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Abstract
<p>In the framework of the ELEGANCY project we performed an integrated techno-environmental assessment of hydrogen production from natural gas and biomethane, combined with CO₂ capture and storage (CCS). We have included steam methane reforming and autothermal reforming for syngas production. CO₂ is captured from the syngas with a novel vacuum pressure swing adsorption (VPSA) process, that combines hydrogen purification and CO₂ separation in one cycle. As comparison, we have included cases with conventional amine-based technology. We have extended standard attributional LCA following ISO standards with a detailed carbon balance of the biogas production process (via digestion) and its by-products.</p> <p>The key outcome of our contribution is summarized as follows. Process simulation results show a clear advantage of ATR against SMR regarding overall CO₂ capture rates and the fact that configurations with the novel VPSA technology reach higher CO₂ capture rates than those with MDEA. However, higher CO₂ capture rates come along with higher electricity requirements. While adding a low-temperature WGS for SMR hardly makes a difference in terms of process efficiencies and capture rates, it is crucial for ATR, which performs rather poorly with high-temperature WGS only. Regarding reduction of direct CO₂ emissions, ATR with a low-temperature WGS and VPSA turns out to be the optimal configuration with an overall CO₂ capture rate of almost 100%.</p> <p>From the life cycle perspective, adding CCS results in clear benefits regarding impacts on climate change. In this respect, ATR performs substantially better than SMR due to higher CO₂ capture; adding a low-temperature WGS improves the life-cycle performance in general. However, SMR and ATR with CCS perform worse than without CCS regarding other environmental burdens as a result of increasing energy consumption and the (comparatively small) burdens associated with transport and storage of CO₂. Life cycle environmental performances of commercial CO₂ capture technologies (MDEA) and second-generation technologies (VPSA) are similar for equivalent CO₂ capture rates.</p> <p>The full work can be found in D5.3.1 (for the journal article see reference (1)). Additionally, the technical description of the VPSA process is summarized in a dedicated publication (2).</p>

1. Antonini C, Treyer K, Streb A, Van der Spek M, Bauer C, Mazzotti M. Hydrogen production from natural gas and biomethane with carbon capture and storage – a techno-environmental analysis. *Sustain Energy Fuels*. 2019;Submitted.
2. Streb A, Hefti M, Gazzani M, Mazzotti M. Novel Adsorption Process for Co-Production of Hydrogen and CO₂ from a Multicomponent Stream. *Ind Eng Chem Res*. 2019;58(37):17489–506.

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Hydrogen from wood gasification with CCS - a techno-environmental analysis of production and use as transport fuel[†]

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The use of biomass as a resource for hydrogen production can contribute to the transition towards carbon neutral or carbon negative energy systems. This paper offers a comprehensive investigation of the technical performance and life cycle environmental footprint of the three most common gasification technologies for H₂ production, using dry biomass (wood) as input. These are compared with H₂ production from reforming of natural gas or biomethane and electrolysis as presented in our previous work.¹ This is followed by an evaluation of the use of H₂ as fuel for passenger cars and trucks. The quantity of biomass required for the production of 1 MW H₂ is calculated with an integrated process simulation approach on the basis of Aspen Plus simulations and real-plant literature data. We observe that all the technologies analysed provide negative CO₂ emissions when coupled with CCS. However, the sorption enhanced reforming and the entrained flow gasifiers are more suited to this scope than the dual fluidized bed gasifier, because higher overall CO₂ capture rates can be achieved. As CO₂ is from biogenic sources, the life cycle carbon footprint of the produced H₂ is negative (with CCS) or only slightly positive (without CCS). This negative carbon footprint is not obtained at the cost of important trade-offs with regards to ecosystem quality, human health or resource depletion, with the exception of high forest land use. Fuel cell electric vehicles using hydrogen from biomass (both wood and biomethane) with CCS as fuel turn out to be the most climate friendly among all options, with even possible negative total greenhouse gas emissions. However, limited biomass resources and its current and potential alternative uses need to be considered.

1 Introduction

Effective climate change mitigation limiting global warming to 1.5-2°C in line with the Paris Agreement requires substantial reduction of greenhouse gas (GHG) emissions, aiming for a “net-zero” economy by the middle of this century². This implies a major shift from fossil to renewable primary energy resources in all economic sectors as well as the deployment of negative emission technologies to compensate for GHG emissions difficult to avoid². Hydrogen not only plays a crucial role in an economy compatible

with the Paris Agreement, but is also a major player in the EU's commitment to reach carbon neutrality by 2050³. Hydrogen is a flexible product and can be used as chemical feedstock, fuel or as energy carrier with many applications in industry, transport and power sectors. Its use does not cause any direct greenhouse gas emissions and offers the co-benefit of zero air pollution as opposed to the centralized and distributed combustion of fossil fuels. However, from an overall system perspective, hydrogen production must be associated with very low GHG emissions in order to contribute to climate change mitigation. This is currently not the case, as the vast majority of hydrogen production relies on fossil feedstock, mainly natural gas and coal⁴. Low-carbon hydrogen production pathways include water electrolysis with low-carbon electricity supply, fossil feedstock conversion with carbon capture and storage (CCS), and biomass conversion, i.e. reforming of biomethane (a natural gas equivalent from biogenic origin) or thermochemical conversion of solid biomass³. Hydrogen from biomass conversion processes with CCS can even lead to so-called “negative” GHG emissions, i.e. permanent removal of greenhouse gases from the atmosphere⁴. The portfolio

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[†] Electronic Supplementary Information (ESI) available: Inventories and LCIA results of LCA, including corresponding Jupyter Notebooks (“AntoniniTreyer et al H2 from wood gasification ESI LCA.xlsx”, “1 LCI Import.ipynb”, “2 H2 from wood gasification”, “3 contribution analysis wood gasification.ipynb”); “ESI-Technical.pdf”. See DOI: 00.0000/00000000.

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of “biomass-to-hydrogen” pathways is broad and includes use of different biogenic feedstock, different conversion technologies as well as different alternatives for CO₂ capture and storage⁵⁻¹⁵. To identify the feedstock and conversion options most beneficial for climate change mitigation, these need to be evaluated before large-scale implementation from a technological and environmental perspective employing process simulation, and Life Cycle Assessment (LCA). Quantifying technical and environmental benefits as well as potential trade-offs calls for an assessment of not only hydrogen production, but also of its use compared to conventional (fossil) alternatives.

Techno-environmental assessment of hydrogen from biomass – analyses performed so far

The body of literature addressing technical and environmental aspects of hydrogen production from biomass as such is very broad⁶; however, carbon capture has hardly been included in previous assessments and in general, the majority of the LCA studies reviewed by Tian et al.⁶ “failed to explain the robustness (of results) due to the lack of sensitivity and uncertainty analysis, indicating high quality life cycle assessment studies are needed in the future”. We recently aimed at partially filling this stated gap by performing a techno-environmental assessment of hydrogen from biomass, focusing on hydrogen production via anaerobic digestion of waste resources with high water content (“wet biomass”) and subsequent reforming of biomethane with and without CCS and compared the results to hydrogen from natural gas reforming and electrolysis¹. This assessment shows benefits of auto-thermal compared to steam reforming with CCS due to superior CO₂ capture rates, which results in lower life-cycle GHG emissions per unit of hydrogen produced. Furthermore, the use of wet biomass in combination with CCS allows for a net removal of CO₂ from the atmosphere. Hydrogen production using woody (or “dry”) biomass via gasification and including CO₂ capture has been evaluated from a techno-environmental perspective by two recent studies^{16,17}. Hybrid poplar as feedstock for the gasification process and a low-pressure indirect gasifier consisting of dual fluidised bed (DFB) reactors – the gasifier itself and a char combustor – was evaluated by Susmozas et al.¹⁷. The syngas was fed into a water gas shift (WGS) section involving high- and low temperature shift reactors, followed by a purification section; a pressure-swing-adsorption (PSA) unit is used to separate hydrogen from other gases. CO₂ was captured from the exhaust gas of the boiler with a two-stage gas separation polymeric membrane process. Conversion of Canadian pine wood with two different biomass gasification processes with CO₂ capture was analyzed by Salkuyeh et al.¹⁶: an atmospheric-pressure, air blown, DFB gasifier, (indirect fired system); and a high pressure, oxygen-blown entrained flow (EF) gasifier (direct fired system). The design of the DFB gasification hydrogen production pathway was similar to the one in Susmozas et al.¹⁷. The EF gasification analyzed required high purity oxygen instead of air as input, thus allowing for a higher process efficiency. The electricity required to run the process auxiliaries and eventually the CO₂ capture unit is provided internally; additional fuel is burned with oxygen to generate pro-

cess steam via heat integration. This steam is then sent to the power island to produce the electricity needed. Both studies are based on Aspen Plus simulations and perform a process-based, attributional LCA based on the simulation results. While Salkuyeh et al.¹⁶ reports only LCA results in terms of life-cycle GHG emissions and selected air pollutants, Susmozas et al.¹⁷ quantifies life-cycle GHG emissions, non-renewable cumulative energy demand and selected midpoint impacts applying the CML method¹⁸. Both studies show negative GHG emissions for hydrogen from biomass gasification with CCS and some trade-offs for other environmental burdens due to CO₂ capture. However, neither of them includes geological storage of CO₂ following CO₂ capture. The use of hydrogen is not included in any of these evaluations.

Scope and novelty of this study

To the best of our knowledge, we perform the first complete LCA of hydrogen production via gasification of woody biomass with CO₂ capture and permanent geological storage, directly linked to the outcomes of a detailed technical assessment, and its subsequent use as fuel in fuel cell vehicles. This allows for quantification of the environmental benefits and potential trade-offs not only of a broad set of hydrogen production pathways, but also of its application as vehicle fuel compared to other power-train options. In terms of biomass conversion technologies, we model the main wood gasification technologies currently available: i) the dual fluidized bed gasifier, ii) the sorption enhanced reforming gasifier, and iii) the entrained flow gasifier. These three gasifiers differ in terms of process conditions and feedstock pre-treatment requirements. Therefore, even if the feedstock used is the same, the composition of the product gas and the amount of electricity required for the operation differ substantially in the three cases. The downstream chain that follows the gasification process is case specific; in fact, the gasifier product gas specifications such as composition, temperature and pressure are different for each gasification technology implemented. Furthermore, we calculate the net energy requirement of the systems by calculating the electricity needed by the various process steps and subtracting the electricity co-produced by means of heat integration. In this way it is possible to compare the three technologies in terms of biomass input required for the production of 1 MW H₂, net energy requirements and net CO₂ emissions. The results of the wood-based hydrogen production chains are then compared with the natural gas/biomethane cases studied in our previous work (see Figure 1 for an overview on the different production pathways considered in this analysis)¹. The process analysis provides the main indicators required for the integrated techno-environmental modelling framework, which directly connects mass and energy flows from the process simulation with the Life Cycle Inventories (LCI). This integrated approach allows for the quantification of the environmental performance of many different cases based on consistent and physically sound data. The comparative evaluation of hydrogen use in fuel cell electric vehicles (FCEV) and the comparison with other vehicle fuels and drivetrains builds upon recent work of some of the authors, which is particularly valuable for its transparency and completeness^{19,20}.

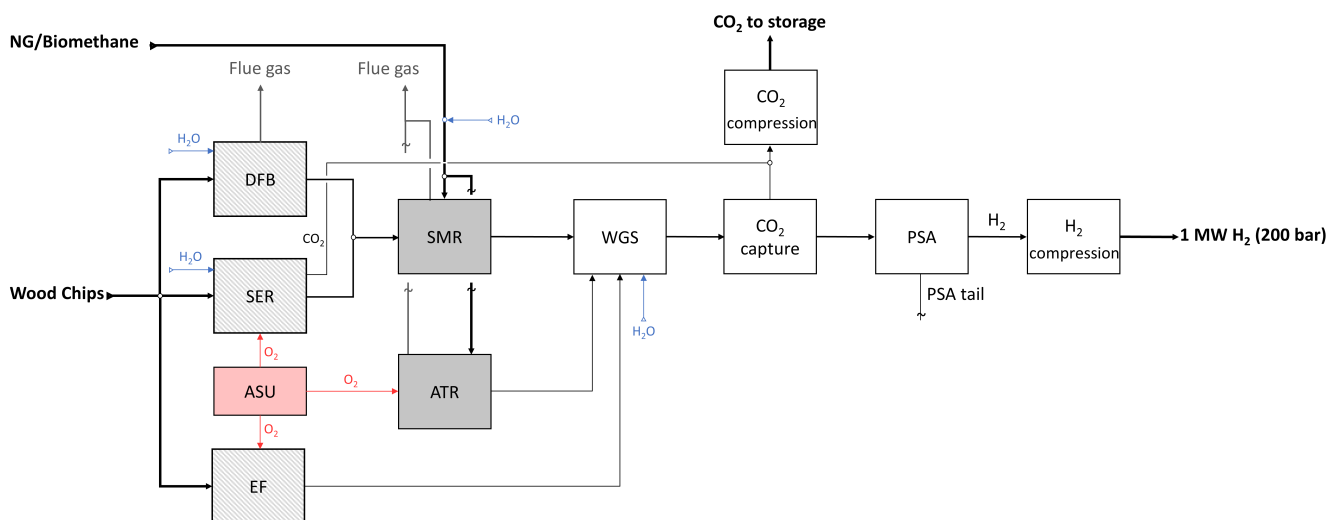


Fig. 1 Schematic representation of all hydrogen production pathways modelled and analysed in this work.

2 Technologies

2.1 Hydrogen production via woody biomass gasification

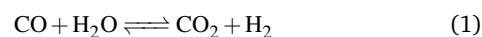
A valuable alternative to classical hydrogen synthesis pathways that involve fossil fuels is the production *via* dry biomass gasification. A variety of gasifiers exists and in the framework of this analysis we selected the following three: i) the dual fluidized bed steam gasifier (DFB), ii) the sorption enhanced reforming gasifier (oxySER), and iii) the entrained flow gasifier (EF) (see Table 1). All the gasification technologies have been tested in relevant fields and have reached a technology readiness level (TRL) higher than 6.

Table 1 Gasification technologies description.

Name	Design	Gasification agent	Heating
DFB	fluidized bed	steam	indirect (air)
oxySER	fluidized bed	steam	indirect (O ₂)
EF	entrained flow	oxygen	direct

Dual fluidized bed. The DFB technology selected is the one developed primarily at TU Wien, Austria, and tested at a demonstration-plant scale in Güssing, Austria.²¹ Examples of existing biomass-fired industrial-scale power plants using the design of the Güssing plant are the HGA Senden plant in Germany, and the GoBiGas plant in Gothenburg, Sweden.^{22,23} The DFB gasifier is an indirect heated gasifier and it consists of two beds: the gasification reactor, where the wood chips are gasified with steam, and the combustion reactor (see Figure 2a). The heat required to perform the endothermic steam gasification reactions is indirectly provided by the combustion of char with air occurring in the combustion reactor; char is a co-product of the gasification process and it is transported from the gasification reactor to the combustion reactor with the recirculating bed material. The bed material is looped between the two beds and acts as heat carrier: it heats up in the combustion reactor and it releases heat in the gasification reactor. To prevent the contamination of the

gasification gases with the combustion flue gas that might occur because of the bed material recirculation, fluidized seals are used. Therefore, the syngas produced is almost nitrogen free. The bed material used at industrial scale is Olivine.²⁴ As shown in Figure 3a, wood gasification is only one part of the hydrogen production chain. In fact, after the gasification process the product gas needs to be cleaned from particulates and contaminants that could interfere with the downstream processes. The particulates are removed using cyclones and filters, whereas to eliminate the sulphur components a desulfurization section is required. Usually, desulfurization occurs at medium-low temperature, but to prevent tar condensation (that might cause fouling and blockage of the pipes), the temperature has to be kept above 350°C.²⁵ Although high-temperature desulfurization processes are preferred, they are at an early stage of development and further research is necessary to determine their performance and feasibility.²⁶ Additionally to sulfur, other contaminants might be present in the product gas (e.g. chlorine and alkali metals). However, the detailed analysis of the product gas cleaning process goes beyond the scope of this study. Therefore, we only consider particulates and sulfur removal; among the different options reported in the work of Cheah et al.²⁶, we assume that the desulfurization process occurs at around 650°C, using zinc titanate-based sorbent material.^{27,28} After the cleaning and desulfurization section, the hydrocarbons present in the product gas are reformed in a steam reforming reactor. Depending on the type of tar formed during the gasification process, reforming might not be enough to completely eliminate it hence additional treatments might be necessary (e.g., using rapeseed oil methyl ester tar scrubber²³). However, since this issue is case specific and thus hard to generalize, for the sake of simplicity in this analysis we assume that total tar conversion is attained after reforming. The reformed gas is cooled down and then shifted in a water gas shift (WGS) section.



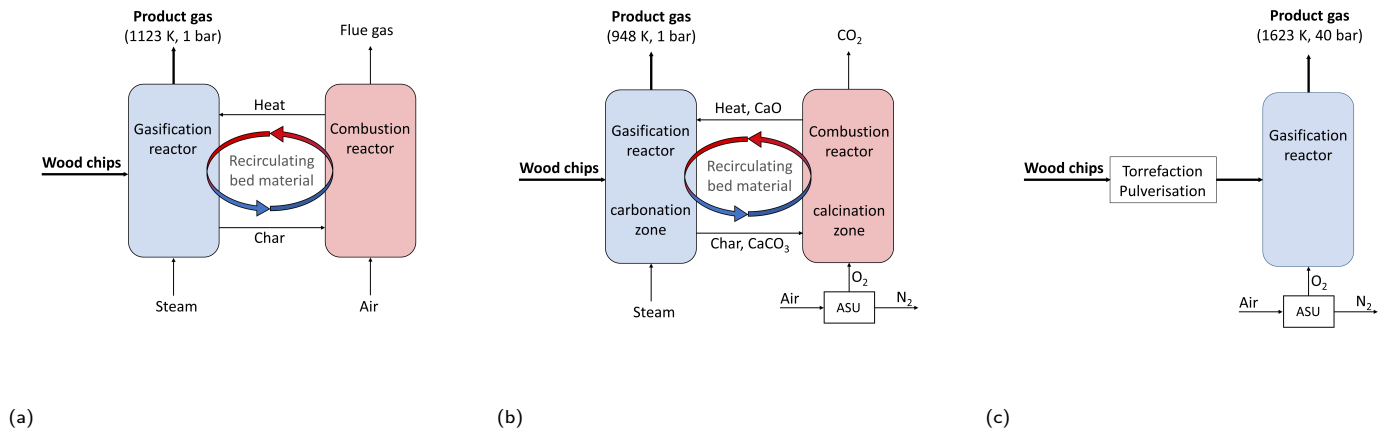
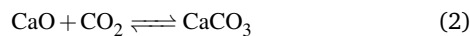


Fig. 2 Schematic representation of the gasification technologies considered in this work: (a) Dual fluidized bed gasifier (DFB), (b) Sorption enhanced reforming gasifier (oxySER) and (c) entrained flow gasifier (EF).

After the WGS section the product gas is compressed and dehydrated. If the plant is equipped with a pre-combustion CO₂ capture unit, the shifted syngas is then compressed and CO₂ is separated. The raw hydrogen is purified in a pressure swing adsorption (PSA) unit, and it exits with a purity of $\geq 99.97\%$ and it is successively compressed to 200bar, while the rest (also called PSA tail gas) is combusted with air in a combustor. Figure 3a shows the hydrogen production chain considered with the DFB gasifier, with and without a pre-combustion CO₂ capture unit.

Sorption enhanced reforming gasifier. The second gasification technology considered is the oxySER gasifier (see Figure 2b). It also consists of a gasification and a combustion reactor; the bed material is recirculating from one bed to the other, with the difference that in an oxySER process limestone (CaCO₃) is used instead of Olivine. Limestone is calcined in the combustion reactor and CaO is looped into the gasification reactor, and it reacts with the CO₂ present in the product gas. Therefore, in-situ CO₂ capture occurs.



The removal of CO₂ shifts the equilibrium of the WGS reaction towards the products, therefore the volumetric content of H₂ in the syngas is very high (ca. 70%). As in the case of the DFB gasifier, char is transported with the bed material from the gasification to the combustion reactor where it is combusted with oxygen. A CO₂-rich stream is collected at the outlet of the combustion reactor and then dehydrated and compressed to be suitable for geological storage. The O₂ needed in the combustion process is produced with an air separation unit (ASU). As for H₂ production from a DFB gasifier, the product gas has to be cleaned and desulfurized. Therefore, also in this case sulfur and tar removal might be challenging. Figure 3b shows the hydrogen production chain considered with the oxySER gasifier, with and without a CO₂ capture unit.

Entrained flow gasifier. The third gasification technology is a pressurized entrained flow gasifier (Figure 2c). This technology

is widely used at industrial-scale (in plants of more than 100 MW_{th}) for coal gasification in integrated gasification combined cycles (IGCC) and for chemical synthesis applications. Furthermore, the co-gasification of biomass in large scale IGCC has already been tested.^{29,30} In contrast to fluidized bed gasification, where the wood chips are directly fed into the gasification reactor, a thorough pre-treatment is needed. Here we consider pre-drying, torrefaction and pulverisation (to reach an average particle size smaller than 0.5 mm) as suggested in Tremel et al.³¹ A pressurized feeding system is needed and different technologies are available; we select as reference technology the hydraulic piston system with screw.^{32,33}

The product gas is treated in a gas cleaning section and, because of the elevated temperature reached in the gasifier, it contains neither methane nor tar hence reforming is not required. However, the carbon monoxide content is significant and to increase the hydrogen yield a water-gas shift section with two reactors, one at high and one at low temperature, is required. The shifted syngas can be sent to the CO₂ capture plant or directly to the purification unit (see Figure 3c).

In all hydrogen production chains, process steam is co-produced by means of heat integration; some of it is used in the process, while the rest is expanded in a turbine section. The electricity produced is used internally to run the various utilities, and in case of excess it is fed into the grid. Concerning the CO₂ capture unit, an amine-based absorption process is considered, where the solvent used is methyl diethanolamine (MDEA). The CO₂ capture rate selected is 98%.

2.2 Hydrogen production via natural gas/biomethane reforming with carbon capture and storage

Steam methane reforming (SMR) and autothermal reforming (ATR) of natural gas (NG) and biomethane were described and investigated in our previous work¹. In this analysis we compare the technical and environmental performance of producing hydrogen from wet waste biomass converted to biomethane (see Figure 1) with the benchmark production via NG reforming. We consider

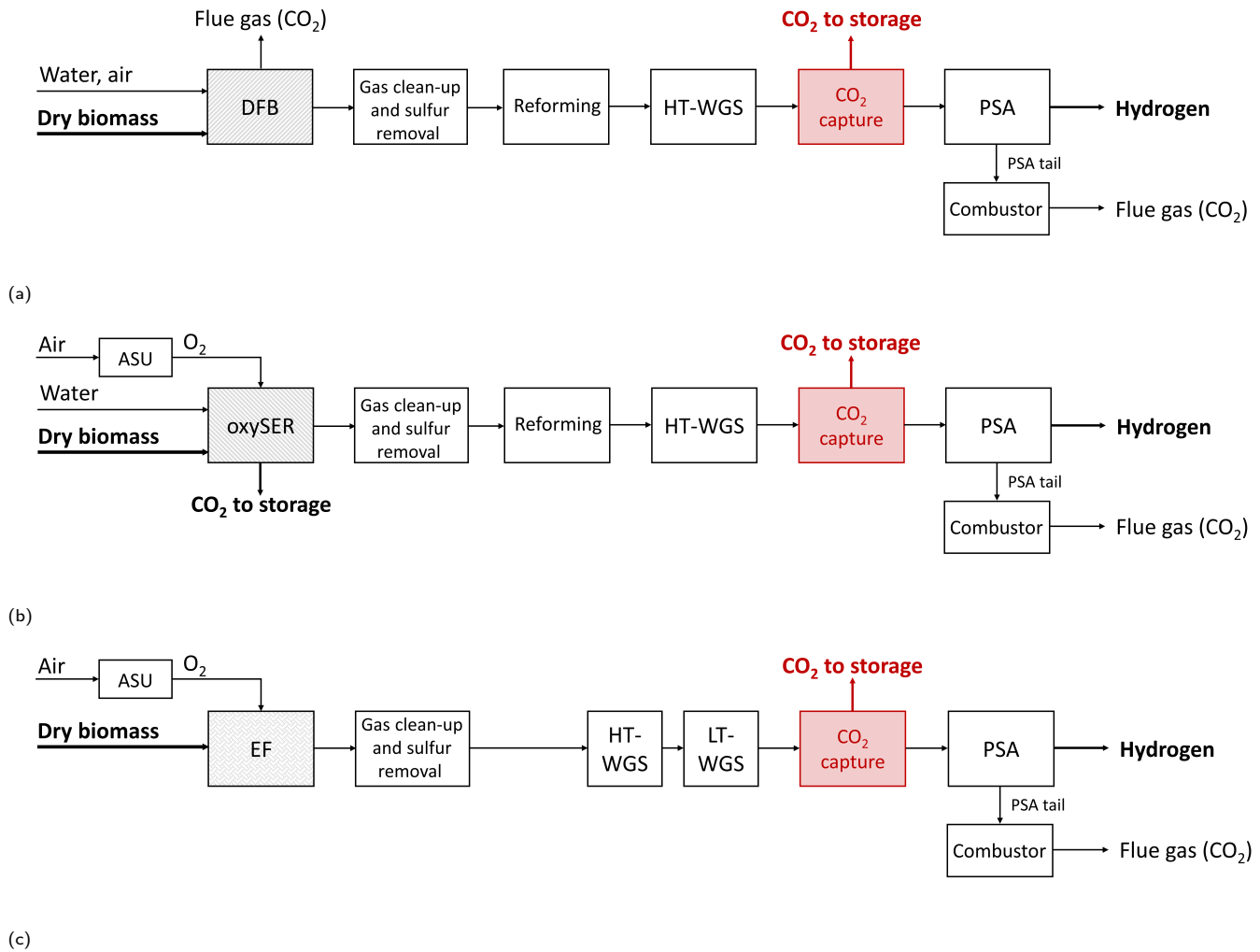


Fig. 3 Wood to hydrogen production chains: (a) with a DFB gasifier, (b) with an oxySER gasifier, and (c) with an EF gasifier. HT/LT-WGS: high/low temperature water-gas shift reactor; PSA: pressure swing adsorption unit; ASU: air separation unit.

biomethane as a starting point for the comparison and neither biogas nor wet biomass because of the following reasons: first of all the availability of wet biomass is generally decentralized and at small scale, and second, the transport of both, precursor (wet biomass) and product (biogas), is challenging. Therefore, we believe that it is generally convenient to produce and upgrade biogas to biomethane locally and then feed it into the natural gas grid. Therefore, what we are presenting here as "H₂ production via biomethane reforming", corresponds to a hypothetical case where the plant is 100% fed with biomethane.

3 Process modelling

In this section a detailed description of the process modelling strategy is provided. The functional unit is "production of 1 MW of hydrogen, with purity of at least 99.97%". To be precise, for the configurations including an EF or an ATR unit, argon (Ar) is introduced into the system as an impurity of the oxygen stream. Argon is a light inert gas, it does not interfere with any conversion or purification process but it leaves the system with the purified H₂ stream. To distinguish between Ar and the rest of the impu-

rities (i.e. CH₄, CO, CO₂ and N₂) we define an "adjusted purity" where Ar is not taken into consideration. Therefore, for all EF and ATR cases we consider an adjusted purity of at least 99.97%, which correspond to an overall purity of at least 99.9%. As feedstock we consider forest wood chips with a molecular composition in line with the LCA according to the ecoinvent report on wood energy³⁴: C 49.4 wt%, O 44.5 wt% and H 6.1 wt% (LHV of 18.9 MJ/kg).

3.1 DFB and oxySER gasifiers

Given the complexity of the dual fluidized bed gasifiers (DFB and oxySER), we adopted a so called "black-box modelling approach"; based on data available in the literature we define a base-case product gas composition (see the electronic supplementary information (ESI) for data and references). Starting from these compositions, we calculate the product gas flow rate needed to produce 1MW of H₂. We assume that 30 mol% for the DFB²⁴ and 63 mol%³⁵ for the oxySER of the carbon present in the biomass is burnt in the combustor. The amount of steam needed as reactant and for heat provision to the CO₂ capture unit is subtracted by

the amount of process steam co-produced; whereas the remaining process steam is expanded in the turbine section. The oxygen used in the oxySER and EF gasifiers is assumed to have a purity of 99.5%, with the make-up being argon, and an energy consumption of 265 kWh per t O₂^{36,37} is considered (see the supplementary information for calculations and modelling assumptions).

3.2 EF gasifier

Contrary to the other two gasification technologies, in this case the biomass needs to be pre-treated; we consider: pre-drying, torrefaction and pulverization. Instead of modelling these processes in detail we assign the corresponding energy consumption based on data available in the literature (see ESI for more details).^{31,38} The EF gasifier is modelled in Aspen Plus V 8.6. following the modeling strategy presented in Meerman et al.³². The gasifier is operated at 40 bar and isothermally at 1623 K. The molar oxygen to carbon (O/C) ratio is 0.32 and O₂ is fed with an over-pressure factor of 1.2 (48 bar); the compression of oxygen occurs in 4 steps with an intercooling temperature of 308 K. The calculations and modelling assumptions are described in the supplementary information.

3.3 Reforming of biomethane

The reforming-based technologies considered in our comparative evaluation (SMR and ATR of natural gas and biomethane) have been comprehensively described earlier¹.

3.4 Sensitivity analysis

Wood gasification is a complex process and, as explained in the previous section, for some of the technologies we used literature data instead of developing a detailed physical model. To assess the dependency of the results on the modelling assumptions and to offer a better overview on the effect of parametric variation on the process output, we performed a sensitivity analysis (see Table 2 for the details). First of all, based on the results available in the literature, we defined a range of possible compositions of the product gas of both DFB and oxySER gasifiers; as lower bound (LB) we defined the composition with the smallest amount of H₂, while as upper bound (UB) the one with the highest (see ESI for data and references). For each of these compositions, we performed a sensitivity analysis on the percentage of carbon that is consumed in the combustion chamber, defined as the molar ratio between the moles of carbon in the combustor flue gas n_C^{cFG} and those present in the biomass inlet stream (n_C^{dBM}):

$$\gamma_C = \frac{n_C^{cFG}}{n_C^{dBM}} \quad (3)$$

In the case of the EF gasifier, the section that is not modelled in detail is the biomass pre-treatment (comprising drying, torrefaction and pulverisation). Therefore, we performed a sensitivity analysis on the energy consumption needed for these pre-processes, and we defined ω_{dtp} as the kW of electricity needed

Table 2 Sensitivity analysis; pp: percentage points, dBM: dry biomass

Parameter	Base case	Range of sensitivity
DFB		
γ_C [%]	30	±5 pp
oxySER		
γ_C [%]	63	±2 pp
EF		
ω_{dtp} [kWel/kg _{dBM}]	0.116	±20%

per kg of dry biomass processed:

$$\omega_{dtp} = \frac{E_{dtp}}{m_{dBM}} \quad (4)$$

4 Life Cycle Assessment

The evaluation of the environmental performance of potential near-future Negative Emission Technologies (NET) needs to be done considering a life cycle perspective, taking into account all environmental burdens occurring during the entire life cycle of a product or service. We perform an ISO 14040 and 104044^{39,40} compliant, attributional Life Cycle Assessment (LCA) of the production of 1 MJ H₂ via gasification of woody biomass at a pressure of 200 bar and a purity higher than 99.97 % (SMR, DFB and oxySER) or higher than 99.9 % (ATR and EF, with Argon representing the additional impurity). Calculations are performed with the open-source software Brightway2⁴¹ and the ecoinvent life cycle inventory database v3.6, system model "allocation, cut-off by classification"⁴². All jupyter notebooks and detailed Life Cycle Impact Assessment (LCIA) results are part of the Electronic Supplementary Information (ESI). The system boundaries and allocation choices are illustrated in Figure 4, and the geographical scope is Europe. The wood chips used as feedstock for gasification are a product from sustainable softwood and hardwood forestry of various species grown in Germany and Sweden (namely beech, birch, oak, pine and spruce), which represent the European market for wood chips in ecoinvent v3.6. It should be noted that the carbon uptake is assumed to be the same for all these species in ecoinvent. Other variabilities in terms of forestry in other European countries, transport distances, regional market compositions, or wood chips imports from overseas could not be modelled within the scope of this paper due to lack of information. All carbon content values are calculated on a dry matter basis. Carbon uptake by trees is accounted for with a characterisation factor of -1 for CO₂, while release of biogenic CO₂ is accounted for using a positive factor of 1 in order to be able to quantify impacts on climate change due to capturing and permanently storing biogenic CO₂. Detailed discussion and a carbon balance of the biomethane chain and corresponding modeling choices are part of our previous work¹. Impacts on climate change of greenhouse gases are quantified according to the IPCC 2013 LCIA method with a 100 years Global Warming Potential timeframe² as implemented in the ecoinvent database. The ILCD 2.0 (2018) LCIA method⁴³

covering environmental impacts such as ecotoxicity, effects on the human health, ozone layer depletion or near-ground photochemical ozone creation, or metal depletion is further used, in addition to the non-renewable cumulative energy demand (CED) as a measure for depletion of fossil, nuclear and non-renewable forest resources. The inventories for the natural gas supply chain as well as all materials, infrastructure, or transports (i.e. the life cycle inventories of the so-called "background processes") are taken from the ecoinvent database, while the biomethane chain, H₂ production plant, and CO₂ transport over 200 km per pipeline and storage in a saline aquifer at a depth of 800 m are all based on data from previous projects of the authors or own project-specific data. Electricity use or electricity fed back to the grid in case of excess electricity is modelled with the European ENTSO-E mix. Our analysis of the use of hydrogen is limited to its application as vehicle fuel. We build upon previous work performed by some of the authors^{19,20} and link the hydrogen production pathways modeled within this analysis and our previous work¹ to vehicle (LCA) models established by^{19,20} to quantify life-cycle environmental burdens of passenger vehicles and trucks of different hydrogen supply options for FCEV compared to conventional gasoline and diesel vehicles as well as battery electric vehicles (BEV). Such modelling of the end use enables understanding the importance of differences in LCIA scores of the various H₂ production pathways from an overall LCA perspective.

5 Results and discussions

The technical performance of the different hydrogen production pathways is evaluated based on four key indicators: product gas molar composition (Figure 5), overall CO₂ capture rate (6b), electricity balance (Figure 7) and net process efficiency. The overall CO₂ capture rate is the ratio between the amount of CO₂ captured and the overall CO₂ produced (sum of the CO₂ captured and emitted). The electricity balance includes the overall electricity consumption of the H₂ production plant, normalized by the amount of H₂ produced (which is constant for all cases presented), while the net process efficiency is defined as the energy content of hydrogen produced, divided by the energy content of the biomass needed to produce it. In Figure 8 the conversion of wood into hydrogen is compared with natural gas/biomethane reforming. The presentation of the LCA results is structured as follows. First, climate change impacts from the six configurations modelling the gasification of wood are compared to selected configurations for hydrogen production via steam methane reforming or autothermal reforming of natural gas or biomethane with and without CCS¹ (Figure 9). Then, we provide a comparison of the performance of the various H₂ production pathways with regards to selected Life Cycle Impact Assessment (LCIA) categories (results for all LCIA categories are included in the ESI) (Figure 10). And finally, the use of H₂ in fuel cell passenger cars and trucks is compared with other fuel supply and vehicle options in terms of impacts on climate change (Figure 11) (all LCIA categories represented in the ESI).

5.1 Gasifiers product gas composition

Figure 5 shows the product gas molar composition of the three gasifiers; for the DFB and oxySER gasifiers the composition refers to the base case. The base-case product gas of the DFB gasifier contains around 38 mol% of hydrogen, 24% of CO, 23% of CO₂, and around 10 mol% of methane. Methane is converted via reforming into syngas (CO and H₂), and to increase the H₂ yield, carbon monoxide is shifted with steam in a WGS section. The oxySER product gas is richer in hydrogen (base-case composition 67 mol%). However, because of the low operating temperature of the gasifier, a substantial amount of methane is generated (ca. 14 mol%, which corresponds to almost 10 wt%), which is converted into syngas via steam reforming. Therefore, although the molar fraction of CO present in the product gas is relatively low (below 10 mol%), it increases after the reforming process and therefore, to increase the H₂ yield, a WGS section is needed. The product gas after the EF reactor contains a substantial amount of CO (ca. 60 mol%); this is because oxygen is used as gasification agent instead of steam. Therefore, a WGS section with two reactors (at high and low temperature) is needed. The advantage of the EF reactor is that, by operating the gasifier at high temperature (> 1300°C), the product gas is free of hydrocarbons (i.e. methane and tar).

5.2 Net efficiency and sensitivity analysis on the carbon balance

Figure 6a shows the net efficiency and the corresponding overall CO₂ capture rate of the different wood to hydrogen production chains. For the configurations with a DFB or an oxySER gasifier, the results of the sensitivity analysis on the γ_C ratio performed on the different product gas compositions (LB, base case (BC) and UB) are enclosed in the range delimited by the transparent area, while the specific results for the base case are shown with black dots (net efficiency) and red diamonds (CO₂ capture rate). Figure 6b instead shows the specific amount of CO₂ captured and emitted; for the configurations with a DFB or an oxySER gasifier, the results for all three product gas compositions (LB, BC, UB) are shown, while the error bar highlights the variation due to the sensitivity analysis on γ_C .

The net efficiency of the DFB configurations ranges from 51 to 60%. The variation of ± 5 pp on γ_C causes a ± 4 pp variation of the net efficiency. Indeed, the less carbon goes to the combustor (γ_C smaller) the less biomass is needed per unit of hydrogen produced. This translates into higher efficiency and lower specific carbon emissions. In fact, as shown in Figure 6b, the specific amount of carbon emitted varies by varying γ_C , whereas the amount of carbon captured is not affected by the sensitivity analysis on γ_C . If a pre-combustion capture plant is added to the chain, the overall CO₂ capture rate goes from 0 to ca. 60%. Finally, it is worth noting that these trends are similar for all three product gas compositions considered (LB, BC and UB).

The net efficiency of the oxySER configurations without and with the addition of a CO₂ capture plant (oxySER S and oxySER CCS, respectively), ranges from 60 to 82%. Unlike the DFB cases, the range of variability is remarkable and it is due to the uncer-

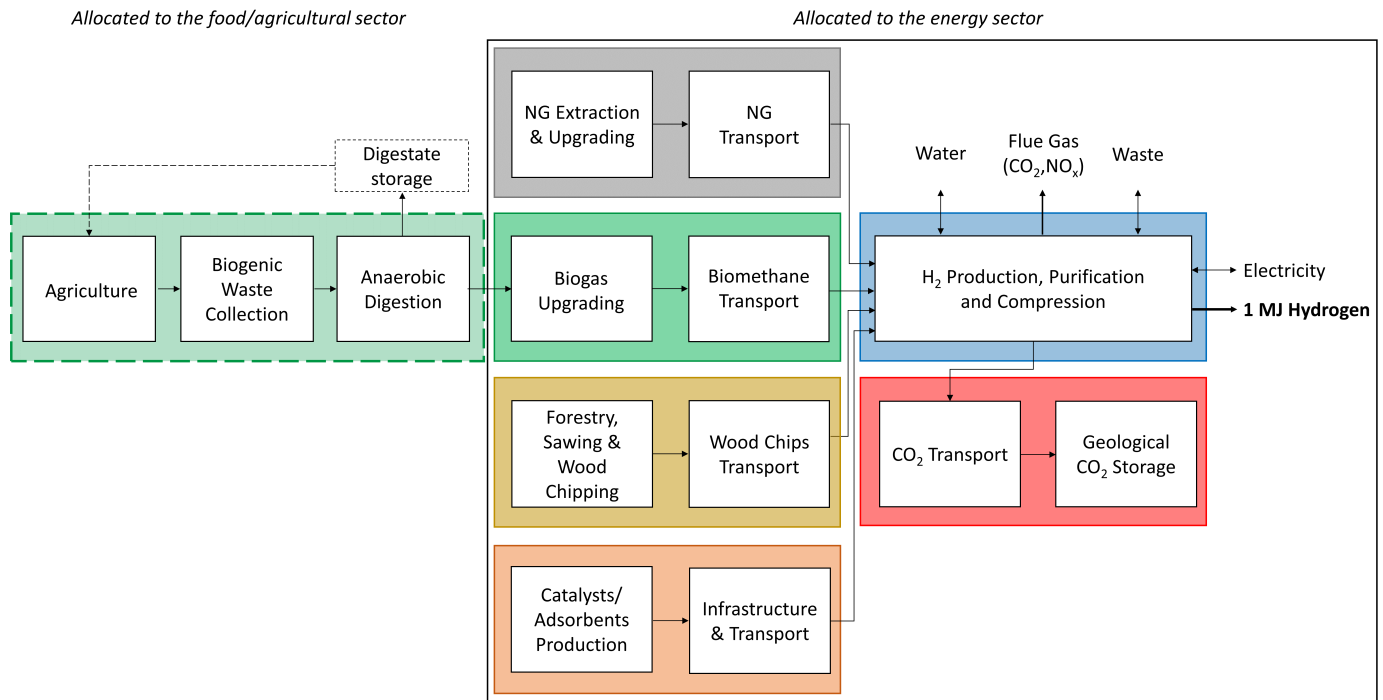


Fig. 4 System boundaries chosen for the LCA of H₂ production from natural gas (NG), biomethane, or wood as feedstock. The chain of the wet waste biomass down to the by-product biogas from treatment of the wet waste biomass in an anaerobic digestion plant is allocated to the food and agriculture sector. Extended from¹.

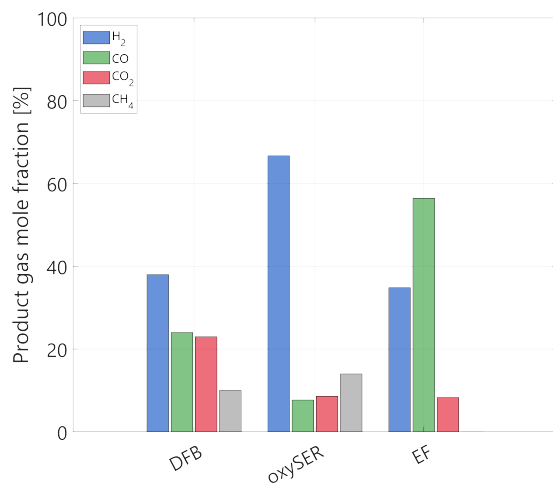


Fig. 5 Gasifiers product gas molar composition; DFB and oxySER: base case product gas composition.

tainty on both product gas composition and amount of carbon entering the combustor. The variation on the net efficiency from the base case composition to the lower/upper bound cases is around ± 7 pp. Additionally, by varying γ_c by ± 2 pp, the net efficiency in all three cases (BC, LB and UB) varies by ± 4 pp. Based on these results we can conclude that the performance of the oxySER configurations strongly depends on the operating conditions of the gasifier (i.e. temperature and residence time). As already explained in section 2.1, because of the oxy-fuel combustion, CO₂ can be recovered at the outlet of the combustion reactor without the need of a dedicated capture unit. Therefore, if we dry, compress and store the CO₂-rich stream produced by the combustor, we can reach a similar overall capture rate as the configuration with a DFB gasifier with pre-combustion capture (ca. 60%). Whereas by adding a pre-combustion capture plant (oxySER CCS) we can capture also the CO₂ present in the syngas, reaching an overall CO₂ capture rate of 92%. The remaining CO₂ is emitted in the flue gas resulting from the combustor after the PSA unit (see Figure 3). As shown in Figure 6b, the variation of γ_c affects the amount of CO₂ captured. However, its effect on the overall CO₂ capture rate is larger for the oxySER S than for the oxySER CCS configuration, because while applying a dedicated pre-combustion capture unit the perturbation on γ_c only affects part of the total CO₂ captured.

The highest overall CO₂ capture rate is reached by the configuration with an EF gasifier with CCS (ca. 98%); concerning the net efficiency both EF and EF CCS cases perform a bit better than the DFB configurations and they are in the range of the oxySER configurations.

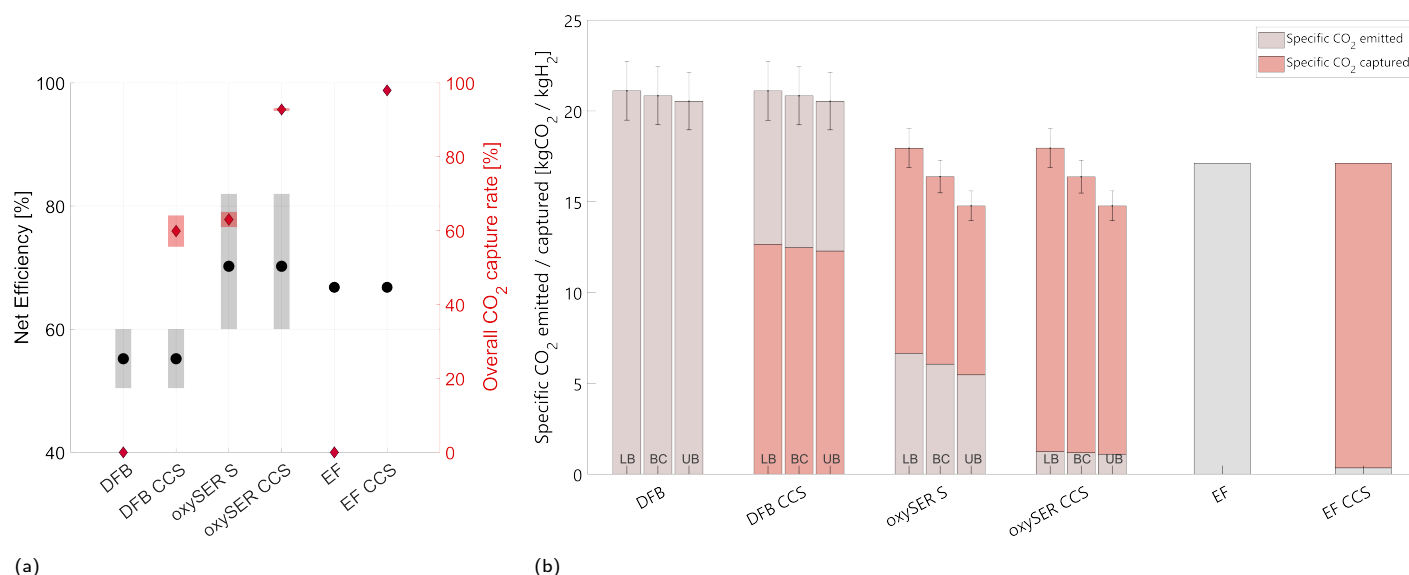


Fig. 6 (a) Comparison of the net efficiency (left y-axis, black dots and gray areas) and of the overall CO₂ capture rate (right y-axis, red diamonds and areas). (b) This figure shows the specific CO₂ captured and emitted in each production chain. For the DFB and oxySER cases the central bar corresponds to the base case (BC) product gas composition, on the left we have the lower bound (LB) while on the right the upper bound (UB). The error bar highlights the variation due to the sensitivity analysis on γ_C . For the DFB cases the change in γ_C affects the specific emissions (light gray), whereas for the oxySER cases it affects the CO₂ captured (light red).

5.3 Analysis on the specific electricity consumption

Figure 7a shows the net efficiency and the electricity balance for all configurations; when the electricity balance is below zero, electricity has to be provided to the system (all cases except of EF without CCS), while if positive, excess electricity is available and can be fed into the electricity grid (i.e. EF HTLT WGS CCS case). Figure 7b shows the specific electricity balance of the 6 configurations (for DFB and oxySER only the base case is considered) divided into eight categories of power usage: CO₂ capture, CO₂ compression, H₂ compression after the PSA unit to 200 bar, H₂ production plant auxiliaries (pumps, compressors, blower,...), air separation unit (ASU), O₂ compression, biomass feed pressurization and biomass pre-treatment.

In general, the addition of a CO₂ capture plant results in an additional energy demand, proportional to the amount of CO₂ captured; both electricity and heat are needed to run the CO₂ capture plant and the dehydration and compression section. The oxySER configurations are the most energy demanding; the variation on the electricity balance shown by the blue shaded area is mainly due to the variation on γ_C : the less carbon goes to the combustor, the less CO₂-rich flue gas is produced and therefore less CO₂ is dehydrated and compressed. Consequently, it results in a reduction of the energy demand.

Regarding the sensitivity analysis on the ω_{dtp} ratio, it only affects the electricity balance. Although the energy consumption of the biomass pre-treatment processes is varied by $\pm 20\%$, the overall electricity balance does not substantially change. Moreover, the only configuration that has a positive electricity balance is the case with a EF gasifier, and if CCS is added to this configuration the balance becomes negative, but it still performs better than all other configurations with CCS.

As shown in Figure 7b, the electricity required to compress

the hydrogen delivered by the PSA unit to 200 bar is more or less equal for all six cases. Focusing on the configuration with a DFB gasifier without CCS, we notice that the electricity balance is driven by the plant auxiliaries consumption (e.g. pumps, compressors, blower). If we add CCS, we can capture around 60% with an electricity consumption increase of around 43%. Regarding the oxySER configurations we also have to account for the electricity consumed by the air separation unit. The majority of the carbon (63 mol%) is combusted with oxygen in the combustion reactor, and thus also in the configuration without CCS (SER SMR WGS S), the electricity required for CO₂ compression is significant. By adding CCS we can increase the capture rate from 63 to 92% of the overall direct CO₂ emissions, which corresponds to an increase on electricity consumption of about 25%.

The configurations with an EF gasifier instead require considerably less electricity to run the plant auxiliaries; indeed, the system is operated under pressure and no compression of the syngas before the purification section is needed. However, they require extra electricity for biomass pre-treatment, O₂ compression and biomass feed pressurization. The specific electricity consumption of the DFB SMR WGS case is similar to the EF HTLT WGS case with the difference that the latter co-produces more electricity, resulting in a positive electricity balance. While adding CCS, because of the single-reactor configuration, an overall CO₂ capture rate of about 98% can be obtained; nevertheless, the electricity consumption is doubled (+ 114%).

5.4 Comparison between hydrogen production from wood, natural gas and biomethane

Figure 8 illustrates the comparison between the hydrogen production chains from wood and natural gas/biomethane. With the exception of the two configurations with an oxySER gasifier, the

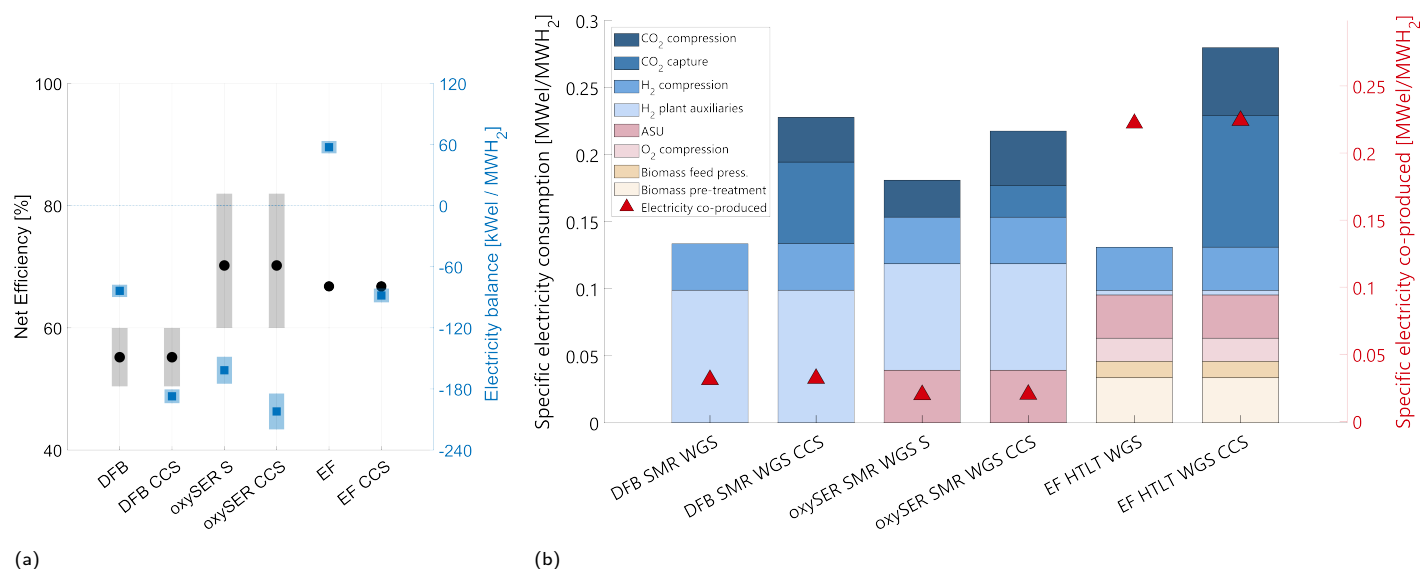


Fig. 7 (a) Comparison of the net efficiency (left y-axis, black dots and gray areas) and of the electricity balance (right y-axis, light blue squares and areas), of all hydrogen production configurations from dry biomass. (b) Specific electricity consumption of the wood to hydrogen production chains; the sum of the coloured bars gives the overall specific electricity consumption of the configuration. The red triangle shows the electricity co-produced by the H₂ production process, which is used internally. For all cases except of one (EF HTLT WGS), the amount of electricity co-produced is below internal consumption and this gap is assumed to be filled by electricity supply from the grid.

net efficiency is higher when using natural gas or biomethane as feedstock. The origin of this variation lies in the different C/H ratio in biomass with respect to methane (1:1.5 vs. 1:4). However, when using an oxySER gasifier, a considerable amount of external electricity is needed; indeed, for each MW of hydrogen produced, 160-200 kW of electricity are needed. The EF CCS configuration has a similar electricity consumption as the ATR HTLT CCS configuration, and the overall capture rate is also comparable (see Figure 8), but the net efficiency is considerably lower. Excluding few specific cases (oxySER upper bound product gas composition and EF configuration), the natural gas/biomethane production chains are performing better in terms of net efficiency and overall electricity balance; in fact, the amount of energy input to the production chain (both feedstock and external electricity from the grid) is considerably higher for the wood cases. However, in order to draw some final conclusion we have to look at the whole life cycle analysis of the different chains.

5.5 Climate change and other environmental impacts of hydrogen production from wood compared with reforming of natural gas or biomethane and electrolysis

Based on the technical modelling results discussed in the previous sections, the environmental impacts of H₂ production from wood gasification in the configurations DFB, oxySER and EF with and without CO₂ capture and storage have been calculated. Results are compared to the environmental impacts of H₂ production from reforming of natural gas or biomethane as well as from electrolysis with varying electricity sources as described in our previous work.¹ Figure 9 shows the contribution of different life cycle phases to the overall impact on climate change per MJ of H₂ produced. Further, it shows the overall CO₂ capture rate at the hydrogen production plant. The feedstock supply of wood

gasification processes, i.e. forestry and wood chipping, comes with net negative emissions from biogenic carbon uptake by trees even when GHG emissions from forestry, transportation, energy use for the chipping, etc. are considered. Due to the low H/C ratio of wood, a rather large amount of wood chips is required as feedstock to produce 1 MJ H₂, which leads to both a rather high amount of CO₂ removed from the atmosphere through the fuel supply, but also to high direct biogenic CO₂ emissions in cases without CCS. The DFB process chain without a pre-combustion CO₂ capture unit exhibits slightly positive impacts on climate change (20 g CO₂-eq/MJ hydrogen). This is due to the electricity required, assumed to be provided by the European grid, which is associated with life cycle GHG emissions of about 400 g CO₂-eq per kWh. The EF configuration exhibits climate neutrality even without CCS, which is due to surplus power generation by this process, assumed to substitute the average European power generation. This substitution is accounted for by corresponding GHG emission credits. OxySER S exhibits negative emissions (ca. -70 g CO₂-eq/MJ) even without a pre-combustion carbon capture unit, thanks to the oxy-combustion process. Even though the oxySER S configuration requires large amounts of electricity, the CO₂ emissions related to the use of electricity are more than compensated by the CO₂ captured from the combustion unit, when subsequent CO₂ transport and storage is assumed (see section 2.1 for the technology description).

When one considers the same H₂ production chains with pre-combustion CCS, the overall GHG emissions become negative in all the cases (between -80 g CO₂-eq/MJ for DFB CCS and -130 g CO₂-eq/MJ for EF CCS). The impacts of the carbon capture, transport and storage processes on the results are negligible, so that the fuel supply chain, CO₂ capture rate and power balance at the plant are decisive. Therefore, the addition of CCS leads

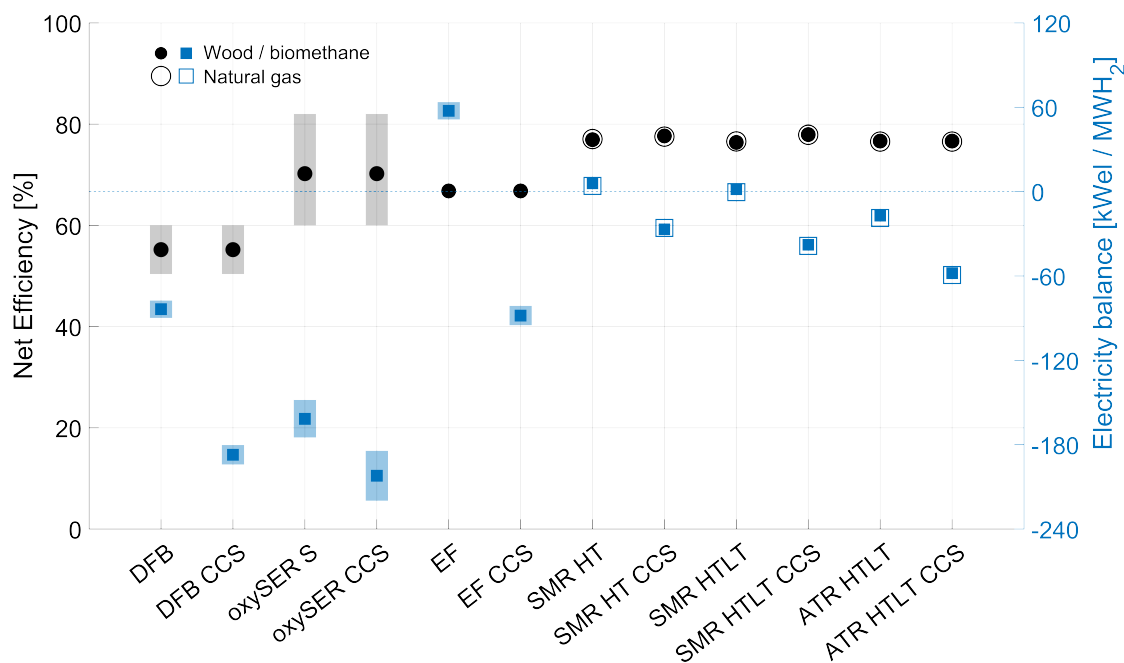


Fig. 8 Comparison of the net efficiency (black) and the electricity balance (light blue) between wood and natural gas/biomethane H₂ chains. The light-blue dotted line illustrates the 0 value for the electricity balance: all cases that are below this threshold have to import electricity while the cases that are above have electricity in excess to be fed into the grid.

to a substantially improved performance of all the process chains analysed with respect to overall greenhouse gas emissions.

Compared to other hydrogen production pathways, wood gasification performs well in terms of impacts on climate change. Electrolysis operated with renewable electricity generates hydrogen with life-cycle GHG emissions in the order of almost zero (with hydropower) to about 50 g CO₂-eq/MJ (with electricity from photovoltaic arrays). Natural gas reforming causes impacts on climate change in a range from about 90 g CO₂-eq/MJ in the case of ATR and SMR w/o CCS down to 20 g CO₂-eq/MJ for ATR with CCS. Using biomethane as input to the same reforming processes decreases the life-cycle GHG emissions to 10 g CO₂-eq/MJ (assuming low carbon uptake into biomass and digestate incineration) without CCS, going negative with CCS to -120 g CO₂-eq/MJ in case of a favourable carbon balance. For a detailed discussion of hydrogen from natural gas and biomethane reforming, we refer to our previous analysis.¹ However, the generally observed trend that wood gasification with CCS generates more negative emissions than biomethane reforming with CCS can be attributed to the higher H/C ratio of methane compared to wood, which leads to higher removal of CO₂ per unit of hydrogen for wood-based hydrogen.

Figure 10 provides LCA results for H₂ production with all woody biomass gasification configurations for a number of selected, representative impact categories (the complete set of results can be found in the ESI). We compare the dry biomass gasification process chains to autothermal reforming in each impact category. The scores of the results are normalized to the worst performing technology in each impact category.

Neither the use of wood nor addition of CCS result in large additional burden regarding non-renewable cumulative energy de-

mand, ecosystem quality and human health impacts compared to hydrogen from natural gas and biomethane. The only exception is land use: forestry for wood chips production is associated with forest land use and this dominates the results in the land use category. The non-renewable cumulative energy demand is highest for the use of natural gas, and shows even a slightly negative result in the case of oxyEF without CCS, which is a result of the substitution of European electricity. In freshwater ecotoxicity, the comparatively high impact of H₂ from wind-based electrolysis is driven by the use of stainless steel, which is not a required material in the other cases. Sources for emissions to air responsible for photochemical ozone creation are diverse and therefore the burdens in this impact category are driven by the use of natural gas, the wood chips supply chain, use of electricity, or material use (wind electrolysis). In general, results for hydrogen from electrolysis in non-climate change impact categories depend mostly on the source of electricity, even within the portfolio of renewables with e.g. considerable differences between hydropower or photovoltaic power. Therefore, generally valid conclusions for these impact categories comparing hydrogen from electrolysis with biomass-based hydrogen (with and w/o CCS) cannot be drawn.

In order to evaluate the environmental performance of hydrogen from different production pathways from an overall life-cycle perspective, we include the end use. We select the mobility sector and quantify environmental life-cycle burdens of passenger or freight transport vehicles in the next section.

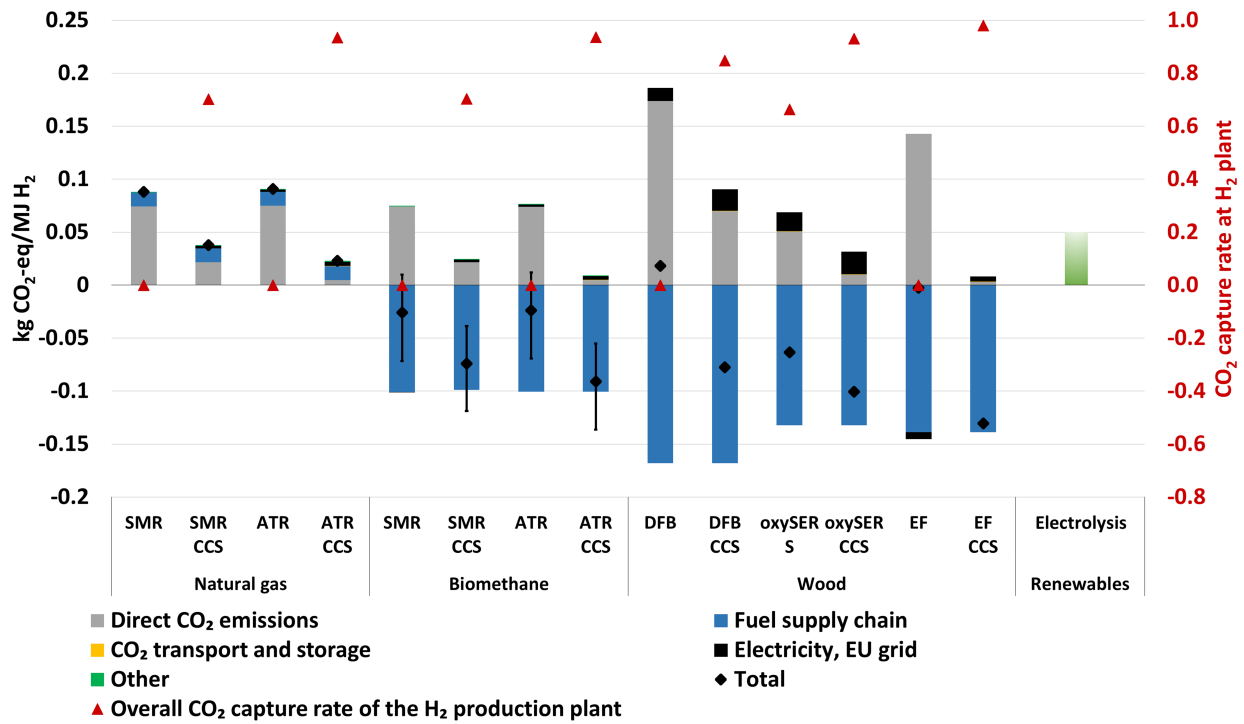


Fig. 9 Contribution of life cycle phases on the impacts on climate change from production of 1 MJ H₂ at 200 bar via selected technologies and system designs. The right y-axis shows the overall CO₂ capture rate at plant level. The category "Other" includes "Catalyst and Adsorbents", "Direct emissions from fuel combustion in furnace", "H₂ production unit infrastructure", and "Water supply".

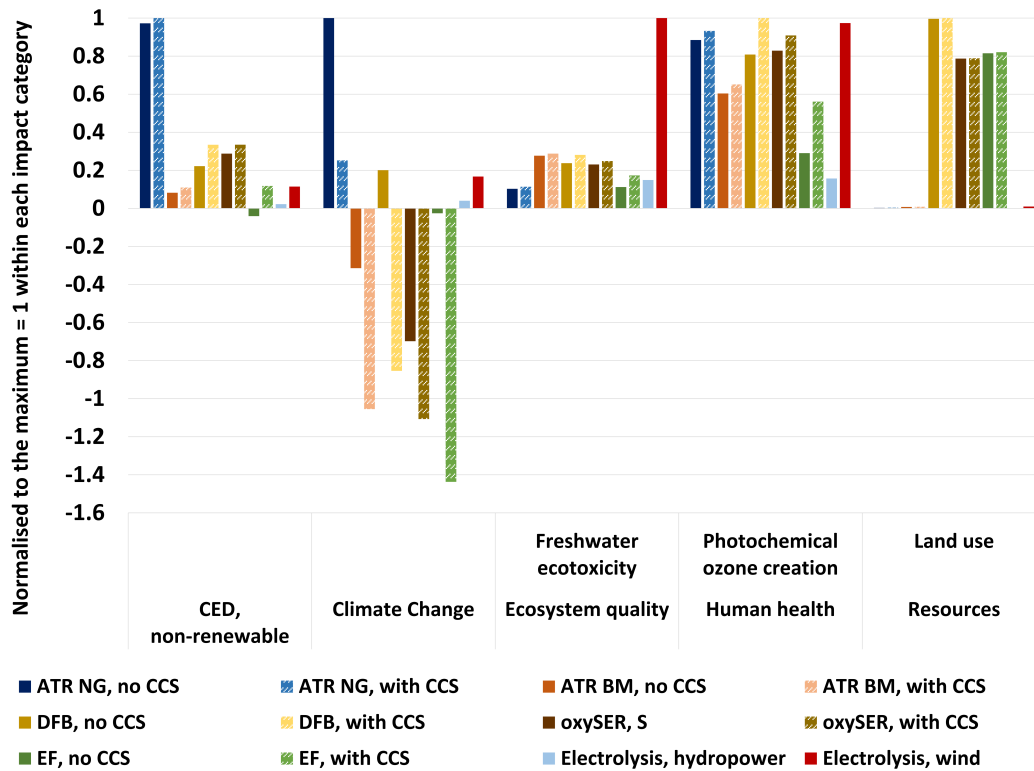


Fig. 10 Performance of selected individual technologies in selected impact categories per MJ of hydrogen. Scores are normalised to 1, which is the maximum score in each impact category, i.e. the technology which performs worst.

5.6 Hydrogen and its role in decarbonization of the transport sector - use of hydrogen in passenger cars and freight trucks

Figure 11 illustrates the climate change impacts and the contributions of various life cycle phases from driving 1 vehicle-kilometer (vkm) in a medium size passenger car of various drivetrain technologies and fuel supply chains (a), and results for 1 ton-kilometer (tkm) with a 7.5t truck in a regional driving cycle and average load factor, as specified elsewhere^{19,20}. Note that due to the different functional units the results for passenger vehicles and trucks are not directly comparable. We select conventional Internal Combustion vehicles (ICEV) with diesel (-d), natural gas/biomethane (-g), and petrol (-p), Fuel Cell Electric Vehicles (FCEV) with several hydrogen supply pathways, and battery electric vehicles (BEV) with different electricity supply options. We only consider steam reforming (SMR) with one (or two) water-gas-shift reactors for reforming of natural gas (NG) or biomethane (BM), as modeled by¹ to reduce the set of alternatives for hydrogen production. The BM chain selected here corresponds to the pessimistic range assuming low carbon uptake and release of carbon in the digestate of anaerobic biowaste digestion to the atmosphere (i.e. no long-term storage of that carbon in soil).¹

For passenger vehicles, only using H₂ from wood EF with CCS results in negative life-cycle GHG emissions, while in case of trucks, life-cycle GHG emissions are negative when H₂ from wood DFB and EF with CCS is used. This difference between passenger vehicles and trucks is due to the fact that fuel supply related contributions to life-cycle impacts on climate change of trucks are larger, since the "vehicle utilization" is higher for trucks than for passenger cars. This means that trucks exhibit a larger number of lifetime-kilometers, which leads to lower contributions from vehicle manufacturing and maintenance (corresponding to glider, powertrain, maintenance, energy storage, end-of-life), since the associated emissions are "amortized" over a larger number of kilometers. In general, the use of biomass as feedstock for fuel supply seems to yield substantially reduced climate change impacts compared to fossil fuel ICEV. Using woody biomass results in the highest carbon removal per km driven due to the different H/C-ratios of wood and biomethane, as discussed above. FCEV fuelled with almost all of the analyzed hydrogen production pathways as well as BEV using low-carbon electricity perform (much) better in terms of climate change than current conventional vehicles (ICEV diesel, natural gas or gasoline). An analysis of all other environmental impact categories (see ESI for the complete set of results) shows that this often does not result in significant negative environmental effects in other impact categories. Wood-based hydrogen supply causes high impacts in the land-use category for FCEV operated with such hydrogen. In addition, battery production for BEV can cause substantial burdens in some impact categories. The use of a carbon intensive electricity mix for charging a BEV or producing H₂ via electrolysis should be avoided due to even higher environmental impacts compared to conventional fuels. Such electricity supply results in high impacts on climate change, while the use of hydropower is most beneficial in all im-

port categories. This latter seems to be a good choice when aiming at decarbonization, together with FCEVs driven by H₂ from biomass, be it with or even without CCS. In contrast, the use of conventional fossil-based H₂ from reforming processes without CCS will be harmful for the climate. Adding CCS with a high capture rate (i.e. specific pre-combustion capture unit CO₂ recovery of 98%) is better suited for decarbonization, and the use of biomethane improves the climate performance even further, thus outperforming the direct use of biomethane in a gas vehicle. The use of wood chips exhibits one strong negative effect compared to other fuels, which is obviously land use. The good environmental performance of using wood or wet waste biomass feedstock for hydrogen supply for FCEV raises questions on the availability of these resources, and trade-offs between the necessity to fulfill a service demand (km driven) and minimising climate change impacts. With a given amount of wood feedstock, a larger distance can be driven in passenger cars than in trucks due to lower fuel demand of passenger cars. This effect increases with increasing size of trucks. However, as seen above, the carbon removal from the atmosphere is higher when using the H₂ in a truck than in a passenger car from a life-cycle perspective. In any case, biomass resource availability is limited, and it needs to be carefully evaluated to which use it should be allocated in terms of social, economic, technological and environmental performance.

6 Conclusions

This analysis represents an extension of our earlier techno-environmental assessment of hydrogen production from natural gas and biomethane with carbon capture and storage.¹ We extended the scope i) by including hydrogen production from woody biomass and ii) by evaluating the environmental performance of hydrogen use as vehicle fuel in comparison to alternative options, thereby filling important research gaps identified in our previous work.¹ We have performed an integrated techno-environmental analysis of the three most common gasification technologies for H₂ production from woody biomass: i) the dual fluidized bed steam gasifier (DFB), ii) the sorption enhanced reforming gasifier (oxySER), and iii) the entrained flow gasifier (EF), each of these with and without pre-combustion CO₂ capture followed by permanent geological storage (CCS). For this purpose, we have linked detailed process models of hydrogen production and Life Cycle Assessment (LCA) taking into account all relevant processes from forestry to end-of-life of vehicles. This procedure allows for a quantification of benefits and potential trade-offs of a range of process configurations from both technical and environmental perspectives in a consistent way. Nevertheless, a few simplifications made in this work should be considered more closely in the future:

- Wood gasification is a complex process and the process efficiency strongly depends on the type of feedstock used and on the operating conditions (e.g., residence time, temperature, wood water and impurities content). In this analysis we consider a standard wood composition which is in agreement with theecoinvent report on wood energy³⁴. However, a different feedstock composition might lead to differ-

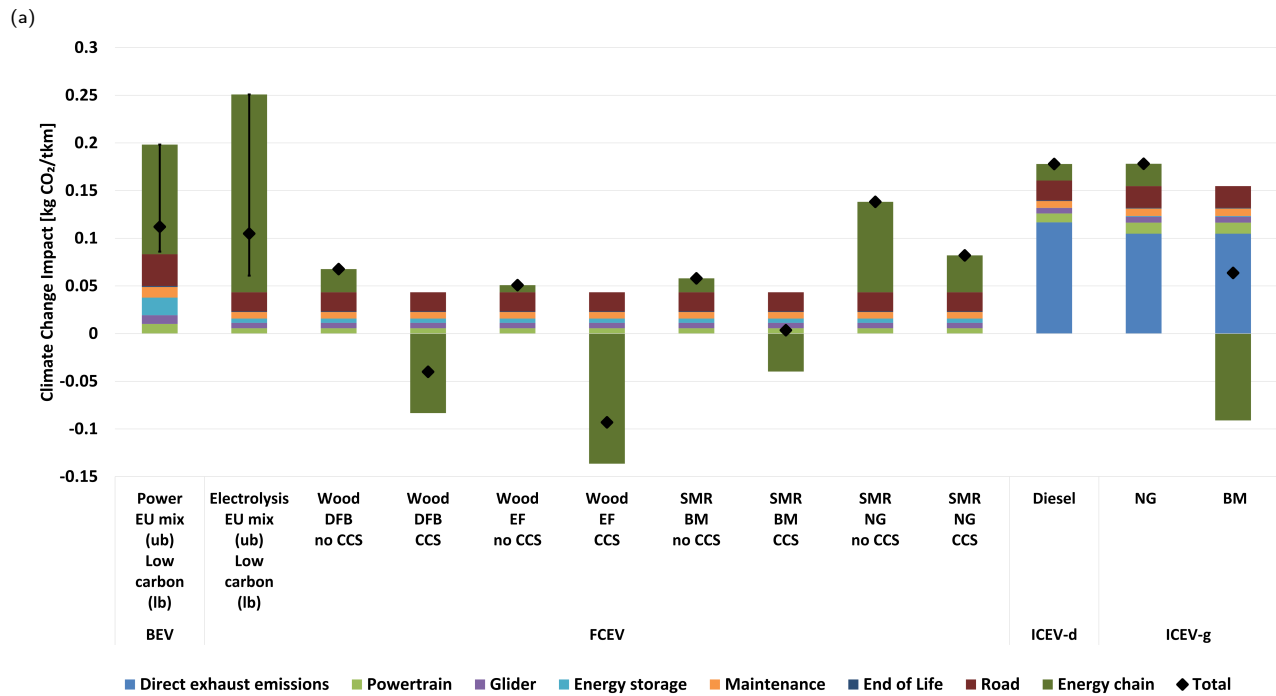
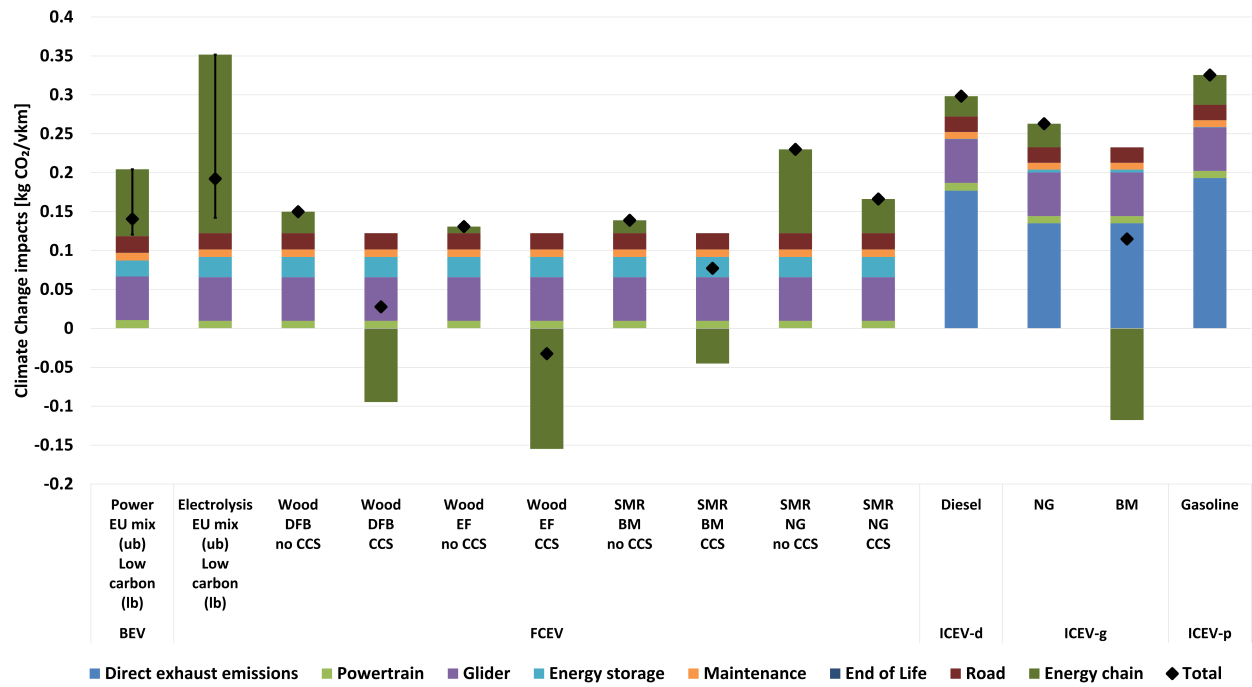


Fig. 11 Climate change impacts including contribution analysis for driving 1 km in a medium size passenger car (a) or a small 7.5 t truck (b) with varying fuel chains and drivetrains. The portfolio includes FCEV driven with H₂ from various sources as presented above; BEV supplied by an average European electricity mix (corresponding to the upper bound of the error bar), hydropower (corresponding to the lower bound of the error bar), and solar PV (corresponding to the "total"); and ICEV fuelled by conventional petrol, natural gas or biomethane, or diesel.

ent results (both in terms of process efficiency and type of pre-treatment required), and the wood quality is region specific. Concerning the operating conditions, it is hard to define standard operating conditions because those technologies are at an early stage of development and employment; nevertheless, in a real application the operating conditions of the gasification technologies should be optimized for the specific feedstock composition.

- Our LCA considers woody biomass feedstock for H₂ production from forestry in Germany and Sweden, as considered to be representative for the European wood chips market in the ecoinvent database.⁴² However, forestry-related environmental burdens depend on regional boundary conditions and wood markets can differ from region to region. Such differences should be addressed.
- The wood supply chain considered in this analysis represents "sustainable forestry", i.e. the use of trees extracted from existing forests in a quantity at or below the natural growth rate. Since the potential of such resources is limited, using wood from dedicated plantations should be analyzed, appropriately reflecting site-specific boundary conditions.
- Our quantification of selected environmental burdens in addition to impacts on climate change represents amounts of emitted pollutants, but not actual damages to human health and ecosystems. Quantifying these impacts would require regionalized or even location-specific impact assessment based upon specific dose-response functions - an issue which would be especially important in the context of mobility, but the LCA community is struggling with.

Acknowledging these limitations, we are still confident that our analysis provides reliable and useful outcomes, which can be summarized as follows. Regarding hydrogen production, the oxySER and EF configurations exhibit better technical and environmental performances than the DFB configurations - biomass-to-hydrogen conversion efficiencies as well as CO₂ capture rates of oxySER and EF configurations are higher, and both factors are key drivers for the environmental life-cycle performance. However, the oxySER process represents the most immature technology among the three configurations analyzed. All three wood-based hydrogen production configurations with CCS result in negative life-cycle GHG emissions, i.e. a "net-removal" of CO₂ from the atmosphere due to the permanent storage of CO₂ absorbed by trees; these results are similar to those of biowaste-based biomethane reforming with CCS. With the exception of land-use, which is substantial for wood supply chains, even under the assumption of "sustainable forestry" in the sense that wood consumption does not exceed natural growth rates, there are no clear negative environmental impacts resulting from the negative GHG emissions of wood-based hydrogen production with CCS. In general, hydrogen from biogenic feedstock used in fuel cell vehicles represents an environmentally sound fuel-powertrain combination, not only in comparison with conventional diesel, petrol and natural gas vehicles, but also in comparison with battery electric vehicles (BEV).

Regarding impacts on climate change, FCEV with hydrogen from biomass without CCS exhibit similar or slightly better (depending on the vehicle type) performance as BEV charged with low-carbon electricity. Adding CCS to biomass-based hydrogen production results in substantially lower impacts on climate change of FCEV compared to BEV, and in certain cases even to negative life-cycle GHG emissions per kilometer driven. However, these results require a careful interpretation. First, biomass resources to be used for hydrogen production are limited and can only provide fuel for a minor fraction of current vehicle fleets. And second, negative life-cycle GHG emissions for vehicles on a "per km" basis partially result from inefficient fuel use: if the fuel supply chain exhibits negative GHG emissions, the more fuel a vehicle consumes, the more CO₂ it removes from the atmosphere. More efficient fuel use would increase life-cycle GHG emissions per kilometer, but from a vehicle fleet perspective it would allow for travelling more km with the same amount of fuel and CO₂ removal from the atmosphere. Therefore, increasing the "negativity" of life-cycle GHG emissions of vehicles per km driven by increasing consumption of fuel associated with negative GHG emissions must not be the goal. The results of our analysis clearly demonstrate that biomass-based hydrogen - with and without CCS - must be considered as an environmentally sound transport fuel and that FCEV fuelled with such hydrogen represent an option to substantially reduce road-transport related impacts on climate change without major adverse environmental side-effects, if biomass is either sourced from waste streams or from sustainable forestry. Resource limitations need to be kept in mind though, and therefore, transition to a low-carbon transport system will require further fuel and vehicle options, e.g., low-carbon electricity used for hydrogen production via electrolysis, hydrogen from natural gas reforming with CCS, and direct electrification using BEV.

Conflicts of interest

There are no conflicts to declare.

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Appendix

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