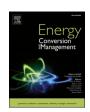
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Life cycle assessment and life cycle costing of hydrogen production from biowaste and biomass in Sweden

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ABSTRACT

In this study, an environmental and economic assessment of hydrogen production from biowaste and biomass is performed from a life cycle perspective, with a high degree of primary life cycle inventory data on materials, energy, and investment flows. Using SimaPro LCA software and CML-IA, 2001 impact assessment method, ten environmental impact categories are analyzed for environmental analysis. Economic analysis includes capital and operational expenditures and monetization cost of life cycle environmental impacts. The hydrogen production from biowaste has a high climate impact, photochemical oxidant, and freshwater eutrophication than biomass while it performs far better in ozone depletion, terrestrial ecotoxicity, abiotic depletion-fossil, abiotic depletion, human toxicity, and freshwater ecotoxicity. The sensitivity analysis of LCA results indicates that feedstock to biogas/pyrolysis-oil yields ratio and the type of energy source for the reforming process can significantly influence the results, particularly climate change, abiotic depletion, and human toxicity. The life cycle cost (LCC) of 1 kg hydrogen production has been accounted as 0.45–2.76 ϵ with biowaste and 0.54–3.31 ϵ with biomass over the plant's lifetime of 20 years. From the environmental impacts of climate change, photochemical oxidant, and freshwater eutrophication hydrogen production from biomass is a better option than biowaste while from other included impact categories and LCC perspectives it's biowaste. This research contributes to bioresources to hydrogen literature with some new findings that can be generalized in Europe and even globally as it is in line with and endorse existing theoretical and simulation software-based studies.

1. Introduction

Bioresources are considered promising for different renewable energy carrier production [1–3] such as liquid biofuels (e.g., biodiesel, biogasoline, and ethanol), hydrogen, and biogas [4–7]. Bioresources based hydrogen (biohydrogen) is viewed as a clean fuel of high energy density, 120 to 142 MJ/kg, with suitability across a range of energy and industrial applications e.g., as a fuel in the steel industry, as a feedstock for fuel cells mobility and as a feed source for ammonia, etc. [8,9]. Thus, from these many various aspects, biohydrogen can be a way forward for a more sustainable transformation of energy systems. However, this transformation also poses some safety challenges related to hydrogen production, storage, transportation, and utilization technologies [10].

As of 2021, approximately 94 million tons (Mt) of hydrogen was produced globally. Of this, 47 % was generated from natural gas (NG), 27 % from coal, 22 % from oil as a byproduct, 3.7 % from water-electrolysis, and<1 % from bioresources [11]. The use of fossil-based hydrogen has raised environmental concerns, which can be addressed

by the development of bioresources-based hydrogen technologies [12]. However, despite the advancement of bioresources-to-hydrogen and other transport fuel conversion technologies, there are still challenges regarding process optimization and scale-up [13]. As such, the sustainability of these routes, particularly in terms of economic and environmental factors, may vary depending on the substrate type, processing technology, energy source, and end-use. These aspects are typically assessed from a system perspective, using tools such as life cycle assessment (LCA) and life cycle cost assessment (LCA) [14].

Several studies have assessed the environmental sustainability of various biohydrogen production pathways [1,3,15–20]. For example, the study [16] examines five waste-to-hydrogen technologies and highlights the need for further research and development to address challenges linked to environmental sustainability and cost. Wulf and Kaltschmitt [21] analyzed six different hydrogen production pathways (i.e., wood gasification, vegetable oil transesterification, coal gasification, fossil methane reforming, and electrolysis with wind electricity and conventional electricity mix) considering global warming potential.

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They conclude that hydrogen produced via electrolysis with green electricity and biomass gasification has the least global warming potential compared to other routes. The study [18] finds that biogas reforming is a sustainable and eco-friendly route of hydrogen production compared to fossil-sourced feedstocks.

Further, the literature on LCA and techno-economic analysis of hydrogen production has primarily focused on biomass gasification and biogas reforming routes. For instance, Kalinci et al. [22] report LCA and LCC of hydrogen production from pine wood with two different gasification reactors considering LCI data from the literature and conclude biomass pre-treatment, hydrogen compression, and transportation as energy-intensive processes in both studied systems. Fossil energy consumption rate differs with the types of processing technology while the capital investment effectiveness is influenced collectively by the amount of indirect energy utilization, and fossil-to-renewable energy output ratio. Similarly, an LCA study [21] recommends renewable sources like wind and biomass for climate-friendly hydrogen fuel compared to fossilsourced hydrogen. A recent study [22], using the Chinese Life Cycle Database, assesses the environmental impact of three hydrogen production processes for biofuel from wheat straw via fast pyrolysis, noting that production system configurations and energy source selection could significantly affect the results.

Furthermore, a LCA of hydrogen production from bio-oil steam reforming and glycerol derived via fast pyrolysis of popular biomass and rapeseed respectively has concluded that the former method is better than the latter in terms of both environmental impact and energy efficiency [23]. The processes were simulated in Aspen Plus to obtain inventory data. In a recent LCA study by Zheng et al., the conversion of bio-oil obtained through fast pyrolysis of wheat straw into hydrogen, biodiesel, and gasoline was investigated using Aspen Plus software. The study's findings indicate that utilizing the bio-oil to produce biodiesel and biogasoline, in combination with an aqueous portion of the bio-oil for hydrogen production, results in significantly lower impacts of primary energy depletion (PED), global warming potential (GWP), and abiotic depletion potential (ADP) compared to fossil fuels, with reductions of 89.81 %, 117.44 %, and 1.74 %, respectively [24].

From an economic perspective, several techno-economic studies [6,25-31] have reported the cost of hydrogen production with a range of feedstocks, production technologies, and end-use. Sanchez et al. [25] examined the overall cost of hydrogen production from wheat straw in advanced biorefineries, utilizing Aspen Plus software. The results showed that the production cost of hydrogen from wheat straw gasification was lower than that of traditional methods, such as coal gasification and methane reforming. This was particularly true when accounting for the cost and credits associated with environmental emissions. Overall, the study suggests that hydrogen production from wheat straw has significant potential as a cost-effective and environmentally friendly alternative to conventional production methods. Brown et al. [26] estimated the levelised cost of H2 production from wood chips gasification on an energy basis with inventory data from twelve different studies and reported it as \$25.06- \$40.21 per GJ. The study concludes that producing hydrogen via biomass gasification and Fischer-Tropsch synthesis could be more cost-effective than from synthetic gasoline and diesel. In a study by Wang et al., using Aspen Plus, the cost of hydrogen production from wheat straw and coal gasification was analyzed. The study found that when carbon taxes were introduced, the production of hydrogen from biomass was cheaper, with a cost of $$0.09/m^3$ compared to the cost of $$0.11/m^3$ for coal gasification [29]. Further, a recent study [9] used the lifecycle monetization concept where endpoint LCA emissions damage costs are added to the levelised cost of hydrogen production from steam methane reforming (SMR), coal, and biomass gasification, methane pyrolysis, and electrolysis powered by wind, solar and nuclear energy with LCI from literature. They found that steam methane reforming with carbon capture and storage has the lowest monetize cost followed by methane pyrolysis and water electrolysis powered by wind and nuclear.

The above literature review indicates that studies on the environmental and economic assessment of hydrogen production have focused on the ranges and variations of techno-economics and environmental indicators. Biomass gasification has been studied with different reactor designs and feedstocks. Even though biomass gasification is a common route for hydrogen, and globally several biomass gasification plants of sizes 6 MW are in operation with notable successes [32], still, the direct biomass gasification technology is confronting some technical issues (primarily tar caused by inorganic compounds) which is not the case with fast pyrolysis as most of the inorganic elements are retained in the char which is combusted normally in the large-scale pyrolysis plants and the residual ash is easily separated. This makes biomass-fast pyrolysis technically less complicated and more flexible to feedstock and thus analyzed in this paper. Further, this route is assumed will grow in Sweden as the development of direct biomass gasification has been slowed after the shutdown of the GobiGas 20 MW [33,34] biomass gasification plant in 2018.

There is hardly any LCA and LCC study has been found that covers sawdust biomass for hydrogen production via fast pyrolysis and monetized cost of life cycle environmental impacts to LCC. Further, in the published studies of LCA and LCC of hydrogen production, results rely on a high degree of secondary LCI data obtained either from simulation software (i.e., Aspen plus, GREET, etc.), lab experiments, or literature. Additionally, most of those studies lack sensitivity analysis. This led to that previous work ending up with varying conclusions on the most sustainable routes of hydrogen production. Additionally, the studies ask for caution in generalizing their findings as they depend on several technical, market, and geographic variable parameters such as the source of energy supply, feedstock characteristics, plant size, operational costs, etc. In such a situation, case studies with primary life cycle inventory (LCI) and detailed results sensitivity analysis can considerably lower the uncertainty and lead to more realistic and robust conclusions.

Thus, the objective of this study is to contribute to the above-highlighted knowledge gap with detailed environmental and economic performance analysis of hydrogen production from biomass which is sawdust representing forest industry residue and biowaste obtained from municipal solid waste from a life cycle perspective, and to identify where the improvement efforts should focus to make the systems more efficient. This is done using primary LCI data on materials, energy, and investment flows, and the LCC model includes the monetized cost of environmental impacts. The results of this study can work as a good example for other Swedish regions and similar regions in Europe. The results shall also contribute to the literature on the inventory of LCA and LCC of hydrogen production from renewable resources as this study to a high degree uses primary LCI and introduces environmental emissions damage remediation cost to LCC analysis.

2. Methodology

The study employs a gate-to-gate Life Cycle Assessment (LCA) and Life Cycle Costing (LCC) methodology, which is guided by the ISO 14040–44 [35,36] and ISO15686-5 [37] standards, respectively.

ISO 15686–5:2017 defines Life Cycle Cost (LCC) as the total cost or cash flow incurred over the entire life cycle or selected stages for products, services, or assets under analysis. LCC considers both financial costs and externalities costs, including environmental costs. The financial cost can be divided into capital investment expenditures (CAPEX) and operational expenditures (OPEX), whereas the environmental cost includes economic loss of well-being (e.g., human health and biodiversity) due to an impact as well as the mitigation cost of any necessary intervention [38,39]. In this study, to be consistent with the LCA system boundary, a steady-state model of life cycle cost assessment was

 $^{^{\}rm 1}$ Personal communication with Pyrocell AB represented, Katarina Persson (Dated: 2021–01-20).

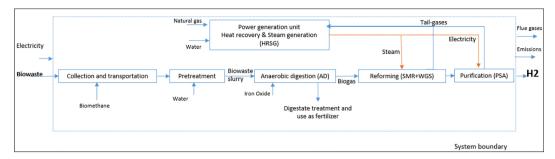


Fig. 1. System boundary for biowaste to hydrogen (BwH2).

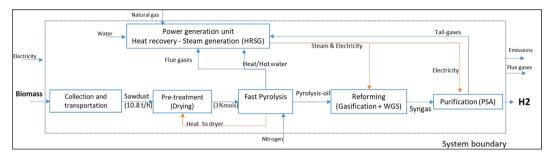


Fig. 2. System boundary for biomass to hydrogen (BmH2).

employed which includes financial and environmental costs. Hydrogen storage is not included as the production rate is the same in both cases and it is assumed that the LCA and LCC of hydrogen storage would be the same. By excluding hydrogen storage, the study focuses specifically on the production of hydrogen and allows for a more detailed analysis of the environmental and economic performance of the production process itself.

The choice of the assessment methods and technology routes was motivated by the specific relevance in the perspective of the hydrogen production technology mix, particularly at the regional but also at Swedish national levels. The rationale is further discussed in section 2.1.1.

2.1. Goal and scope definitions

This study aims to evaluate and compare the environmental and economic performance of hydrogen production from two partially established waste management/utilization systems in a northern region (Gävleborg) of Sweden from a life cycle perspective (gate-to-gate). These two systems refer to hydrogen production from biowaste and biomass. The goal is to identify the hotspots where improvement efforts should be focused to make the systems more efficient from both environmental and economic points of view. The functional unit is 1 kg of hydrogen produced with a purity level of 99.99 % at standard temperature and pressure. The system boundary of BwH2 is presented in Fig. 1 while that of BmH2 in Fig. 2.

2.1.1. System description: biowaste to hydrogen

Biowaste to hydrogen (BwH2): the case study plant is the Ekogas Forsbacka biogas plant, in Sweden (Gävleborg region). The plant uses solid-state anaerobic digestion technology to treat the biowaste of proximate analysis given in Appendix Table 2, with iron-hydro oxide as additives, ² and produces biogas at the rate of 9700 m³/day. Using biogas

as a resource for green hydrogen and electromobility is on the agenda of the producers and the local authorities.³ Data inventory for the biogas production system is based on the information from the plant manager whereas the inventory on biogas reforming is based on [18,40]. Major processing steps include biowaste collection and transport, pretreatment, anaerobic digestion, biogas reforming, and purification of the produced hydrogen. A schematic of biowaste to hydrogen is shown in Fig. 1. The biowaste is transported on average 50 km by waste collection trucks (fueled with biomethane), then shredded in the plant, and heated to 40–45 °C in the pretreatment process. During this process, plastic and other unwanted materials are removed and water (4.36 L/ton waste) is added, and then pumped to the AD tank operated at 57 °C to produce biogas of that composition is given in Appendix Table 1.

The digestate from the AD tank is treated and separated into liquid and solid parts. The solid part is stored on-site while the liquid is in the satellite-monitored sealed tank at farmer's sites. The liquid part is transported on average 20–30 km for use as biofertilizers, while the solid part 10–15 km as a soil layer in parks. The produced raw biogas is desulfurized via fixed bed activated carbon and ZnO beds to avoid catalyst poisoning before reforming for hydrogen production. ⁴ For the steam reforming, high-temperature (700–800 °C) steam produced at heat recovery and steam generation (HRSG) unit is fed to the reformer to convert biogas methane to $\rm H_2$.

After SMR, the produced syngas is cooled and goes through a water gas shift reaction (WGS) where CO reacts with water to increase H_2 yield (Eq.A2). The biogas reforming process parameters (temperature, pressure, steam to methane ratio) were assumed like [18] and adjusted to present case biogas composition for the calculation of process inputs (energy, steam to biogas ratio) and outputs (e.g., the conversion ratio of biogas to H_2 and biogenic CO_2 emissions). Subsequently, the syngas stream is condensed and fed to the PSA unit to separate H_2 . Whereas condensate is reused for steam generation and tail gases consisting of H_2 (around 1 %, considering PSA efficiency 99 %) and CH_4 (around 14 %,

 $^{^{2}}$ Personal communication with the plant manager, Nyquist Henrik [dated: 2022–06-20].

 $^{^{3}}$ Personal communication with the plant manager, Nyquist Henrik [dated: 2022–06-20].

⁴ Biogas reforming for hydrogen production is modeled based on [18,40] as the facility does not exist on site.

Table 1 Summary of life cycle inventory inputs per FU (1 kg H_2). The values were calculated based on yearly average data for the year 2021.

Life cycle stage	Unit	Inputs	Inputs	Data source
		BwH2	BmH2	
Collection and transportation				
Biowaste (food waste 86 %, green waste 12 %, and food slurry 2 %)	kg	197		Ekogas AB
Biomass -moisture 55 %wt	kg		46	BtG-bioliquids
Biomethane-as transport fuel	MJ	2.48		Ekogas AB
Electricity	kWh		0.06	Ekogas AB
Pretreatment				
Biowaste	kg	197		Ekogas AB
Biomass	kg		46	BtG-bioliquids
Water	Liter	0.0856		Ekogas AB
Electricity	kWh	0.153		Ekogas AB
Heat	MJ	1.65	N/A	Ekogas AB
AD/pyrolysis				
Biowaste slurry	kg	198		Ekogas AB
Biomass (3 %moisture)	kg		24	BtG-bioliquids
Additives/catalyst-Iron Oxide	kg	0.02		Ekogas AB
Nitrogen gas	kg		0.43	BtG-bioliquids
Electricity	kWh	0.451	9.31	Ekogas AB, BtG
Heat	Mj	3.661		BtG-bioliquids
Diesel-transport of digestate	Mj	5.52		Ekogas AB
Reforming				
Biogas	kg	3.79		[a]
Pyrolysis-oil	kg		14.4	[b]
Water (as steam)	kg	5.91	5.40	[a, b]
Electricity	kWh	1.25	5.96	[a, b]
Natural gas	MJ	20.4	17.0	[a, b]
Purification				
Syngas (assuming density 0.95 kg/m ³)	kg	9.6	13.70	[a, b]
Electricity	kWh	0.827	0.82	[c]
Residues				
Liquid digestate	kg	15.89		Ekogas AB
Solid digestate	kg	3.972		Ekogas AB

Ekogas AB- calculated based on information from the plant manager (Henrik Nyquist).

BTG-bioliquids- calculated based on information from the technology manager (Ardy Toussaint).

Note: Heat exchange efficiency has been taken as 70% in all calculations where heat exchange occurs from one medium to another. Swedish electricity mix (medium voltage) is considered for all cases.

considering reformer efficiency around 86 %) are directed to the power generation. Some assumptions taken in the LCA of BwH2 are:

- 1. The biowaste is collected and transported with municipal solid waste and thus only 20 % of the resources (biomethane) used in this process are allocated to biowaste collection and transportation. The allocation is based on annual fuel consumption.
- 2. Uncontrolled methane leakage from the biogas plant is estimated to be $2\,\%$ of the produced biogas and the methane concentration is taken to $53\,\%$.
- Liquid digestate is used as a biofertilizer with N, P, and K equal to 6.6, 0.6, and 2.4 kg/ton⁵ respectively, assumed to replace mineral fertilizer that is produced in Europe and transported to Sweden by ship.
- 4. The emissions of nitrogen compounds (NH_3 , N_2O , NO_x , and NO_3), phosphate, and heavy metals from the use of biofertilizer in agricultural practices are estimated with the AGRAMMON model presented in [41]. Details are given in Appendix A.2. Both upstream and downstream emissions linked to biofertilizer production and use are considered.
- 5. The emissions of ammonia and methane from the storage of digestate were considered the same as in [42] which are 4.465 and 0.115 kg/t AD feedstock, respectively. Further, ammonia release during the

- 6. The efficiency of the reformer unit has been considered 86 % while the hydrogen purification (PSA) process to 99 % [18].
- 7. Considering current biogas production (9700 m^3 /day) of the Forsbacka biogas plant, a H_2 production unit of a capacity of 40,000 m^3H_2 /day is assumed.

2.1.2. System description – biomass to hydrogen

Biomass to hydrogen (BmH2): the case study plant is the pyrolysis oil production facility located at the Setra sawmill in Gävle, Sweden where biomass which is sawdust produced at the adjacent sawmill is used for pyrolysis oil production. The ultimate analysis of the considered biomass and produced pyrolysis oil is given in Appendix Table 3. The plant technology is fast pyrolysis developed by the BtG-biofuels⁶ and produces pyrolysis oil at the rate of 85 tons/day. Currently, the produced pyrolysis oil is transported to a refinery in Lysekil, Sweden, to produce biodiesel and biogasoline, but its use as a resource for green hydrogen is foreseen.⁷ Data inventory for the pyrolysis oil production is

[dated: 2022-06-20].

 $^{\rm 6}\,$ The company provided the technology plant to Pyrocell AB. The company is

[[]a]-In BwH2- calculated bed on the information from [18].

[[]b]-In BmH2- calculated based on the information from [32].

[[]c]-In both cases- values were calculated based on the information from [43].

spreading process was assumed 0.677 kg/t AD feedstock [18]. For details on the calculation, and procedure see Appendix $\rm A.2$

AD feedstock, respectively. Further, ammonia release during the

a biomass-fast pyrolysis technology developer and consulting firm based in the Netherlands (https://www.btg-bioliquids.com).

⁷ Personal communication with Preem Lysekil Business development engi-

neer [Dated: 2021–01-20].

⁵ Based on personal communication with the plant manager, Nyquist Henrik

Table 2Overview of the investment costs used for CAPEX calculation.

Equipment	Value	Unit	Source
Pyrolysis plant (3.6-ton oil/hr)	40.0	MEUR	BtG-bioliquids
Biogas plant (405 m ³ /hr)	20.0°	MEUR	Ekogas AB
Biogas reformer unit (405 m ³ biogas/hr)	5.26 ^a	MEUR	[a]
Pyrolysis oil gasification unit (3.6-ton oil/hr)	2.22	MEUR	[b]
PSA unit (6.33 m ³ /hr)	1.76 ^b	MEUR	[a]

Ekogas AB- Information provided by the plant manager (Henrik Nyquist).

-bioliquids- information provided by the company managing director (Gerhard Muggen).

[a]: calculated based on the information from [40].

[b]: calculated based on the information from [51] and adjusted according to Eq.2.2.

- ^a Includes the steam reformer, the WGS unit, the compressor, the PSA unit, and the steam generation system.
- ^b Includes the compressor and PSA units.
- c including desulphurization.

based on the information from the case study company, Pyrocell AB,⁸ and the plant technology provider, BtG-bioliquids.⁹ Whereas inventory for pyrolysis oil reforming for hydrogen production is based on [32,40]. A schematic flow of biomass to hydrogen is shown in Fig. 2.

The Setra sawmill produces high-quality plank from the regional forest pine trees with bark, woodchips, and sawdust as by-products. In this study, only the energy (heat and electricity) used in timber processing (i.e., barking, sawing, sorting, drying, and grinding) at the sawmill is allocated to biomass used on a mass basis. In Fig. 2 this process is referred to as collection and transportation.

According to BtG-bioliquids, the biomass sized (<1mm) is fed into the dryer at the rate of 10.8 tons per hour and dried up to 3 % moisture content. The heat from the biochar combustion unit is used in the drying process. The dried biomass (3 % moisture) is then fed to the fast pyrolysis unit (which operates at temperatures 700–800 °C) at a rate of 5.0 tons per hour where it is mixed and heated with hot sand in a rotating cone reactor in the absence of oxygen, resulting pyrolysis vaporous and char. Sand, char, and inorganic compound particles are removed from the vaporous gas in separators and cyclones. Pyrolysis vapors gas is condensed in a condenser which results in pyrolysis oil at the rate of 3.35 tons per hour. Separated Sand is sent back to the Pyrolyzer while char and non-condensable gases are burnt internally in the fluidized bed combustor to deliver heat and electricity to the pyrolysis plant. However, additional electricity is supplied to the plant from the national electricity grid.

Here, like BwH2, it is assumed that the pyrolysis plant is integrated with the hydrogen production facility where the pyrolysis oil is gasified in an entrained flow gasifier powered with a Swedish electricity mix to produce syngas. The gasification operating parameters (pressure, temperature, residual time, etc.) are assumed as in [32]. The produced syngas goes through catalytic steam reforming where a nickel-based catalyst is used. After that, the gas stream is cooled and goes through water gas shift reaction processes (low and high temperatures) to further increase the yield of hydrogen. For syngas reforming, high temperature (700-800 °C) steam is delivered by the heat recovery and steam generation (HRSG) unit where, like BwH2, natural gas was assumed burnt together with tail gases. After reforming, the gas stream is cooled and fed to the PSA unit to separate hydrogen which is assumed to be stored at standard conditions. The unconverted methane (around 14 %, assuming 86 % conversion efficiency of the reformer) in the syngas and hydrogen (around 1 %) are directed to the power production unit. Some assumptions taken in the LCA of BmH2 are.

- In BmH2, energy and materials input to forest trees cultivation and harvesting are not included as the considered biomass is assumed a waste resource of the plank production process. Only sawmill energy use is considered where allocation is made on a mass basis.
- 2. All the char produced is combusted internally to produce heat and electricity which is used to run the plant.
- The recovered heat from the combustion of tail gases in the HRSG unit was assumed to replace natural gas.
- 4. It is assumed that the compressed nitrogen (purity 98 %) used in the pyrolysis process has been transported 3,000 km by ship.
- 5. Based on the current pyrolysis oil production (85 tons/day), a $\rm H_2$ production unit of capacity of 40,000 $\rm m^3H_2/day$ at normal temperature and pressure was assumed.

2.2. Life cycle inventory (LCI)

The main inputs used for environmental and economic assessment in the biogas and pyrolysis oil production cases are shown in Table 1. The case study plant managers provided data on mass, energy, fuels, and chemical flows. However, there was a lack of primary data on biogas and pyrolysis oil reforming, so the data was calculated based on previous studies [18,40]. Data on the purification process was obtained from a separate study conducted by [43].

The outputs, such as emissions to water, air, and soil, were generated using Simapro LCA software version 9.13 with a predefined dataset Ecoinvent 3.8. A detailed LCI of emissions in both cases can be found in Appendix Table A–4.

2.3. Life cycle impact assessment (LCIA)

In this study, for LCIA CML-IA, 2001, method, developed by the Institute of Environmental Sciences, Leiden University, the Netherlands [44] is applied to evaluate the mid-point environmental impacts. Simapro V9.3.0.3 software with Ecoinvent database V3.0 as a library for a background data source is used. Several LCA of hydrogen production studies e.g., [18,45,46] have used CML-IA and therefore CML-IA was chosen to compare this study results with published literature. However, to test the sensitivity of the results to impact assessment method, ReCiPe is selected for sensitivity scenario S3(4.3).

The most common mid-point categories, global warming potential (GWP), freshwater eutrophication potential (EP), terrestrial acidification potential (AP), ozone layer depletion potential (ODP), photochemical oxidant formation (POP), terrestrial ecotoxicity potential (TEP), human toxicity potential (HTP), fresh water ecotoxicity potential (FEP), Abiotic depletion potential (ADP) and abiotic resources depletion potential (ADP-fossil) [44] are included. The impact assessment results excluding infrastructure and long-term emissions are explained in detail in each impact category.

⁸ Personal communication with Pontus Friberg, Chairman of the Board of Pyrocell AB [2022–02-02].

⁹ Personal communication with BtG-biofuels technology manager, Ardy Toussaint [Dated: 2022–02-02].

Table 3The impact categories and unit prices used for environmental cost calculation. The environmental cost unit values are taken from [52].

Impact categories	Unit price (ϵ)
Climate change (kg CO _{2 eq})	0.06
Ozone depletion (kg CFC-11 _{eq})	30.00
Terrestrial acidification (kg SO _{2 eq})	5.00
Freshwater eutrophication (kg P eq)	1.90
Human toxicity (kg 1,4-DB _{eq})	0.10
Photochemical oxidant formation (kg NMVOC)	1.20
Terrestrial ecotoxicity (kg 1,4-DB _{eq})	8.70
Freshwater ecotoxicity (kg 1,4-DB eq)	0.04
Abiotic depletion (kg fossil oil _{eq})	NA

Table 4
Sensitivity analysis scenarios.

Scenarios	Description
S0	Base case scenario i.e., BwH2 and BmH2
S1	Feedstock to biogas and pyrolysis oil yields decreased by 25 %
S2	Wood chips as fuel instead of natural gas in reforming
S3	The impact assessment method changed to ReCipe 2016

2.4. Life cycle cost assessment (LCC)

As described in the methodology (section 2.1), the LCC assessment includes financial and environmental costs following ISO 15686–5:2017 guidelines. Equation (2.1) was used to estimate the average yearly LCC of hydrogen production. Based on techno-economic studies [16,40,47,48], it is assumed that the plants will operate for 8,000 h annually and have an operational lifespan (n) of 20 years. A straight-line discounting approach was implemented with a discount rate (r) of 10 % [49,501]

$$CC_{hydrogen} = \sum_{n=0}^{n=20} \left(\frac{CAPEX + OPEX}{(1+r)^n} + Environmental \cos t \right)$$

$$\div Hydrogen \ produced$$
2.1

2.4.1. Capital investment cost (CAPEX)

The capital investment costs for the biogas plant (BwH2) and pyrolysis oil plant (BmH2) were obtained from the respective company. Whereas, investment costs for pyrolysis oil gasification, reforming, and PSA units were estimated based on [40,51] and adjusted using order-of-magnitude estimates and capacity rationing according to Eq.2.2 with scaling factor "m" equal to 0.8 to consider the whole plant [40]. Table 2 gives an overview of the investment costs of plants and equipment used for the calculation of CAPEX.

$$\frac{Capital1}{Capital2} = \left(\frac{Capacity1}{Capacity2}\right)^{m}$$
2.2

2.4.2. Operational costs (OPEX)

Operational costs consist of fixed and variable costs. Fixed costs include labor, repair and maintenance, insurance, laboratory, and plant overhead while variable costs include raw materials, utilities, and waste management costs. Equation (2.3) was used for OPEX calculation. Appendix Table B.5 gives an overview of the variables and their price values used for OPEX calculation. The fixed costs were calculated based on the estimates provided by the case companies whereas variable cost was calculated based on the actual 2021 average prices of materials, energy, and waste streams according to the amounts as calculated for LCA. The prices of electricity, heat, fuels, and water were assumed same in both base cases as both case study plants were operating in the same region.

$$OPEX = C_R + C_U + C_{R\&M} + C_{OL} + C_{EB} + C_{LAB} + C_{I\&T} + C_{POH} - R_{DS}$$
 2.3

In equation (2.3), C_R is for raw materials cost, C_U is utility cost, $C_{R\&M}$ is for repair and maintenance cost, C_{OL} is operating labor cost, C_{EB} is the cost of employee's benefits, C_{LAB} is lab running cost, $C_{I\&T}$ is for insurance and taxes, C_{POH} is for plants overhead cost. Whereas R_{SD} is for revenue from byproducts of the systems.

2.4.3. Environmental costs

For the environmental cost calculation, the absolute values of the impact categories were multiplied by their respective environmental unit price (Table 3). These unit prices indicate the loss of welfare due to the emission of one additional kilogram of pollutant to the environment and are set to use at the level of the EU28 as monetary weighting factors [52] to normalize the LCA impacts into a comparable common unit, in this study, euro (\mathfrak{E}).

3. Results and discussion

To increase transparency, a breakdown of the results based on the contributions from different compartments, including feedstock collection, biogas/pyrolysis-oil production, reforming, and purification is provided. In addition, sensitivity analysis is conducted to improve the generalization of the findings. These sensitivity scenarios are presented in Table 4.

3.1. Life cycle environmental impacts

The LCA results are presented in Fig. 3, Fig. 4, and Fig. 5, where each environmental category is plotted with its absolute value per 1 kg of hydrogen produced. A negative bar represents environmental benefit whereas a positive bar indicates the environmental burden of that impact.

The global warming potential of BwH2 (Fig. 3a) is 28 % higher than that of BmH2 where AD accounts for 73 % reforming 25 % and others 2 %. While in the BmH2 case, reforming accounts for 87 %, pyrolysis 8 %, and other processes 5 %. In the 73 % global warming potential of AD, 96 % links to process byproduct digestate treatment and use as biofertilizer attributed to energy inputs to the digestate treatment, transport, and emissions from its storage and spreading on arable land. Looking at the life cycle emissions inventory (Appendix Table B.4) almost 60-70 % of the impact is attributed to direct air emissions of CO2 (2.6 kg), fossil methane (7 g), Ammonia (0,3 mg), etc., sourced to the production and use of transport fuel (diesel), reforming fuel (natural gas) and ammonia from the anaerobic digestion tank and digestate storage (i.e., 4.4 kg/ton AD feedstock). Fast pyrolysis of biomass is reported as a highly energyintensive process [32,53] but this study (BmH2) shows a low contribution (14 %) of the process to the overall global warming of the system (BmH2). This is attributed to the self-energy sufficiency of the process generated through biochar combustion. However, reforming of pyrolysis oil accounts for 85 % attributed to higher energy input to pyrolysis oil reforming compared to biogas reforming.

In both cases, the global warming potential of the reforming process is sourced from natural gas production and combustion used to produce high-temperature steam for reforming. The global warming potential emissions inventory of reforming is mainly attributed to the direct air emissions of fossil CO_2 (277 g) and methane (1.45 g) in the case of BmH2 and direct air emission of CO_2 (144 g) and methane (7,1g) in BwH2 case.

The photochemical oxidant (POP) and freshwater eutrophication (EP) impacts (Fig. 3b, d) of BwH2 are almost double than of BmH2. Like climate change, in these impact categories, AD and reforming processes contribute the most. In POP of BwH2, digestate handling and use as fertilizer contribute 97 % attributed to air emissions of NMVOC (419 mg), CO (770 mg), etc., sourced to the production and use of transport fuel (diesel). Further, emissions of phosphorus and sulphate compounds like phosphorus (5.8 mg) to the soil, sulphate (8.8 g) to water etc., from the use of digestate as fertilizer attribute 70–80 % EP of the AD process. In BmH2 eutrophication potential (EP), pyrolysis accounts for 23 %

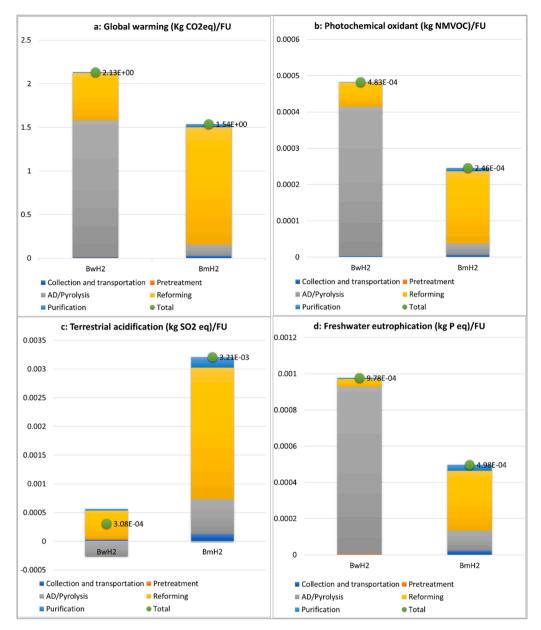


Fig. 3. The GWP, POP, EP and AP of 1 kg hydrogen production of BwH2 and BmH2 systems.

while reforming 67 %. The low contribution of the pyrolysis process is due to the lower external energy input which was electricity and the process's excess heat energy (1.35 MJ/ton oil) that has been assumed to replace natural gas. Whereas the high contribution of oil reforming links to the production and use of natural gas used for steam production.

Further, the AP (Fig. 3c) of hydrogen production from biowaste (BwH2) is 90 % lower than biomass (BmH2). In BwH2, biogas reforming contributes the most while AD has a negative value attributed to the use of digestate as a biofertilizer which is considered to avoid mineral fertilizer production. Even though the substitution of mineral fertilizers has been accounted for but due to the very low quantity of fertilizer minerals in the digestate (N = 6.6 kg, P = 0.6 kg, and K = 2.4 kg per ton), the substitution effect is not significant. Whereas, in BmH2, reforming has a high positive contribution to AP mostly attributed to the air emissions of SO₂ (1 g) that are probably linked to the production and combustion of natural gas use in the process. The AP of collection and transportation and purification processes is attributed to the production of electricity.

The negative toxicity impacts (i.e., TEP, HTP, and FWEP) of BwH2 (Fig. 4a, b, c) are mostly (around 97 %) linked to the AD process,

attributed to the use of digestate as fertilizer. The output emissions inventory of this process indicates that the use of digestate as biofertilizer avoids the direct and indirect release of toxic metals to water such as Aluminum (-45 mg), Calcium (-660 mg) Phosphate (-49 mg), Zinc (-3.6 mg), etc., and acidic chemicals such as Sulphate (-12 g), Sulfuric acid (-603 mg), etc., that could otherwise happen if mineral fertilizers were produced and used. It should be noted that the release of heavy metals and ammonia compounds through field application of digestate depends on weather and soil conditions such as soil permeability and thus, it may affect the LCA results which has also been reported elsewhere [54].

Whereas, in case of BmH2, oil reforming process accounts for $67\,\%$ in TEP, $54\,\%$ to HTP and $21\,\%$ FWEP attributed to the production and combustion of natural gas used for steam production. The emissions inventory of this process shows release of toxic metals and substances to water like iron ($84\,\text{mg}$), Lithium ($410\,\text{mg}$), Nitrate ($92\,\text{mg}$), Phosphate ($36\,\text{mg}$), etc. Pyrolysis is the second largest contributor of that contribution attributed to the release of acidic gases to air such as Benzene ($20\,\text{mg}$), Butane ($31\,\text{mg}$), Ethane ($130\,\text{mg}$), Hydrogen chloride ($9\,\text{mg}$), Hydrogen sulfide ($8\,\text{mg}$), etc.

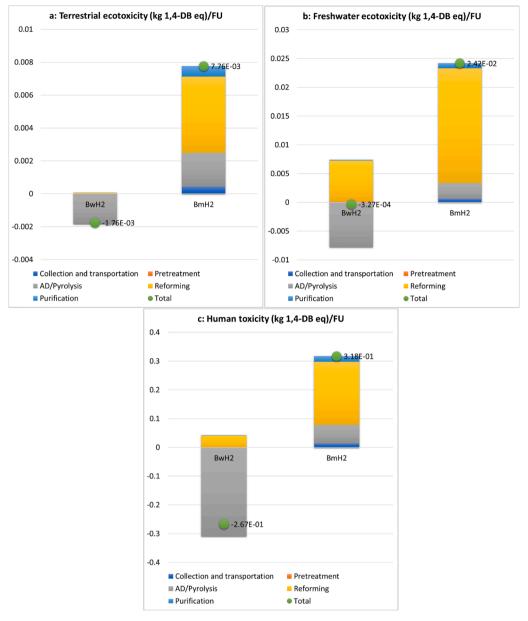


Fig. 4. The toxicity impacts of BwH2 and BmH2 systems.

In Ozone depletion potential (Fig. 5a), the reforming process shows the highest contribution in both cases attributed to direct air emissions of mainly hydrocarbon aromatic compounds that are (4 mg) in BwH2 while (15 mg) in BmH2. The output emissions inventory of pyrolysis oil reforming shows that the emissions of benzene compounds (Benzene 20.3 mg, Butane 27.2 mg, Ethane 141 mg, etc.) to air deemed responsible for ODP. Whereas, in BwH2, AD has a negative contribution due to added advantages of avoided emissions attributed to mineral fertilizer production replaced with the digestate used as biofertilizer.

The positive contribution of BmH2 system in abiotic depletion categories (ADP, ADP-fossil oil) is dominated by the natural gas use in reforming of pyrolysis oil. Even though natural gas has also been used for biogas reforming in BwH2 system, but the positive contribution of biogas reforming is encountered by avoided emissions attributed to avoided mineral fertilizer production due to the use of digestate as biofertilizer. In the same impact categories, in BmH2, the use of electricity in biomass pyrolysis and purification processes show positive contribution attributed to the indirect use of fossil resources linked to electricity production.

Digestate treatment and its use as biofertilizer, and reforming of biogas and pyrolysis oil have been identified as hotspots. Thus, to improve the environmental performance of the systems, efforts should focus on, i) energy-efficient treatment of digestate (i.e., energy use in its separation to solid and liquid, and for the killing of harmful bacteria) and its transportation for use as biofertilizer. There could be several ways to do this but here a simple solution is to reduce water content. ii) substitute natural gas used in the reforming process with a biobased energy source such as biogas or biomethane. iii) Technology improvements are needed both from energy input and output of biogas /pyrolysis oil per unit of feedstock. As evaluated in [13], utilization of noncondensable (NC) off-gases from the pyrolysis, oil gasification, and reforming processes for synthetic natural gas production may further improve the systems' environmental sustainability and LCC efficiencies. Such improvement can substantially reduce the environmental impacts of the systems.

As presented in the introduction, some LCA studies indicate that H_2 is better produced via biomass pyrolysis than biomass direct gasification, biowaste AD, and fossil methane reforming. For example, Susmozas

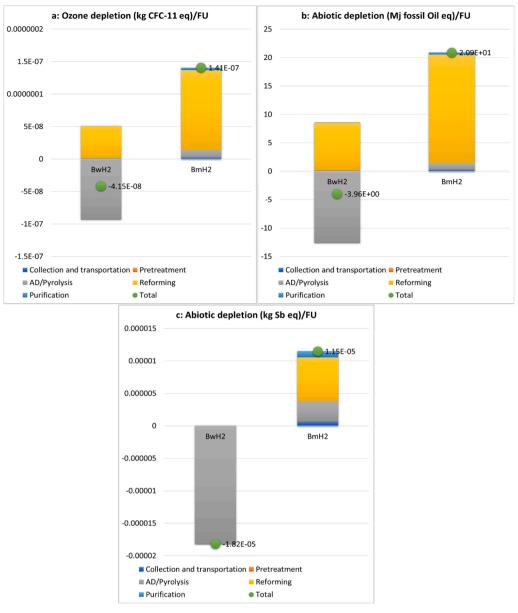


Fig. 5. Abiotic and Ozon depletion potentials of hydrogen production of BwH2 and BmH2 systems.

et al. [46] found that hydrogen production from popular biomass is better than fossil methane reforming from climate change and nonrenewable energy demand perspectives. Hajjaji et al. [18] conclude that biogas-based hydrogen is much better than that natural gas from climate change and fossil energy input. Further, the study [23] concludes that biomass-derived bio-oil for hydrogen production is more favorable than glycerol and natural gas in environmental impact and energy recovery efficiency. Overall, the study's life cycle assessment (LCA) results are in line with previous research in terms of global warming potential (GWP), eco-toxicity potential (EP), freshwater eutrophication (FWEP), and photochemical oxidant potential (POP). However, for other impact categories examined, this study shows that hydrogen production from biowaste via anaerobic digestion (AD) outperforms biomass via fast pyrolysis. It is important to note that life cycle assessment (LCA) results can be influenced by several factors. These factors include the selection of production system boundaries, processing technology, impact assessment method employed, and the types of input data [55]. From a sustainability perspective, the conversion efficiency of biomass carbon and the energy efficiency of the conversion technology are crucial parameters that need to be considered [55].

Investigating these aspects can improve the comparability of this study's results and enhance understanding of the sustainability performance of the systems being studied. To evaluate the influence of the impact assessment method on the LCA results, a sensitivity analysis was performed, which is discussed in section 4.3.

3.2. Life cycle cost (LCC)

The LCC of hydrogen is calculated based on the cost inventory data given in (Appendix Table B.5), and monetization of life cycle environmental impacts using environmental cost unit price given in Table 3. With discounting factor (r = 10 %), the annual average LCC of 1 kg hydrogen production from biowaste and biomass is accounted as 0.45 – 2.76 ε and 0.54–3.31 ε respectively over the plant lifetime, 20 years. Hydrogen production from biowaste via anaerobic digestion (BwH2) is 20 % cheaper than that of biomass fast pyrolysis (BmH2). This is attributed to low variable OPEX that is mainly influenced by feedstock price, the capital investment which is 50 % less than BmH2, and fixed OPEX that again links to CAPEX. To the total LCC, in BwH2, environmental cost accounts for 3 % while in BmH2 its 13 % (Fig. 6).

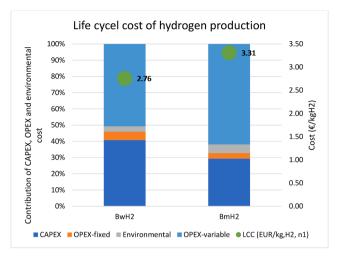


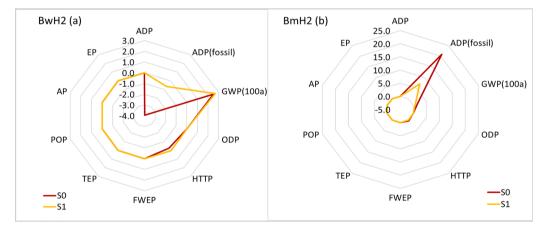
Fig. 6. The LCC of 1 kg hydrogen produced and the cost distribution, n1 represents the first year.

If the price of biowaste is set equal to the biomass price, the difference in LCC would only be 1 %. Further analysis of variable OPEX indicates that, in BwH2, utility cost contributes most that link to electricity, natural gas, and transport fuel but in the case of BmH2, feedstock contributes the highest (54 %) and utilities 30 %. This shows variable OPEX in both cases highly sensitive to feedstock price. The fixed OPEX, in both cases highly sensitive to CAPEX as this based on a fixed

percentage of CAPEX (Appendix Table B.5). Further, the environmental cost of hydrogen production from biomass (BmH2) is 10 % higher than that of biowaste (BwH2) even though the global warming potential of BmH2 is 28 % low compared to BwH2. This is because of the LCA values of HTTP, TEP, and EP of BmH2 are higher than biowaste which in terms of environmental cost contributing 15 %, 33 %, and 8 % respectively. Overall, to the environmental cost, climate change contributes the highest which is 135 % in BwH2 and 43 % in BmH2.

The discounting factor (r) has been reported between 5 and 10 % [2,40,51]. With r=5 %, the LCC of 1 kg hydrogen production increased by 5 % compared to the base case (r = 10 %) and resulted in 0.78–2.83 $\[mathebel{e}$ /kg in BwH2 and 0.94–3.40 $\[mathebel{e}$ /kg in BmH2. Whereas, at r=7 %, the LCC increased by 3 % (1.14–2.89 $\[mathebel{e}$ /kg in BwH2 and 1.37–3.47 $\[mathebel{e}$ /kg in BmH2).

From the LCC perspective, to make biomass-based hydrogen competitive, the CAPEX of BmH2 system technology should be reduced to 50 % of its present value. And, to reduce variable OPEX and environmental costs, low-cost feedstock and energy-efficient biomass pyrolysis and oil reforming are vital. These parameters have also been reported most influential in biomass to liquid fuels production cost and GHG emissions [56]. Whereas in the case of BwH2, to reduce both variable OPEX and environmental costs improvements should focus on the reduction of natural gas and transport fuel consumption. This can be done by several means, but the simple explanation could be to replace natural gas with a low-cost renewable energy source and reduce transport distances or use more fuel-efficient transport services. Use of blend feedstocks could be a way to reduce feedstock cost which may increase intermediate products (biogas and pyrolysis oil) yields as evaluated in [561].



 $\textbf{Fig. 7.} \ \ \text{Sensitivity scenario S1.}$

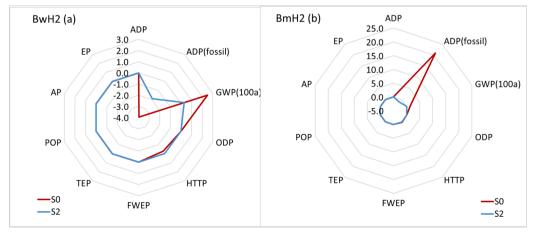


Fig. 8. Sensitivity scenario S2.

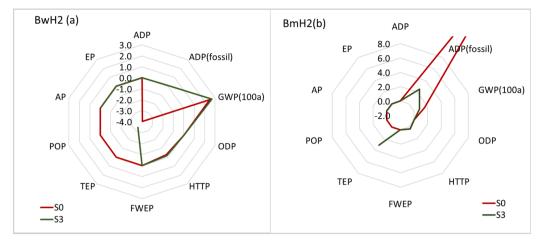


Fig. 9. Sensitivity scenario S3. In the ReCiPe 2016 midpoint, Hierarchist perspective the LCI parameter Non-methane volatile organic compounds (NMVOCs) is not given and thus the value of POP category is not plotted in S3.

Table B1The composition of biogas produced at the Forsbacka biogas plant.

CH ₄	50–55 %
CO ₂	40–45 %
H ₂ S	up to 250 ppm
O_2	0.9 %

Yukesh et al. [57] report that the cost of hydrogen production largely depends on the size of the production plant, initial investment, and feedstock price. For example, in the biochemical conversion of biowaste to $\rm H_2$ such as dark and photo fermentation, the overall cost (excluding environmental cost) of 1 kg $\rm H_2$ production is reported as 10 $\rm \ell$ (adjusted to 2022 USD to Euro price) from industrial wastewater [58], 2.7 $\rm \ell$ from beverage wastewater [48] and 2.57–2.80 $\rm \ell$ from the agricultural residue with a plant capacity of 300–400 m³ $\rm H_2/hr$ [59]. In this study, the LCC of hydrogen production from biowaste (BwH2) is calculated as 2.76 $\rm \ell$ which is in the range of reported values. However, in this study environmental cost is included, while in the reported studies it's not.

Considering biomass, the LCC of 1 kg $\rm H_2$ production from biomass via fast pyrolysis and hydro-processing has been reported as $1.85-2.82 \in [60]$ and $1.06-2.05 \in [61]$ with agricultural residue, price $0.092 \in \rm per$ kg. Helf et al. [62] have reported the cost of hydrogen production from beech wood gasification to be $3 \in \rm kg \, H2$. Whereas IEA [11] reports the cost of renewable hydrogen production as $1.8-4.5 \in \rm kg$. Keep in mind that these studies included only CAPEX and OPEX, not environmental costs. This case study results are $0.54-3.31 \in \rm which$ confirms the reported LCC of hydrogen production from biomass of different types. Findings of influencing parameters to LCC are also in line with previous studies i.e., CAPEX, plant size, and biomass feedstock price.

4. Sensitivity analysis

In LCA studies uncertainty in LCI of some parameters may exist which can further increase with change in technology and geography. Sensitivity analysis of the LCA outcomes with process parameters that deemed sensitive to technology or geography can reduce the uncertainty and improve the generalization of LCA study outcomes. To enhance this study generalization and get a better insight into the investigated systems' result sensitivity to the used inputs inventory and impact assessment method, three sensitivity analysis scenarios (S1, S2, S3) are developed (Table 4).

Feedstock-to-output yields of biogas and pyrolysis oil largely depend on the type of feedstock and the operating conditions of the production

Table B2Proximate analysis of Biowaste (Source: Personal communication with Ekogas AB).

Biowaste composition	Unit (%)
Household food waste	80.76 %
Slaughterhouse waste	0.11 %
Waste oil from restaurants	1.06 %
Animal waste	7.15 %
Garden green waste	10.92 %

Table B3The Ultimate analysis of the biomass and Pyrolysis oil (source: personal communication with Pyrocell AB).

Ultimate analysis of Biomass		Ultimate analysis of Pyrolysis oil	
Moisture content	55 %wt	Carbon	42 %wt
Ash	0.3 %TS	Hydrogen	8 %wt
Sulphur	30 %TS	Nitrogen	<0.1 %wt
Nitrogen	40 %TS	Oxygen	50 %wt
		Sulphur	<0.05 %wt
		Ash	<0.05 %wt

system. To analyse the feedstock-to-yield ratio sensitivity, it was assumed that biowaste-to-biogas yield in BwH2 and biomass-to-pyrolysis oil yield in BmH2 in S1 decreased by 25 % in relation to the base case (S0). Similarly, the reforming of biogas and pyrolysis oil has been reported as an energy-intensive process [1,9,18,51] which means the amount and type of fuel used could influence the results and conclusion. As in the present study, site-specific data for the reforming process was lacking, and energy inputs were calculated based on literature, where it was assumed that natural gas is used as fuel in biogas and pyrolysis oil reforming. Thus, to analyze the sensitivity of this study's results to this assumption, in S2, natural gas was replaced with energy from wood chips-based 10 heat and electricity production plant.

CLM-IA and ReCipe 2016 are the most used impact assessment methods for LCA of hydrogen production systems. In this study, CML-IA

 $^{^{10}}$ The process is in SimaPro where the dataset represents the production of heat and electricity with wood chips in a co-generation plant with a capacity of 6667 kW (referring to fuel input) in Sweden. It is assumed that the plant is operated mainly to produce heat, i.e., following the heat demand. Heat is therefore considered the main product and therefore the reference product, whereas electricity is a by-product. Wood chips are burned in a boiler at a temperature of 800–1300 $^{\circ}\mathrm{C}$ under excess air conditions and turned into carbon dioxide and water. [source: Simapro].

Table B4

Outputs inventory of important elements emissions to water, air, and soil. Miner emissions are omitted.

Substance	Compartment	Unit	BwH2	BmH2
BOD5 (Biological Oxygen Demand)	Water	mg	-445.6	264.0
Bromine	Water	mg	24.1	84.2
Calcium	Water	g	-44.7	1.4
Chloride	Water	g	-130.7	15.1
COD (Chemical Oxygen Demand)	Water	mg	-440.4	384.2
DOC, Dissolved Organic Carbon	Water	mg	-113.0	120.9
Iron	Water	mg	-39.7	84.4
Lithium	Water	mg	132.3	418.8
Magnesium	Water	mg	-1.1	269.2
Nitrate	Water	mg	-40.4	92.3
Nitrogen, atmospheric	Water	mg	-269.5	5.0
Oils, unspecified	Water	mg	-125.6	66.3
Phosphate	Water	mg	-48.2	36.7
Potassium	Water	mg	-1.7	22.9
Silicon	Water	mg	-18.2	7.2
Sodium	Water	g	-99.3	4.4
Solids, inorganic	Water	mg	-134.6	137.6
Sulfate	Water	g	-14.6	1.5
Suspended solids, unspecified	Water	g	5.2	17.5
TOC, Total Organic Carbon	Water	mg	-128.8	122.1
Aluminium	Air	mg	-4.0	17.1
Ammonia	Air	mg	0.3	58.3
Benzene	Air	mg	46.6	22.4
Butane	Air	mg	18.2	31.2
Calcium	Air	mg	48.4	16.5
Carbon dioxide, fossil	Air	kg	2.6	1.1
Carbon monoxide, fossil	Air	mg	470.8	651.2
Dinitrogen monoxide	Air	mg	15.6	51.2
Ethane	Air	mg	203.2	143.7
Hydrocarbons, aliphatic, alkanes, unspecified	Air	mg	0.9	4.0
Hydrocarbons, aromatic	Air	mg	4.0	15.8
Hydrogen chloride	Air	mg	-29.8	11.1
Hydrogen fluoride	Air	mg	-3.0	2.4
Hydrogen sulfide	Air	mg	-6.6	13.3
Magnesium	Air	mg	0.0	1.1
Methane, fossil	Air	g	15.3	5.1
Nitrogen oxides	Air	g	-1.3	1.1
Nitrogen, atmospheric	Air	mg	970.4	7.0
NMVOC, non-methane volatile organic compounds	Air	mg	-199.3	410.3
Ozone	Air		3.1	19.7
Particulates, < 2.5 um	Air	mg	-166.4	98.6
Particulates, < 2.3 min	Air	mg	-100.4 -317.9	225.7
· · · · · · · · · · · · · · · · · · ·	Air	mg	-317.9 -401.9	58.8
Particulates, > 2.5 um, and < 10um Potassium	Air	mg	7.0	62.6
Sulfate		mg		
Sulfur dioxide	Air	mg	-8.3	7.0
	Air	g	-1.3	1.0
Toluene	Air	mg	6.5	24.2
Xylene	Air	mg	2.7	11.5
Aluminium	Soil	mg	-3.0	12.7
Calcium	Soil	mg	13.2	158.9
Carbon	Soil	mg	-17.6	14.6
Carbon dioxide, to soil or biomass stock	Soil	mg	-22.9	13.2
Iron	Soil	mg	-16.5	16.9
Magnesium	Soil	mg		18.0
Manganese	Soil	mg	1.5	11.1
Oils, biogenic	Soil	mg		3.5
Oils, unspecified	Soil	mg		55.6
Phosphorus	Soil	mg		5.5
Potassium	Soil	mg	3.5	30.3
Silicon	Soil	mg	1.7	49.3
Sulfur	Soil	mg	-4.9	5.8

is used. To test the impact of the choice of impact assessment method, in S3, ReCipe 2016 was selected as the impact assessment method and results have been compared with the base case S0.

4.1. Scenario S1

Up to 25 % decreases in yields of biogas and pyrolysis oil per unit of feedstock (i.e., S1) significantly influence ADP (fossil), GWP, and HTTP. In BwH2, S1 results (Fig. 7a) show an increase of 85 % ADP (fossil), 17 % GWP and 31 % HTTP. This can be explained by transport activities

linked to the biowaste collection, and the use of the digestate as a biofertilizer. The lower yield of biogas per unit of feedstock means more biowaste would be required to generate a unit (m³) of biogas and more generated digestate would also have to be transported a long distance for use on agricultural land. At the same time, ammonia emissions from the digestate storage would also increase which would eventually increase GWP, HTTP, and ADP impacts. While in BmH2, S1 shows a decrease of 67 % ADP (fossil), 48 % GWP, 30 % HTTP, and 5 % FWEP.

This significant environmental gain could be explained by an increased amount of biochar that is combusted internally for energy that

Table B5
Parameters and cost values used for OPEX in both base case scenarios. For raw materials and utilities, average prices for the year (2021) were used [conversion rate. 1Euro = 10SEK].

Parameters	Euro (€)	Unit	Source
Feedstock (biowaste) price	0.0*	kg	Primary
Feedstock (biomass) price	0.06	kg	[a]
Price of additives	0.0006	kg	Primary
Price of nitrogen	1.5	kg	[b]
Electricity price	0.15	kWh	Primary
Heat price	0.10	kWh	Primary
Water price	0.005	kg	Primary
Diesel price	2.8	L	Primary
Natural gas price (LHV = 43 MJ/kg)	0.082	kWh	[c]
Biomethane price (LHV = $36Mj/m^3$)	0.0396	MJ	Primary
Liquid digestate as biofertilizer selling price	0.0001	kg	Primary
Solid digestate as biofertilizer selling price	0	kg	Primary
Repair and maintenance cost (1.5 % of CAPEX)-BwH2	0.378	MEUR	Primary
Repair and maintenance cost (1.5 % of CAPEX)-BmH2	0.678	MEUR	Primary
operating labors cost (5 people with an average salary of 3500 €/month)	0.21	MEUR	Primary
Employees' benefits (22 % of operating labor)	0.0462	MEUR	[d]
Laboratory cost (6 % of CAPEX)-BwH2	1.512	MEUR	[d]
Laboratory cost (6 % of CAPEX)-BmH2	2.712	MEUR	[d]
Insurances and taxes (3 % of CAPEX)-BwH2	0.756	MEUR	[d]
Insurances and taxes (3 % of CAPEX)-BmH2	1.356	MEUR	[d]
Plants overhead cost (1 % of CAPEX)-BwH2	0.252	MEUR	[d]
Plants overhead cost (1 % of CAPEX)-BmH2	0.452	MEUR	[d]

Primary- calculated based on information the information from the case study plant (company-Ekogas AB).

- [a]-Calculated based on the information from [65].
- [b]-Based on the information from https://www.svenskafoder.se/ [assessed 20 Jun 2022].
- [C]- Adapted from https://ec.europa.eu/eurostat [assessed 20 Jun 2022].
- [d]- Calculated based on the information from [40].

has been assumed to replace natural gas. Hence, feedstock to yield (biogas and pyrolysis-oil) ratio is a sensitive parameter that could significantly influence LCA results, and this has also been reported in research [4,63,64].

4.2. Scenario S2

The S0 and S2 LCA results show that natural gas substitution in the reforming of biogas and pyrolysis oil with wood chips (see, Table 4), significantly influence the ADP (fossil) and GWP in both cases (Fig. 8).

In BwH2 (Fig. 8a), environmental gains over impact categories ADP-fossil, GWP are 44 % and 88 % respectively. Whereas, in BmH2, a large gain can be observed in ADP-fossil (-106 %), GWP (-100 %) that could be explained with reforming process as the only fossil energy (natural gas) dependent process. The improvements in the other impact categories are small (30 – 60 %) and thus it can be said that S2 has a big advantage over S0 from ADP-fossil and climate change perspectives in both cases.

4.3. Scenario S3

In this scenario, the only difference concerns the impact assessment method, which is ReCiPe 2016 midpoint, Hierarchist perspective. It is obvious from the figures (Fig. 9) that the difference in LCA results over some impacts categories like ADP (fossil), TEP, and GWP is significant. In BwH2, the difference between S0 and S3 impacts values of ADP-fossil, GWP and TEP is $102\,\%$, $7\,\%$ and $>1000\,\%$ respectively while in BmH2, this is $88\,\%$, $47\,\%$, and $>1000\,\%$. A large variation in TEP is unclear and may be relevant to the background methodology of the ReCiPe method. Although, this scenario results indicate that the choice of impact assessment method in reporting LCA results does matter particularly in climate change, abiotic depletion-fossil, and terrestrial ecotoxicity. Thus, comparing products' sustainability performance with LCA results generated using different impact assessment methods could be misleading.

5. Conclusions

This study compares two scenarios for hydrogen production from environmental and economic perspectives. In the first scenario, BwH2, the use of fossil fuels and emissions from digested storage and spreading on arable land are the main contributors to climate change, photochemical oxidant, and freshwater eutrophication. However, using the digestate as biofertilizer can credit the system on other impact categories. In the second scenario, BmH2, most of the pyrolysis process energy demand is met internally, which credits the system on climate change, photochemical oxidant, and freshwater eutrophication. However, the positive environmental impacts of BmH2 are mostly due to energy input to pyrolysis oil gasification and reforming. Therefore, from the perspective of impact categories, hydrogen production from biomass is better for climate change, photochemical oxidant, and eutrophication, while hydrogen production from biowaste is better for the other seven included impact categories.

The lifecycle cost of 1 kg hydrogen production is calculated as $0.55-2.76\varepsilon$ with biowaste and $0.45-3.31\varepsilon$ with biomass. The capital investment cost (CAPEX) of BmH2 is almost double that of BwH2, resulting in a similar situation on OPEX-fixed. Variable OPEX in both cases is affected by feedstock and utility prices. In BwH2, the feedstock price is zero, resulting in a 32 % lower variable OPEX compared to BmH2, where a positive feedstock (biomass) price accounts for around 54 % of the system's variable OPEX. From an LCC perspective, hydrogen production from biowaste is a better option than biomass.

The study also identifies hotspots for environmental and economic efficiencies in both scenarios. In BwH2, the reforming of biogas and digestate handling are hotspots, while in BmH2, it is pyrolysis oil reforming. Improvements can be made by replacing natural gas with biogas or bio-oil, enhancing the biofertilizer density (i.e., the mass of NPK per ton of liquid digestate), and using biofuel instead of fossil diesel in digestate transportation. From an LCC perspective, technology prices, lab expenditures, and insurance and taxes are hotspots. Improvements focused on these hotspots, particularly on CAPEX in the BmH2 case, can

^{*} A waste treatment fee (0.06€) paid to the company by the municipalities.

significantly reduce the cost of hydrogen production.

This study contributes to existing research by providing primary LCI data, addressing previously unreported impact categories, and providing useful insights that can be generalized beyond Sweden to Europe and globally. The findings endorse existing research results and thus contributing in reducing uncertainty in the generalization of those theoretical and simulation software-based studies.

CRediT authorship contribution statement

Muhammad Arfan: Conceptualization, Investigation, Methodology, Data curation, Formal analysis, Writing – original draft. **Ola Eriksson:** Supervision, Conceptualization. **Zhao Wang:** Supervision, Conceptualization. **Shveta Soam:** Supervision, Conceptualization, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial

interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A

A.1. Steam methane reforming (SMR)

The SMR process is generally presented with the following chemical equations [1,18,21].

$$CH4 + H2O \rightleftharpoons CO + 3H2 \qquad \Delta H^{\circ}_{298k} = +206kJ/mol \tag{A1}$$

$$CO + H2O \rightleftharpoons CO2 + H2 \qquad \Delta H^{\circ}_{298k} = -41.17kJ/mol \tag{A2}$$

$$CH4 + 2H2O \rightleftharpoons CO2 + 4H2 \quad \Delta H^{\circ}_{298k} = (+206 - 41.17)kJ/mol$$
(A3)

A.2. Emission from fertilizers

The use of mineral and biofertilizers in agriculture practices involves emissions of nitrogen compounds (NH3, N2O, NOx, and NO3), phosphate, and heavy metals depending on their composition, soil, climate, and application method (see assumption section). In the life cycle impact assessment, both upstream and downstream emissions are considered.

For the calculation of NH₃-N emissions from the application of digestate as a biofertilizer AGRAMMON model (Eq.A4) presented in [41] was used.

$$NH_3 - N = TAN.(EF + C_{app}).C_x$$
(A4)

 NH_3 - $N=quantity\ of\ ammonia\ [kg\ NH3-N]\ emissions\ to\ air.$

TAN = total ammonium nitrogen of the organic fertilizer [kg N], here N, equals to 6.6 kg/ton. 11

EF = emission factor for the organic fertilizer [%TAN/100].

C_{app} = correction factor influencing the emission factor (applies only for liquid manure and thus here it is 0).

cx = correction factor considering impacts of technical equipment used for liquid fertilizer application, time of fertilizer application, and weather conditions. Here it is assumed as 1.

Phosphorus (P) leaching to groundwater from the application of the digestate as biofertilizer was estimated using the method presented in [41]. Here, a simplified version of the model equation (A6) was used.

$$P_{\rm gw} = P_{\rm gwl}.F_{\rm gw} \tag{A5}$$

$$P_{gw} = 1 + \frac{0.2}{80} * P_2 O_{5sl}$$
 A6

P_{gw} = quantity of P leached to groundwater (kg/(ha*year))

 $P_{gw}l = average$ quantity of P leached to groundwater for a land use category (kg/(ha*a)), for this study assuming the same conditions as considered by [41] and here taken 0.07 kg P/(ha*year) for arable land and 0.06 kg P/(ha*year) for parks.

 $F_{gw}=\mbox{correction factor for fertilization by slurry (-). Here, it is assumed as 1.}$

 $P_2^{\circ}O_{5sl}$ = quantity of P_2O_5 contained in the slurry. The values of P2O5-content as P were taken from the case study company which is 0.6 kg/ton. ¹²

 $^{^{11}}$ Based on personal communication with the plant manager, Nyquist Henrik [dated: 2022–06-20].

 $^{^{12}}$ Based on personal communication with the plant manager, Nyquist Henrik [dated: 2022–06-20].

Appendix B

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