electricity consumed per day was then multiplied by the residual mix emission factor (in kg CO_2/kWh) to determine total CO_2 emissions. The emissions from CH_4 and N_2O were not included.

CO₂ transportation

The compressed CO_2 was assumed to be transported from Uppsala vatten AB's biogas station to Bäsingen lake (Uppsalavatten n.d.; *Carbfix Atlas* n.d.). The total distance was found using google maps and the shortest route possible was chosen, 208 km a roundtrip journey. The transport truck is expected to weigh 18 tonnes and the compressed CO_2 will be able to be carried in a single trip each day (Viktbestämmelser 2021).

   The GHG emission from transportation was calculated according to Stolpe 2021. By finding out the fuel consumption for diesel trucks (in l/km) and the density of the diesel (kg /l). As well as emission factors from production, distribution, and

usage of diesel with 5% RME for CO_2, CH_4, and N_2O (in GHG/MJ). Then the lower heating value of diesel (in MJ/kg fuel) was used (Stolpe 2021).

CO_2 mineralization

The CO_2 storage site was assumed to be placed in the Swedish region of Bäsingen which is 104 kilometers from Uppsala and should be a feasible location for CO_2 mineralization according to the Mineral Storage Atlas (*Carbfix Atlas* n.d.). When the compressed CO_2 has been transported to the CO_2 storage site in Bäsingen it is suctioned into the injection well's down-flowing water at a depth of 350 meters. The optimal temperatures for carbon mineralization are between 25 and 170 degrees Celsius, with substantial CO_2 mineralization expected when the fluid reaches 6 pH (Snæbjörnsdóttir et al. 2022). The CO_2 mineralization requires large amounts of water, around 27 tonnes of water to fully dissolve one tonne of CO_2 charged water at 25 bar pressure and at 25 °C (Snæbjörnsdóttir et al. 2014). To determine the water consumption for the mineralization and the emission from pumping water, the daily consumption to dissolve the captured CO_2 was found. The water to dissolve the injected CO_2 was assumed to be pumped up from the Bäsingen lake, where the average deep is 15 meters (*VISS* 2021). According to Cotton info 2015, the electricity needed to lift 1 ML a height of 1 meter would use 4.55 kWh. This is a theoretical calculation based on 70% pump efficiency, with 95% drive train efficiency, and that the average electrical motor efficiency is 90% (Cottoninfo 2015). The electricity demand to pump water up to 15 m in height was calculated and multiplied by the daily water consumption (in ML). To calculate the CO_2 emission of water pumping of 15 m, the total electricity consumption (in kWh) was then multiplied by the Nordic residual mix (in kg CO_2/kWh). The GHGs CH_4 and N_2O were not included.

The compressed CO_2 arrives at the storage site pressurized at 13.5 bar, however, CO_2 must be dissolved during injection at a minimum pressure of 25 bar. Therefore, a second compression up to 25 bar is necessary for which the same specific electricity consumption was used as a conservative approach. The main energy demand for carbon mineralization storage is to pressurize the CO_2-charged water, which is 75 kWh for 1 tonne of CO_2-charged water (Snæbjörnsdottir 2022; *FAQ* n.d.). The amount of GHG emission from electricity consumption of the CO_2 storage was calculated. The overall amount of CO_2 captured per day from the bioH$_2$ process (in kg CO_2/d) was multiplied by the residual mix emission factor (in kg CO_2/kWh) and the specific electricity consumption (in kWh).

When water and captured CO_2 are injected together, the CO_2 dissolves into a mineral, and after two years, roughly 95% of the carbon is permanently deposited as carbonate minerals (Matter et al. 2016; *How it works* n.d.). To figure out the amount of CO_2 mineralized per kg $bioH_2$ produced, the amount of CO_2 released per H_2 produced from the $bioH_2$ process (in kg CO_2/kg H_2) was multiplied by the capture efficiency rate and the rate of CO_2 mineralized over two years.

3.3   Sensitivity analysis

By modifying one parameter at a time, a sensitivity analysis was performed to examine the impact of various assumptions stated in the thesis results:

- The electricity source was modified by using certified renewable energy instead of the Nordic residual mix. According to EI, certified renewable energy production such as wind- and solar energy can be reported with zero carbon dioxide emissions (EI n.d.).
- The methane leakage rate from the biogas plant was modified, by increasing the rate from 1.4% to 2.8% and then reduced to 0.7%.
- The capture efficiency of the carbon capture was changed to the maximum, which is 90% (Timmerberg et al. 2020).
- The electricity source and the capture efficiency rate were modified at the same time, to certified renewable energy and a 90% capture efficiency rate.
- Rather than a GWP of 100 years (GWP_{100}), the GWP time horizon was changed to 20 years (GWP_{20}). Then the GWP of $bioCH_4$ changes from 28 CO_2-eq., to 84 CO_2-eq. (Myhre et al. 2014). This was done because CH_4 has a relatively shorter atmospheric lifetime, between 12-15 years when compared to the tens of thousands of years of the lifetime of atmospheric CO_2 and the 120 years of atmospheric N_2O (Doble et al. 2007).
- Along with changes in the electricity source and the capture efficiency rate, the scope of the CO_2 capture was also increased, where the CO_2 was additionally captured from the biogas upgrading process.

4. Results

In this chapter results of the system, and simulations are presented and described, as well as the GHG flows within the system boundary and the climate impact of bioH$_2$ production deployed with and without CCS. Finally, the results of the sensitivity analysis are outlined.

4.1 GHG flows within the system boundary

Table 2 illustrates daily GHG emissions flows from bioH$_2$ production with and without the CCS technology, using GWP$_{100}$ to convert emitted GHG to CO$_2$-equivalents. The table shows the GHG emissions impact from each process, where the main contributor to GHG emissions is the power consumption of the system's technology, which results in the release of fossil CO$_2$. The table also gives a better picture of how much GHG is emitted if CO$_2$-eq. are not captured and long-term stored. The bioH$_2$ production stage cannot reach carbon neutrality because the overall emissions from the bioH$_2$ production are higher than the amount of CO$_2$ absorbed by the biomass. However, the CCS stage can result in more than carbon neutrality, when the CO$_2$-eq. are mineralized the total net emissions results in negative emissions.

Table 2. GHG flows for the hypothetical bioH$_2$ plant with and without CCS per day. The green color represents the biogenic flow of CH$_4$ and CO$_2$.

Process	BioH$_2$ without CCS [kg CO$_2$-eq.]	BioH$_2$ with CCS [kg CO$_2$-eq.]	GHG emissions
Feedstock production	-24320.29	-24320.29	◦ CO$_2$ uptake in biomass
Biogas production	2092.15	2092.15	◦ BioCH$_4$ from biogas reactor + upgrading [2]
Biogas upgrading	11404.36	11404.36	◦ BioCO$_2$ from a biogas reactor ◦ CO$_2$ from power consumption
SMR+WGS+ PSA	15929.73	15929.73	◦ BioCO$_2$ from breaking down CH$_4$ ◦ CO$_2$ from power consumption
CO$_2$ capture	-	556.69	◦ CO$_2$ from power consumption
CO$_2$ compression	-	525.45	◦ CO$_2$ from power consumption
CO$_2$ transportation	-	184.49	◦ CH$_4$ + CO$_2$ + N$_2$O from fuel usage
CO$_2$ storage	-	225.95	◦ CO$_2$ from power consumption
CO$_2$ mineralization	-	-7392.46	◦ CO$_2$ from non – mineralized fraction ◦ Mineralized CO$_2$
Net emission	5105.96	-793.92	

Biohydrogen production spans the Feedstock production through SMR+WGS+PSA rows (blue). *Carbon capture and storage* spans the CO$_2$ capture through CO$_2$ mineralization rows (yellow).

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[2] It was not possible to allocate different CH$_4$ leakage from biogas production and biogas upgrading.

4.2 Climate impact of bioH$_2$

Figure 4 indicates GHG emissions per kilogram of H$_2$ produced. By deploying CCS with bioH$_2$ production the GHG emissions were reduced from 3.19 kg CO$_2$-eq. per kg bioH$_2$ to a negative emissions of 0.50 kg CO$_2$-eq per kg H$_2$, resulting in 2.69 kg CO$_2$-eq. per H$_2$ difference.

The bioH$_2$ process (including SMR, WGS, and PSA), is the biggest contributor to the total GHG emissions, emitting 9.95 kg CO$_2$-eq./kg H$_2$ per day. Followed by the biogas upgrading of 7.12 kg CO$_2$-eq./kg H$_2$ per day. Freshwater needed for CO$_2$ injection is approximately 219 tonnes per day, however, the required energy for pumping the water is rather low. The total GHG emissions from the whole CCS stage is 0.60 kg CO$_2$-eq./kg H$_2$ per day, which is significantly low compared to the amount of CO$_2$-eq. that is mineralized into the reservoir per H$_2$ produced, approximately 4.62 kg CO$_2$-eq./kg H$_2$.
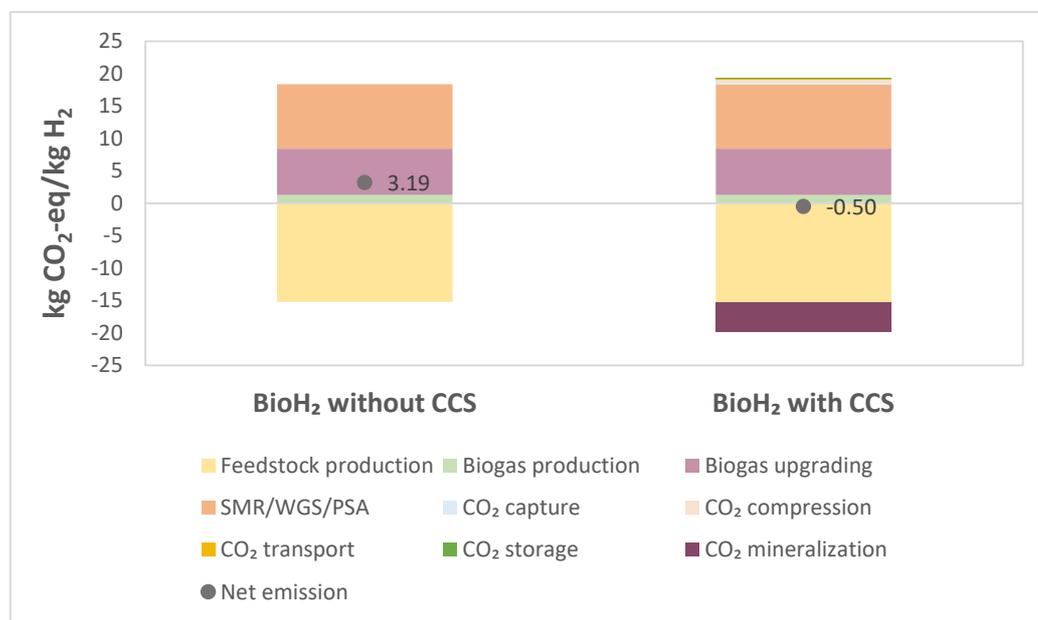

Figure 4. GHG emissions in CO$_2$-equivalents from the hypothetical production of bioH$_2$ with and without CCS. Each color represents different processes within the system.

4.3 Sensitivity analysis

By modifying the electricity source, methane leakage rate, CO_2 capture efficiency, the GWP time horizon, and the scope of CO_2 capture the system's sensitivity was determined and presented.

Changing the electricity source from Nordic residual mix to certified renewable energy has a major impact on the climate impact of H_2 production with CCS. The total carbon removal of the CCS process increases, from -0.50 kg CO_2-eq./kg H_2 to -3.20 kg CO_2-eq./kg H_2, as shown in figure 5. It also has an impact on the total GHG emissions from the bioH$_2$ process and the biogas upgrading. Therefore, bioH$_2$ production with and without CCS technology is particularly sensitive to the source of electricity.
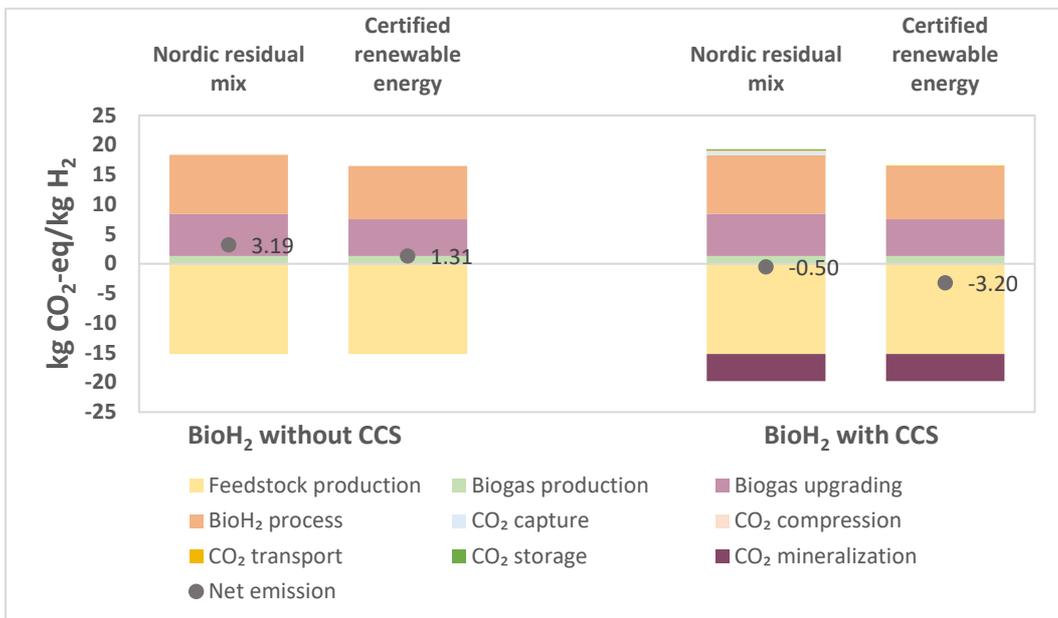

Figure 5. Systems sensitivity on electricity sources. Changes made by using certified renewable energy source instead of the thesis Nordic residual mix.

When the leakage rate is increased to 2.8%, the GHG emissions from bioH$_2$ production with and without CCS increase significantly, as shown in figure 6. The bioH$_2$ production with CCS no longer results in net negative emissions when producing H_2. When the leakage rate is reduced to 0.7%, the net climate impact from the whole life cycle decreases. When the production is coupled with CCS, the total GHG emissions removal increases to a net negative emission of 1.15 kg CO_2-eq./kg H_2. This demonstrates that CH_4 leakage has a considerable high impact on H_2 production.
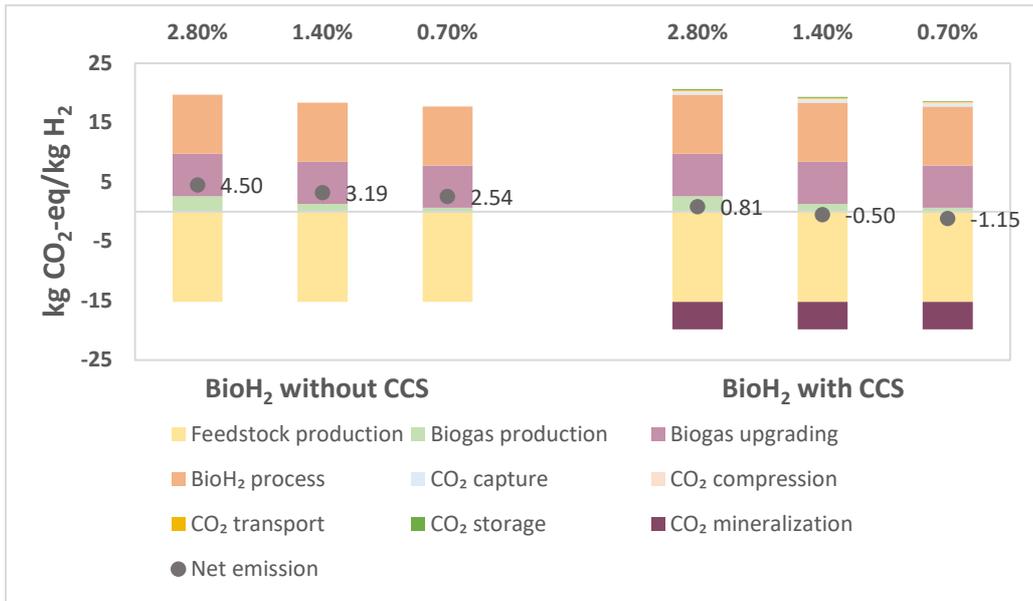
Figure 6. Systems sensitivity on CH$_4$ leakage rate. Changes made by using the thesis rate 1.4%, then the leakage rate 2.8% and 0.7%.

When the CO$_2$ capture efficiency is increased from 54% to 90%, the amount of CO$_2$ mineralized per kg H$_2$ increases. Biogenic H$_2$ production without CCS is unaffected. By increasing the capture efficiency, in the CCS stage, the amount of CO$_2$ injected per kg H$_2$ increases to the negative emissions of 3.03 kg CO$_2$-eq./kg H$_2$, shown in figure 7.
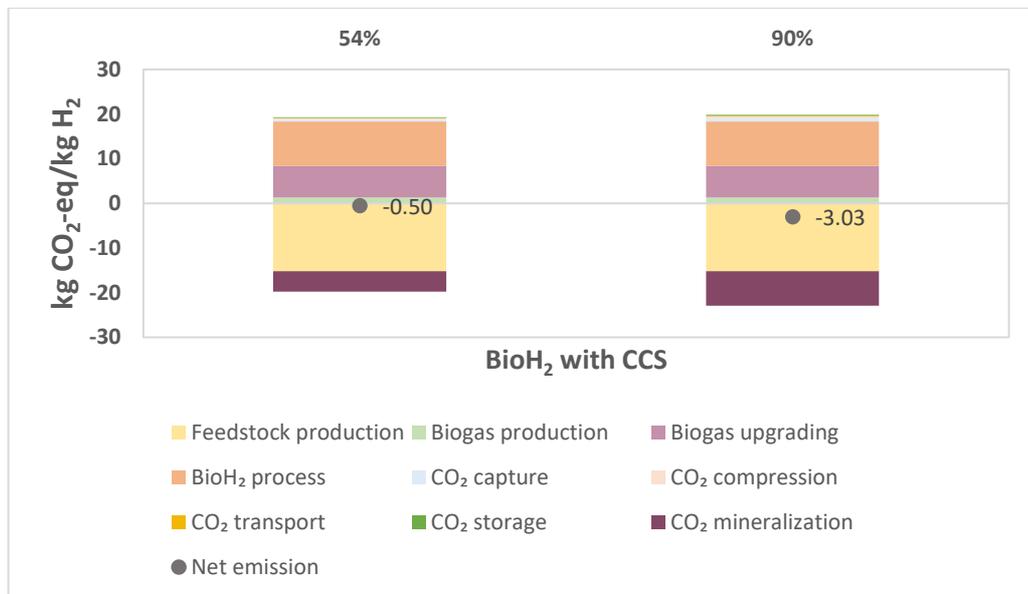

Figure 7. Systems sensitivity on CO$_2$ capture efficiency. By changing the capture efficiency in the bioH$_2$ production with CCS from 54% up to 90%.

By increasing the amount of CO_2 captured and avoiding fossil-based electricity the climate impact decreases drastically, see figure 8. The amount of CO_2 injected per kg H_2 produced increases to the negative emissions of 6.28 kg CO_2-eq./kg H_2.
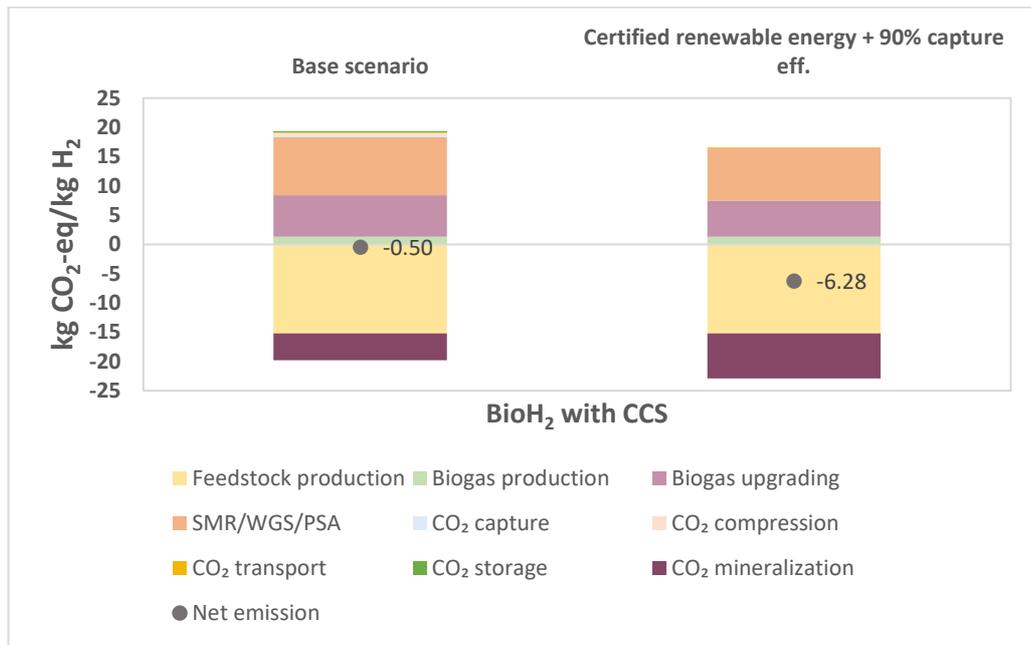

Figure 8. Systems sensitivity on both electricity source and capture efficiency. Changes made on bioH$_2$ production with CCS by using certified renewable energy sources and 90% capture efficiency of CO$_2$ from the bioH$_2$ process.

Figure 9 illustrates the system's sensitivity to the GWP time horizon, by changing the thesis GWP of 100 years to GWP of 20 years. The two processes that are affected in the system, are the biogenic CH_4 that is leaked from biogas production and the fossil CH_4 that is emitted from CO_2 transportation. Both bioH$_2$ productions, with and without the CCS, change significantly when the parameter is modified. Even when CCS is deployed, the GHG emissions increase to the point where the CO_2 mineralization is insufficient, where it no longer results in net negative emissions. This means that the system is highly sensitive to GWP's time horizon.
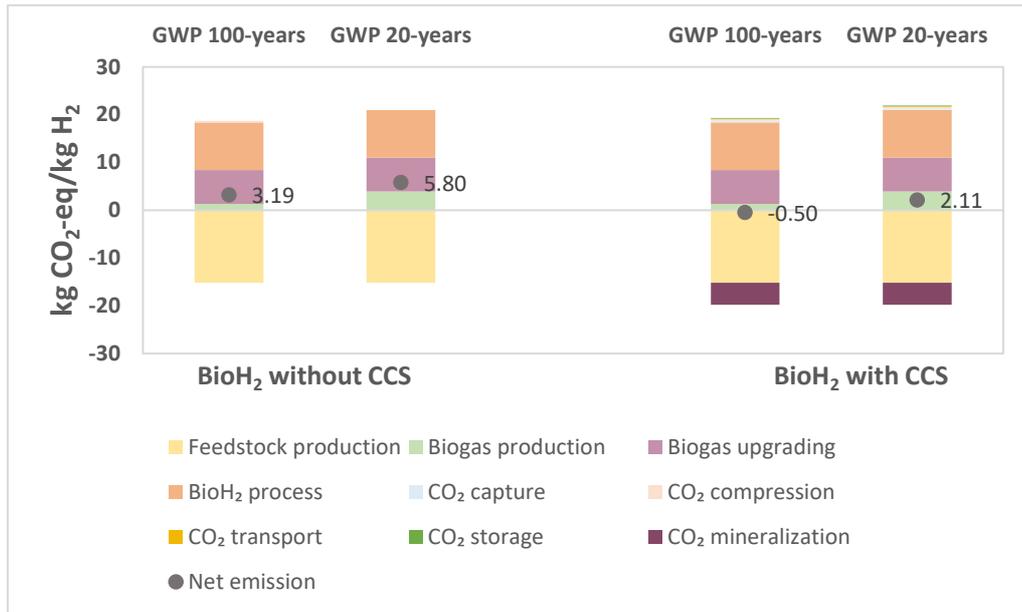
Figure 9. Systems sensitivity on GWP time horizon. Changes made by using 20 years instead of the thesis time horizon of 100 years.

Figure 10 demonstrates the climate impact when the scope of the CO_2 capture is expanded, so CO_2 is also captured from biogas upgrading, while also using certified renewable energy and maximizing CO_2 capture efficiency. The amount of CO_2-eq. mineralized per kg H_2 produced increases and the climate impact improves, from negative emissions of 0.50 kg CO_2-eq./kg H_2 to negative emissions of 12.47 kg CO_2-eq./kg H_2. This sensitivity analysis indicates how efficient the system can be when it is used to its greatest capacity.
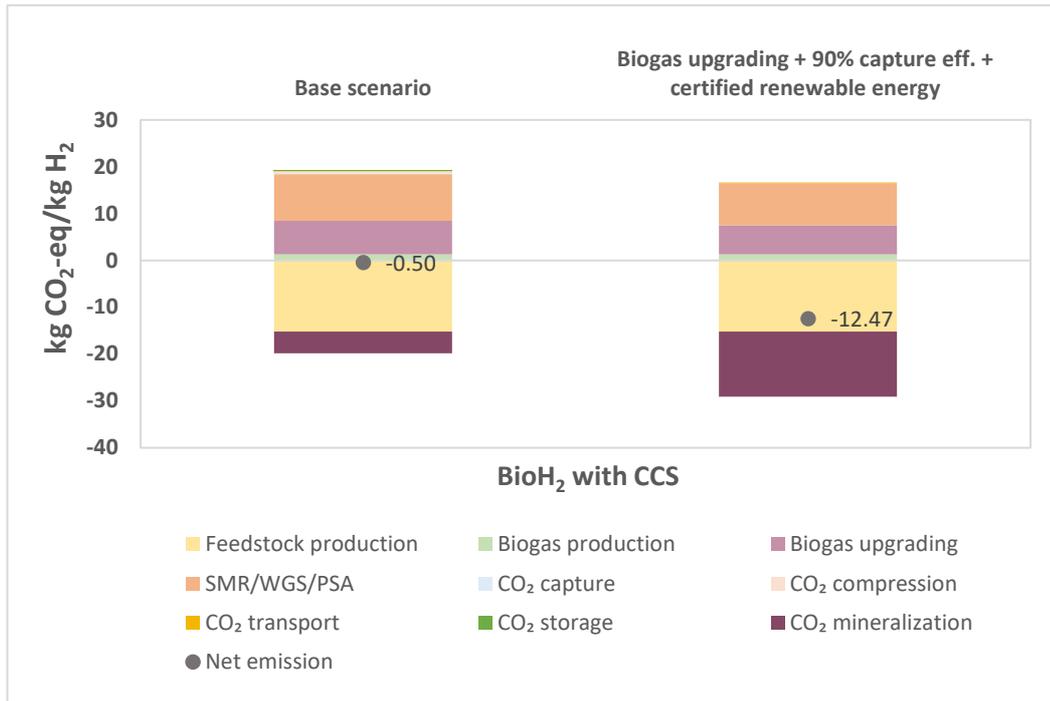
Figure 10. Systems sensitivity when the electricity source, capture efficiency, and CO₂ capture scope is changed. Changes made on bioH₂ production with CCS by using a certified renewable energy source, the capture efficiency of 90%, and CO₂ is additionally captured from biogas upgrading.

5. Discussion

To be able to meet the Paris agreement targets, technologies like CCS and H_2 production are crucial. This would be a good step toward decarbonizing the fossil fuel-based industry, where $bioH_2$ could potentially replace fossil fuels in many energy sectors.

The results of this thesis indicate that when producing $bioH_2$ using CO_2 mineralization for CCS the production can be climate neutral, or even reduce the climate impact from the production. The $bioH_2$ production has the potential to decrease climate impact even further in different scenarios. When the system is operated with certified renewable energy, all the process's input emissions are reduced, resulting in a greater net negative emission per kg H_2 produced. The CO_2 storage through mineral carbonation is solely limited by the efficiency of CO_2 that can be captured during $bioH_2$ production. The biggest system improvements happen when increasing CO_2 capture efficiency and scope along with using renewable electricity sources. It demonstrated how advantageous $bioH_2$ production with CCS could be if it is used to its maximum potential, where the climate impact was drastically reduced, resulting in significantly high net negative emissions.

However, a shift in CH_4 leakage can have a large effect on GHG emissions from $bioH_2$ production. If the leakage rate rises above what this thesis estimates, the CCS when producing $bioH_2$ is no longer beneficial since it no longer decreases the climate impact to net negative. The results also show that the GWP time horizon that is applied will impact the results significantly. The GWP of 20 years is sometimes used when calculating emissions from short-lifetime gases. Then it is based on energy absorbed over 20 years and does not count the impacts after 20 years from emission. Since CH_4 has the capacity to absorb substantially more energy than CO_2 gives, it will result in a higher GWP impact (Vallero 2019). The $bioH_2$ production with CCS will no longer have net negative emissions if the GWP time horizon is modified to 20 years.

The climate impact of $bioH_2$ production found in this study is higher than the impact reported in other studies of H_2 production. According to the Hydrogen

council report, the emissions of GHG from SMR using $bioCH_4$ from waste without the CCS is 1.0 kg CO_2-eq./kg H_2 (Using 2030 global average grid electricity) (Hydrogen council 2021). That is a significantly lower climate impact than the results of $bioH_2$ production without CCS in this study. That report also indicates that the SMR process using natural gas with CCS technology has the GHG emissions amount of 1.5 kg CO_2 eq./kg H_2 (The gas was transported over 1700 kilometers and using the conventional CCS method, but with a 75% capture rate) (Hydrogen council 2021).

Another study found that H_2 production using $bioCH_4$ as feedstock but gasification instead of SMR and without CCS technology resulted in -0.43 kg CO_2-eq./kg H_2 (Again using 2030 global average grid electricity) (Valente et al. 2020). That indicates that a method other than SMR for producing H_2 would possibly be more beneficial.

It is important to mention, as previously indicated, that CO_2 mineralization is geologically dependent. The rock formation must be right, and the method requires a large amount of freshwater. That could face potential usage conflict in areas where fresh water is limited. Sweden, however, currently has good access to freshwater resources compared to other countries in Europe, when the potential effects of climate change are not considered (Johansson 2020). As well as good access to renewable energy sources and biomass (Sveriges miljömål 2020; Anderson et al. 2022). Using seawater instead of freshwater could be an option. The use of seawater to dissolve CO_2 is being developed, although field site research has not yet been completed, but will be in 2022 (Voigt et al. 2021). Rock formations, other than basalt, also have the potential for mineral carbonation, although it is unknown which formations have high enough concentrations of the required cations for successful carbon mineralization (Snæbjörnsdóttir et al. 2022). Outside of Iceland, this technique of CO_2 mineralization has been utilized in a few projects, but not on a large scale (Blondes et al. 2018; Snæbjörnsdóttir et al. 2020; *Current operation* n.d.).

5.1.1 Uncertainties

All LCA studies have many assumptions which can lead to uncertainties in results. One uncertainty is pressurizing the CO_2 two times in the system, which could lead to inaccurate results. The pressure could be unrealistic and then electricity consumption to compress CO_2 at injection could be higher or lower, leading to different climate impacts and lower or greater freshwater consumption. Another significant uncertainty is that, while the Carbfix Atlas implies that CO_2 storage through mineral carbonation might be possible in Bäsingen or even Sweden, it does not mean it is 100% feasible. The Carbfix atlas is simply a first indicator of

geological viability; it overlooks other important aspects such as bedrock permeability.

6. Conclusion

The advantages of utilizing the carbon mineralization method in Sweden when producing $bioH_2$, are that Sweden has good access to freshwater and the production, and the use of biogas is common in the country. According to the Carbfix Atlas, more locations could potentially work for CO_2 mineralization throughout Sweden.

The CCS method of carbon mineralization is efficient and has a great potential to remove GHG emissions when producing $bioH_2$. The climate impact for producing 1 kg H_2 with the CO_2 mineralization method is -0.50 kg CO_2-eq., compared to the climate impact of the same $bioH_2$ production without carbon mineralization being 3.19 kg CO_2-eq./kg H_2. The climate impact is negative when using carbon mineralization due to CO_2 binding in biomasses feedstock and the injection of CO_2 into the basaltic rock. With the possibility of a higher reduction in climate impact when electricity is generated from renewable energy sources, as well as a larger scope of CO_2 capture and higher CO_2 capture efficiency, the net negative emissions of 12.47 CO_2 eq./kg H_2 can be achieved. The production is sensitive to changes in CH_4 leakage rate from the biogas process, as well as when the GWP time horizon is limited to 20 years, leading to higher GWP for biogenic CH_4. It becomes clear that, with the right approach, $bioH_2$ production using CO_2 mineralization for CCS could on a bigger scale and at a faster rate potentially contribute to climate change mitigation and help meet the target of the Paris Agreement.

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Appendix I

Biogas process: Feedstock production + biogas production + biogas upgrading

Value	Unit	Description	Source
30	GWh/year	Biogas plant output	Assumption
15.4	kWh/kg CH_4	The amount of CH_4 produced per kWh	Stolpe 2021
1948051.9	kg CH_4/year	The amount of CH_4 produced by the biogas plant per year	Calculation
5337.13	kg CH_4/day	The amount of CH_4 produced by the biogas plant per day	Calculation
0.717	kg/m^3	Density of CH_4	Engineering ToolBox 2003
7443.69	m^3 CH_4/day	The amount of CH_4 in the biogas	Calculation
60	%	Fraction of CH_4	Assumption
40	%	Fraction of CO_2	Assumption
4962.46	m^3 CO_2/day	The amount of CO_2 in the biogas	Calculation
12406.16	m^3 biogas/day	Biogas needed to produce bioCH_4	calculation
1.997	kg/m^3	Density of CO_2	Engineering ToolBox 2003
-15.19	kg CO_2/kg H_2	CO_2 uptake during feedstock production	Calculation
0.33	kWh/m3	Power consumption for biogas upgrading	Stolpe 2021

Value	Unit	Description	Source
1.4	%	Leakage rate of CH_4	Stolpe 2021
0.365	kg CO_2/kWh	Nordic residual mix emission factor	EI n.d

BioH$_2$ process: SMR+WGS+PSA

Value	Unit	Description	Source
9910.04	kg CO_2/day	CO_2 released from biogas upgrading	Calculation
6.19	kg CO_2/kg H_2	The amount of CO_2 released per kg H_2	Calculation
0.3	kg H_2/kg CH_4	The amount of H_2 that is produced based on 1 kg CH_4	Stolpe 2021
1601.14	kg H_2/day	The amount of H_2 produced per day	Calculation
2.7	kg CO_2/kg CH_4	Combustion relation of CO_2 from CH_4	Stolpe 2021
14410.25	kg CO_2/day	CO_2 released from SMR/WGS/PSA	Calculation
9	kg CO_2/kg H_2	The amount of CO_2 released per kg H_2	Calculation
2.6	kWh/kg H_2	Power consumption of SMR	Stolpe 2021

Carbon capture and storage

Value	Unit	Description	Source
54	%	Capture rate of CO_2	Timmerberget.al 2020
0.196	kWh/kg CO_2	Power consumption of CO_2 capture	Stolpe 2021
0.185	kWh/kg CO_2	Power consumption for CO_2	Janke et.al 2022
0.332	l/km	Fuel consumption for a diesel truck	Stolpe 2021
43.1	MJ/kg fuel	LHV diesel (5%RME)	Stolpe 2021
0.8	kg/l	Density of diesel	Stolpe 2021

208	km	Distance (roundtrip)	Stolpe 2021
0.00003	kg CH_4/MJ	Emission factor from production, distribution, and usage of diesel with 5% RME	Stolpe 2021
0.076	kg CO_2/MJ	Emission factor from production, distribution, and usage of diesel with 5% RME	Stolpe 2021
0.0000022	kg NO_2/MJ	Emission factor from production, distribution, and usage of diesel with 5% RME	Stolpe 2021
18	Tonnes/truck	Size of truck	Viktbestämmelser 2021
0.075	kWh/kg CO_2	Power consumption for CO_2 mineralization	Snæbjornsdóttir 2022
95	%	CO_2 mineralization rate	Matter et al. 2016
27	Tonnes	Amount of water to fully dissolve one tonne of CO_2	Snæbjornsdottir et. al 2014
4.55	kWh	Electricity consumption of 1 ML lifted 1 meter of height	Cotton info 2015
15	meters	Height of water pump	Assumption

Appendix II

Questions asked Sandra Snæbjörnsdóttir at Carbfix via Email (Snæbjörnsdottir 2022):

1. Is it correct that the amount of CO_2 injected every day is 32.88 tonnes CO_2 or 12000 tonnes CO_2 per year, as stated on your website? Can I assume that injecting 15 tonnes of CO_2 per day could be possible in my case?

2. I was also curious about the amount of electricity required to inject 15 tonnes of CO_2 every day. According to the Carbfix website, the main energy demand for Carbfix technology is to pressurize the CO_2 charged water, which is 75 kWh for 1 tonne of CO_2 charged water. Is this electricity amount a standard? Is it safe to assume that the 15 tonnes of CO_2 injected per day will require the same amount of electricity per tonne?

3. Does the process require any thermal energy, or does it only operate on electricity? If so, how much thermal energy would the injection of 15 tonnes of CO_2 require every day?

4. Regarding the water consumption, according to Snæbjörnsdóttir the water demand to fully dissolve one tonne of CO_2 at 25 bar pressure and 25°C is 27 tonnes of pure water. Is it reasonable to assume that this can also apply to my case study?

5. Could you inform me what the estimated capital expenses (CAPEX) and operating expenditures (OPEX) for a 15 tonne CO_2 injection per day could be?