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Evaluation of the environmental impact of HCNG light-duty vehicles in the 2020–2050 transition towards the hydrogen economy

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ABSTRACT

As the world intensifies its efforts to reduce the adverse effects of global warming, the shift towards a fully developed hydrogen-based economy is emerging as a core strategy. This transition involves the strategic blending of hydrogen with conventional fossil fuels such as natural gas (HCNG), allowing for adaptation to hydrogen availability. Nevertheless, the environmental impact of HCNG vehicles in realistic scenarios with variable hydrogen content remains unexplored. This study focuses on evaluating the global warming impact of the transition of light-duty passenger cars from CNG to H_2 vehicles using HCNG blends from 2020 to 2050 and different realistic scenarios. The results in the present study were obtained through a combination of an experimental testing campaign that allowed obtaining how the performance and emissions of HCNG vehicles change with the H_2 content and a life cycle assessment methodology. Based on the findings, the scenario in which hydrogen was mostly produced from SMR-dominant, was found to have the potential to reach and outperform the zero-emission concept due to the utilization of biogas. From the results of this study, the recommended H_2 content in HCNG blends that offers low environmental impact while avoiding the overdemand of hydrogen in the short term for the 2020–2030 decade is 25% H_2 content, increasing to 50% by 2030, and to 75%–100% during the 2040–2050 decade, thus reaching the transition towards pure-H_2 technology that minimizes environmental impact.

1. Introduction

The current issue of climate change presents a complex array of interconnected problems. One of the primary concerns is the escalating local temperatures, which have far-reaching implications for ecosystems, human health, and agricultural productivity [1]. Rising temperatures also contribute to the disruption of oceanic and atmospheric currents, leading to altered weather patterns and increased occurrences of extreme events such as hurricanes, droughts, and heat waves. These changes pose significant threats to vulnerable communities [2] and ecosystems [3] worldwide.

Addressing the multifaceted challenges of climate change requires comprehensive strategies and significant investments [4]. A crucial initial step is to curb our reliance on fossil fuels, which are the primary contributors to greenhouse gas emissions. Transitioning towards renewable energy sources, such as solar, wind, or hydroelectric power, can help reduce carbon emissions and mitigate the adverse impacts of climate change. This transition necessitates a shift towards sustainable transportation systems, increased energy efficiency measures, and the promotion of clean technologies. This interest in reducing fossil fuel consumption has led to the exploration of various solutions for the energy-intensive road transportation sector [5]. Electric vehicles (EVs) are considered a promising option due to their high efficiency in energy utilization. However, the widespread adoption of EVs poses challenges related to the availability of charging infrastructure, the demand for rare-earth elements, less fossil fuels dependent electricity mix [6], and the environmental impact of their production and disposal [7].

Hydrogen (H_2) has emerged as a promising alternative fuel, offering the potential to reduce the material requirements in vehicles. As a carbon-free fuel, it can be seamlessly integrated with renewable energy [8,9], resulting in significantly lower emissions during its production. However, the current state of hydrogen production is not yet fully environmentally friendly, as the industry is still in the process of scaling up renewable energy sources. Consequently, the availability of green hydrogen remains limited, suggesting that its widespread use may be constrained due to the current low supply. To overcome this challenge, the concept of blending hydrogen with fossil fuels has gained attention, aiming at ensuring equitable and accessible energy while transitioning towards a hydrogen-based economy. Recent research has focused on studying fuel blends of hydrogen with gasoline, diesel, ammonia, and compressed natural gas (CNG) [10,11].

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The addition of hydrogen has shown promising results in improving combustion performance and mitigating carbon dioxide (CO_2) tailpipe emissions. For instance, H_2 and CNG blends (HCNG) have demonstrated significant potential as an alternative fuel for internal combustion engines (ICE), delivering notable improvements in efficiency levels compared to pure CNG engines [12]. Furthermore, these blends offer the opportunity to reduce carbon monoxide (CO) and hydrocarbons (HC) emissions [13]. By enabling higher air dilution ratios, HCNG blends facilitate lower combustion chamber temperatures, thereby reducing nitrogen oxides (NO_X) emissions. Extensive research has also been conducted to evaluate the overall emission performance of vehicles running on HCNG blends, revealing substantial improvements compared to conventional fuels. In a previous study, Molina et al. [14] investigated the performance and emission implications of an engine fuelled with HCNG blends and pure hydrogen. This investigation simulates the hydrogen transition across various stages of infrastructure and green hydrogen production deployment by varying the percentage of hydrogen in the fuel blend. Results show an improvement in performance and emissions as the hydrogen substitution percentage increases, especially in terms of efficiency and NO_x.

However, this study only considers the effects caused by vehicle operation whereas other relevant aspects such as vehicle and fuel production are not pondered. In this sense, other studies evaluated the problem including vehicle operation, production, and fuel refinement processes. Numerous studies have been conducted to compare single fuels, whether they are conventional or unmixed, in the realm of alternative energy sources. For example, Karman et al. [15] conducted a comparative analysis between CNG and diesel fuels. Additionally, liquefied natural gas (LNG) has garnered significant attention in recent years, with several researchers examining the specific case of liquefied biomethane for heavy-duty transport [16–18]. Regarding passenger car applications, Desantes et al. [19] estimated greenhouse gas (GHG) and NO_x emissions for various fuel types, including hydrogen-fuelled vehicles (both fuel cells and internal combustion engines), battery electric vehicles (BEVs), compressed natural gas internal combustion engine vehicles, and conventional fuel vehicles.

These studies often involve scenarios that may not reflect long-term realities, potentially introducing bias into the results. For instance, in the case of hydrogen fuel, these scenarios frequently assume its exclusive production from biomass or biowaste [20] or from renewable energy sources [21]. Regarding natural gas (NG) based fuels, current studies aim to evaluate the environmental benefits of sourcing this fuel from various origins, including conventional extraction [22], natural resources such as sugar cane bagasse [23], or renewable hydrogen and waste sources [24,25]. While this research is undoubtedly valuable for understanding the potential of hydrogen and natural gas to reduce the environmental impact of transportation technologies, it may not adequately represent short-term conditions. Some of these technologies may not yet be available at the scale required to supply the existing vehicle fleet. Therefore, they cannot be used to assess the environmental impact of current vehicle technologies without considering a realistic evolution of the hydrogen and CNG mixtures over time in a specific geographical area.

Limiting consideration to a single fuel neglects a degree of freedom necessary to minimize the environmental impact of a particular vehicle application, as it restricts the ability to leverage fuel availability. For instance, in the case of HCNG fuel blends, the current limited availability of hydrogen can be offset by excess natural gas production. Moreover, because both fuels are gaseous, they can be utilized with relatively minor modifications to the propulsion system. Consequently, the adoption of HCNG blends offers a promising avenue to enhance vehicle performance and mitigate environmental impact.

In this field, Gupta et al. [26] conducted a comprehensive life cycle analysis of a heavy-duty vehicle powered by HCNG. The study focused on evaluating a 20% gaseous hydrogen blend in terms of net energy ratio, GHG emissions, and cost-effectiveness throughout the entire well-to-wheel cycle. Additionally, in their study [27], Gupta et al. analysed the well-to-wheel performance of a light-duty truck, considering different blends of hydrogen (0%, 15%, and 30%) within the HCNG mixture.

In the research conducted by Candelaresi et al. [28], they investigate the use of Hythane blends (20% hydrogen and 80% methane by volume) in ICE vehicles (ICEV) and hybrid-EV vehicles. The findings highlight vehicle infrastructure as the primary contributor to the environmental impact. However, vehicles using hydrogen blends with natural gas or gasoline show promise in promoting short-term hydrogen use with emissions reductions compared to conventional vehicles. In another study [29], they compare the environmental life cycle assessment (LCA) of eight-passenger car fleets. These fleets utilize renewable hydrogen and a conventional fuel (natural gas or gasoline) under the same total energy input and hydrogen-to-mixture energy ratio. The proposed strategies achieve a reduction in carbon footprint ranging from 7% to 35% compared to conventional fleets. Nevertheless, none of these studies optimized the hydrogen content in the fuel to minimize the environmental impact of HCNG vehicles by simultaneously considering both fuel production and tailpipe emissions. Therefore, future research and development efforts should focus on optimizing blend compositions and investigating their long-term effects on engine performance, emissions, and overall system compatibility.

1.1. Knowledge gaps

Building upon the findings of previous studies, the literature highlights several knowledge gaps, as summarized below:

1. Research on fuel blends is limited: The majority of studies projecting the future environmental impact of fuels primarily concentrate on single, unmixed compositions [15–19]. There is a notable absence of extensive literature dedicated to investigating the effects of fuel blends, especially in the case of HCNG, across a broad spectrum of mixtures.
2. Heavy-duty Application Focus: The majority of studies on HCNG predominantly centre around heavy-duty applications [26,27]. There is a scarcity of research exploring the potential of HCNG in other sectors or applications.
3. Relying on Literature-Based Estimations: A few of the identified studies rely on experimental results when estimating fuel consumption and emissions in the operational phase of the life cycle assessment. Instead, estimations are predominantly based on existing literature, indicating a need for more empirical data in this aspect [28,29].

Based on the previous considerations, it becomes evident that combining engine experiments with life cycle assessment is essential to predict a realistic scenario where HCNG-powered engines for automotive applications could play a significant role in global decarbonization. For this reason, this study aims to evaluate the environmental impact of HCNG light-duty vehicles from 2020 to 2050, using realistic scenarios that can indicate the optimal transition strategy for decarbonizing the transportation sector with HCNG, hydrogen, and CNG-fuelled vehicles.

1.2. Contribution and objectives

The motivation behind this study is to assess the environmental impact of HCNG-powered vehicles in realistic scenarios and to optimize this technology for the purpose of decarbonizing the transportation sector during the transition towards a hydrogen-based economy. The specific contributions aimed at achieving this objective include:

- Quantifying the impact of using different blends of Hydrogen-Compressed Natural Gas (HCNG) on fuel consumption and emissions in a light-duty vehicle's internal combustion engine (ICE).

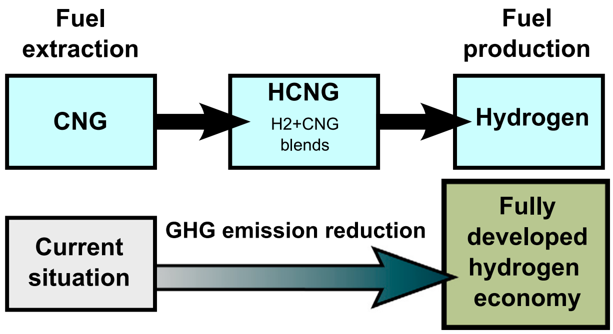

Fig. 1. Diagram of the proposed method based on the fuel transition.

- Analysing the potential cradle-to-grave emissions of HCNG vehicles depending on the energy, NG and H_2 mix evolution.
- Determining the optimal HCNG blend for minimizing cradle-to-grave emissions between 2020 and 2050 and provide recommendations regarding the optimal transition from CNG to H_2.
- Identifying the biogas requirements for achieving zero cradle-to-grave emissions, if feasible.

2. Methodology

Recognizing the imperative of transitioning from fossil fuels to hydrogen, it is essential to underscore the gradual nature of this process, emphasizing that such a transformation cannot be instantaneous. A noteworthy challenge lies in the absence of a dedicated hydrogen infrastructure; however, the existing natural gas infrastructure holds promise as a viable interim solution. Particularly noteworthy is the capability of the natural gas grid in the European Union to transport up to 12% of the hydrogen volume and, in the United States, up to 15% [30]. Given these considerations, strategic utilization of the current natural gas infrastructure emerges as a prudent approach to expedite the shift away from fossil fuels towards a hydrogen-based energy paradigm. A gradual approach is sought through blends with fossil fuels, with CNG acting as the representative fossil fuel, as presented in Fig. 1.

Observations have indicated that HCNG offers advantages in terms of reducing typical emissions such as CO, NO_x, and unburned hydrocarbons. However, the performance of HCNG varies depending on specific circumstances, engine types, and operational modes.

Five scenarios are proposed to characterize this transition: pure CNG, a blend of 75% CNG and 25% H_2, a blend of 50% CNG and 50% H_2, a blend of 25% CNG and 75% H_2 by mass, and finally, pure H_2. These scenarios aim to define a gradual progression towards hydrogen economy.

2.1. Experimental tools

The experimental study was conducted on a research engine with a single cylinder and a spark ignition system. The engine was equipped with a port fuel injection system. The specifications of the engine used in this campaign were identical to those employed in previous research works [11,14]. The most relevant data are listed in Table 1 for reference. The test cell used in this research is illustrated in Fig. 2, which features a modified layout to accommodate the implementation of dual gaseous fuel strategies.

To enhance precision in the injection and mixing procedures, a pair of Zavoli JET Injectors were utilized to separately inject the fuels into the intake manifold. These injectors boast a maximum pressure of 4.5 bar and have been engineered to operate within a temperature range of −40 °C to 120 °C. The discharge nozzle of each injector measures 3 mm in diameter.

Table 1
Main engine specifications.

Number of cylinders	1
Displaced volume	454.2 cm^3
Stroke	86.0 mm
Injection systems	PFI
Ignition system	Spark plug
Cylinder diameter	82.0 mm
Compression ratio	10.7
Connecting rod length	144.0 mm
Valves per cylinder	2 intake, 2 exhaust
Engine management system	AVL PREMS GDI
Combustion system	4-valve pent roof GDI
IVO[a]	−380 CAD
IVC[a]	−135 CAD
EVO[a]	−600 CAD
EVC[a]	−338 CAD

[a] With respect to the firing TDC (0 CAD).

Exhaust gas composition measurements, including O_2, CO, CO_2, HC, and NO_x, were conducted using a gas analyser HORIBA MEZA 7100 DEGR. To facilitate the analysis, a sample of exhaust gases from the settling chamber was directed through a pre-heated pipe maintained at 150 °C to the gas analyser. For the measurement of instantaneous in-cylinder pressure, a piezoelectric sensor was used. Piezoresistive sensors, on the other hand, were employed to measure intake and exhaust pressures. To control the mass flow rate of both fuels, two flowmeters were employed: the Bronkhorst F-113AC-1M0-AAD-55-V for hydrogen and the F-113AC-M50-AAD-55-V for CNG. The experimental facilities provided comprehensive control over all relevant parameters. Boost conditions were achieved through an external compressor, and exhaust back pressure was regulated by a knife-gate valve positioned in the exhaust line.

The accuracy of the instrumentation used in the study can be found in Table 2. Table 3 presents the precision of the gaseous pollutant measurements using the HORIBA device.

Compressed natural gas used in this study was sourced directly from the Spanish natural gas network. The composition of CNG consisted of 89.95% methane, 6.27% ethane, and other impurities. On the other hand, hydrogen was provided in pressurized tanks. Additional information regarding the characteristics of both fuels can be found in Table 4.

Engine and combustion-related output parameters, such as indicated mean effective pressure (IMEP), cycle-to-cycle variability expressed by the IMEP covariance (COV_{IMEP}), emission levels, and indicated efficiency, were computed using an in-house combustion diagnostics tool [31]. The tool was adapted for hydrogen, CNG, and HCNG fuel blends. The combustion diagnosis tool referenced in this study employs an estimation approach to calculate the energy released during combustion. This estimation is achieved by solving the energy equation using the measured in-cylinder pressure while making certain simplifying assumptions. One such assumption is the consideration of uniform pressure and temperature distributions throughout the entire combustion chamber. Additionally, simplifications are made to estimate the heat transfer to the cylinder walls and other related factors. To enhance accuracy in the calculations, the combustion diagnosis tool has undergone modifications, as described in previous work by Benajes et al. [32]. These modifications were implemented to reduce errors associated with the estimation process. Originally, the model considered three distinct components: air, fuel, and combustion products. However, the current version of the model treats the fuel component as a single hydrocarbon entity ($C_xH_yO_z$) with equivalent properties to those of the actual fuel mixture.

2.2. Life cycle assessment

The environmental impact of HCNG vehicles according to their H_2-CNG mixture is evaluated by means of a life cycle assessment (LCA).

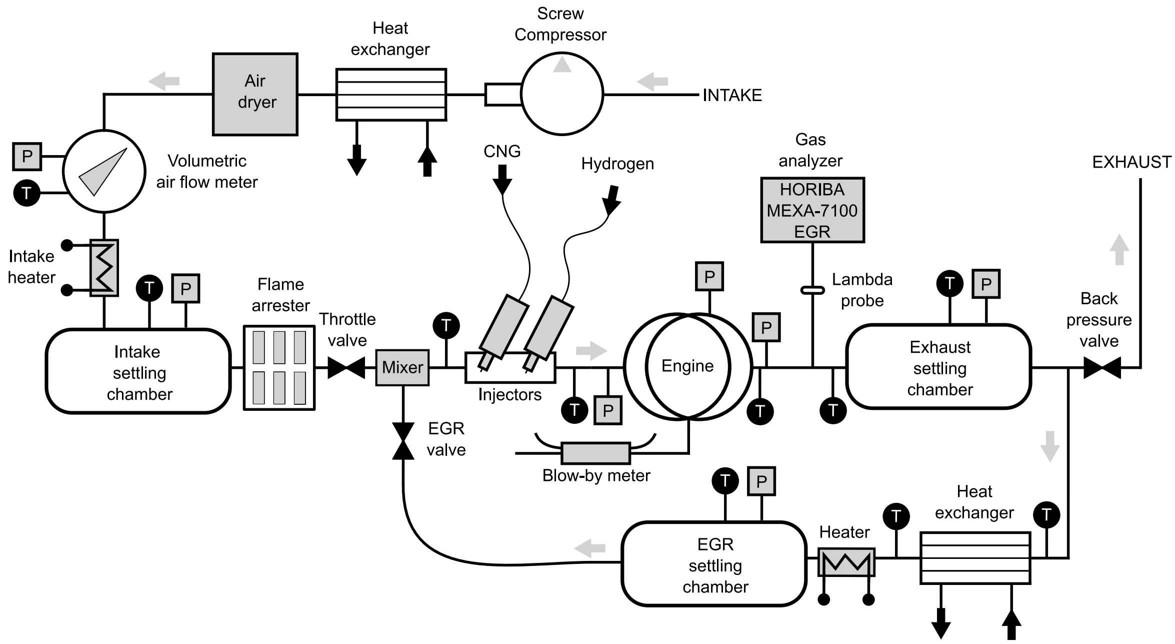

Fig. 2. Test bench layout.

Table 2
Instrumentation accuracy.

Signal (high frequency)	Sensor	Specification
In-cylinder pressure	Piezoelectric sensor	0 to 250 bar ± 0.3% linearity
Intake pressure	Piezoresistive sensor	0 to 10 ± 0.001 bar
Exhaust pressure	Piezoresistive sensor	0 to 10 ± 0.001 bar
Variable (low frequency)	Sensor	Specification
Engine Speed	Optical angular encoder	1 to 6000 ± 1 rpm
Engine Torque	Strain-gauges torque-meter	−200 to 200 ± 1 N m
Intake pressure	Piezoresistive transducer	0 to 10 bar ± 1%
Exhaust pressure	Piezoresistive transducer	0 to 10 bar ± 0.3%
Intake temperature	Thermocouple type K	0 to 1000 ± 0.5 °C
Exhaust temperature	Thermocouple type K	0 to 1000 ± 0.5 °C
Fluid temperature	Pt100 thermoresistance	−200 to 850 ± 0.3 °C
Air mass flow	Air flow meter	0.6-100 m^3/h ± 1%
Hydrogen mass flow	Thermal mass flow meter	200-1600 l/min (based on N_2) ± 0.5%
CNG mass flow	Thermal mass flow meter	200-1600 l/min (based on N_2) ± 0.5%

Table 3
Accuracy levels of HORIBA MEXA 7100 DEGR for measurements of gaseous pollutants.

Pollutant	Analyser	Range	Accuracy
HC	FID	min. 0 to 10 ppm C max. 0 to 50 kppm C	± 3%
NO_x	CLD	min. 0 to 10 ppm max. 0 to 10 kppm C	± 3%
CO	NDIR	min. 0 to 3 kppm C max. 0 to 12 vol%	± 3%
CO_2	NDIR	min. 0 to 5 kppm C max. 0 to 20 vol%	± 3%
O_2	PMA	min. 0 to 5 vol% max. 0 to 25 vol%	± 3%

Table 4
Specifications of CNG and H_2 fuels.

Properties	CNG	H_2
RON	120	130
AF_{st}	16.00	34.3
LHV	46.87 MJ/kg	119.9 MJ/kg
H/C	3.79	–
O/C	0.026	–
Molar mass	17.77 g/mol	2.01 g/mol
Purity	–	≥99.9%

An LCA consists of evaluating the environmental impact of a given technology by including the emissions produced during its operation, its manufacturing and the production of the fuel required for its operation or any combinations of these processes. This methodology can be applied in different scenarios to understand how a given technology would perform depending on factors such as the carbon intensity in the energy mix of a given country. In the case of the automotive industry, a cradle-to-grave LCA implies calculating the emissions associated with the manufacturing of all the components of the vehicle, quantifying/measuring the emissions produced during its operation and those generated during the production of the energy carrier used in the vehicle, be it electricity, H_2 or any other fuel. Therefore, cradle-to-grave LCAs provide a fair basis for comparison of transportation technologies fuelled with alternative fuels by avoiding any bias coming from the omission of a given stage of the life cycle.

The first step of an LCA is to define the scenarios under study in terms of the fuel mixtures, time, and geographic location since these factors influence vehicle manufacturing and fuel production emissions. Then, the boundaries and environmental flows are selected since they define the processes included in the LCA which emissions are to be considered as part of the life cycle of the vehicle as well as the inputs and outputs that are taken from and released to the environment. Then,

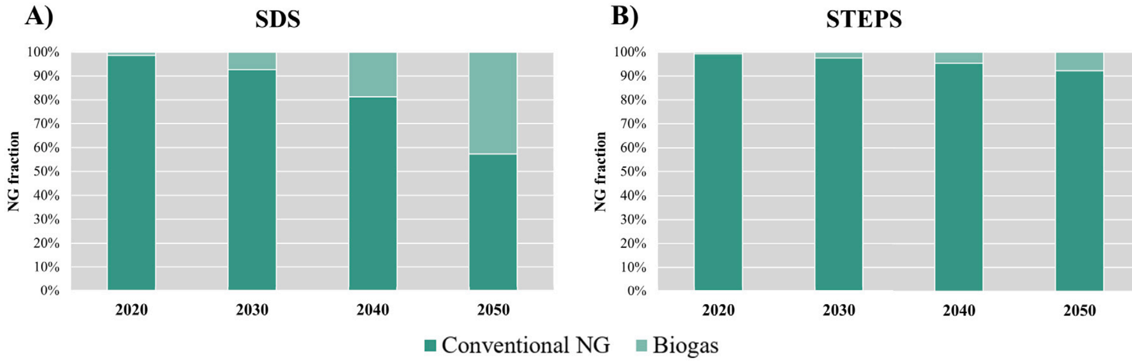

Fig. 3. Time evolution of the natural gas fraction obtained from conventional gas and biogas in the SDS (A) and STEPS (B) scenarios.

the functional unit is selected as the unit corresponding to the amount of emissions provided in the study. For instance, if the functional unit is 1 vehicle and 150000 km, then it means that for the overall LCA all the emissions results will correspond to the manufacturing of 1 vehicle and its operation during 150000 km. The impact categories are then defined as the type of environmental impact quantified in the study. Finally, the life cycle inventory, which defines the data and its source used for the LCA, is presented.

In this study, this methodology is only applied to evaluate the global warming impact of the technology since the main purpose of transitioning towards these vehicles is to mitigate the GHG. And, since considering other impact categories in the numerous scenarios considered would result in an incomprehensible amount of information with unclear conclusions.

2.2.1. Scenarios definition

The scenarios considered in this study comprise a set of possibilities whose impact on the LCA is sometimes grouped in error bars and sometimes differentiated clearly. These scenarios and their impact on the environmental flows and production pathways are:

- Geographic: Europe.
- Temporal: 2020, 2030, 2040 and 2050.
  - Impact 1: Electricity mix evolution.
  - Impact 2: H_2 mix evolution [33].
  - Impact 3: Natural gas composition evolution [34].
- Strategic towards the use of natural gas: SDS and STEPS.
  - Impact: biogas fraction in the natural gas mix (Fig. 3). [34]
- Strategic towards the H_2 economy: Steam methane reforming (SMR) and Electrolysis dominant.
  - Impact: dominant technologies in the H_2 production pathway (Fig. 4) [33].

The evolution of the electricity mix together with its uncertainty was obtained from the Sphera professional database according to the energy trends, the NI (no improvements) and the SI (significant improvements) scenarios. In the NI scenario, there is no improvement in the sustainability policies with time while the SI scenario implies a significant improvement in the sustainability policies compared to the current policies. The carbon intensity in the electricity mix, i.e., the GHG emissions per kWh of consumed electrical energy, was used as an input for all the processes in the CNG, H_2 and vehicle components production. An average value from the energy trends, NI and SI scenarios was considered while the highest and lowest values were used to compute the emissions uncertainty on such processes.

The source of the natural gas has a significant impact on the well-to-tank emissions. If the source of the natural gas is a byproduct or waste from other chemical processes (water sludge, for example), then its environmental impact is computed as the emissions produced in the process of capturing and processing it minus the hypothetical impact of releasing it to the environment, since that would be representative of an "inaction" scenario. Therefore, since the global warming potential (GWP) of CH_4, which is the major gas in NG mixtures, is significantly higher than that of CO_2, it is possible to find negative emissions in the well-to-tank and even in the well-to-wheel processes. Nonetheless, it is imperative to develop a careful analysis of these negative emissions to avoid any undesirable bias towards biogas.

Given the relevance of accounting for these negative emissions, it is imperative to identify the fraction of the natural gas that comes from conventional gas (extraction from natural resources) and from biogas (from processes of natural origin that otherwise would be released to the environment). This was considered in this study by means of two potential scenarios, defined as STEPS and SDS, following the report issued by the International Energy Agency about the prospects for the evolution of the natural gas and biogas mix worldwide [34]. The natural gas mix and its expected evolution with time is presented in Fig. 3. As with the energy mix, the average natural gas mix between these scenarios is used as an input for the H_2 and CNG production pathways while the upper and lower values in terms of emissions are considered as the uncertainties.

The H_2 mix evolution depending on whether the electrolysis-dominant or SMR-dominant scenarios are considered was obtained from the Hydrogen Roadmap for Europe [33]. The breakdown in the H_2 production pathway source is presented in Fig. 4.

The combination of this set of scenarios implies that the final results in terms of cradle-to-grave emissions account for the uncertainty of the environmental impact to the energy, natural gas and H_2 mixes in the major pathways, thus providing a clear picture about the future trends and the expected uncertainty. This is expected to permit the identification of the optimal HCNG mixture and provide recommendations about how the HCNG vehicles should evolve in the coming decades to imply lower emissions than CNG and H_2 ICEV if possible.

2.2.2. Boundaries and environmental flows

The boundaries of an LCA define the processes involved in the production pathways that contribute to the environmental impact under study. The inputs and outputs that come directly from the environment are called environmental flows and can be raw materials, water, or pollutants, among others. The boundaries and environmental flows considered in this study are presented in Fig. 5. As it can be seen, the boundaries comprise the extraction of raw materials, their processing, and their use in the vehicle during the operation phase. In Fig. 5, it is possible to differentiate the different processes associated with the H_2 production pathways attending to the colours. The reader should note that in this study there are several H_2 production pathways according to the mixes in Fig. 4. One of the key novelties of this study is that, despite not proposing any novel hydrogen production method, a combination

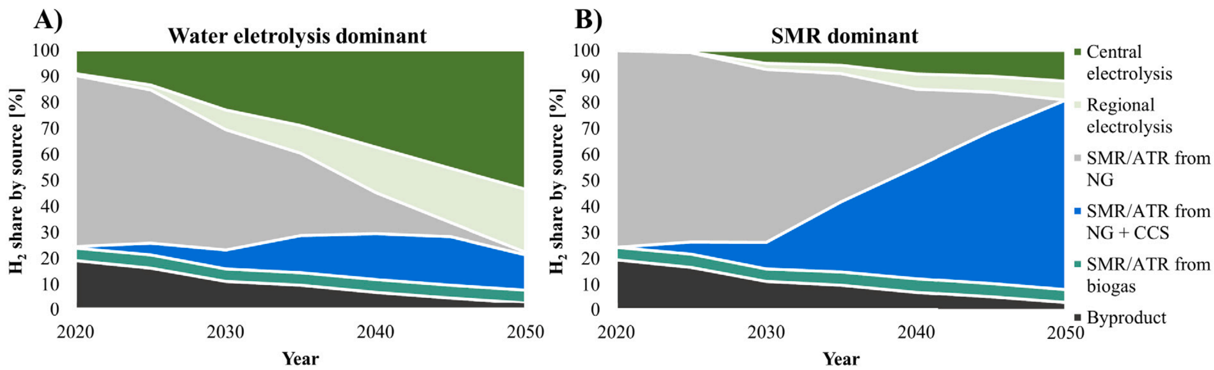

Fig. 4. Evolution of the H_2 production pathway breakdown from 2020 to 2050 following the electrolysis-dominant (A) and SMR-dominant (B) scenarios proposed by the EU [33].

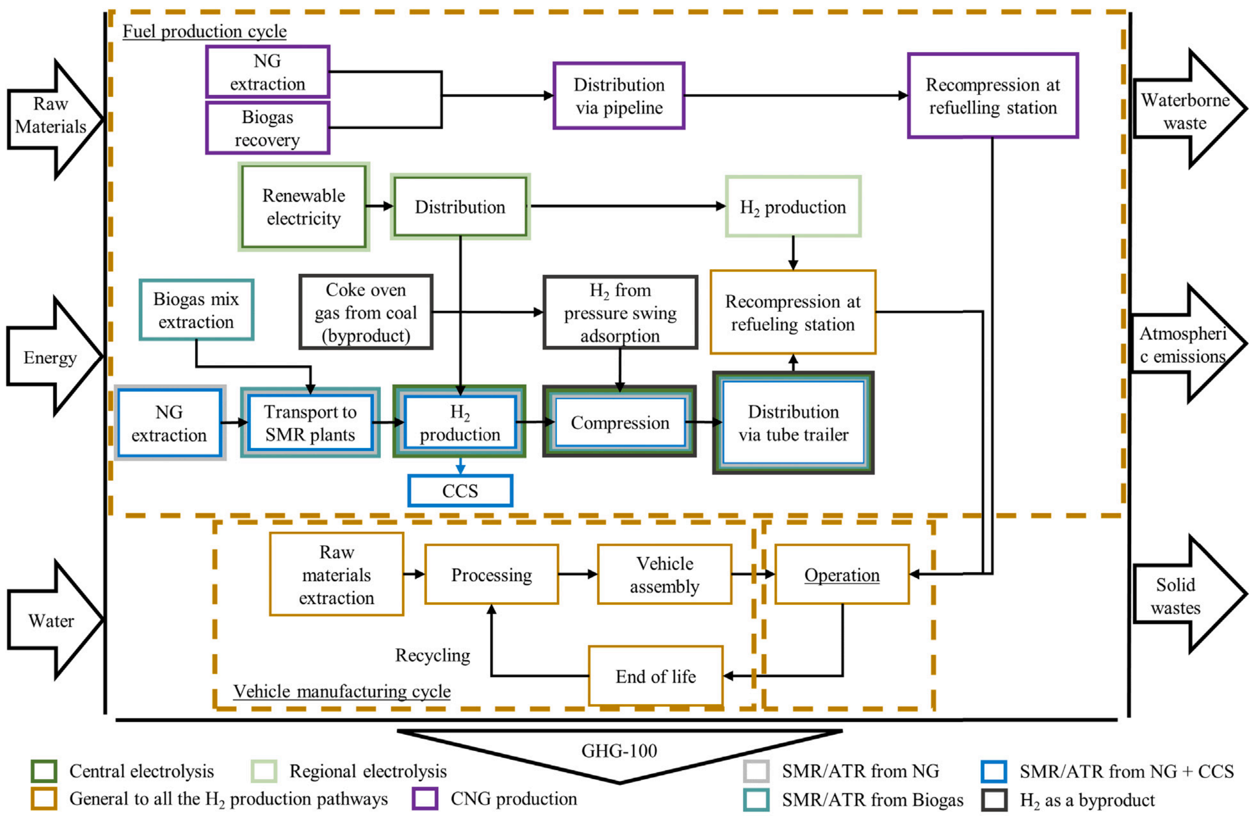

Fig. 5. System boundaries and environmental flows.

of hydrogen production pathways that is representative of the current and future scenarios, following the hydrogen roadmap in Europe, was proposed. Furthermore, these production pathways are combined with different electricity mix and natural gas mix scenarios, thus providing an integral LCA framework that can be used to identify the potential cradle-to-grave emissions of HCNG vehicles in the 2020–2050 period and the uncertainty associated with the evolution of such mixes. The processes included in this study are presented in Fig. 5, the hydrogen mix scenarios in Fig. 4, the natural gas mix scenarios in Fig. 3 and the electricity mix scenarios were extracted from LCA for experts (Sphera professional) database and described in Section 2.2.1.

2.2.3. Functional unit

The functional unit of this study depends on the part of the life cycle under analysis. For the well-to-tank part, the functional unit is 1 kWh of energy contained in the fuel, considering its lower heating value. In the cradle-to-gate, the functional unit is 1 vehicle produced. For the tank-to-wheel and well-to-wheel phases the functional unit is a lifetime of 150000 km. Finally, for the cradle-to-grave process, all the emissions are referred to 1 vehicle produced and a lifetime of 150000 km.

2.2.4. Impact categories

The impact category considered is global warming by means of greenhouse gases and their effect in a 100-year horizon (GHG-100). Other impact categories were not considered since an extensive analysis of this pollutant is explored in so many dimensions and scenarios that including further impact categories may affect the readability of this study. Furthermore, GHG were considered since mitigating the global warming impact is the main motivation behind transitioning towards the Hydrogen Economy and more sustainable fuels. Therefore, considering this impact category is completely in line with the objectives of this study.

2.2.5. Life cycle inventory

The life cycle inventory used in this study is based on the databases of GREET® v2022 and Sphera Professional. First, the software Sphera Professional (LCA for experts) together with the professional database was used to obtain the carbon intensity of the EU scenarios from 2020 to 2050. The carbon intensities considered in each of the timeframes are three: those predicted from the energy trends report (ETS) according to the European Commission forecasts [35], those in a scenario where there are no improvements in the sustainability policies, and those in the scenario where there are significant improvements in the sustainability policies as defined in the Sphera Professional database.

Then, the electricity from these mixes and their associated carbon intensity was used as an input for the GREET® model in all the production pathways, including those for CNG, H_2, and the vehicle components. The combination of both tools enabled obtaining the cradle-to-gate and well-to-tank emissions of the HCNG vehicles and the associated uncertainty caused by the variability in future scenarios.

2.2.6. Limitations and hypotheses

The main scope of the paper is to understand the environmental impact of HCNG vehicles and its uncertainties in the long term with the scenarios proposed by the EU in terms of the electricity, NG, and H_2 mixes. This will provide an understanding of which HCNG mixture should be used as time goes by to minimize the environmental impact of light-duty vehicles and what these scenarios mean in terms of actual GHG emissions. Nonetheless, this study comprises the same hypothesis and assumptions behind these scenarios. The main assumption is that there is enough energy, natural gas, biogas, and H_2 to cover the fuel demand of the HCNG vehicle fleet, be it because the production is increased or because the fuel demand of the fleet is small. Therefore, it is not within the scope of the paper to indicate whether the considered scenarios are realistic, but rather to identify the implications of environmental impact for the HCNG vehicles in case they are fulfilled. Furthermore, the analysis is mainly focused on the environmental impact of HCNG vehicles, so there is no cost analysis depending on the blend. This is out of the scope of this study and is left for future work.

Other relevant hypotheses included in the LCA are:

- The investigation primarily centres on mid-size passenger vehicles, given their predominant representation within the contemporary vehicular fleet.
- The constancy of fuel production and engine technologies over time is assumed, notwithstanding fluctuations in the significance of individual production pathways. Consequently, the emissions projections associated with these factors in the EU 2050 scenario may exhibit slight under or overestimation.
- The emissions stemming from the manufacturing processes of the machinery and devices employed in energy source generation and vehicle production are omitted from consideration. The emissions generated during the transportation of materials between factories for the vehicle manufacturing cycle are neglected as well. These omissions are deemed inconsequential in the context of the overall vehicle life cycle emissions, as the machinery in question is consistently utilized in the industry for the production of alternative vehicles or the generation of H_2.

3. Fuel consumption of HCNG vehicles

This section shows the process of obtaining the fuel consumption of CNG, HCNG blends, and H_2 vehicles, starting from the experimental data to the derivation of the fuel consumption values.

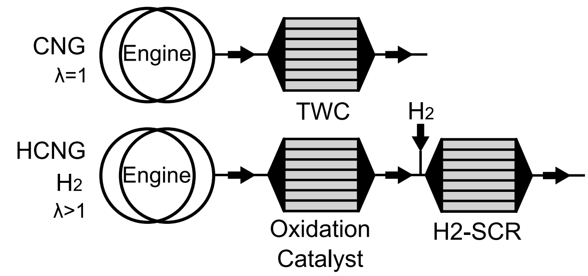

Fig. 6. Schematic representation of the after-treatment systems.

3.1. Engine fuel consumption and emissions

To evaluate the fuel consumption and performance of a representative engine, the authors referred to previous investigations that utilized a contemporary engine platform designed for light-duty applications. The experimental database was constructed using a single-cylinder version of a turbocharged spark-ignition engine equipped with port fuel injection and a variable valve timing (VVT) system [11]. The engine fuel consumption was measured across various conditions, including different fuel compositions, air-based dilutions, ignition timings, and engine loads. Specifically, the study in [14] measured HCNG fuel blends containing 25%, 50%, and 75% hydrogen (by mass percentage), along with CNG and hydrogen fuels. The operating conditions, determined by engine speed and load, were set at 1500 rpm and loads ranging from 4 to 10 bar of IMEP.

Within the entire measured range, operating settings were optimized to minimize fuel consumption. In this context, air dilution and ignition timing were fine-tuned for each combination of fuel composition and operating conditions. This approach enabled the estimation of optimal fuel consumption for each HCNG fuel under varying operating conditions.

For CNG, only stoichiometric conditions (no dilution) were employed to operate a three-way catalyst (TWC) in the vehicle, a state-of-the-art technology for pollutant reduction and regulatory compliance. Blends containing hydrogen necessitated higher dilution ratios due to the unique combustion characteristics of hydrogen. This phenomenon has been previously noted by several researchers [36–38], who argued that thermal losses are the primary contributors to reduced efficiency and performance when comparing hydrogen to other fuels. To maintain high engine efficiency, significant dilution, whether with air or exhaust gas recirculation, is essential. Consequently, an alternative after-treatment technology is required instead of the traditional TWC. Here, an oxidizer catalyst should be employed to reduce unburned fuel, accompanied by a selective catalytic reduction (SCR) system to mitigate NO_x emissions, as illustrated in Fig. 6. Recent studies suggest that these catalysts can be supplied with H_2 [39,40], potentially eliminating the need for liquid urea refuelling and addressing ammonia slip issues.

A sample from the referenced database is presented in Figs. 7 and 8. These graphs illustrate the impact of hydrogen blending on CNG, revealing an enhancement in IMEP and GIE as hydrogen content increases in the fuel blend. This improvement primarily stems from the higher λ achieved due to the combustion properties of hydrogen, enabling ultra-lean, stable combustion (COV_{IMEP} > 3%). For pure hydrogen fuel, the optimal air dilution reaches $\lambda = 3$, resulting in a reduction in thermal losses. Similar optimal λ values are reached for 25%, 50%, and 75% H_2. However, efficiency gains are slightly reduced for 50% and 75% HCNG blends, and the performance is diminished compared to the 25% HCNG blend.

Increased air dilution also has a positive impact on pollutant emissions, diminishing levels of CO and NO_x emissions. Carbon monoxide undergoes oxidation and transformation into carbon dioxide, while lower combustion temperatures impede the formation of NO_x due

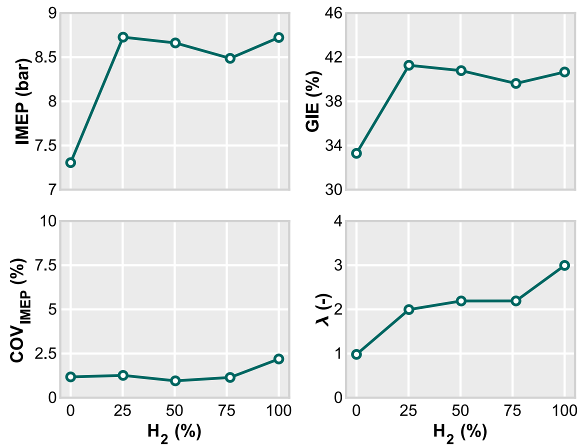

Fig. 7. Impact of hydrogen substitution percentage on engine performance.

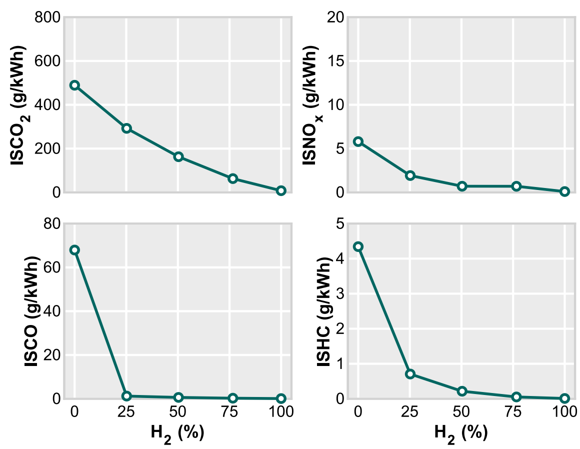

Fig. 8. Impact of hydrogen substitution percentage on exhaust gas emissions.

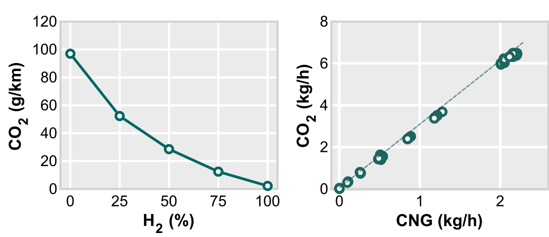

Fig. 9. Correlation obtained for CO_2 emissions estimation as a function of H_2 percentage in the fuel and consumed amount of CNG.

to thermal generation mechanisms. Specific carbon monoxide (ISCO) emission levels decrease from approximately 70 g/kWh with pure CNG to around 1 g/kWh at 25% HCNG, while NO_x emissions exhibit a reduction of approximately 35% under these conditions. This effect is evident in Fig. 8, where both CO and NO_x emission values decrease due to the higher optimal dilution.

Furthermore, the substitution of CNG with hydrogen results in a reduction in all carbon-based emissions (CO, CO_2, and HC). However, even in the case of pure hydrogen, the net CO_2 emission is not zero due to the combustion of lubricant, as highlighted in a previous study [41].

3.2. Vehicle fuel consumption and emissions

The vehicle utilized for the overall fuel consumption estimation was the FIAT Panda Natural Power, and the World Harmonized Light-duty Vehicle Test Procedure (WLTP) was employed to represent real driving conditions. This vehicle is equipped with an engine platform designed for CNG operation, sharing similar geometrical characteristics with those used in the preceding section to characterize the engine fuel consumption under steady bench conditions.

Table 5
WLTP Fuel consumption of the H_2 and CNG.

Design N°	1	2	3	4	5
% H_2	0	25	50	75	100
% CNG	100	75	50	25	0
H_2 (kg/100 km)	0	0.52	0.83	1.06	1.17
CNG (kg/100 km)	3.5	1.55	0.83	0.35	0

Table 6
Capacity of the H_2 and CNG tanks for a range of 400 km.

Design N°	1	2	3	4	5
% H_2	0	25	50	75	100
% CNG	100	75	50	25	0
H_2 capacity [kg]	0	2.07	3.31	4.42	4.67
CNG capacity [kg]	14	6.21	3.31	1.41	0

The estimation of vehicle fuel consumption took into account the engine database described in the previous section and the stoichiometric CNG operation reference point obtained from [42]. Consequently, the vehicle fuel consumption, considering both fuel components (CNG and H_2), under realistic conditions was scaled based on the difference obtained from this reference point to the conditions measured for each fuel blend composition. The summarized results are presented in Table 5.

Moreover, a correlation was established between the quantity of hydrogen present in the fuel and the amount of CO_2 emitted during the process. This data is depicted in Fig. 9, enabling a rapid estimation of CO_2 emissions as a function of the hydrogen content. Additionally, the relationship between the consumed CNG and the generated CO_2 was also established, revealing a clear linear trend.

4. Environmental impact of HCNG vehicles

This section is aimed at presenting the results related to the environmental impact of HCNG vehicles in each phase of the life cycle: cradle-to-gate (Section 4.1), well-to-tank (Section 4.2), tank-to-wheel (Section 4.3), well-to-wheel (Section 4.4) and cradle-to-gate (Section 4.5).

4.1. Cradle-to-gate emissions

The cradle-to-gate emissions are those associated with the manufacturing phase of the life cycle. In the case of the HCNG vehicles considered in this study, they are expected to operate with the same CNG-H_2 mixture during the whole operation. Therefore, the capacity of their CNG and H_2 tanks should be fixed for each mixture. Considering the fuel consumption results derived from the experimental data in Section 3 and a target range of 400 km the capacity of the tanks was estimated as in Table 6.

These capacities were used to calculate the mass of the tanks to estimate the emissions associated with their production. For the CNG tanks, their mass was correlated to their capacity following the database in [43] for CNG cylinders of type 2. For the H_2 the mass was calculated considering a gravimetric capacity of 0.055 kg H_2/kg system for type IV tanks that store gas at 700 bar based on the technical targets provided by the Department of Energy [44].

Fig. 10 shows the GHG emissions and how they would evolve with the expected energy mix decarbonization from 2020 to 2050 for the CNG-pure and the H_2-pure vehicles. The emissions associated with the production of 1 vehicle powered only with CNG (Fig. 10A) are lower in all the time scenarios than those associated with the production of a

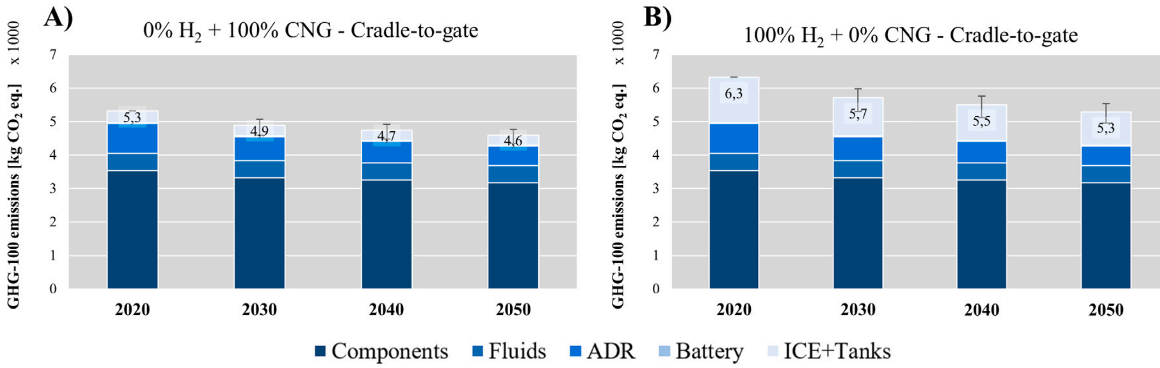

Fig. 10. Cradle-to-gate emissions of the target vehicle only with 0% H_2 and 100% CNG (A) and with 100% H_2 and 0% CNG (B). Evolution from 2020 to 2050.

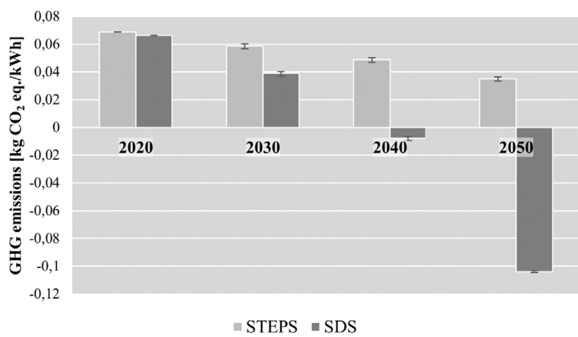

Fig. 11. Well-to-tank emissions of CNG production from 2020 to 2050 in the SDS and STEPS scenarios.

vehicle running purely on H_2 (Fig. 10B). This is due to the additional materials required by the H_2 tanks since they need to resist higher gas pressures. Between these two vehicle concepts, the cradle-to-gate emissions of the H_2-fuelled vehicle compared to the CNG vehicle are 18.9% higher in 2020 while they are only 15.2% higher in 2050, meaning that the production of H_2 tanks is more energy-consuming than the production of CNG tanks since they are more affected by the energy mix carbon intensity.

In the case of pure CNG vehicles, the emissions in the manufacturing phase range from 5.3 tonnes of CO_2 eq. in 2020 to 4.6 tonnes in 2050 while for H_2-pure vehicles this phase implies 6.3 tonnes in 2020 and 5.3 tonnes in 2050. As explained previously, the carbon intensity has a greater impact on the H_2-powered vehicles (1 tonne of CO_2 of difference against 0.7 tonnes for the CNG vehicle) since the manufacturing of the storage system requires more energy.

4.2. Well-to-tank emissions

The well-to-tank emissions are those related to the processes in the "fuel production cycle" box in Fig. 5. They involve all the processes required to produce either H_2 or CNG, treat them, and distribute them to the refuelling station. In this case, since the H_2 comes from a mix of production pathways it is necessary to consider all the production strategies independently and estimate the well-to-tank emissions as a weighted average.

In this study, CNG and H_2 are considered as different fuels for different reasons. The first reason is that NG is used as an input for SMR processes in the H_2 production. Therefore, if any modification should be added to the NG mix used in the vehicle, it should consistently be considered in any other process as well. The second reason is that it provides a higher degree of flexibility to generate mixes of each fuel according to the production pathways and mixes of both fuels combined (HCNG).

Regarding the CNG production, the well-to-tank GHG emissions per kWh of CNG are presented in Fig. 11 for the SDS and the STEPS scenarios. These two scenarios were defined in Fig. 3A and Fig. 3B, being the former the scenario representing a higher share of biogas in the natural gas mix. As a consequence, the well-to-tank emissions in the SDS scenario are always lower than in the STEPS scenario. From 2040, the biogas percentage in the natural gas mix reaches 19% and the well-to-tank emissions turn negative, reaching a value close to −0.1 kg CO_2/kWh in 2050. The reader should note that, despite the negative emissions in the well-to-tank process, consuming CNG during the operation phase implies releasing CO_2 emissions. Therefore, it does not mean that the well-to-wheel nor the cradle-to-grave emissions are negative. The small uncertainty in this figure comes from the energy mix. These uncertainties are low since CNG production requires relatively low electricity.

The well-to-tank GHG emissions of each H_2 production pathway and how they change with the time scenarios that include changes in the electric and NG mixes are found in Fig. 12. In this figure, the negative emissions when producing H_2 are only achieved when it is produced from biogas, either partially or totally. In this sense, the lowest emissions are found when the H_2 is produced from SMR fed with biogas since it is considered that a significant amount of biogas is consumed instead of released to the atmosphere. The SMR pathways are classified as grey (without carbon capture and storage or CCS) or blue (with CCS) and are evaluated in the STEPS and SDS scenarios. In the case of grey H_2, the well-to-tank emissions are higher than for most of the production pathways except in the 2050 SDS scenario, in which the amount of biogas in the NG mix reaches 43%. The reader should note that these scenarios are those proposed in the different roadmaps to improve the sustainability of the current and alternative fuels, but they may not be realistic unless significant investment is produced while the technology is widely integrated into all the processes where biogas can be retrieved.

Blue H_2, i.e., produced from SMR with CCS, implies less than half the amount of GHG emissions than SMR in the 2020 scenario and far more negative emissions than grey H_2 since around 90% of the CO_2 released in the reforming process is captured.

Green H_2 is divided into H_2 produced from central electrolysis (CE) or regional electrolysis (RE). In both pathways, since the axiom of green H_2 is that it is produced from renewable energy, it is assumed that there is any amount of energy where the H_2 is produced and all the on-site processes are powered with renewable energy are. Therefore, in the case of CE, the H_2 production, treatment and compression to be loaded into the tube trailers for distribution is powered with renewable energy while there are CO_2 emissions produced by the truck transporting the fuel and the recompression at the refuelling station is powered with the electricity mix. In contrast, in this study, RE implies the production of H_2 at the refuelling station. Hence, all the processes, including the total compression of the fuel can be covered with renewable energy

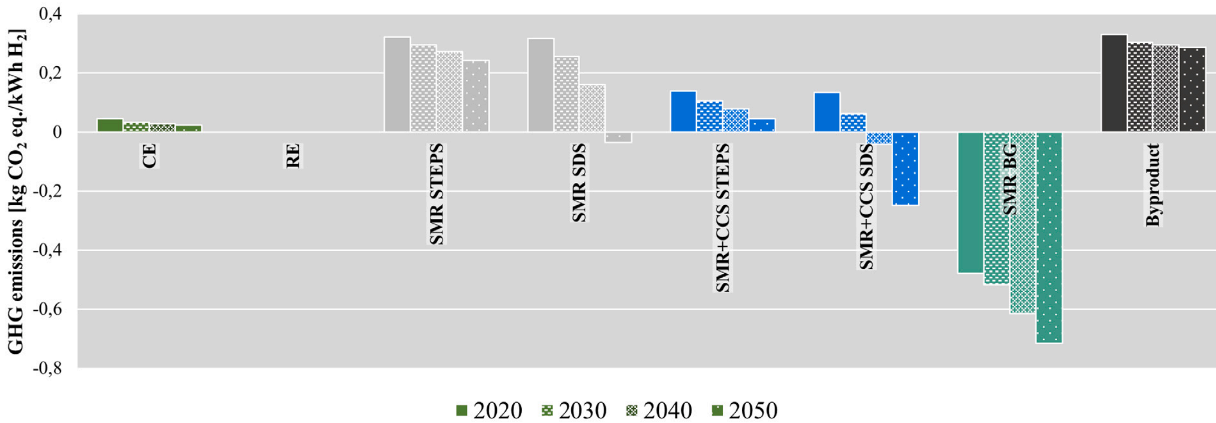

Fig. 12. Well-to-tank emissions the different H_2 production pathways proposed by the EC from 2020 to 2050.

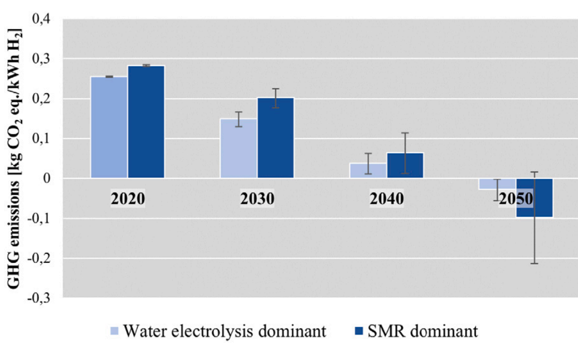

Fig. 13. Well-to-tank emissions of H_2 production from 2020 to 2050 in the electrolysis-dominant and SMR-dominant scenarios.

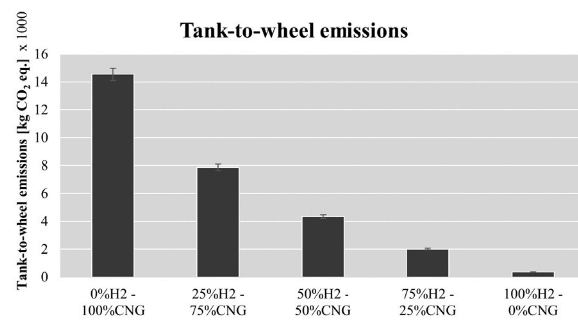

Fig. 14. Tank-to-wheel emissions for the different HCNG mixtures for a lifetime of 150000 km.

since there should be a renewable energy production facility nearby for it to be green. Hence, the environmental impact of H_2 produced from RE is nearly zero. Again, the reader must note that this is only due to the fact that the manufacturing of the equipment to produce the H_2 is left out of the boundaries as in Fig. 5, which is the usual approach for cradle-to-grave LCA for transport applications.

Among all the production pathways, that in which the H_2 is produced as a byproduct implies the highest emissions since its production implies the release of emissions from the same source it is produced. Such is the case when it is produced from coke oven gas, captured and then purified.

The information in Fig. 12 combined with the evolution of the H_2 mixes in the water electrolysis and SMR dominant scenarios (Fig. 4) is then used to estimate the well-to-tank emissions per kWh of H_2 produced in Fig. 13. As it can be noted, here the STEPS and SDS scenarios for the NG mix are condensed into average values and error bars to identify the variability in the actual well-to-tank emissions for H_2 production. The results in Fig. 13 show how in the early stages of the transition towards the Hydrogen Economy (2020–2040), the water electrolysis dominant scenario implies on average lower emissions than the SMR-dominant since the use of green H_2 implies lower emissions than considering a higher share of H_2 coming from SMR with or without CCS. Nonetheless, in the 2040 scenario, the combination of the SMR pathway together with the SDS scenario, implying a high content of biogas in the NG mix, makes that the SMR-dominant scenario could offer lower emissions than the water electrolysis dominant scenario, provided that they are not negative yet. It is not until 2050 that the well-to-tank emissions coming from the production of H_2 turn negative for both scenarios, being them lower in the SMR-dominant one due to the high use of the SMR production pathway together with the high share of biogas in the NG mix. Nevertheless, the error bars indicate that depending on the evolution of the electricity mix and the biogas share, the well-to-tank emission of both scenarios could be either similar and lower than zero or the SMR-dominant could provide significantly lower emissions. The negative results in emissions are only possible due to the use of biogas to produce H_2. Therefore, even though green H_2 could imply close-to-zero emissions, the use of biogas and SMR technology must be considered to achieve negative emissions in the well-to-wheel phase and zero emissions in the cradle-to-grave cycle. However, the use of biogas and NG depends on the natural resources availability and on the amount of waste and processes that can be used to retrieve biogas. Despite the increased attention towards green H_2 and the urgent need to enhance its application to reduce the emissions emanating from its production, the energy production sector must shift its focus towards the production and utilization of biogas, provided that there are abundant natural resources available for such production. This shift is critical in pursuit of the ultimate goal of achieving zero-emission vehicles.

Finally, if Figs. 11 and 13 are compared, it is possible to identify how in 2020–2040, the well-to-tank emissions per kWh for H_2 are higher than for CNG, and this trend changes from 2040, depending on the NG mix scenario. This will be critical to compute the well-to-wheel emissions, since it may influence the optimal HCNG mixture as a trade-off between the well-to-tank and the tank-to-wheel emissions.

4.3. Tank-to-wheel emissions

The tank-to-wheel emissions are mainly derived from the experimental results by applying the process described in Section 3. These GHG emissions are presented in Fig. 14 for different HCNG mixtures considering a lifetime of 150000 km. As it is expected, there is a direct

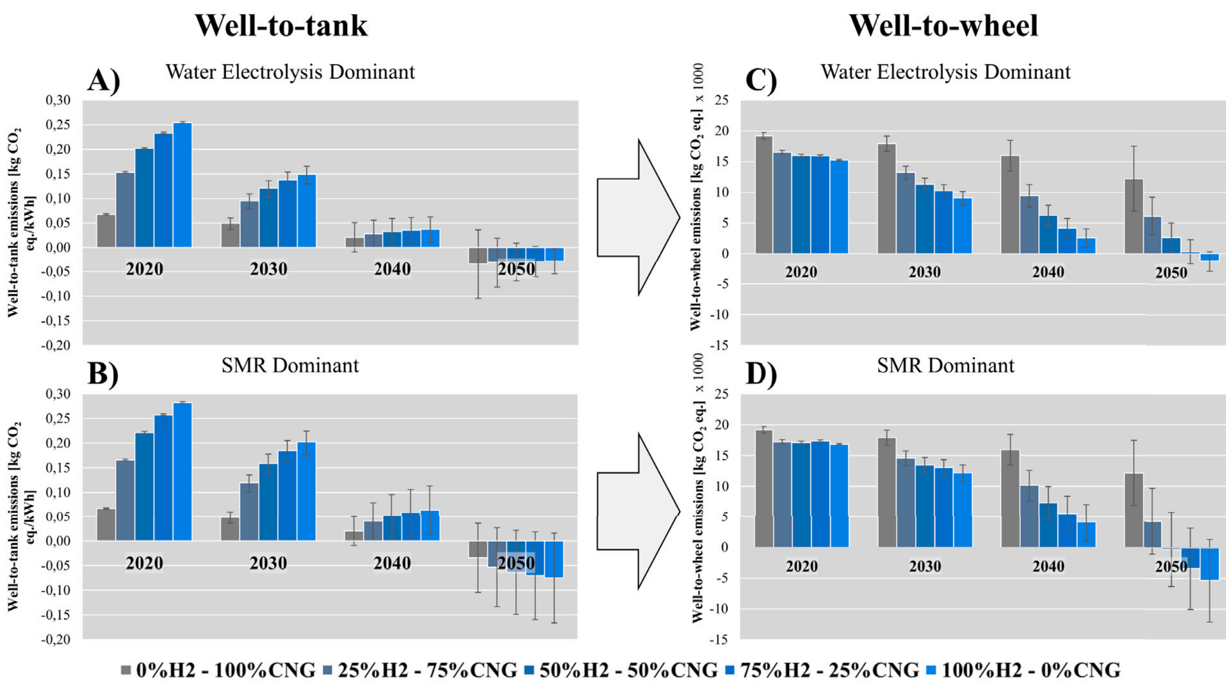

Fig. 15. Well-to-wheel and well-to-tank emissions of HCNG mixtures from 2020 to 2050 according to both the electrolysis-dominant and the SMR-dominant scenarios. The functional unit for the well-to-tank phase (graphs A-B) is 1 kWh of fuel referenced to their lower heating value while for the well-to-wheel results (graphs C-S) the functional unit is 150000 km of lifetime.

correlation between the CO_2 emissions and the carbon content of the fuel. In this sense, the highest change in the tailpipe GHG emissions occurs when comparing pure CNG with the mixture containing 75% of CNG and 25% of H_2 for two reasons. On one hand, although the percentages are in mass terms, the criteria for the mixtures in the experimental campaign was to introduce the same amount of energy at each operating condition so that all the results are comparable and, in case of obtaining higher useful energy, isolate the increase in the efficiency in the combustion process from the addition of more energy in the fuel. Following this methodology with a fuel such as H_2 with a high energy content implies that the amount of CNG introduced in the mixture actually decreases more than 25% compared to the pure CNG case, thus implying a much significant decrease in the fuel carbon content. On the other hand, there is a significant increase in the indicated efficiency motivated by the addition of H_2, which decreases the fuel consumption for a given driving cycle.

The emissions for the case of 100% H_2 are not zero since there is some CO_2 content produced from burning the lubricant that is required in the combustion engine. Therefore, although these emissions may seem negligible, it would not be correct to address the H_2 ICE as a carbon-free tank-to-wheel emission technology.

This graph, together with Figs. 11 and 13 indicates that there would be a trade-off in the well-to-wheel emissions due to how the HCNG mixture affects both the tank-to-wheel and the well-to-tank cycles.

4.4. Well-to-wheel emissions

The results in terms of the well-to-tank and well-to-wheel emissions for different HCNG mixtures and vehicles are presented in Fig. 15. Both phases are presented in the water electrolysis dominant scenario (graphs A and C) and for the SMR-dominant scenario (graphs B and D).

The emissions associated with the production of the HCNG mixtures show a unique trend for both scenarios from 2020 to 2040. In this timeframe, the well-to-tank emissions are lower for high CNG content mixtures since the share of H_2 produced from biogas or renewable energy is not enough and the additional energy consumption required to produce H_2 outweighs the progress towards a more sustainable H_2 production mix. Nonetheless, in 2050 the fuel production emissions in the electrolysis-dominant scenario are similar for all the HCNG mixtures while the trends in the SMR-dominant scenario indicate that the H_2 production could imply even lower emissions than the CNG production. This can only be analysed in terms of trends since the uncertainty in the SMR-dominant scenario in 2050 is so high that identifying the HCNG mixture in terms of well-to-tank emissions would be incorrect.

It is observed that the overall uncertainty in the well-to-tank phase of an HCNG mixture increases with time irrespective of the scenario. However, the increase in this uncertainty is significantly different in the two scenarios (electrolysis-dominant and SMR-dominant). In the electrolysis-dominant scenario, the uncertainty in the HCNG mixture decreases in 2050 with an increase in the H_2 content of the mixture. This is because H_2 relies mainly on the electrolysis technology, and the uncertainty produced by the biogas share in the NG mix is minimized. On the other hand, in the SMR-dominant scenario, increasing the H_2 content of the HCNG mixture increases the uncertainty since the H_2 production relies significantly on the SMR process from NG. Therefore, it can be deduced that in terms of reducing emissions in the well-to-tank process, the SMR-dominant scenario is superior, provided that there is enough conventional NG and biogas to support this scenario. In contrast, the electrolysis-dominant scenario also has the potential of offering negative emissions with much lower uncertainties, provided that there is enough renewable energy to support the H_2 production. It should be noted that this study does not aim to evaluate the feasibility of these scenarios as it depends on natural resources availability, investments to advance towards the Hydrogen Economy, and politics.

The well-to-wheel GHG emissions (graphs C and D of Fig. 15) are obtained as the combination of the results presented in Fig. 15 (graphs A and B) with those in Fig. 14. Here, it is possible to identify how in 2020 there is not a significant difference between the HCNG mixtures in terms of well-to-wheel emissions except for the pure-CNG case, where the emissions are 11.9–20.6% higher in average, since the low well-to-tank emissions are compensated by the high GHG tank-to-wheel emissions. In this case, it seems that for the short term even a moderate amount of H_2 added to the CNG (25%, for instance) could provide great benefits in terms of well-to-wheel emissions, but adding more H_2 does not significantly benefit the sustainability of HCNG vehicles.

In 2030, the impact of adding a higher fraction of H_2 to the HCNG mixture has a noticeable impact on the well-to-wheel GHG emissions. In this sense, the emissions of the pure-H_2 technology are 49.2% lower than a pure-CNG vehicle in the electrolysis-dominant scenario and 31.8% in the SMR-dominant scenario. This difference is lower in the latter case since the biogas fraction is still small and thus producing H_2 from SMR does not decrease significantly the environmental impact. Still, the difference in emissions between the HCNG mixtures and the uncertainties are moderate.

In the following decade (2040), the electrolysis-dominant scenario still offers lower emissions than the SMR-dominant one, but no HCNG mixture or even the pure-H_2 solution offers zero well-to-wheel emissions within the possible scenarios, as can be seen from the error bars. In both scenarios, the difference between any HCNG mixture and the pure-H_2 vehicle is significant, thus indicating that a complete transition towards H_2-fuelled vehicles could bring noticeable environmental benefits. In this case, the transition from a pure-CNG technology towards the pure-H_2 vehicles could reduce the well-to-wheel emissions by 83.9% in the electrolysis-dominant scenario and by 74.2% in the SMR-dominant scenario.

It is not until 2050 that the trend between these two scenarios is reversed, namely due to the significant increase in the biogas share in the NG mix following the SDS scenario in 2050 (43% as in Fig. 3A). This is most likely produced since the 2050 timeframe is perceived as a long-term goal, so the targets in terms of sustainability are ambitious and may be unrealistic. Nonetheless, the reader should note that analysing the feasibility of such targets is not within the scope of this study and is left for future work.

Following the scenarios proposed by the EU, zero well-to-wheel emissions are only reachable after 2040 and close to 2050, but this will ultimately depend on how the production pathways evolve and, in the case of HCNG vehicles, on the availability of both biogas and H_2 to transition to the pure-H_2 solution. In this case, following the possible scenarios considered in this study, no pure-CNG vehicle could reach zero well-to-wheel GHG emissions by 2050. Nevertheless, as explained previously, the further it is predicted in time, the higher the uncertainty. The error considered in this study when assessing the possible scenarios in terms of electricity, NG and H_2 mixes brings to light how these emissions may change since at some points the uncertainty is several times higher than the average values. Nevertheless, the lowest values are representative of the potential of these strategies and scenarios if combined correctly. In this sense, it is important to note how until 2040, the data showed how it was more environmentally beneficial to put efforts into increasing the renewable energy share in the electricity mix to produce green H_2 and transition to pure-H_2 vehicles rather than increasing the amount of biogas in the NG mix. Nonetheless, in 2050, the lower emissions presented by the SMR-dominant scenario indicate how in the long term increasing the biogas share in the NG mix is critical to achieving negative emissions in the well-to-wheel phase and potentially zero emissions in the cradle-to-grave cycle.

It is important to note at this point that the negative emissions in Fig. 15 may be misleading. One may think that this is environmentally beneficial with these results, but this is only under the assumption of producing enough energy and biogas, which may not be the case in the long term for large HCNG vehicle fleets. Furthermore, the energy efficiency of using biogas to produce H_2 and then consuming it to power a vehicle may not be the ideal solution to decarbonize the economy, but it indeed is to decarbonize the transportation sector.

Finally, in order to obtain meaningful conclusions about the environmental impact of HCNG vehicles, it is imperative to include the vehicle manufacturing process (cradle-to-gate), thus offering a comprehensive cradle-to-grave process as in Section 4.5.

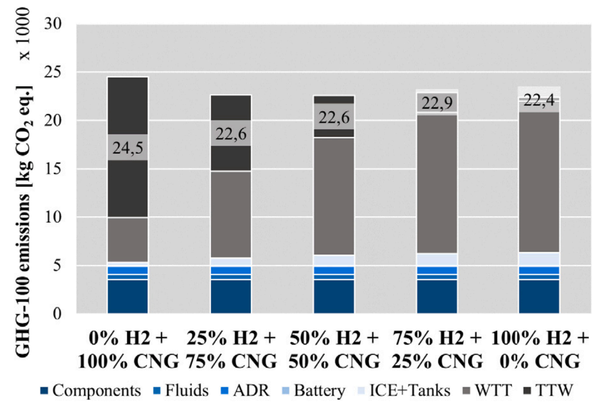

Fig. 16. Cradle-to-grave GHG emissions breakdown of HCNG vehicles in 2020.

4.5. Environmental impact and optimal HCNG mixtures

This section comprises the environmental analysis in the cradle-to-grave cycle, thus including both the well-to-wheel emissions (Section 4.4) and the cradle-to-gate or vehicle manufacturing emissions (Section 4.1).

4.5.1. Current scenario

The cradle-to-grave emissions of HCNG light-duty vehicles in 2020 are presented in Fig. 16. The results in this figure are averaged and do not include the error bars for simplicity and since they are included in Fig. 17.

The results in this figure show the trade-off between the decrease in well-to-wheel GHG emissions presented in Fig. 15 and the increase in cradle-to-gate emissions due to the bigger H_2 tanks (Fig. 10A and Fig. 10B) required when increasing the H_2 content of the fuel. For these vehicles, the well-to-wheel phase dominates the overall cradle-to-grave emissions since they are responsible for most of the environmental impact and follow a similar trend, i.e., they decrease the higher the H_2 fuel content. Nonetheless, a local minimum is found around an HCNG mix of 25%–50% of CNG and 50%–75% of H_2 due to the non-linearities of the fuel consumption and tailpipe emissions produced from the experimental results (Fig. 9) coupled with the comparatively low emissions of manufacturing a CNG tank against those of manufacturing a H_2 tank. Nonetheless, the cradle-to-grave emissions are nearly constant and lower than the pure-CNG case if H_2 is added to the fuel mixture due to the increase in the combustion efficiency and the decrease in the carbon content of the fuel. Therefore, with the H_2 mix of the electrolysis-dominant scenario in 2020 it is recommended to operate with HCNG mixtures with low-H_2 content since the H_2 availability is low and there are no significant environmental benefits from increasing the H_2 fraction.

In the current scenario, the vehicle manufacturing phase produces 21.7% of the total GHG emissions in the pure-CNG vehicle while this fraction increases to 28.3% for the pure-H_2 technology. This change is mainly due to the decrease in the well-to-wheel emissions and the increase in the vehicle manufacturing environmental impact when increasing the H_2 fuel fraction. As can be seen, the cradle-to-gate phase constitutes a significant fraction of the overall life cycle and, although it may not be the major impact, it needs to be indeed considered to achieve the zero-emission vehicle. This concept is only achievable if the vehicle manufacturing emissions are compensated with negative emissions in the well-to-wheel phase.

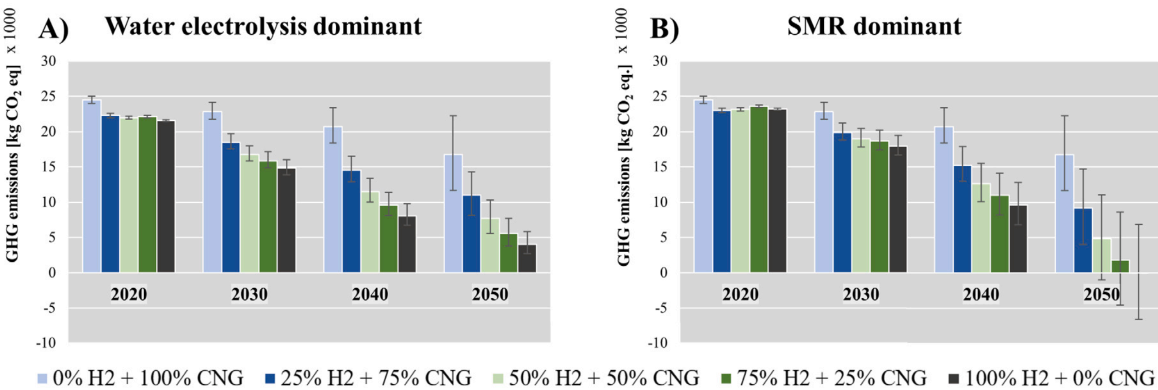

Fig. 17. Evolution of the cradle-to-grave GHG emissions in the electrolysis-dominant (A) and SMR-dominant (B) scenarios from 2020 to 2050.

4.5.2. Future scenarios

The evolution of the cradle-to-grave emissions in Fig. 16 is projected to 2030, 2040 and 2050 in both the electrolysis-dominant (Fig. 17A) and the SMR-dominant (Fig. 17B) scenarios. The information contained within these results describes the potential of each scenario for different HCNG mixtures, thus allowing the identification of the optimal mixture according to the scenario and timeframe, thus providing valuable information to support the evolution of HCNG vehicles that minimizes their environmental impact.

In the electrolysis-dominant (Fig. 17A) and the SMR-dominant (Fig. 17B) scenarios, the short-term results (2020) indicate how adding any fraction of H_2, among those considered in this study, implies a noticeable change with respect to the cradle-to-grave emissions produced with the pure-CNG vehicle but they remain similar despite increasing the H_2. Therefore, the 2020 timeframe could be defined as the starting point for the transition towards pure-H_2 vehicles by adding a small amount of H_2 with a positive impact on the environment impact. This is additionally in line with the low H_2 production in the early stages of the transition. For this set of reasons, the recommended H_2 fraction in the mixture is 25% for 2020.

The 2030 timeframe is similar for both scenarios but the differences between HCNG mixtures are more noticeable in the electrolysis-dominant scenario since the biogas fraction in the NG mix is still small. In this case, the transition towards the pure-H_2 vehicles implies a decrease in GHG cradle-to-grave emissions of 35% for the electrolysis-dominant scenario and of 21.4% in the SMR-dominant scenario. Nonetheless, in any case, the difference in cradle-to-grave emissions falls below 6% when increasing the H_2 fraction from 50% to 75% when the electrolysis production pathway dominates and below 2% when the SMR process does, which can be considered small. Therefore, this timeframe could be suitable for progressively increasing the H_2 share to 50% in HCNG mixtures.

The 2040–2050 timeframes indicate that the transition towards the 75% H_2 mixture or pure-H_2 could bring significant benefits in terms of environmental impact. In the electrolysis-dominant scenario, the uncertainty increases the further away it is looked in time but it decreases the higher the H_2 content in the mixture since the dominance of green H_2 implies that the uncertainty is mainly due to the possible scenarios in the electricity mix, rather than in the NG mix. In this sense, despite not reaching negative emissions the decrease in emissions has a lower uncertainty. For this reason, the electrolysis-dominant scenario ensures lower emissions the higher the H_2 content in the HCNG mixture but it is not suitable for reaching the zero-emission vehicle concept since for that a higher amount of negative emissions coming from the use of biogas is required.

In contrast, the SMR-dominant scenario shows the potential of reaching and even surpassing the zero-emission concept due to the high share of biogas in the NG mix. In this sense, only the HCNG mixture with more than 50% of H_2 could approach this concept, but this will depend mainly on the investment and politics that foster the capture and usage of biogas. In the 2040–2050 decade, it is recommended to transition towards the pure-H_2 technology to minimize the environmental impact of these vehicles since, despite the increasing uncertainty, the trend indicates that this technology should offer lower emissions.

In conclusion, the strategy to minimize the environmental impact of HCNG vehicles without putting too much stress on the H_2 indicated that the transition towards a H_2 content of 25% should be carried out in the 2020–2030 decade. From 2030, the H_2 mixture should increase to 50% and in 2040–2050 the transition should aim towards pure-H_2 vehicles. The zero-emission concept is only feasible if enough biogas is introduced in the NG mix combined with the use of H_2. Therefore, in the early stages of the transition towards the H_2, the electrolysis-dominant is encouraged since it offers lower cradle-to-grave GHG emission through the use of green H_2 but in the long term, once there is enough renewable energy to produce H_2 the focus must change towards increasing the biogas share in the NG mix, thus approaching the SMR-dominant scenario in 2050. Following this hybrid strategy would not only allow minimizing the environmental impact of HCNG vehicles in terms of global warming but to reach the zero-emission concept in the long term-

5. Conclusion

This study examined the impact of transitioning to a fully developed H_2 economy on GHG emissions and considered multiple hydrogen blends on CNG in different scenarios. The most significant novelty in this study are the identification of the optimal blend of HCNG that minimizes emissions in the 2020–2050 period and the determination of the timeframe and HCNG blend that enables zero cradle-to-grave emissions for these vehicles. The findings suggest that hydrogen blending has the potential to reduce emissions and provide an alternative fuel source for cleaner and more efficient transportation systems. Increasing hydrogen content in blends improves engine performance but requires higher air dilution ratios to maintain efficiency and minimize thermal losses. As the H_2 content in blends increases, exhaust gas emissions reduce due to higher air-fuel equivalence ratios and lower combustion temperatures. Using hydrogen instead of CNG lowered carbon emissions, but some carbon dioxide is still generated from lubricant oxidation.

Based on the results of this study, it is demonstrated that HCNG vehicles possess a significant potential to decarbonize the transportation sector in both water electrolysis and SMR-dominant scenarios. In the water electrolysis scenario, the cradle-to-grave emissions have been observed to decrease significantly from [24.5, 21.6] to [16.8, 4.1] tonnes of CO_2 eq. by the year 2050, with pure-H_2 vehicles experiencing the lowest value and CNG vehicles experiencing the highest. In the SMR scenario, there is a notable increase in the uncertainty of cradle-to-grave emissions, but it offers the possibility of reducing emissions to

−0.1 tonnes of CO_2 eq., depending on the biogas share in the NG mix and the electricity mix.

The optimal HCNG blend evolution implies incorporating 25% H_2 content in 2020, 50% in 2030, and 75%–100% between 2040 and 2050 to reduce greenhouse gas emissions in the transportation sector. While these suggested H_2 shares may not yield the lowest emissions in 2030 and 2040, they are chosen pragmatically due to potential feasibility constraints in H_2 availability. Achieving zero cradle-to-grave emissions in 2050 necessitates an SMR-dominant scenario with fuel mixtures exceeding 50% H_2 content, requiring increased use of biogas to promote negative emissions and offset environmental impact throughout the vehicle life cycle. The SMR-dominant scenario has the potential to either yield significant negative emissions or decrease environmental impact by 72% compared to pure-CNG vehicles in the least-optimistic scenario.

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CRediT authorship contribution statement

S. Molina: Conceptualization, Funding acquisition, Supervision, Visualization, Project administration. **J. Gomez-Soriano:** Conceptualization, Formal analysis, Methodology, Software, Supervision, Writing – review & editing. **M. Lopez-Juarez:** Conceptualization, Formal analysis, Investigation, Methodology, Resources, Software, Writing – original draft, Writing – review & editing, Supervision. **M. Olcina-Girona:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Glossary

AF_{st} Stoichiometric Air-fuel Ratio

BEVs Battery Electric Vehicles

BTE Brake Thermal Efficiency

CAD Crank Angle Degrees

CCS Carbon Capture and Storage

CCV Cycle-to-Cycle Variation

CE Central Electrolysis

CFD Computational Fluid Dynamics

CH_4 Methane

CLD Cadmium Luminescence Detector

CNG Compressed Natural Gas

CO_2 Carbon Dioxide

COV_{IMEP} IMEP covariance

EGR Exhaust Gas Recirculation

ETS Emissions Trading Scheme

EVs Electric Vehicles

EVC Exhaust Valve Closing

EVO Exhaust Valve Opening

FID Flame Ionization Detector

GIE Gross Indicated Efficiency

GWP Global Warming Potential

GHG Greenhouse Gases

H/C Hydrogen–Carbon ratio

H_2 Hydrogen

HC Hydrocarbons

HCNG Hydrogen-CNG fuel blends

ICE Internal Combustion Engine

IMEP Indicated Mean Effective Pressure

ISCO Indicated specific CO

$ISCO_2$ Indicated specific CO_2

ISHC Indicated specific HC

$ISNO_x$ Indicated specific NO_x

IVO Intake Valve Opening

IVC Intake Valve Closing

LCA Life Cycle Assessment

LHV Lower Heating Value

MBT Maximum Brake Torque

NDIR Non-Dispersive Infrared Spectroscopy

NI Scenario where there are no improvements in the sustainability policies

NO_x Nitrogen Oxides

O/C Oxygen–Carbon ratio

O_2 Oxygen

PFI Port Fuel Injection

PM Particulate Matter

PMA Magneto-Pneumatic Analysis

RE regional electrolysis

RON Research Octane Number

SCR Selective Catalytic Reduction

SMR Steam Methane Reforming

SI Scenario where there are significant improvements in the sustainability policies

SDS Sustainable Development Scenario [34]

ST Spark Timing

STEPS Stated Policies Scenario [34]

TWC Three-Way Catalyst

WLTP World Harmonized Light-duty Vehicle Test Procedure

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