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Prospective Life Cycle Assessment of Early-Stage CO_2-Removing Artificial Photosynthesis Producing Synthesis Gas, Hydrogen Peroxide, and Methane

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Due to the urgent need to mitigate the consequences of climate change, emerging technologies such as artificial photosynthesis could be crucial. To enable an environmental sustainability assessment of this early-stage technology, recent methods in prospective Life Cycle Assessment are applied and extended with break-even sensitivity analysis to evaluate the minimum requirements of the technical parameters as well as Monte Carlo simulations to include uncertainties in the model. This study focuses on two CO_2-removal technologies based on artificial photosynthesis: photocatalytic carbon dioxide reduction for producing synthesis gas (H_2+CO) and photoelectrochemical carbon dioxide reduction for producing hydrogen peroxide and methane (H_2O_2&CH_4). The results indicate that while artificial photosynthesis could be environmentally beneficial, it is highly dependent on the scenarios used in the prospective background database and system design parameters, both of which can alter the technology ranking. While prospective Life Cycle Assessment incorporating break-even sensitivity analysis and uncertainty analysis can guide technology development by identifying minimum parameter requirements and theoretical competitiveness, these analyses can significantly affect the outcomes by producing a wide range of results, making interpretation challenging. Thus, these analyses are crucial and highlight the need for further research, particularly to improve result interpretation and communication.

1. Introduction

Because of unsustainable practices leading to the exceedance of Earth's planetary boundaries, humanity is facing the risk of shifting the Earth's system to a much less hospitable state for the development of human societies.[1,2] Among these environmental challenges, climate change and biosphere integrity were recognized as fundamentally important.[2] The Intergovernmental Panel on Climate Change[3] report indicates that to avoid surpassing the critical threshold of 1.5 °C in global warming, a significant reduction in greenhouse gas (GHG) emissions is required, achieving net zero around 2050 and transitioning to net negative emissions thereafter. While measures to halt and reverse biodiversity loss are more diversified,[4] efforts to mitigate climate change can simultaneously benefit biodiversity and vice versa.[5,6]

Globally, the primary activities contributing to climate change are GHG emissions from the energy sector, accounting for 34% of the global total, followed by the industry sector (24%).[7] The chemical industry is the largest industrial energy consumer, using 28% of industrial energy and 10% of global final energy. This sector also contributes about 10% of total global direct industrial emissions.[8,9] Most of this energy consumed by the chemical sector stems from oil, gas, and coal with half of the energy input consumed as feedstock.[10] Consequently, even if process energy is replaced with renewable energy, the demand for oil is expected to increase driven by the forecasted growth of chemical production.[11,12]

Therefore, beyond energy efficiency measures and electrification, which enable the use of renewable energy sources for process energy, the adoption of biomass feedstock and carbon capture technologies that can transform carbon dioxide (CO_2) from a waste product into a valuable resource is required.[9] The IPCC report reaffirms this, stating that the 1.5 °C global warming level is likely to be exceeded in all scenarios except in the Shared Socioeconomic Pathway 1–1.9, which assumes "very low and low GHG emissions and CO_2 emissions declining to net zero around or after 2050, followed by varying levels of net negative CO_2 emissions".[13] This implies that even with an immediate GHG reduction, CO_2 removal from the atmosphere will most likely be necessary to keep global warming below the 1.5 °C level. Therefore, carbon capture and direct utilization technologies such as artificial photosynthesis (AP)—particularly CO_2-utilizing AP—could offer a solution for the production of chemicals and the reduction of GHG at the same time. AP technology, which avoids additional fossil-based process energy and fossil precursors, harnesses solar energy to convert

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CO_2 into useful chemical products. The sun is a particularly advantageous energy source, as solar resources are orders of magnitude larger than all other renewable sources combined,[14] exceeding global energy consumption by over 1000 times (assessed by assuming a global primary energy consumption of around 179000 TWh, which corresponds to around 20.43 TWyears and solar energy received of around 23000 TWyears per year).[14,15]

AP is derived from natural photosynthesis, the biological process by which plants, algae, and some bacteria convert light energy from the sun into chemical energy.[16] However, the maximum theoretical efficiency of natural photosynthesis is relatively low (4.6–6%)[17,18] with practical global averages ranging from 0.1 to 0.2%,[19] limiting its ability to meet annual human energy consumption.[19] AP systems potentially offer higher efficiencies (solar-to-hydrogen conversion) reaching up to 30% depending on the system configuration.[20,21] The principle of AP involves the photoexcitation of a semiconductor, the most common material used for AP, which, when exposed to light of an energy greater than or equal to the bandgap energy, leads to the promotion of an electron from the valence band to the conduction band and thus generates a reduction and oxidation potential. For a thorough description of AP principles, technologies, and history, see Hoffmann et al.[17] and Zamfirescu et al.[22] Furthermore, comprehensive overviews of AP technology and system design options are provided, assessing the benefits, challenges, and the state of the art for hydrogen production by AP.[21–23] Song et al.[23] concluded that while photoelectrochemical (PEC) and especially photovoltaic (PV) electrolysis systems exhibit high efficiencies, their potential costs and environmental impacts—measured in global warming and acidification Potential—are higher due to their complexity. In contrast, photocatalytic (PC) and photobiological systems could be more beneficial from an environmental and cost perspective, though their efficiencies are currently low. PEC systems already combine acceptable efficiencies with low environmental impacts, but their durability, similar to PC and photobiological systems, remains unsatisfactory.

Compared to hydrogen production by AP, the reduction of CO_2 provides the advantage of using CO_2 as a building block for a wide variety of organic chemicals and fuels,[24] enabling the recycling or removal of CO_2 from the atmosphere, either directly or via biomass production as an intermediate. However, CO_2 reduction poses an additional challenge due to the thermodynamic stability of CO_2.[17,24–27] Hoffmann et al.[17] indicate that while the production of fuels and industrial chemicals through CO_2 reduction holds significant promise, it is challenging from a technical perspective. Sun[16] confirmed that despite ongoing research into novel photocatalysts, the state of the art in PC CO_2 reduction remains far from commercialization.

Given its potential to reduce climate change impacts, this study focuses on PEC and PC AP for CO_2 reduction. The terminology around AP is still debated.[28–30] In this study we use the term PC also for energetical uphill reactions. To verify and assess the potential environmental benefits of carbon capture and utilization through AP, including upstream and downstream processes, Life Cycle Assessment (LCA) is applicable as it provides a systematic method for identifying a wide range of environmental impact categories of products or services.[31–34]

Studies investigating AP product systems using LCA have shown that the efficiency of the conversion is crucial for the impact assessment results. Efficiencies exceeding 2–5% are necessary to match the environmental performance of reference product systems, depending on the impact categories and products considered.[35–39] Moreover, according to Sathre et al.[37] a cell lifetime of at least five years is necessary to achieve a positive net energy value. Song et al.[23] summarized LCA results of two selected impact categories, global warming potential (GWP) and acidification potential. They found that conventional H_2 production from steam methane reforming has the largest emissions, while carbon capture and storage technology can reduce these emissions to a certain extent. PC, PEC, and photobiological approaches demonstrated the lowest GWP and acidification potential. Photothermal catalytic H_2 production exhibited the highest GWP and acidification potential among solar-powered methods, though this was based on using fossil methane. PV–electrochemical systems also showed high GWP and acidification potential due to the manufacturing process of silicon-based PV panels. Song et al.[23] identified a trade-off between the cost of H_2 production and environmental impacts, recommending further research to either reduce the costs of PC, PEC, and photobiological systems, or to decrease the GWP of cost-competitive technologies such as PV–electrochemical, solar thermochemical, and photothermal catalytic systems.

Most of the identified literature focused on cumulative energy demand and climate change impact categories, with occasional consideration of acidification potential. Up to the time of this study and to the best of our knowledge, no studies have applied broader midpoint impact assessment categories that could reveal potential trade-offs. Moreover, LCA studies specifically addressing the CO_2 reduction pathway in AP systems could not be found beyond previous work by the authors. No studies applying recent prospective or ex-ante LCA to AP were identified.

Since AP is in its early stage of development, no industrial-scale facility is available to provide data for an environmental assessment. To assess the stage of development of AP, the technology readiness level (TRL) concept provides a systematic metric for assessing the maturity level of a given technology, even though it is not standardized and allows for some interpretation.[40] Within this framework, AP could be associated with a TRL of 1–3,[40,41] representing a very early stage in technology development (TRL 3: experimental proof of concept). As a result, there are a lack of standardized methods for addressing technological, economic, environmental, and social aspects using common indicators for comparison.[40] Particularly in early stages of technology development where a technology can still be changed, it is recommended to ensure environmental, social, and ethical compatibility.[42]

LCA is a standardized method and was originally developed to assess the environmental performance of existing technologies.[33,34] Typically, technology assessments are conducted assuming present techno- and biosphere conditions, without considering differences due to scaling or potential future elementary flows and characterization factors. This approach can introduce unprecedented uncertainties when assessing

emerging technologies with a low TRL,[43–47] such as AP, which is unlikely to be commercially viable in the near future. Furthermore, data collected for emerging technology are often at the lab scale level, based on idealized calculations or estimates, leading to high uncertainties. Parvatker and Eckelman[48] found that stoichiometric calculations could underestimate actual global warming results by 35–50%. To address these challenges, prospective, early-stage, ex ante or anticipatory LCA approaches have been proposed, applying future scenarios to create probable future solution spaces and discussing the scaling of emerging technologies to an industrial scale and its uncertainties.[49–75] Ready-to-use open-source tools have also been provided[76–78] along with recommendations and lessons learnt.[50,51,79,80] A straightforward approach to incorporating uncertainties into LCA, which can also be seen as part of prospective LCA, is to parametrize the Life Cycle Inventory (LCI) and couple it with a Monte Carlo simulation.[81–85] A simplified LCA approach, considering the peculiarities of AP and applying TRL, was proposed by another study.[86]

Beyond methods for scaling, handling of uncertainties, and future scenarios in the case of emerging technologies, different intensity levels can be applied in prospective LCA regarding future solution spaces by modifying foreground and background model data.[49,79] Methods, tools, and the application for AP were covered in a previous paper.[87] We found that climate change impacts in the emerging technology may be underestimated compared to the reference technology if the background database is not adjusted according to results from integrated assessment models (IAM). Moreover, a reduced trade-off between climate change and land use was observed.[87]

The terminology surrounding future-oriented LCA for emerging technologies is not fully determined yet and under discussion.[88,89] Following the recommendation of Arvidson et al.[89] we use the term "prospective LCA" in this study, as our product system is modeled at a future point in time relative to the study's timeframe. To enable a prospective environmental assessment of AP, despite its early stage of development, this study aims to 1) build a simplified LCI of AP based on theoretical and lab-scale data for PC and PEC systems, 2) parameterize the LCI according to unknown technical parameters for sensitivity and uncertainty analysis, 3) implement recent adjustments for scaling the background model data over time to allow for a prospective assessment, 4) develop a sensitivity analysis to determine the minimum requirements for selected technical parameters, and 5) extend the uncertainty analysis by probabilistic simulations to provide a range of possible results depending on the unknown technical parameters. To reach these objectives, the following key contributions are made by this article. 1) A simplified process design calculation and consequently a LCI for the emerging technology AP according to its current state of development are presented, focusing on two product systems: a) synthesis gas; and b) hydrogen peroxide and methane, including the LCI for competing and reference product systems; 2) A methodological approach to enable and enhance prospective LCA of very early-stage emerging technologies with uncertain parameters is evaluated, using partly lab-scale data set in the future, applied with an open-source graphical user interface (GUI) software; 3) Sensitivity analysis is conducted to assess the modification needed to achieve comparable performance to reference or competing product systems (break-even-sensitivity analysis); 4) LCA software is coupled with a Monte Carlo simulation, enabling the assessment of theoretical competitiveness and providing the distribution and range of Life Cycle Impact Assessment (LCIA) results; and 5) LCIA results of AP with fixed foreground and modified background data are presented compared to a reference and competing product system as well as a contribution analysis identifying optimization needs within the considered AP technologies.

2. Experimental Section

LCA served as primary method for this study. Thus, we followed LCA methodology, adapting it to a prospective LCA approach. A brief description of the LCI is provided in the Experimental Section. A comprehensive description, along with the complete LCI, enabling the reconstruction of the model, is available in the Section S2, Supporting Information.

2.1. Goal Definition

The LCA was conducted to a great extent in compliance with the DIN ISO standards 14 040, 14 044, and ILCD recommendations.[31–34,90] It aims to assess the environmental performance of the chemical intermediates: 1) synthesis gas; and 2) hydrogen peroxide and methane, produced through AP, utilizing recycled or captured CO_2. The selection of these final products and the design of the product system, including the choices such as photocatalyst materials and reactor selection, was based on the data collected in two research projects. Consequently, the scope of the LCA was limited by the data availability and the specific pathways defined in these projects.

Synthesis gas, a mixture of hydrogen (H_2) and carbon monoxide (CO), is a key intermediate for producing value-added industrial chemicals, synthetic fuels such as methanol, dimethyl ether, and Fischer–Tropsch diesel.[91–93] Hydrogen peroxide (H_2O_2) is widely used across various industrial areas as bleaching agent, precursor for chemicals in chemical industry, and detoxifier in environmental protection. It is also discussed as a potential renewable energy carrier.[94–96] Methane (CH_4), the primary constituent of natural gas, is utilized as fuel, as a source for synthesis gas and hydrogen production through steam methane reforming (SMR).[97] Currently, these products are predominantly derived from fossil fuels.

Synthesis gas is primarily produced from natural gas, accounting for 62% of global production,[98] and serves as a precursor of hydrogen, which is typically realized by SMR to an extent of 95% in the United States[99] and in the European Union.[100] This leads to a significant amount of GHG emissions, ranging from 1 to 2.3 kg CO_2-equivalents (eq) per kg of synthesis gas (climate change impacts are not directly comparable because the H_2-to-CO ratio varies as a function of production technology and feedstock).[101–104] Hydrogen peroxide is primarily produced via the anthraquinone process, which also relies on hydrogen produced by SMR, resulting in a cumulative energy demand of 23 MJ kg^{-1} of hydrogen peroxide and climate change impacts of 1.13 kg CO_2-eq per kg.[105] Methane is mainly produced from natural gas.[97]

To meet global demand while mitigating climate change and avoiding the exceedance of other planetary boundaries, nonfossil-based production routes for these chemical intermediates are crucial. AP technologies offer an alternative production process based on renewable feedstock materials such as biogas, CO_2, and water, using light as a renewable energy source. Utilizing recycled CO_2 from industrial activities like ammonia or cement production, or CO_2 captured directly from the atmosphere through Direct Air Capture (DAC), AP can contribute to temporary or permanent CO_2 removal as an additional benefit. This makes AP a promising approach for environmentally advantageous chemical production.

It is essential to analyze the overall environmental performance of chemical intermediates produced by AP compared to conventional product systems to understand the environmental benefits and potential drawbacks of AP. Given that AP is an emerging technology, this study is based on idealized calculations and lab-scale experiments corresponding to a TRL of 2–3 and is therefore subject to high uncertainties (see also Section S2.1 and S2.2, Supporting Information).

The LCA results are intended to provide insights into the environmental benefits and drawbacks of AP compared to conventional systems, identify bottlenecks, and highlight optimization potential within AP technology. The study aims to support decision-making in the field of chemical intermediate production and inform AP developers, particularly in the scientific community, about key impact drivers and areas for improvement.

Due to the relatively small production volumes of the model compared to global production of reference products, this LCA reflects small-scale or "microlevel decision support" according to ILCD recommendations.[32] Small-scale consequences are unlikely to overcome thresholds and trigger large-scale consequences in the market. Consequently, an attributional approach was used for this LCA.

2.2. Product System

Two product system groups were defined: synthesis gas as H_2+CO and the combined products hydrogen peroxide and methane as H_2O_2&CH_4. These groups encompassed the emerging technology AP, more precisely the PC and PEC as well as the respective reference (REFERENCE) and competing (COMPETING) product systems. Additionally, the H_2+CO group included the upstream supply chain for carbon dioxide and methane while the H_2O_2&CH_4 group included the upstream supply chain for carbon dioxide and water (**Figure 1** and **2**).

The product systems incorporated a photoreactor combined with a photocollector, photocatalysts, and, in the case of PEC, photoelectrodes, and a PEC cell, along with the associated manufacturing processes. The photoreactor technology for the PC product system was assumed to be based on concentrated parabolic trough collectors. In the PEC product system, a gas diffusion photocell was integrated, assumed to be connected to a PV collector frame. In the PC product system, pressure swing adsorption was assumed to enable the recovery of unreacted CO_2 and compression to offset pressure losses in the parabolic trough collector system. For the PEC product system, methanation was assumed to convert residual gases to methane.

Several scenarios were considered within the product systems regarding the photocatalyst choice, the CO_2 sources, and the IAM scenarios. For the PC photocatalyst material, three options were evaluated based on laboratory-scale production: TiO_2 with gold (Au/TiO_2) and a catalyst based on graphitic carbon nitride (WO_3—C_3N_4—Rh—Cr_2O_3). A third option involved an optimized version of graphitic carbon nitride (WO_3—C_3N_4—Ni), which was considered likely for future implementation. Four different CO_2 sources were analyzed: CO_2 captured from the atmosphere by DAC technology, CO_2 from biogas purification, and CO_2 from ammonia or cement production.

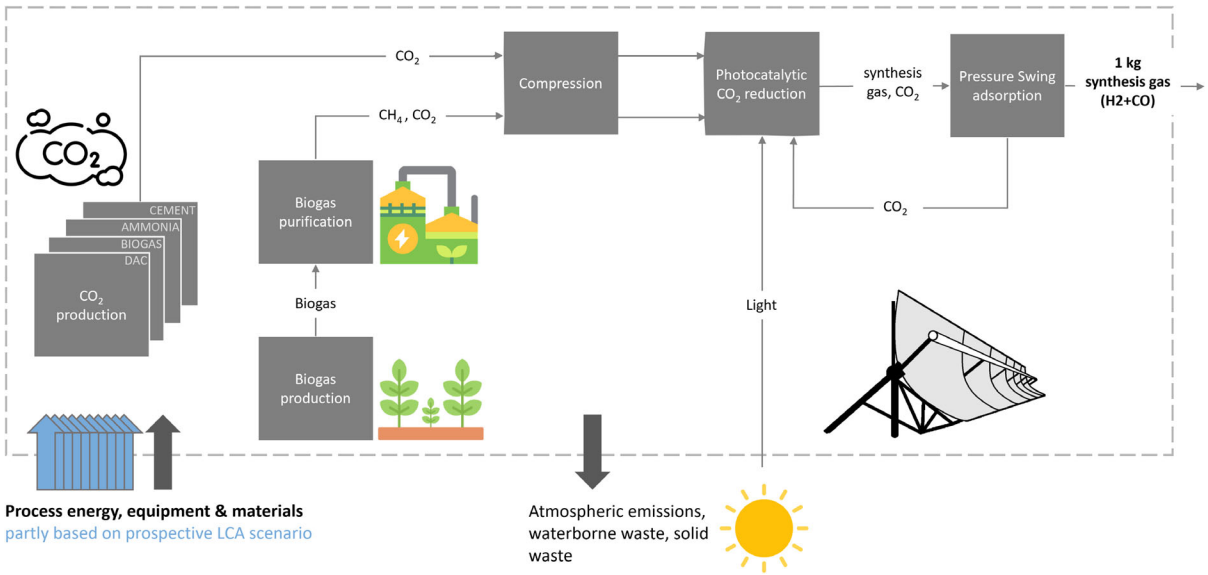

Figure 1. Schematic diagram of PC production of synthesis gas (H_2+CO) from biogas and CO_2. Upstream and downstream processes, materials, services, and emission flows are included in the depicted process. Icons created by http://flaticon.com.

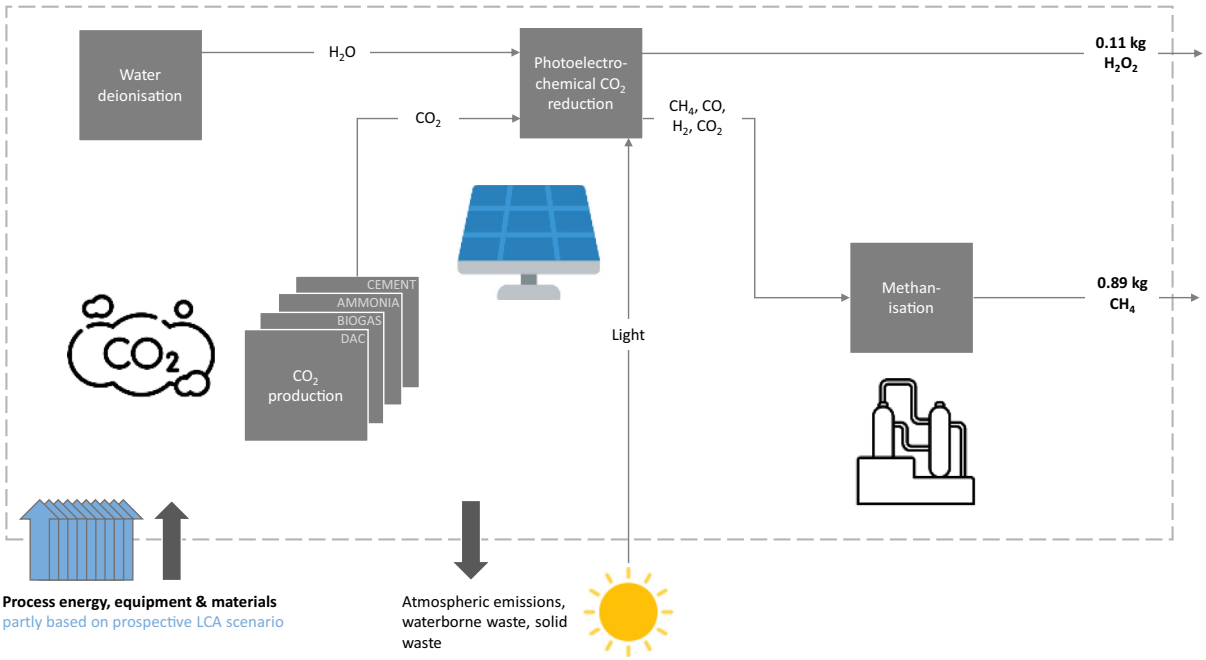

Figure 2. Schematic diagram of PEC CO_2 production of hydrogen peroxide and methane ($H2O_2$&CH_4) from water and CO_2. Upstream and downstream processes, materials, services, and emission flows are included in the depicted process. Icons created by http://flaticon.com.

The IAM scenarios corresponded to results of the IAM REMIND model,[106,107] integrated with the premise tool.[78] REMIND calculates the electricity mix and the industrial sector composition needed to meet specific climate goals, such as the Paris Agreement objectives in the PkBudg500 scenario. The IAM scenarios were utilized to project future conditions and maintain consistency within the prospective LCA. Based on REMIND outputs, energy-intensive sectors (power generation, cement, steel production, freight and passenger road transport, and the supply of conventional and alternative fuel) were adjusted in the ecoinvent database with updated efficiencies.[78] Further details on modifications made by the premise tool are available in its documentation.[77] The scenarios considered are summarized in **Table 1**, with detailed descriptions of the corresponding scenario processes provided in the LCI (Section 2.10 and S2, Supporting Information).

2.3. Reference and Competing Product System

The reference product systems were modeled with results from process simulations,[103,108–110] which represented mature technologies, along with data from the ecoinvent[105] inventory database. For the H_2+CO product system group, SMR was used as reference, with an additional reverse-water–gas-shift reaction to maintain the required H_2-to-CO ratio. For the H_2O_2&CH_4 product system, the anthraquinone process as provided in ecoinvent

Table 1. Overview of scenarios for H_2+CO PC, H_2O_2&CH_4 PEC, REFERENCE and COMPETING product systems. The default scenario choices for the impact assessment and sensitivity analysis are underlined.

	H_2+CO PC	H_2O_2&CH_4 PEC	REFERENCE	COMPETING
Photocatalyst scenarios	<u>Au/TiO_2</u> WO_3-C_3N_4-Rh-Cr_2O_3 WO_3-C_3N_4-Ni	<u>$CuNb_{13}O_{33}$</u> & <u>$BiVO_4$</u>		N/A
CO_2 scenarios		<u>Direct Air Capture (**DAC**)</u> **Biogas** purification **Ammonia** production **Cement** production		
REMIND **IAM scenarios**		<u>**2030/2050 (for each IAM scenario):**</u> <u>**Base**: Business as usual, ≈3.5 °C</u> **NPi**: National Policies Implemented, ≈3.3 °C **NDC**: Nationally Determined Contributions, ≈2.5 °C **PkBudg1150**: Paris Agreement objective, ≈2 °C **PkBudg500**: Paris Agreement objective, ≈1.5 °C		

was utilized. Competing nonfossil-fuel-based product systems were added to the LCA to broaden the comparison base, including another low TRL. This approach allows for a better comparison of nonclimate-change-related impact categories, in which fossil fuel systems might have an advantage over renewable product systems. For the competing H_2+CO product system, dry methane reforming (DRM),[78,111] assuming biogas as feedstock, and electrolysis[112] were modeled. For the H_2O_2&CH_4 product system, the anthraquinone process was modeled with hydrogen produced via electrolysis.[105]

2.4. System Boundaries & Allocation Procedures

In the background model data, economic allocation according to the ecoinvent standards was used alongside the ecoinvent cutoff system model for burdens and credits related to recycling.[113] Consequently, we applied economic allocation to the foreground modeling and assessed the product systems from an attributional perspective. However, we also integrated system expansion using the avoided burden approach as an alternative in the uncertainty analysis to account for potential result deviations due to the allocation method. System expansion is recommended by ISO 14 040[33] and 14 044[34] but adds additional challenges and dependencies in the results.

2.5. Functional Unit & Reference Flow

Based on the described goal and product system of the LCA, two functional units and reference flows are defined. For the first case we focused on the production of H_2+CO: 1 kg of synthesis gas (side products are not separated but included in the functional unit in H_2+CO because further processing of product is not defined; therefore, product composition and allocation of coproducts is unclear) with a molar H_2:CO ratio of ≈0.89:1, including side products and for the second case, focused on $H_2O_2 + CH_4$: 1 kg of the combined products of 0.89 kg of hydrogen peroxide and 0.11 kg of methane, provided either by the emerging AP technology or by a reference or competing product system.

For the assessment of photocatalyst materials in the case of H_2+CO (S3.1.3, Supporting Information), the functional unit corresponds to 1 kg of the respective photocatalyst material: TiO_2-based (Au/TiO_2), g-C_3N_4-based (WO_3—C_3N_4—Rh—Cr_2O_3 or WO_3—C_3N_4—Ni). The functionality is simplified and assumed to be equivalent across materials, with the same total mass of photocatalyst material being used.

2.6. Life Cycle Impact Assessment Methods

For the LCIA, ILCD midpoint 2.0[114] impact assessment method collection was used which categorizes impacts based on the uncertainty of the method. Additionally, we included global temperature potential (GTP) for 100 years from IPCC, cumulative energy demand (CED), and total land occupation (own implementation) to calculate the area occupied per year by the product system. GTP provides a different balance between short and long-term GHGs compared to the common GWP 100 approach which is widely applied but lacks a scientific basis for exclusive use.[115] The CED helps estimate process efficiency, while land use, including urban and agricultural areas, shows the total area used, offering a straightforward assessment. This is particularly useful compared to the land use impact category in ILCD, which additionally considers land quality and thus can pose challenges if the characterization factors are not individually adjusted to the specific regional land use and transformation. In the context of AP applications, land use coverage is crucial due to the dependency on land for solar irradiation collection, making land use an essential additional metric. A comprehensive overview of the impact categories and the impact assessment methods is provided in the Supporting Information (S1).

2.7. Sensitivity and Uncertainty Analysis

In the sensitivity analysis, we focused on evaluating key parameters to understand their influence on the results and additionally to determine the break-even point with the reference and competing product systems. This approach allows us to provide recommendations on the parameter values at which the assessed product system could become competitive. The parameters that were varied for the PC system included solar-to-chemical efficiency, lifetime, Direct Normal irradiation, reaction conversion rate, and parabolic trough concentration factor. For the PEC system, the parameters included solar-to-chemical efficiency, Global Horizontal Irradiation, lifetime, rooftop installation, and PEC cell material input. Solar-to-chemical efficiency represents a modified form of the usual solar-to-hydrogen efficiency but is not limited to hydrogen.

Given the number of parameters with uncertain values, a prospective LCA for this product system involves numerous modeling options. To address these uncertainties and avoid arbitrary parameter value selections, we conducted a Monte Carlo simulation with 17 system design and methodological parameters, each varied across 1,000 runs. Additionally, the IAM scenarios were varied by year (2030/2050) and mitigation scenario, resulting in a total of 10 000 runs.

To facilitate both sensitivity and uncertainty analyses, we developed a Python script that generates scenario files and enables subsequent data analysis. This script (Jupyter Notebook), including details on the probability distribution of each parameter, is provided in the Supporting Information.

2.8. Software Tools and Databases

For the LCA and LCIA, we utilized the open-source software Activity Browser,[116] which is based on the brightway2[117] framework connected with the ecoinvent 3.8 database[105] using the cutoff system model. The ecoinvent database was modified using the premise tool[78] (version 1.4.2) and premise-gwp (version 0.7) to integrate the IAM results from REMIND into the background inventory database. For sensitivity and uncertainty analyses, we used Python 3.8.4 and additional python libraries. A full list of libraries is presented in the Jupyter Notebook available in the Supporting Information.

2.9. Data Sources, Quality, and Representativeness

Process design data of the AP process was derived from theoretical considerations and ideal calculations, for the H_2+CO

product system established by LIKAT Rostock (Leibniz Institute for Catalysis, heterogeneous photocatalysis: https://www.catalysis.de/en/research/heterogeneous-photocatalysis) and TU Berlin (Technical University of Berlin, reaction engineering: https://www.tu.berlin/en/reaction-engineering). In the case of the $H_2O_2 + CH_4$ PEC product system, the DECHEMA research institute (https://dechema-dfi.de/en/Topics+_+Projects/Photocatalysis.html) provided theoretical considerations for the calculations. Photocatalyst material data for the H_2+CO PC product system was collected from the Carl von Ossietzky University of Oldenburg (https://uol.de/tc-wark). Electrode materials in $H_2O_2 + CH_4$ were provided by the DECHEMA research institute and TANIOBIS (https://www.taniobis.com/de/standorte/taniobis-gmbh/). Materials for a prototype reaction cell and a methanation unit in $H_2O_2 + CH_4$ were supplied by the neoxid group (https://www.neoxid-group.de/en/). Potential materials for the membrane and electrolyte were provided by the DECHEMA research institute. Additional processes for the production of the electrodes, catalyst materials, solar reactors, piping, and compression were based on literature and database data. Supply chain data was sourced from literature and databases. Detailed information on these processes and data are provided in the following Section (2.10) and in the Supporting Information (Section S2.1 and S.2.2). Further information on the technology behind the H_2O_2&CH_4 PEC product system is available in the project report and publications from the project partners.[118–121] A report on the H_2+CO PC products system will also be published and accessible through the Leibniz Information Centre for Science and Technology Unversity Library (TIB) using the project name "PRODIGY" as a keyword.[122]

2.10. LCI

The LCI for the H_2+CO PC and H_2O_2&CH_4 PEC product systems was developed based on theoretical considerations. The PC product system was estimated to be at TRL 2, while PEC was at 3, as a prototype was already been developed. Thus, both product systems are at the basic technology research level. Creating an LCI for emerging technologies at this TRL requires prospective LCI generation methods, as plant or industry data is not yet available. We followed the hierarchy of methods proposed by Parvatker and Eckelman[48] and the decision tree developed by Tsoy et al.[72] Based on data, provided by the project partners described in Section 2.9, a stochiometric calculation of the dry reforming of methane reaction was conducted for H_2+CO considering the conversion rate and potential side reactions. The required light-capturing area, coated with the photocatalyst, was determined based on the reaction enthalpy. For $H_2O_2 + CH_4$ a stoichiometric calculation and a Faradaic calculation was applied to determine the mass and energy flows. A full description of both calculations is available in the Supporting Information (Section S2.1 and S2.2). These calculations also guided the selection and estimation of hardware and infrastructure, which remained theoretical at this stage. Further details can be found in Section S2.1.2–S2.2.6, Supporting Information. Additionally, laboratory data provided by the project partners was used for the production of photocatalyst and photoelectrodes, as detailed in the Supporting Information (Section S.2.1.3, S.2.2.3–2.2.5). Further supply chain materials such as carbon dioxide, methane, biogas, and water which were not available in the scope of the research projects were sourced from literature and databases (Section S.2.3 and S.2.4, Supporting Information). Data for the reference and competing product systems were derived from literature and databases and can be found in Section S2.6 and S2.7, Supporting Information.

Table 2. Default values of key parameters for LCIA and sensitivity analysis. The defined values are based on assumptions.

Parameter	H_2+CO	H_2O_2&CH_4
Solar-to-chemical efficiency	5%	5%
Parabolic concentration	50	–
Reaction conversion rate	60%	–
Direct Normal Irradiation	1074 kWh m^2 a^{-1}	
Global Horizontal Irradiation	–	1321 kWh m^2 a^{-1}
Lifetime (system)	30 yrs	40 yrs
Lifetime components (photocatalysts or PEC cell lifetime)	10 yrs	20 yrs
Roof mounted	–	No
PEC cell material input	–	10%

From a methodological perspective, recent advancements in prospective LCA were incorporated regarding LCI data generation methods and background data modification. Several scenarios were constructed, including complete database modifications with IAM results (Table 1 and Section S2.5, Supporting Information).[78] This approach enabled the evaluation of both new and reference technologies within a prospective future context. Further methodological advancements in prospective LCA were implemented: Due to the low TRL, the LCI was partly parameterized, allowing for modified sensitivity and uncertainty analyses. The default parameter set is presented in **Table 2**. If idealized calculations or proxy processes were used, scale-up techniques would not be considered. In the case of photocatalyst material production, we utilized both idealized calculations and lab-scale data provided by project partners, with no scaling applied. For the PEC cell in the H_2O_2&CH_4 PEC product system, a simple scale-up was assumed for the photocell by input material mitigation. Recycling of the plants was not separately considered, except for the aspects already integrated into ecoinvent via input materials. Beyond the LCI description provided in the Supporting Information (Section S2), the full LCI is available in the Supporting Excel file, which can be used to reconstruct the model.

3. Results

The LCIA results and sensitivity analyses are provided for the default scenario and default parameter set (Table 2). A complete set of figures covering all midpoint impact categories is included in the Supporting Information (Section S3).

3.1. LCIA of H_2+CO Production

3.1.1. Climate Change

The largest single contribution in the climate change impact category within the PC product system is from biogas production

and CO_2 capture (**Figure 3**a). The latter has a negative balance, thereby reducing overall climate change impacts. The production of the parabolic trough collector follows distantly. The environmental impacts of manufacturing the photocatalysts are minimal when using Au/TiO_2 (0.5%), or WO_3—C_3N_4—Ni (3%) and higher with WO_3—C_3N_4—Rh—Cr_2O_3 (11%). The largest contributions in photocatalyst production stem from electricity, heat, and ethylene production (Figure S3, Supporting Information). For Au/TiO_2, the manufacturing process itself, as well as the provision of titanium dioxide, are particularly significant. In the context of the CO_2 scenarios, the PC product system shows the lowest environmental impacts in the DAC and biogas CO_2 scenarios (Figure 3a).

In comparison to the fossil REFERENCE, SMR product system, the PC product system demonstrates lower environmental impacts in the climate change impact category (Figure 3). Depending on the scenario, the PC system often outperforms the competing product system, which produces synthesis gas via electrolysis (COMPETING, electrolysis). However, in the future IAM scenarios for 2050, the electrolysis product system offers an advantage, because it relies on electricity as an energy source, which is expected to enhance its environmental performance, driven by the anticipated expansion of renewable energy. The second competing product system (COMPETING, DRM), which assumes dry reforming of biogas, generally exhibits a slightly more favorable environmental performance in most scenarios. In the PkBudg500 IAM scenario, the advantage of the dry reforming-based product system (COMPETING, DRM) doubles in terms of negative CO_2-eq emissions compared to the PC product system (Figure 3b). However, CO_2-eq of electrolysis (COMPETING, electrolysis) is even lower in this scenario.

3.1.2. Further Impact Categories

The PC product system outperforms the reference product system in the impact categories freshwater ecotoxicity, nonrenewable energy resources, water use, ionizing radiation, ozone depletion, and photochemical ozone formation (Figure S1 and S2, Supporting Information). The competing product system based on dry reforming (COMPETING, DRM), which was most beneficial in the climate change impact category, exhibits the most favorable environmental performance in only one other impact category: CED. In contrast, the reference product system demonstrates the lowest environmental impacts across most scenarios in the impact categories of acidification, freshwater eutrophication, terrestrial eutrophication, human toxicity, land use, and particulate matter. The most significant contributor to environmental impacts in the other impact categories is the production of biogas, followed by the manufacturing of the parabolic trough collector.

3.2. Sensitivity Analysis of H_2+CO Production

3.2.1. Climate Change

In the climate change impact category, sensitivity analysis of the parameters solar-to-chemical efficiency, reaction conversion rate, and lifetime exhibits an exponential trend and thus a significant range of results (**Figure 4**). At a solar-to-chemical efficiency of 0.1%, the impact is 3.13 kg CO_2-eq, while at 20% efficiency, it decreases to −0.27 kg CO_2-eq. Substantial changes in results occur when the solar-to-chemical efficiency is below 1%, the lifetime below 10 years, and the reaction conversion range below 40%. The PC product system surpasses the fossil reference product system when the solar-to-chemical efficiency reaches ≈0.3%,

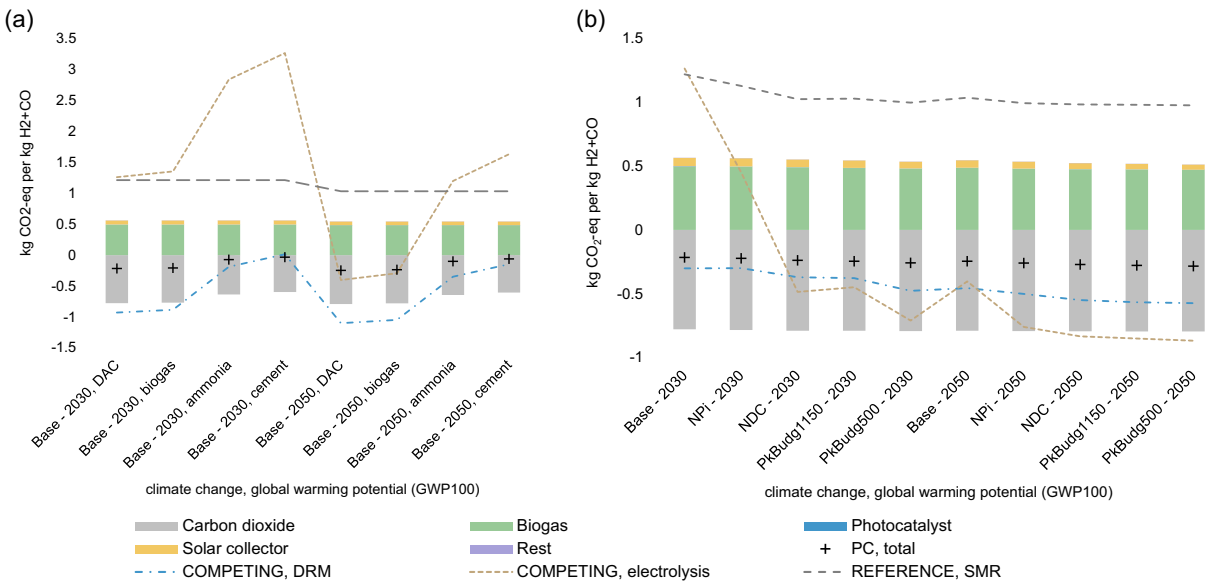

Figure 3. Results of the impact assessment in the climate change impact category for the H_2+CO PC, REFERENCE, and COMPETING product systems, including a) contribution analysis of the PC product system in the Base IAM scenario in the years 2030 and 2050, considering all CO_2 scenarios, and Au/TiO_2 photocatalyst scenario b) taking into account all IAM scenarios in the years 2030 and 2050, DAC CO_2, and Au/TiO_2 photocatalyst scenario. Default values of LCI parameters are used (Table 2). The colored bars represent the contributions of upstream and downstream processes to the total impacts, with markers indicating the overall environmental impacts. The lines are for illustrative purposes only; there are no data points between the scenarios as depicted.

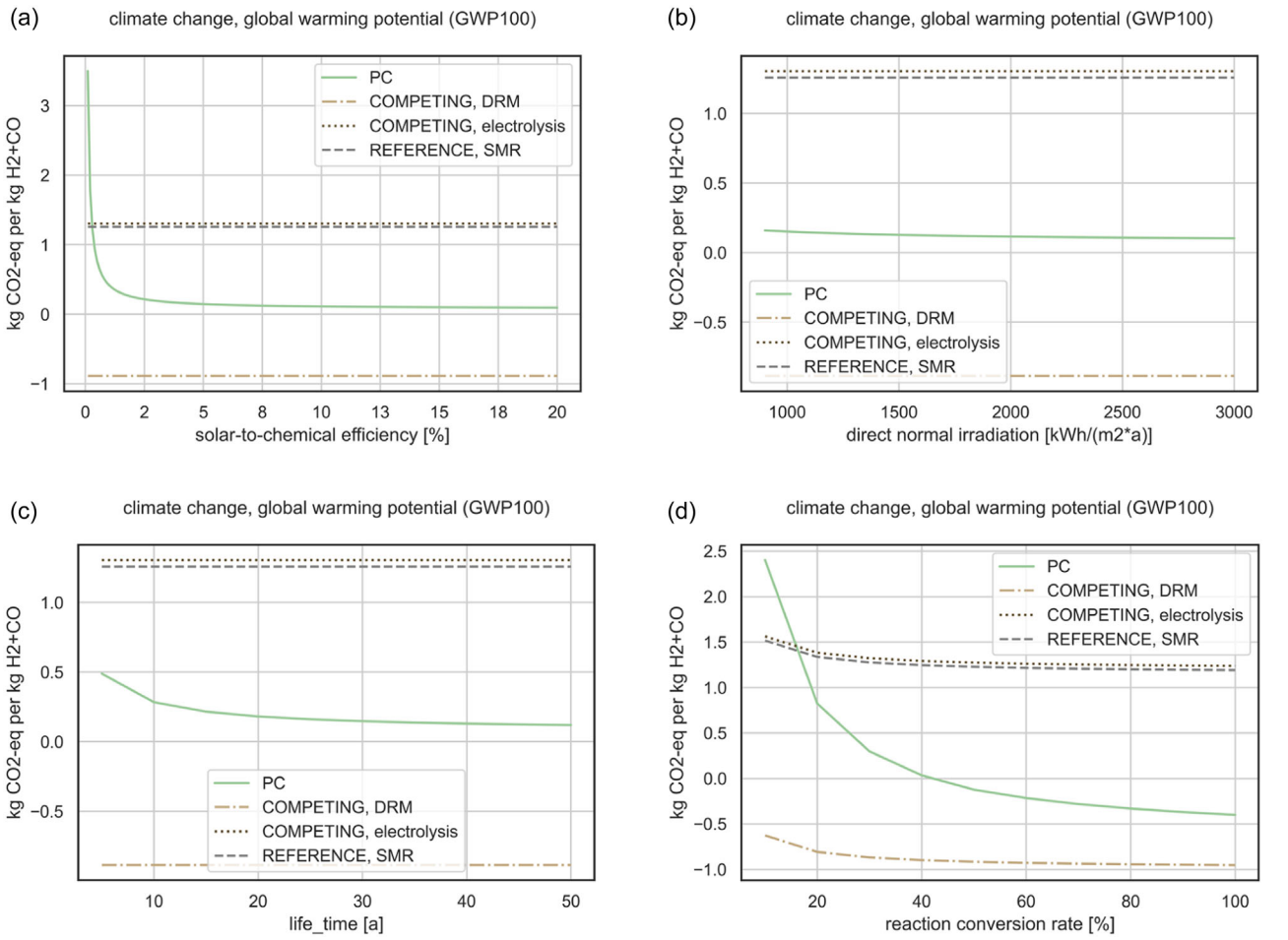

Figure 4. Sensitivity analysis of the LCA in the climate change impact category, illustrating the effects of varying the parameters a) solar-to-chemical efficiency, b) Direct Normal Irradiation, c) lifetime, d) reaction conversion rate. Default scenarios (Table 1) and values of LCI parameters were used (Table 2).

the lifetime more than 5 years, the reaction conversion rate more than 20%, or the Direct Normal Irradiation more than 900 kWh m^2 a^{-1}. A variation in the concentration factor of the parabolic trough collector (Section S3.1.4.2, Supporting Information) has no significant impact on the results.

The competing product system based on DRM cannot be matched by the PC product system, even under optimistic parameter values. Conversely, the electrolysis-based competing product system generally shows less favorable performance compared to the PC product system. Exceptions occur with parameters such as reaction conversion rate and solar-to-chemical efficiency. At very low reaction conversion rates (<20%) and efficiencies (<0.3%), the PC product system may result in higher environmental impacts than the electrolysis-based product system. However, this applies to the GWP but not to the global temperature potential impact category.

3.2.2. Further Impact Categories

When analyzing additional impact categories, the sensitivity curves generally follow a similar trend to those observed in the climate change impact category (Figure S4–S8, Supporting Information). Sensitivity is particularly pronounced within a solar-to-chemical efficiency range of up to ≈5%, a reaction conversion rate below 40%, and a lifetime of less than 10 years. Differences to the climate change impact category mainly arise at the points where the environmental performance of the PC product system intersects with that of reference or competing product systems, if such intersections occur at all. In several impact categories (e.g., acidification impact category, using solar-to-chemical efficiency as a parameter), the PC product system does not achieve comparable or favorable environmental performance relative to the reference product system, even under optimistic parameter values. In contrary, in several other impact categories (e.g., freshwater ecotoxicity, using lifetime as a parameter), particularly under higher parameter values, the PC product system demonstrates favorable or approximately similar environmental performance compared to the competing product systems.

3.2.3. Minimum Requirements

Based on the break-even sensitivity analysis, the threshold values of the parameters, required to achieve competitiveness, are presented in **Table 3**. For additional impact categories, the corresponding

Table 3. Climate change impact threshold values of the parameters. Assessed under the DAC CO_2 and base 2030 IAM scenario.

	Climate change impact treshold		
Parameter	H_2+CO PC to REFERENCE	H_2+CO PC to COMPETING DRM	H_2+CO PC to COMPETING electrolysis
Solar-to-chemical efficiency	0.3%	Not possible[a)]	0.3%
Parabolic concentration	30%[a)]	Not possible	30%[a)]
Reaction conversion rate	≈20%	≈20%	Not possible
Direct Normal Irradiation	900 kWh m^2 a^{-1}[a)]	Not possible[a)]	900 kWh m^2 a^{-1}[a)]
Lifetime (system)	5 yrs[a)]	Not possible[a)]	5 yrs[a)]

[a)]The results are limited by the considered scope and resolution (e.g., lifetime starting from 5 years); threshold values might be different or existent out of scope.

values can be derived from the graphs provided in Section S3.1.4, Supporting Information.

3.3. Uncertainty Analysis of H_2+CO Production

In the climate change impact category, the PC product system ranges from a minimum of −0.36 kg CO_2-eq per kilogram of product to a maximum of 0.66 kg CO_2-eq per kilogram (**Figure 5**a). The reference product system has a similar range (0.84–1.91 kg CO_2-eq), while the competing product systems show a broader range (DRM: −1.46 to 0.02 kg CO_2-eq; electrolysis: −1.60 to 3.27 kg CO_2-eq per kg of product). Due to the lack of access to specific parameters for these competing technologies, their result ranges are primarily influenced by background model data, such as the power source, particularly for electrolysis. The range of results for the PC product system in the climate change impact category is similar to, or smaller than, that of the competing and reference product systems.

The analysis of further impact categories (Figure S9, Supporting Information) indicates that in some impact categories, such as water use, an optimistic scenario might result in an environmental performance comparable to or even better than the reference product system. In certain impact categories, such as ozone depletion, the competing electrolysis product system shows a wider range of results than PC. In contrast, the DRM and reference product systems exhibit a narrower range across several impact categories including land use (Figure 5b).

In terms of median climate change values, DRM exhibits an advantageous environmental performance, followed by electrolysis, which has a much larger spread. PC ranks third, followed by the reference product system at a considerable distance. In the land use impact category, the reference product system shows the best environmental performance (Figure 5b). Considering all impact categories, PC demonstrates advantageous environmental performance in the median across most impact categories. However, PC also has the greatest range of results in these impact categories, while the competing DRM and reference product systems have the narrowest ranges.

3.4. LCIA of H_2O_2&CH_4 Production

3.4.1. Climate Change

The largest contributor to the climate change impact within the PEC product system is the manufacturing of solar panels, particularly due to the production of the mounting systems (**Figure 6**a). The share of anode production can be reduced by 72% in 2050 through an optimized energy mix. Additionally, the shares of the

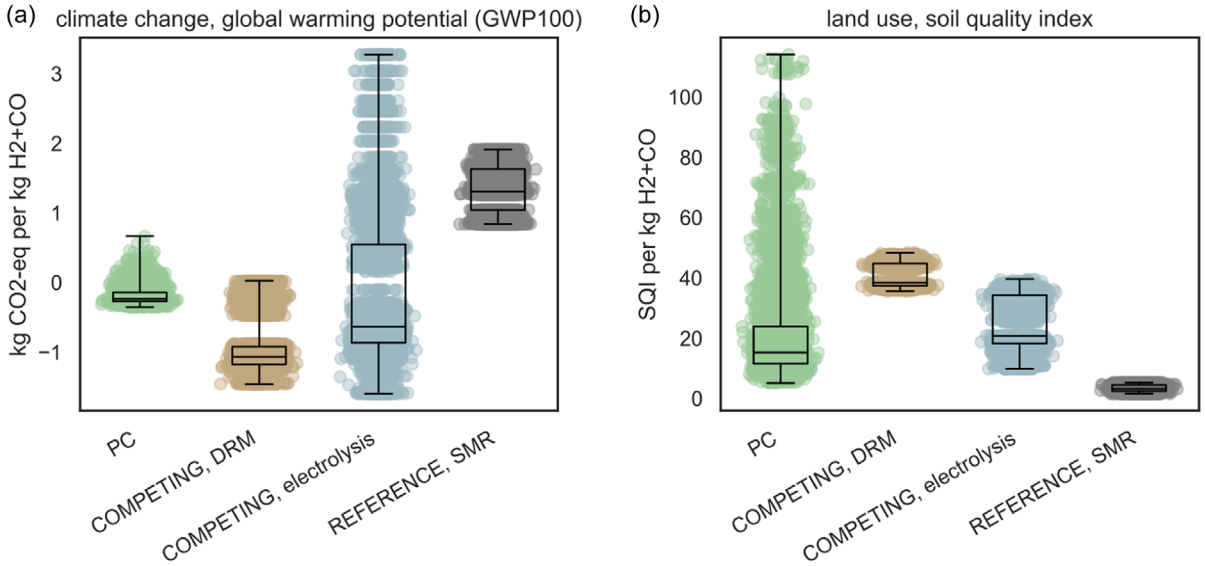

Figure 5. Uncertainty analysis results combined with impact assessment for the climate change a) and land use b) categories. The analysis compares the PC product system with two competing technologies: DRM and electrolysis, as well as a reference product system (REFERENCE, SMR) based on SMR of natural gas. The scatter plots display the distribution of results, while the error bars indicate the full range of outcomes. The horizontal bars represent the lower and upper quartiles, with the median value marked in between.

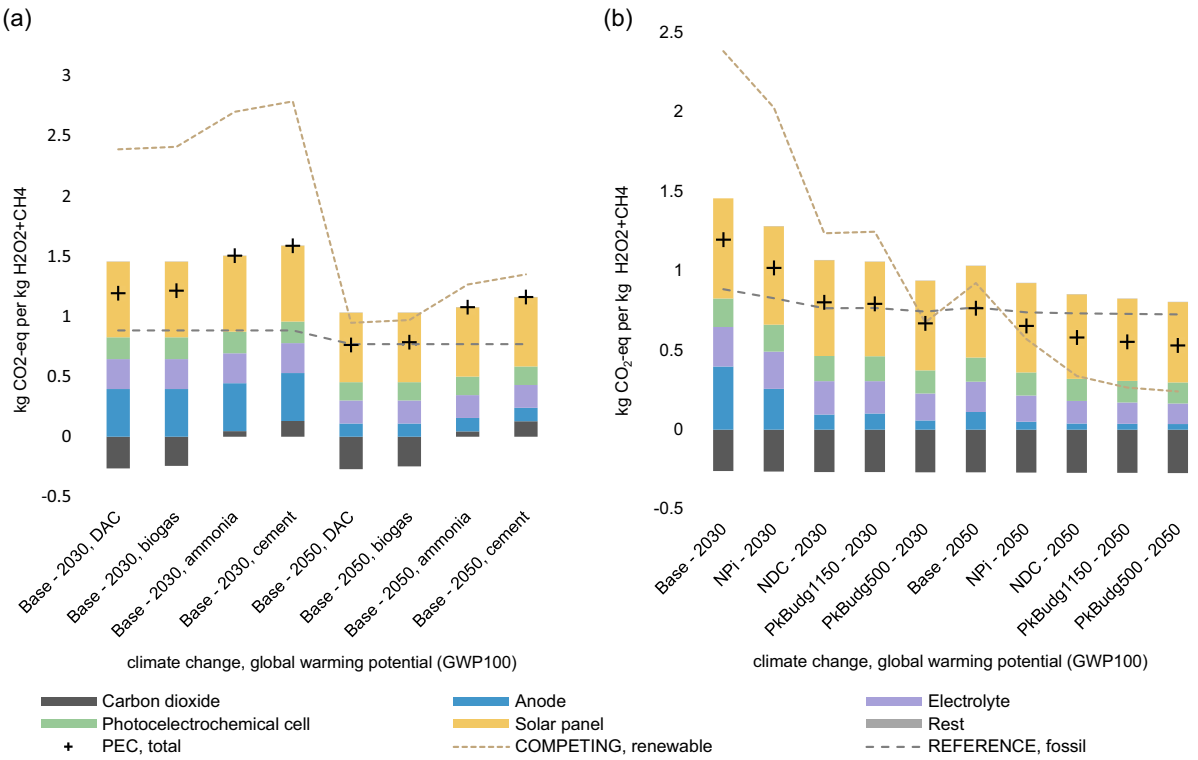

Figure 6. Impact assessment results in the climate change impact category for the PEC, REFERENCE, and COMPETING product systems, including a contribution analysis of the PEC product system. a) Assessment under the Base IAM scenario, considering all CO_2 scenarios. b) Assessment under the DAC CO_2 scenario, considering all IAM scenarios. Default values of LCI parameters were used (Table 2). The colored bars represent the contributions of upstream and downstream processes to the total impacts, with markers indicating the overall environmental impacts. The lines connecting the bars are for illustrative purposes only; there are no intermediate values between the scenarios.

electrolyte and the PEC cell are expected to be reduced by 23% and 14%, respectively. The PEC product system demonstrates the lowest environmental impacts in the DAC and biogas CO_2 scenarios.

When compared to the reference product system, the PEC product system exhibits ≈35% higher environmental impacts in 2030 (Base scenario) under the basic assumptions. However, by 2050, with an improved energy mix, particularly in the CO_2 scenarios involving DAC and biogas, the PEC product system's impacts are expected to closely align with those of the reference product system. Considering all IAM scenarios (Figure 6b), the PEC system can outperform the reference product system by ≈27% in the optimistic PkBudg500-2050 scenario.

The competing product system, on the other hand, shows nearly double the environmental impact in the climate change category in the 2030 scenario (Figure 6). This impact is reduced to about 20% higher CO_2-eq in the 2050 scenario. However, the competing technology is more dependent on the energy mix; thus, in optimistic energy scenarios, the competing product system demonstrates a significant advantage, with ≈55% lower environmental impacts in the PkBudg500-2050 IAM scenario. This advantage becomes apparent starting from the NDC-2050 IAM scenario.

3.4.2. Further Impact Categories

The largest contributions to environmental impacts arise from the solar panels, followed by the PEC cell. The electrolyte plays a particularly significant role in the impact on freshwater ecotoxicity. The differences in results across various CO_2 scenarios are relatively minor in most other impact categories, with the exception of water use and nonrenewable energy resources (Figure S10, Supporting Information). In several impact categories, the PEC product system does not demonstrate a significant advantage over the reference product system across all IAM and CO_2 scenarios (Figure S10 and S11, Supporting Information). This trend is also observed when comparing the PEC with the competing product system, particularly for 2050. However, exceptions include the categories of nonrenewable resource consumption, which follow a trend similar to the climate change impact category, and water use, where the PEC product system shows a reduction in environmental impacts compared to both the reference and competing product systems. Additionally, in the material resources impact category, the PEC product system proves advantageous over the competing product system.

3.5. Sensitivity Analysis of H_2O_2&CH_4 Production

3.5.1. Climate Change

In the climate change impact category, a sensitivity analysis of solar-to-chemical efficiency, Global Horizontal Irradiation, solar panel lifetime, as well as PEC cell lifetime reveal a significant range of results (**Figure 7**). The environmental impact is

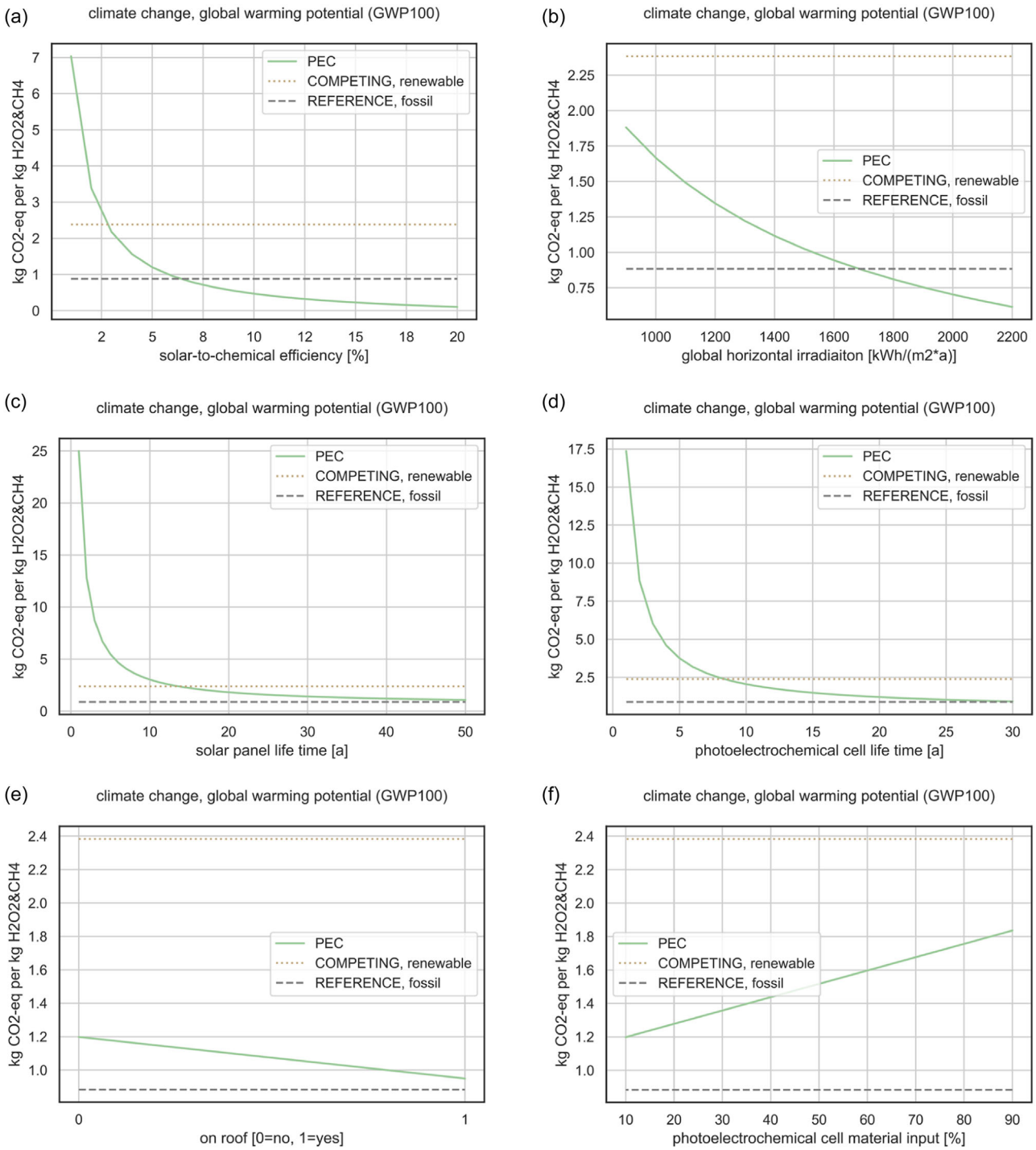

Figure 7. Results of the sensitivity analysis of the LCA in the climate change impact category, considering variations in the parameters a) solar-to-chemical efficiency, b) Global Horizontal Irradiation, c) solar panel lifetime, d) photocell lifetime, e) on-roof or ground-mounted installation, and f) PEC cell material input. Default scenarios (Table 1) and values of LCI parameters were used (Table 2).

25 kg CO_2-eq for a lifetime of one year, reducing to about 1.1 kg CO_2-eq at 50 years. The sensitivity curves, particularly of the solar-to-chemical efficiency, solar panel, and PEC cell lifetime, follow an exponential trend. In solar-to-chemical efficiency below 5% and lifetimes lower than 10 years the change in the results is high.

A similar environmental performance advantage compared to the fossil-based reference product system in the climate change impact category is achieved at a solar-to-chemical efficiency of 7%, a Global Horizontal Irradiation of more than 1700 kWh m^{-2} a^{-1}, or a lifetime of more than 50 years. Even with significant material

savings (PEC cell material input), environmental impacts cannot be reduced below those of the reference product system. A rooftop installation improves environmental performance, approaching that of the reference product system. Compared to the competing product system at a solar-to-chemical efficiency of 3%, a solar panel lifetime of more than 14 years, or a PEC cell lifetime of more than 9 years the environmental performance of the PEC product system becomes beneficial.

3.5.2. Further Impact Categories

The analysis of additional impact categories reveals that the sensitivity curves generally follow a similar trend as observed in the climate change impact category (Figure S12–S17, Supporting Information). Sensitivity is particularly pronounced in the solar-to-chemical efficiency range of 1–5% and for lifetimes of 1–8 years (Figure S12 and S14, Supporting Information). However, differences arise in the points of intersection with the environmental performance of the competing and reference product systems. In some impact categories, these intersections occur within solar-to-chemical efficiency ranges of 11–20% (Figure S12, Supporting Information). Notably, in the categories of land use and ozone depletion potential, achieving a more advantageous environmental performance is not possible, even at high efficiencies. When considering Global Horizontal Irradiation (Figure S13, Supporting Information), it is possible to achieve a favorable or approximate environmental performance relative to the reference product system in several impact categories. Varying the lifetime parameter (Figure S14, Supporting Information) for solar panels and the PEC cell (Figure S15, Supporting Information) shows that, in some impact category, matching the environmental performance of the reference product system is either unachievable or only possible with very long lifetimes. Assuming an on-roof installation (Figure S16, Supporting Information) instead of a ground-mounted one does not lead to significant changes in the technology ranking between the PEC, competing, and reference product systems, in most impact categories. Scaling down material consumption (Figure S17, Supporting Information) for the PEC cell results in a shift from disadvantageous to advantageous environmental impacts relative to the competing product system in several impact categories.

3.5.3. Minimum Requirements

Based on the break-even sensitivity analysis, the threshold values of the parameters required to achieve competitiveness are presented in **Table 4**. For additional impact categories, the corresponding values can be derived from the graphs provided in Section S3.2.3, Supporting Information.

3.6. Uncertainty Analysis of H_2O_2&CH_4 Production

The uncertainty analysis highlights the variability in PEC results, which stems from the low TRL and the associated lack of complete or reliable parameter values (**Figure 8**). The findings suggest that, in most impact categories, the PEC system could potentially approximate the performance of reference and competing product systems under optimistic conditions. However, results based on less favorable parameter sets could be substantially higher.

Table 4. Climate change impact threshold values of the parameters. Assessed under the DAC CO_2 and base 2030 IAM scenario.

	Climate change impact treshold	
Parameter	H_2O_2&CH_4 PEC to REFERENCE	H_2O_2&CH_4 PEC to COMPETING
Solar-to-chemical efficiency	7%	3%
Global horizontal irradiation	1700 kWh m^2 a^{-1}	Not possible[a)]
Solar panel lifetime	Not possible[a)]	14 yrs
Lifetime components (photocatalysts or PEC cell lifetime)	Not possible[a)]	9 yrs
Roof-mounted	Not possible	Not possible
PEC cell material input	Not possible[a)]	Not possible

[a)]The results are limited by the considered scope and resolution (e.g., lifetime starting from 5 years); threshold values might be different or existent out of scope.

In the climate change impact category, the estimated minimum impact is ≈−0.11 kg CO_2-eq per kg of product, while the maximum could reach as high as 180.64 kg CO_2-eq per kg. In contrast, the uncertainty range for the reference and competing product systems is significantly narrower, ranging from 0.24 to 2.38 kg CO_2-eq per kg for the former and 0.73 to 0.88 kg CO_2-eq per kg for the latter. Although we did not have access to specific parameter data for these technologies, nevertheless, the reference and competing product systems are based on a well-established and mature process (the anthraquinone process). A similar pattern of high uncertainty is observed across other impact categories for the PEC product system. Considering the medians, the impacts of the PEC product system are about three times higher than those for the competing renewable and reference fossil product systems.

4. Discussion

4.1. Results

For the H_2+CO PC product system, our assessment based on fixed parameters indicated that the climate change impacts ranged from −0.28 and −0.03 kg CO_2-eq, depending on the IAM and CO_2 scenario. This suggests that the production of synthesis gas could be carbon negative or carbon neutral if the assumptions are valid. In ecoinvent and the literature, the result range for synthesis gas climate change impacts is between −3.3 and 5.4 kg CO_2-eq,[101–105,112,123–126] depending on the technology and not considering any methodological, data, or impact assessment specific differences, which cannot be accounted for in such a crosscomparison without access to the original models. Furthermore, different H_2 to CO ratios, which could significantly alter the results, were not modifiable. Particularly, synthesis gas production based on biomass and as product from electrolysis with a beneficial electricity mix such as wind power provided a negative to carbon-neutral GWP. Since our

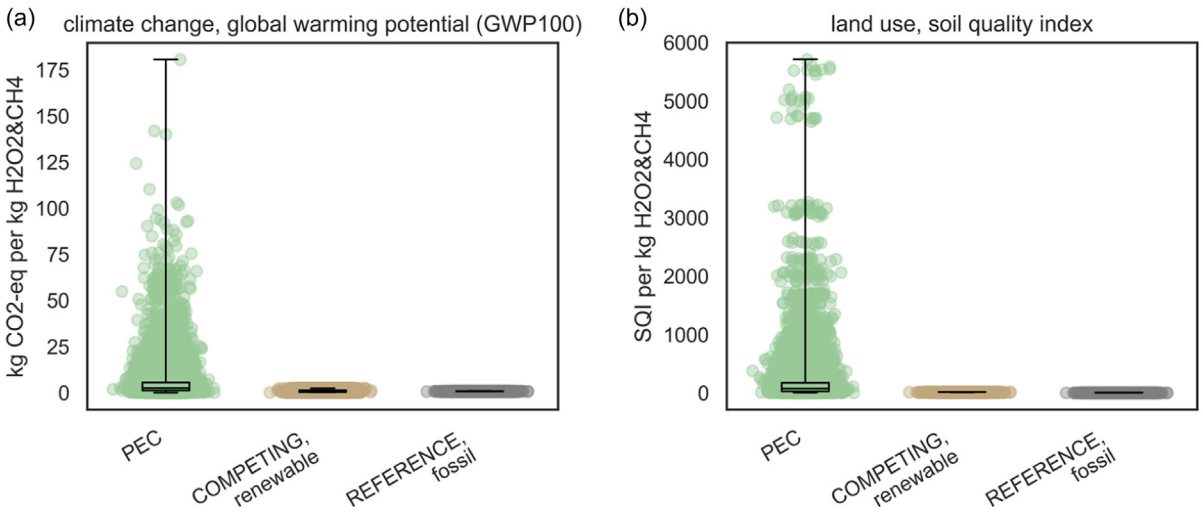

Figure 8. Results of the uncertainty analysis combined with the impact assessment for the climate change a) and land use b) impact categories. The analysis compares three product systems: PEC, a COMPETING renewable, and a REFERENCE product system based on fossil resources. The scatter plots illustrate the distribution of the results, with error bars representing the result range, and horizontal bars indicating the lower and upper quartiles, as well as the median values.

H_2+CO PC approach is also based on biomass, the results align with the literature, even when considering the result range from the uncertainty analysis.

In the contribution analysis, a high share of environmental impacts is attributed to one of the reactants (biogas), especially compared to the parabolic trough collector and the relatively low impacts of photocatalysts. This outcome is most likely due to the impacts associated with the biogas production process and the concentration of light in the parabolic trough collector, which reduces its impact share. This indicates that the outcome is highly specific to the considered product system.

Using IAM scenarios revealed that the relative performance (ranking) of the technology can change significantly depending on the chosen IAM scenario in particular when compared to competing product systems. This effect also partially applies to the choice of CO_2 scenarios. As already shown in our previous work, IAM scenarios can be decisive for technologies that are not competitive with current energy mixes or materials produced through carbon-intensive fossil fuel-based processes.[87] Since IAM scenarios prioritize decarbonization, their effects are most evident in the climate change impact category and in related impact categories (e.g., land use, which impacts expand with the deployment of renewables).

Focusing on the H_2+CO pathway, we identified a low threshold (0.3%) for the solar-to-chemical efficiency at which it becomes competitive with the reference product system in terms of climate change impacts. This value is lower than those for the other product system group (H_2O_2&CH_4) and values reported in the literature which were based on assumptions for modeling.[37–39,41] This threshold was already reached in PC water splitting,[21] and similar efficiencies seem achievable for CO_2 reduction, especially with light concentration.[127] Furthermore, short lifetimes (below five years) and relatively low Direct Normal Irradiation (under 900 kWh m^{-2} a^{-1}) were sufficient to outperform the reference product system. These low requirements likely can be explained by the notable contribution of non-PC processes such as biogas production and the parabolic trough collector's concentration of light. As a result, even increases in solar-to-chemical efficiency or longer operational lifetimes have limited impact on the results, since the corresponding sensitivity curve flattens with higher values.

Our findings on component lifetimes align with literature suggesting a minimum of 5 years.[37] However, this is also the lower bound in our sensitivity analysis, and even shorter lifetimes, though not in our assumed parameter range, could still lead to competitive results. The same applies for parabolic concentration which started at a factor of 30. Reaction conversion rates and parabolic concentration had relatively low threshold values, likely for the reasons already outlined. To the best of our knowledge, no comparable literature data exist for these parameters.

We were unable to determine threshold values for several parameters when compared to the COMPETING DRM product system, because this alternative performed better across all tested parameter ranges. Threshold values were calculated only for climate change; they tend to be higher for other impact categories. Corresponding graphs can be found in the Supporting Information (Section S3.1.4).

By integrating the uncertainty analysis of the H_2+CO PC product system, we found that even though the technology remains at an early stage, with many parameters still highly uncertain, its overall range of results is comparatively narrow. One of the competing product systems (H_2+CO COMPETING, electrolysis) showed a larger result range in the climate change impact category, most likely because this product system relies on electricity whose impacts vary widely depending on the IAM scenarios and their respective energy mixes. As the main impact driver of H_2+CO PC lies in the supply chain, the range of results is most likely comparatively lower. In addition, the selection of parameters and their associated probability distributions, which was not always defined as uniform and covering the full possible

range, could also play a role. Regarding the data itself, availability is not robust for either the reference or the competing product systems. For the reference product system, multiple data sources were used, contributing to higher variability, while the competing product systems were based on literature data originating mostly from laboratory-scale or simulation-based studies. The range of results for H_2+CO PC is notably narrower than of the H_2O_2&CH_4 product system discussed below.

In the H_2O_2&CH_4 product system, climate change impacts ranged from 0.53 to 1.59 kg CO_2-eq per kg of product. Ecoinvent and literature values show impacts of 0.04 kg CO_2-eq per kg of H_2O_2 and between −0.04 and 0.3 kg CO_2-eq per kg of CH_4.[102,103,105,128–131] Electrolysis-based product systems powered by wind energy showed climate-negative to climate-neutral performance. Consequently, a competing product system with a favorable energy mix might outperform the PEC product system in terms of climate change impacts, as was also observed when using an advantageous energy mix from the IAM scenario. However, the assumed PEC solar-to-chemical efficiency was conservative, since literature suggests potentially higher efficiencies.[21,23] The sensitivity analysis, assuming a solar-to-chemical efficiency of 20%, showed that PEC would perform similarly to the competing product system, potentially understating its performance slightly.

The contribution analysis revealed that the solar panel is the largest impact contributor, followed by anode production, electrolyte use, and the PEC cell itself. The relatively high impact of the solar panel stems mainly from the aluminum framing. Conversely, material reduction was already considered for the PEC cell; thus, its impact contribution could increase if those assumptions do not hold. Anode material and electrolyte choices also affect overall impacts.

Sensitivity analysis further suggested that the H_2O_2&CH_4 PEC product system should achieve at least a 7% solar-to-chemical efficiency, which was already reached for water splitting and a Global Horizontal Irradiation of at least 1700 kWh m^{-2} a^{-1} (comparable to Madrid, Spain). Notably, efficiencies up to 19.3% have been reported, though they were achieved for water splitting and not for CO_2 reduction.[21,23] Interestingly, extending system lifetimes did not reduce impacts enough to outcompete the reference product system, and similar results appeared for on-roof mounting and PEC cell material inputs. These outcomes, however, depend on the scope, resolution, and initial assumptions for unvaried parameters. Thus, the importance of further efforts, in particular to increase lifetime for competitiveness remains necessary.

The H_2O_2&CH_4 PEC product system displays a wide range of possible results, as indicated by the uncertainty analysis. This is most likely because its primary impact hotspot lies more in the foreground model than in the supply chain making parameters such as solar-to-chemical efficiency—the main drivers of the result. Because climate change impacts could vary from −0.11 to 180.64 kg CO_2-eq per kg of product, it is difficult to draw any conclusions. The median impact of 2.44 kg CO_2-eq places the PEC system near the upper range of the competing product system, however, approximately three times higher than the reference product system's maximum value. Assuming the median is representative, the PEC system for H_2O_2&CH_4 appears unlikely to be competitive in the climate change impact category and most of the other impact categories but may be competitive in water use.

4.2. Impact of Method Extensions on the Results

In the H_2+CO product system group, we observed that the outcomes varied depending on the method extensions applied. Implementing different CO_2 scenarios, following a standard LCA approach, we found that the H_2+CO PC product system was competitive with the reference and, to some extent, with competing product systems. However, when using IAM scenarios, these results shifted, as competitiveness was only observed against the reference product system in the climate change impact category. In contrast to that, conclusions in further impact categories remained largely unchanged. Furthermore, sensitivity analysis challenged these findings by questioning the validity of the assumed parameter values. It demonstrated that certain parameters could change the outcomes. Additionally, this approach provided minimum threshold values for each parameter, which can be particularly valuable in setting development goals for the technology. Further, when analyzing multiple parameter variations, the uncertainty analysis revealed that competitiveness and the potential shifts identified by the sensitivity analysis may not represent the majority of cases. In fact, based on the median values, the H_2+CO PC product system no longer appeared competitive with competing product systems in the climate change impact category. On the other hand, the uncertainty analysis identified that in optimistic scenarios, the competitiveness against both the competing and reference product system could be possible.

For the H_2O_2&CH_4 product system group, we observed a similar pattern in the changes of results. When considering only CO_2 scenarios, the H_2O_2&CH_4 PEC product system appears competitive with competing product systems. However, under IAM scenarios, competitiveness shifts more toward the reference product system and is only partially maintained against the competing product system. Similar trends are observed in the sensitivity and uncertainty analyses. In the uncertainty analysis, the result shift is more pronounced, as median values indicate no competitiveness. Furthermore, the result range is so wide that drawing conclusions becomes challenging. Thus, the methodological extensions, particularly the uncertainty analyses providing result ranges, challenge the findings of the standard LCA while also introducing new possible outcomes and issues to be addressed.

4.3. Data Limitations and Assumptions

The findings of this study, derived from the application of a prospective LCA in the field of emerging technologies, are subject to limitations due to the methodology used, the assumptions made, and the available data. These limitations are particularly pronounced given the low TRL of the technology assessed. Consequently, the inventory was not based on an industrial process or a functioning prototype and thus represents a preliminary estimate of likely materials and energy flows. The system design and product selection were predetermined; thus, we did not include a prescreening to evaluate other options.

The LCI generation methods applied fall into the midrange of the hierarchy, as indicated by the comparison of Parvatker and Eckelman,[48] meaning that the accuracy is low. They highlight that, for example, stoichiometric calculations can underestimate climate impacts and either under- or overestimate the land use impacts. However, higher-ranked methods such as those based on plant data are not yet available for the assessed technology.

Parameters assumed such as the solar-to-chemical efficiency of 5% have not been confirmed. Efficiencies achieved, for example, in PC hydrogen production (water splitting), which is further developed than PC CO_2-reduction, range from 0.1% to 3.3%. PEC devices have achieved higher efficiencies, up to 19.3%.[21] However, solar water splitting as well is more advanced than solar CO_2 reduction which adds additional challenges (described in Introduction).

Due to data limitations, the materials and manufacturing efforts for the catalysts were based on lab-scale data and thermodynamic models for substances where data was unavailable. Components such as pipes, valves, and pumps were considered for a different application (thermal oil) and were only scaled based on simple calculations rather than being specifically adjusted for AP application. Decommissioning was not taken into account, and recycling was included solely within the input materials based on the ecoinvent methodology.[113] Due to data availability, successful use in other applications, and its potential technoeconomic benefits, parabolic trough collectors were used as photoreactors for photocatalysis, even though alternative collectors might be more suitable for AP.[132]

For the reference product systems, combinations of pathways were assumed that might not be used in the real world to maintain the functional unit of the product mix provided by the AP technology. Additionally, partly competing product systems were assumed that have not yet been tested in application.

To partially address these common drawbacks in prospective LCA, we shifted the sensitivity analysis to investigate what is necessary to achieve similar performance as competing and reference product systems (break-even analysis) and initiated a first approach to implement epistemic uncertainties, running a Monte Carlo Simulation to show result ranges. These analyses could help identify hotspots early in the development stage and estimate whether this technology could ever achieve similar performance to other product systems. If the product system already performs unfavorably compared to the reference product systems, it is highly likely that environmental impacts will increase with a more complex assessment.

4.4. Methodological Limitations

From a methodological perspective, additional limitations exist. The cradle-to-gate approach of the LCA limits the validity of the climate change impact assessment results due to the unknown fate of the CO_2 bound in the potential products. If the products are further processed into long-lasting items, the net negative climate change effects presented could apply. However, if they are further synthesized, for example, into fuels and combusted, the CO_2 would be released, nullifying any negative carbon footprint.

Moreover, since the functional unit did not account for time, the AP product system has the disadvantage of being dependent on fluctuating solar energy, making product output per hour not regulated by demand. However, conventional technologies have the disadvantage of depending on exhaustible fossil energy sources, which was also not included in the assessment.

The prospective IAM scenarios were based on different assumptions and modeling approaches, leading to further limitations and uncertainties, which could accumulate. The scenarios depend on the REMIND model results, its modeling and data assumptions, as well as the modifications made by the premise tool such as the allocation to ecoinvent.

Due to limitations in result analysis and presentation capacity, we had to narrow down the LCIA and sensitivity analysis to a default scenario, so the results must be interpreted within this context and may change if other scenarios or scenario combinations are selected. A limitation of the sensitivity analysis is that single parameters are varied while all others remain at their assumed values. If any of those assumptions are inaccurate, it could mask or distort the importance of the parameter being tested. In the uncertainty analysis the probability distributions of the input parameters in the Monte Carlo Simulation were individually decided, meaning there was no predefined set of rules.

4.5. Software Limitations

For the LCIA calculations, we used the Activity Browser software. For the sensitivity and uncertainty analysis, we developed custom scripts to prepare the data for the scenario function of the Activity Browser and process it afterwards. We also used the scenario function for the Monte Carlo Simulation to test the capability of a GUI-based software like Activity Browser for such analyses. However, due to the high volume of calculations performed instantaneously, significant memory (RAM) was required. Either modifying the Activity Browser to calculate in batches or investigating alternative tools should be considered. For sensitivity analysis, further approaches such as global sensitivity analysis could be beneficial to avoid the partly subjective selection of parameters by the modeler. Due to the integration of script- and GUI-based software approaches, flexibility was naturally limited to a certain extent. Nevertheless, providing a GUI could be advantageous for many users, especially when the number of parameters to be investigated is limited.

4.6. Future Research

Based on our contribution and sensitivity analyses, we assume that the comparatively low minimum requirements for H_2+CO may stem from the high impact share of biogas and the use of parabolic concentration. However, to confirm this definitively, future research should isolate individual processes and vary not only parameters but also collector types and biogas production pathways to obtain a more comprehensive view of impact ranges and their primary drivers. Likewise, for H_2O_2&CH_4, further work could investigate the aluminum framing of solar panels by exploring alternative materials with equivalent functionality. Within the H_2O_2&CH_4, PEC technology itself, alternative anode materials and electrolytes should be developed and evaluated through LCA. The clear trend showing

that higher lifetimes improve competitiveness suggests that technology development should prioritize stability to extend operational life.

In the sensitivity analysis, it became evident that our predefined scope and resolution could be insufficient. For instance, the minimum threshold for H_2+CO PC lifetime may actually be lower than five years. Hence, both scope and resolution should be expanded and refined. Moreover, beginning parabolic concentration at 30 may be too high to adequately compare with nonconcentrating AP approaches. Exploring lower concentration factors may yield insights into how concentrating and nonconcentrating approaches compare. Although it cannot be directly inferred, parabolic concentration in H_2+CO PC appears to reduce facility impacts compared to H_2O_2&CH_4 PEC. Although this comparison is not strictly equivalent since the technologies and materials differ, further investigation of concentration factors for H_2+CO PC and concentrating approaches for H_2O_2&CH_4 PEC should be considered, since these could significantly lower the environmental impacts. Given that certain parameters were highly influential while others showed little effect, future research should employ an objective method such as global sensitivity analysis to identify key parameters while considering concurrent parameter changes.

Additional uncertainties, approximations, and assumptions stem from the databases used, database modifications, and LCIA methods. These factors could affect outcomes and should be addressed in future studies, especially regarding the sensitivity of each individual uncertainty source.

Most of these considerations require more flexible software solutions which need to be addressed. However, these advancements may broaden the overall range of results and therefore challenge result interpretation. Therefore, future work should also address methods which allow to narrow the result ranges.

5. Conclusion

In alignment with the objectives of this study, prospective LCA including a parametrized LCI, break-even sensitivity analysis, and uncertainty analysis, the following conclusions can be drawn regarding the emerging technology of AP, specifically in the context of PC and PEC CO_2 reduction. Considering current limitations and assuming the default scenario, the H_2+CO PC product system producing synthesis gas has the potential to outperform the reference product system in the climate change impact category, as well as in several other midpoint impact categories. The competing product system may offer benefits, particularly in the climate change category, while the reference product system might be beneficial in other impact categories such as land use. The most significant impact drivers are found in the production of biogas and the parabolic trough collector, which both are part of the supply chain rather than the modeled processes themselves. The impact of photocatalyst production is relatively minor, although material-specific impacts can still be significant. For example, Au/TiO_2 demonstrated clear benefits; thus, future research could explore additional options beyond those considered in this study. Additionally, the IAM scenarios show a significant effect on the impacts, particularly on competing product systems which are relying on high amounts of electricity. The CO_2 scenarios considering DAC and biogas show advantages over CO_2 sourced from ammonia or cement production.

Sensitivity analysis revealed that solar-to-chemical efficiency, lifetime and reaction conversion rates are critical parameters. Specifically, achieving a solar-to-chemical efficiency greater than 5%, a reaction conversion rate above 40%, and a lifetime exceeding 10 years could enhance the competitiveness of the PC product system. These findings can guide further research to providing minimum requirements. For certain impact categories, comparatively low thresholds for solar-to-chemical efficiency (0.3%), lifetime (<5 years), and irradiation ($<900\,kWh\,m^{-2}\,a^{-1}$) were observed, likely due to the high impact of preproducts and the high light concentration achieved by the parabolic trough collector. This indicates that light concentration may be crucial for enabling competitiveness of AP product systems. Under optimistic parameter assumptions, the uncertainty analysis suggests that the PC system could be competitive with both the reference and the competing product systems in multiple impact categories (e.g., water use). Notably, the H_2+CO PC product system exhibited a comparatively narrow result range in the climate change category, especially relative to the H_2O_2&CH_4 PEC product system.

The performance of the H_2O_2&CH_4 PEC product system producing hydrogen peroxide and methane is highly dependent on the IAM scenario. In less ambitious scenarios, the climate change impacts of PEC are higher than those of the reference and competing product system, whereas in more ambitious scenarios, PEC is beneficial. However, this trend may not apply to other impact categories, which would require system design enhancements, such as increased solar-to-chemical efficiency, to achieve similar benefits. The largest contribution to climate change comes from the mounting system of solar panels, followed by the anode, electrolyte, and PEC cell production. Nevertheless, in optimistic IAM scenarios, the climate impacts from the production of anode, electrolyte, and PEC cell can be significantly reduced.

In the default scenario, the H_2O_2&CH_4 PEC product system could be competitive with the reference product system in the climate change impact category, depending on the IAM and CO_2 scenario assumptions, but it remains less competitive in other impact categories. However, sensitivity analysis indicates that with higher efficiencies, potentially around 20%, competitiveness could be achieved. Solar-to-chemical efficiency, irradiation, and the lifetime of the solar panel and PEC cell were identified as key sensitive parameters. The required thresholds for solar-to-chemical efficiency and irradiation appear achievable, but extending the lifetime is critical for full competitiveness. Uncertainty analysis indicates a wide result range for the H_2O_2&CH_4 PEC system. Taking the median results, H_2O_2&CH_4 PEC is not competitive. Nevertheless the broad result range shows potential for competitiveness in specific conditions. Hence, drawing definitive conclusions for H_2O_2&CH_4 PEC at this early stage remains challenging.

Methodological developments in prospective LCA for the considered emerging technology are crucial, as they significantly influence both results and interpretation. Thus, further methodological improvements are needed. In both product system groups examined, future research should address not only the

technology itself but also the modeling of supply chain options. Enhanced modeling with more flexible software solutions can help to identify critical parameters, allow for dynamic parameter handling, and enable multiple supply chain configurations to identify more environmentally favorable production routes. Such flexibility is also needed to capture uncertainties in LCA and LCIA more thoroughly.

Unlike the H_2+CO PC product system, H_2O_2&CH_4 PEC did not utilize light concentration. Investigating concentration effects in PEC may be worthwhile, as it could reduce total impacts or lower solar-to-chemical efficiency requirements for competitiveness. Even if thresholds such as solar-to-chemical efficiency can be achieved, lifetime should also be addressed, particularly in H_2O_2&CH_4 PEC, where these factors strongly affect competitiveness.

The outcome of a prospective LCA at this early stage depends heavily on the specific product system and modeling approach. The main advantages of conducting an early-stage assessment lies in highlighting target parameters that developers need to meet and indicating potential competitiveness in various scenarios. However, there is a clear need for further work to refine methods in order to improve modeling. Because further methodological developments can expand the range of potential results, making interpretation more complex, there is a parallel need for approaches that narrow this range and enhance interpretability.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The author declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

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artificial photosynthesis, carbon capture utilizations, emerging technologies, photocatalysis, photoelectrochemical CO_2 reduction, prospective Life Cycle Assessment, uncertainty analysis

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