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Highlights

Comparative global warming impact and NO_X emissions of conventional and hydrogen automotive propulsion systems

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- Well-to-wheel GHG and NO_X emissions for H_2 and conventional vehicles were estimated

- The only impact category was Global Warming, although NO_X were also estimated

- EU 2017 & 2050 energy mixes and water greenhouse effect were considered

- Target HICEVs fuel consumption is around 30 kWh/100 km to compete with BEVs

- The most efficient strategy to reduce the transport emissions in EU was devised

Comparative global warming impact and NO_X emissions of conventional and hydrogen automotive propulsion systems

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Abstract

With the rise of cleaner technologies for transport and the emergence of H_2 as a fuel, most of the emissions in the well-to-wheel process are shifting towards the energy carrier production (fuel or electricity). The objective of this study is to perform a simplified cradle-to-grave Life Cycle Assessment (LCA) that compares the greenhouse gases (GHG) and NO_X emissions of H_2, electric and conventional technologies for the automotive sector in Europe and to devise the optimum strategy of vehicle fleet renewal to reduce the emissions. In this study the effect of water as GHG was considered and, unless other studies, the current European energy mix and that meeting the objectives for 2050 were considered (while technology level was kept constant) since H_2 from electrolysis and electric vehicles' well-to-wheel emissions are sensitive to the energy mix. To estimate the emissions, the fuel, vehicle production and operation cycles were considered independently for each technology and then put together. For H_2, the best production and distribution strategy was steam methane reforming (SMR) with CO_2 sequestration for GHG-100 gases and without capturing CO_2 for NO_X, both with central plant production and tube trailer transport. Fuel cell vehicles (FCV) with optimum H_2 production always produce the lowest GHG-100 emissions and slightly higher NO_X than battery electric vehicles (BEV) in the EU 2050 scenario. In contrast, HICEV would need to reach a fuel consumption of around 30 kWh/100 km to be competitive in emissions against BEV, for that, direct injection (DI) combined with a range extender (REx) hybrid architecture is the recommended powerplant concept. Finally, the optimum strategy to reduce emissions that Europe could follow is presented for the short, mid and long term.

Keywords: LCA, Hydrogen, Fuel cell, HICE, Hybrid vehicles, Electric vehicles

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1. Introduction

Nowadays, there is a major concern about pollution and global warming. Many experts and international organizations claim that it is necessary to decrease greenhouse gases (GHG) in all energy sectors [1, 2]. However, CO_2 emissions worldwide are expected to keep growing with population [1, 3]. In Europe, 19.4% of GHG come from road transport (792 million tonnes of CO_2 equivalent) [4]. Another focus of major concern is NO_X emissions, whose effect over human health and ozone formation/depletion is not negligible [5].
To solve this problem, Europe is increasing the share of renewable sources in the energy mix and moving towards the hydrogen economy [2, 6]. These two actions must be coupled to produce green hydrogen by using energy from renewable sources and lower GHG emissions in the whole life cycle of hydrogen technologies. Regarding the transport sector, vehicles powered by fuel cells (FC) or hydrogen internal combustion engines (HICE) are viable options to shift towards carbon-free transport [2, 7]. In recent years, the attention of the companies has been focused on FC because of their higher break efficiency compared to HICE. However, HICE are still a good option due to their low manufacturing cost and emissions, so it must not be forgotten.

Life Cycle Assessment (LCA) is a relevant tool to analyze the environmental impact of a given technology considering all aspects along its life. Previous studies show that H_2 PEM fuel cells in Canada and the US could produce less CO_2 emissions if the energy mix is not based primarily on coal combustion [8]. This con-

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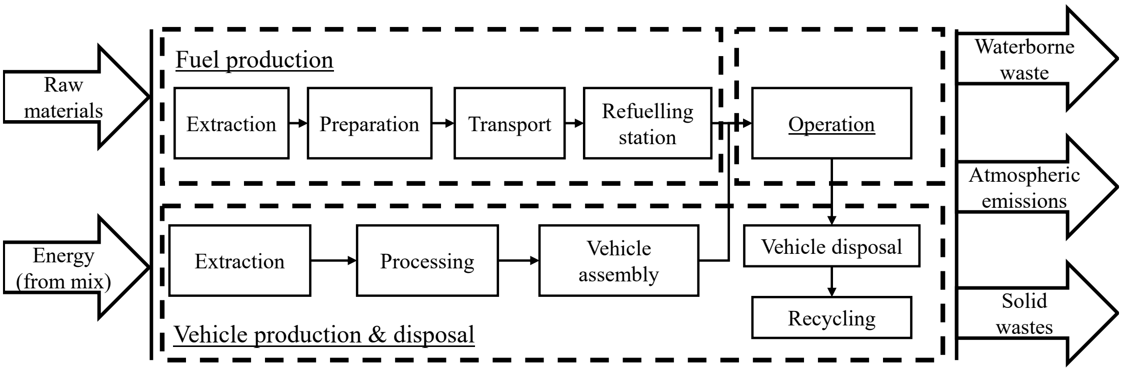

Figure 1: Cradle-to-grave cycle assessment methodology and boundaries

firms how H_2 cradle-to-grave emissions depend greatly upon the energy source that is used to produce it. A similar study [9], this time including HICE, demonstrated that H_2 technologies can produce lower emissions than Spark-Ignition (SI) or Compression-Ignition (CI) ICE fueled with gasoline, Diesel and even methanol if only renewable energy is used for the H_2 production. In none of these studies, the energy mix is representative of Europe's current or future situation, the emissions produced by the H_2 tank manufacturing were included nor the technology is representative of the current state-of-the-art. Other authors have focused their efforts on the analysis of LCA based on modern H_2 technologies and options, but they only analyzed a specific part of the life cycle such as hydrogen production [10, 11] and distribution [12], PEMFC manufacturing and recycling [13] and on-board storage [14]. A study similar to the present work was performed by Garcia et al. [15] considering the Spanish electricity mix in Madrid but it was oriented towards public transport and not towards light-duty passenger vehicles. In all the mentioned studies the effect of emitted-on-surface water vapor was not accounted for. Recently, Sherwood et al. [16] estimated the effective global warming potential on a 100-year horizon (GWP-100) of water ranging from -10-3 to 5·10-4 kg eq. CO_2. These values are low since additional emitted-on-surface water vapor (coming from H_2-fuelled vehicles) cannot reach the troposphere and therefore, the global warming effect of water vapor is compensated by the increase in the reflectance from low-altitude clouds formed with the additional water vapor (cooling effect). With the aim of extending the analysis provided by the already available scientific literature and evaluate the EU objectives of increasing the renewable energy share in the electricity mix, this study intends to be a cradle-to-grave cycle assessment that considers state-of-the-art automotive technologies, including SI and CI ICE fueled with gasoline/Diesel/compressed natural gas (CNG), hybrid systems equipping a SI ICE fueled with gasoline (HEV), battery electric systems (BEV), HICEV, and proton exchange membrane FCVs. This study focuses on passenger cars since all this powerplant portfolio potentially fulfills the requirements of this particular application, and it has also the highest impact on NO_X and CO_2 emissions considering the road transport sector.

The contributions of this paper to the literature are based on estimating the GHG-100 and NO_X emissions for most of the current automotive and hydrogen technologies for passenger cars considering the EU 2017 and 2050 electricity mixes and the water GHG-100 effect. With this estimation the objectives in the following section were accomplished.

2. Objectives

Considering the discussion about the state-of-the-art included in the previous section, the study was divided into a main and general objective and other specific objectives derived from it:

- Estimate and compare the GHG-100 and NO_X produced by H_2 propulsion technologies against those produced by conventional, hybrid and electric powerplants in the whole life cycle with the current and 2050 energy mix EU scenarios.
    - Understand what are the H_2 production and transport strategies that produce lower emissions with European Union (EU) 2017 and 2050 energy mixes.
    - Assess whether the EU objectives to increase the renewable energy share in the energy mix are enough to produce H_2 uniquely from

electrolysis to power the whole vehicle fleet and lower the emissions.

- Estimate the consumption that should be reached by HICEVs in order to be competitive against BEVs and find out which technology could potentially help to achieve it, if any.
- Assess the weight of the water vapour effect as a GHG-100 emission in the operation cycle.
- Establish the most efficient (emissions-wise) strategy to reduce the emissions and reach the H_2 economy in the transport sector.

3. Methodology

Cradle-to-grave cycle assessments for a given transport technology should include fuel production, vehicle production, vehicle disposal and operation cycles. The powerplant technologies and their corresponding fuels considered in this study are included in Table 1.

System boundaries

The system boundaries for each individual cycles are showed, together with the system inputs and outputs, in figure 1. They are those corresponding to a cradle-to-grave LCA, i.e., from the extraction of the raw materials using energy and fuel to the disposal and the recycling of the vehicle. Even though waterborne, solid wastes and other atmospheric emissions such as SO_X were calculated using GREET®, they were not included in the present study.

Functional units

The functional unit was changed for each cycle to improve the understanding of the analysis. In the fuel production cycle (figure 3, 4 and 5), the functional unit was the MJ of fuel since several fuels with different lower heating values and densities were compared. In the vehicle production cycle, the emissions were calculated per manufactured vehicle. Finally, in the cradle-to-grave cycle, including the previous cycles together with the vehicle operation, the functional unit was the life of the each vehicle considering 150000 km as the average common life.

Table 1: Vehicle technologies and fuels considered in the present study.

Engine	Energy source	Fuel production
BEV	Electricity	Electricity mix
FCV	GH_2, LH_2	Electrolysis Steam methane reforming (SMR) SMR with CO_2 sequestration
DI ICE	B10 Diesel	Biodiesel from soybeans + Low sulphur Diesel
PFI ICE	GH_2, LH_2	Electrolysis Steam methane reforming (SMR) SMR with CO_2 sequestration
	CNG	Conventional CNG
	E10 Gasoline	Ethanol + conventional gasoline
	E10 Gasoline (HEV)	Ethanol + Conventional gasoline

Impact category

In this LCA study, Global Warming was the only impact category considered, although NO_X were also estimated, since they are most concerning emissions in recent years. The GHG were calculated by taking into account CO_2, CH_4 and N_2O gaseous emissions. Their GWPs are 1, 28 and 265 kg_{CO_2} equivalent respectively [17].

Life cycle inventory

In this study, all the data, unless otherwise specified, were obtained from the GREET® model version 2019 from the Argonne National Laboratory.

The life cycle inventory is explained in detail for each cycle in sections 3.1., 3.2., and 3.3.

3.1. Fuel production cycle

In the fuel production cycle, also called well-to-pump, all the processes used to generate the fuel were taken into account. This includes from the extraction of the raw materials (oil or gas) or from the generation of raw fuels (H_2 or electricity) to the distribution to the refueling stations after their conditioning to be used (refinement or compression) as described in figure 1. Particularly, alternative fuels differ from conventional ones in the production method. Their main advantage is that they can be generated from renewable energy such

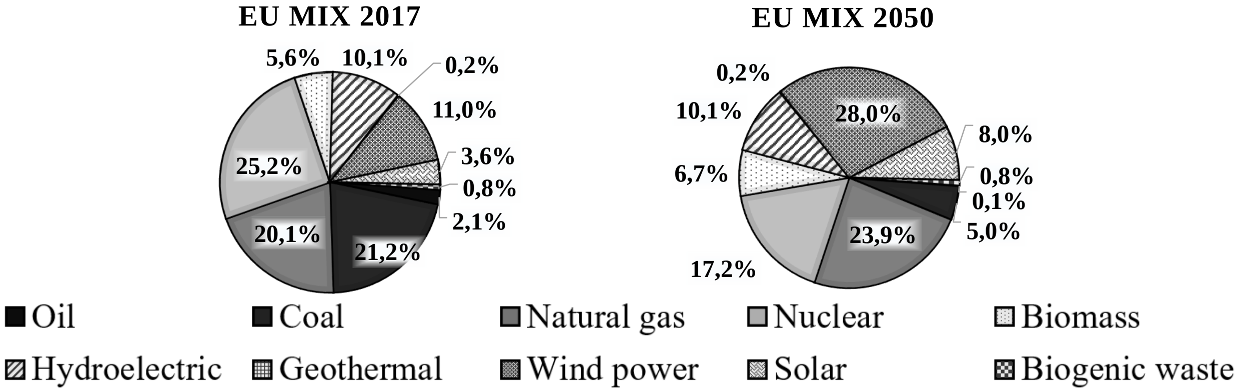

Figure 2: 2017 [18] and 2050 EU energy mixes [6].

as the so-called green hydrogen, so they are virtually unlimited. However, it is not realistic to assume that hydrogen will only be produced from renewable energy and, in the case it was accomplished, the overall effect of increasing the renewable energy share to produce hydrogen would most likely coincide with increasing it to be used in the electric grid. Therefore, if alternative fuels are produced from non-renewable energy, the emissions during the whole life cycle might be even larger than those of fossil fuels. In order to quantify this issue in the current EU situation and to assess the adequacy of EU objectives for 2050, the energy mixes at both scenarios are considered in this study as shown in figure 2.

For H_2, different distribution options to the refueling stations were considered: central plant generation with transport via tube trailer or via pipeline and in-situ production. The emissions of each distribution strategy were then compared and only that with the lowest emissions was used for the whole LCA. This same methodology is also applied to decide if gaseous or liquid H_2 should be used (GH_2 or LH_2).

The raw materials considered as inputs in this cycle were mainly crude oil for fuel processing and organic matter such as soybeans to generate biofuels. In this case, the transportation of the immediate products from the raw materials was also considered.

3.2. *Vehicle production and disposal cycle*

The emissions in the vehicle production and disposal cycle were calculated based on the required raw materials for each component. The mechanical components include the vehicle body (conventional material), the powertrain system, the transmission/gearbox, the chassis, the tire replacements, and the electric motor, controller, and generator (HEV, BEV, and FCV). The mechanical components for the HICE vehicle are the same as for a SI ICE car. Li-ion batteries were considered for BEV while Ni-MH batteries were considered for FCV and HEV vehicles. The emissions produced from the recycling of Li-ion batteries were estimated from [19] considering a pyrometallurgical process. The usage of engine oil, brake, transmission, coolant, windshield and adhesives fluids was included in the production cycle. The manufacturing of the FC and the H_2 tanks (700 bar of storage pressure, type IV carbon fiber) were also included but their recycling was ignored. This was done because the effect of platinum recycling of the fuel cell stack is negligible in the whole life cycle [20] and there is no data about recycling type IV carbon fiber reinforced polymer tanks.

The raw materials for this cycle were mainly steel, aluminum, magnesium, zinc, copper wires, glass, plastic product, styrene-butadiene rubber, carbon-fiber reinforced plastic and other vehicle materials. The emissions associated with the processing of raw materials and the extraction of elementary materials such as bauxite ore, zinc ore, sand water, etc were included while those generated during the transport to the manufacturing plants neglected [21].

3.3. *Operation cycle*

Emissions in the operation cycle depend mainly on fuel consumption and type of fuel. BEV and FCV CO_2 emissions during operation are zero. In the case of a HICE, 3 g CO_2/mile (from oil combustion) and 0.3 g NO_X/mile are emitted based on an FTP 75 cycle [22]. In the case of a CNG ICE, the leakage of CH_4 is also

Table 2: Fuel consumption of similar passenger vehicles with different engine technology [23, 24].

Vehicle	Energy consumption [kWh/100km]	Fuel consumption [Nm^3 (kg)/100 km]
BEV	14.5	-
H_2 FCV	24.4	8.14 (0.73)
Diesel ICE	45.4	$4.54\cdot10^{-3}$ (3.84)
HICE	58.7	19.6 (1.76)
CNG ICE	67.3	6.62 (5.15)
Gasoline ICE	58.7	$6.60\cdot10^{-3}$ (4.87)
Gasoline HEV	39.5	$4.45\cdot10^{-3}$ (3.28)

considered due to its high Greenhouse effect. Table 2 shows the fuel consumption for each technology in terms of fuel energy, mass, and volume. A refueling efficiency of 100% was assumed.

The emissions during the operation cycle were estimated based on the GREET® model but scaled with the consumption data from [25, 24] because the consumptions given in GREET® were abnormally high.

3.4. Cradle-to-grave comparison

Once the emissions per cycle were obtained, they were added to know the total life cycle emissions considering the EU 2017 and EU 2050 energy mixes. The results for each scenario were compared to identify the change in emissions of each technology considering a life of 150000 km and the compatibility of EU objectives with the development of the H_2 economy.

In the case of the emissions produced during the manufacturing and recycling of the vehicle, they are fixed and do not increase with the usage. In contrast, those emitted during the fuel production and operation cycle scale with the life (in km) of the vehicle. Therefore, it is possible that any technology, compared to any other, implies higher emissions during the manufacturing cycle, but they are compensated if the usage is long enough and the ratio emissions/km is lower during this cycle. In order to estimate which technology emits the less as a function of the life (km), the whole life emissions of each technology were plotted against the usage of the vehicle.

4. Limitations

Life cycle assessments are often limited to the amount of information that databases can provide. Therefore, it is necessary to take on certain hypotheses and constraints. This section presents the scope of this LCA study. The limitations of this study are:

- The study is fundamentally based on mid-size passenger vehicles since they compose the majority of the current vehicle fleet.
- Fuel production and engine technologies are assumed to be constant with time. Therefore, the emissions predicted in the EU 2050 scenario associated to these aspects may be under or overestimated.
- Europe and United States technologies for fuel production are assumed to be similar, while the main difference is the energy mix.
- Fuel consumption of HICE and gasoline ICE vehicles were assumed equal. Even though brake efficiency of HICE is higher than that of gasoline ICE, the extra weight of the tanks could compensate for this difference in efficiency.
- The emissions produced to manufacture the machinery needed to extract or produce the fuel are not quantified. This is negligible in emissions/km basis since fuel production plants would generate fuel for a large vehicle fleet.
- Some results are very similar to each other (figures 3,4, 5 and 6) and, even though the tendencies seem correct, an study of uncertainties could provide more value to the analysis. However, not all the data obtained from the literature and from the GREET® model showed the uncertainties in emissions corresponding to each process and pathway. Therefore it was difficult to estimate uncertainties, but the results are expected to be meaningful according to similar literature in the field of study..

5. Results and discussion

5.1. Fuel production cycle

Emissions to produce any fuel may vary largely depending on the production and distribution methods. This fact is highlighted for alternative fuels whose production methodology has not been extensively used and developed in the industry. As such, the recent research was also oriented towards optimizing the hydrogen production and distribution technologies [26, 27]. In the case of H_2, there are mainly two ways of mass-producing it: natural gas steam reforming or steam methane reforming (SMR) and electrolysis. The most extensively used in Europe nowadays is SMR because of the economic and environmental benefits it offers against electrolysis. However, the environmental benefits may no longer be real if the energy mix is mostly composed of renewable energy. In order to understand the sensitivity of these production technologies to the energy mix, the first part of the fuel production cycle analysis was based only on gaseous and liquid H_2 production and distribution strategies. Then, the fuel cycle GHG-100 and NO_X emissions were compared for the fuels in table1.

5.1.1. H_2 production and distribution strategies

As explained previously in this study, for H_2 it is interesting to consider different production and distribution strategies since Europe is still far from the H_2 economy and thus it is not clear what production methodology will be used in the future.

To produce H_2, the processes of SMR with and without CO_2 sequestration and electrolysis where considered. For the SMR process with CO_2 sequestration, it was assumed that 90% of this pollutant was not emitted [28]. Regarding the distribution, central plant production with transport to the refueling stations by means of tube trailers and in-situ production at the refueling stations was considered. Pipeline H_2 distribution was not accounted because it is not a short-term solution since a whole distribution network should be developed along Europe. Natural gas current pipeline network can not be used for H_2 because it is not adapted to contain such a highly diffusive gas, although an option could be to distribute H_2 blended with natural gas. From the raw fuel, compressed gaseous H_2 or liquid cryogenic H_2 were considered. Liquid H_2 was not used in the following analyses nor considered for the scenario of central production with distribution because for road transport it is not feasible to keep any fuel at cryogenic conditions for long periods of time. All these scenarios with the EU 2017 and EU 2050 energy mixes are contemplated in figures 3 and 4.

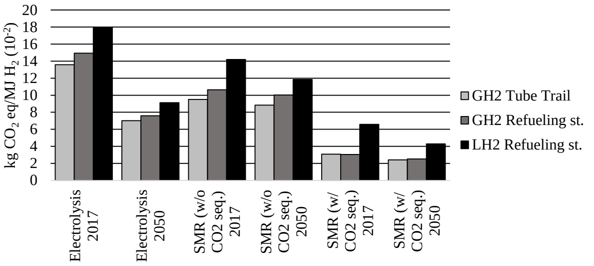

Figure 3: GHG-100 emissions for gaseous and liquid H_2 fuel cycle.

Based only on greenhouse emissions, H_2 production via SMR with CO_2 sequestration is indeed the best option (figure 3). With the EU 2017 energy mix, fuel production via electrolysis is the worst option regarding GHG emissions. In contrast, with the EU 2050 energy mix electrolysis implies lower emissions than SMR without CO_2 sequestration. This is because most of the energy required to produce H_2 through electrolysis is electrical energy while for SMR most of the energy comes from natural gas combustion the heat up the steam reformer. This makes electrolysis highly sensitive to the energy mix. Unless other studies, H_2 mass production from only renewable energy is not included because it is not realistic to have a solar field near every electrolyzer, so in the future, the most probable approach is to cleanse the energy mix and use the energy directly from the general power line.

Regarding NO_X emissions (figure 4), electrolysis is in both energy mix scenarios the worst option because the share of energy produced from fossil fuels through combustion is still significant. In the case of SMR, NO_X emissions are independent of the energy mix because they are mostly produced during the steam reforming where 5-10% of air is needed and is at high temperature during a long time [29]. NO_X emissions are higher in SMR with CO_2 sequestration probably because capturing CO_2 implies higher energy consumption.

According to the results in figures 3 and 4, central plant H_2 production and distribution via tube trailers is a better option than in-situ production. Producing H_2 in each refueling station implies greater water consumption than central production because of economies of scale. This water must be pre-treated, which means higher energy and resource consumption, thus producing higher emissions than central production [25].

Liquid H_2 could provide a higher vehicle range for the same tank capacity than gaseous H_2. However, its liquefaction process requires around 30% of its higher heating value. This high energy demand increases substantially the emissions to produce LH_2 and makes them more sensitive to the energy mix. If not for the difficulty of storing LH_2 at cryogenic conditions and the amount of energy required to liquefy it, LH_2 could be a suitable long-term fuel option.

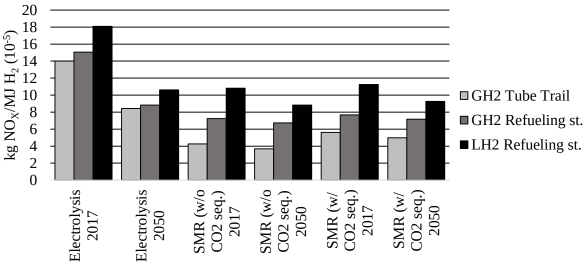

Figure 4: NO_X emissions for gaseous and liquid H_2 fuel cycle.

In sight of the GHG emissions in figure 3, H_2 production via SMR with CO_2 sequestration and distribution via tube trailer is the best option in the short-term (2017) and mid-term (2050). In contrast, due to the additional energy required to capture CO_2, figure 4 shows that the best option to minimize NO_X emissions is SMR without CO_2 sequestration instead. In order to address this problem, it is possible to use a NO_X trap or catalyst at the exhaust of the SMR process to further reduce NO_X emissions. Finally, due to the high sensitivity of electrolysis to the energy mix, this technology has the highest potential for the long-term when a mostly renewable energy mix is expected. In-situ SMR is not considered since it is not feasible to have a H_2 production plant at each refueling station, but distribution via pipelines could be a good solution for the mid to long-term.

5.1.2. Comparative fuel cycle

Once the H_2 production and distribution strategies were analyzed, they must be compared against the production routes of other conventional fuels. In this section, the aforementioned comparison is presented in figure 5. Again, the data is produced for the EU 2017 and EU 2050 energy mixes so that the effect of more-renewable electricity is reflected in the analysis.

According to the results in figure 5, H_2 production generates significantly more GHG-100 and NO_X emissions than B10 Diesel, E10 gasoline or CNG fuels. If H_2 is produced by means of electrolysis, the emissions are the highest while if it is produced through SMR with CO_2 sequestration, the emissions may be lower than using electricity directly in an electric vehicle.

EU 2050 scenario is characterized by a higher renewable energy share in the energy mix (figure 2). As such, all fuel production strategies produce lower emissions. Depending on the grade of dependence on the energy mix, the emissions may change significantly between both scenarios. Electricity directly used as a fuel and H_2 produced by electrolysis present the highest sensitiveness. However, electrolysis, even in 2050, is expected to generate far more emissions than current fuels. In the case of electricity to power electric vehicles, the emissions during the fuel production cycle will always be lower than H_2 produced by electrolysis because it avoids an additional energy transformation with its corresponding irreversibilities. The effect of improving the electrolysis or SMR processes with time is not included in this data. Therefore, lower emissions are expected in the actual EU 2050 scenario in an extent that depends on the level of development of these processes. In contrast, conventional hydrocarbon fuels are almost insensitive to this change since electricity is used as an auxiliary resource to power the machinery to extract and refine the fuel but not as the main energy resource to be converted into fuel.

Finally, it is important to remark at this point that emissions during the operation cycle are almost non-existent for H_2 technologies. Therefore, even though producing conventional fuels may generate lower emissions, the operation cycle must be included to assess the EU objectives and drawing any significant conclusion.

5.2. Vehicle production cycle

Differently from the fuel cycle, the emissions generated during the vehicle production are fixed and do not increase with the usage. Even though these emissions may be a minor part of the whole life cycle, they must be included to quantify the effect of the requirement of components such as H_2 tanks or Li-Ion/Ni-MH batteries. In the case of low emissions technology, such as BEV or H_2 FCV whose operation cycle is characterized by virtually zero emissions, this cycle can be significant.

In order to make the different vehicle production cycles comparable, a common vehicle body of 740 kg without the powertrain system nor the chassis (where the FC or the batteries can be integrated) was considered. The total weight of the vehicles varies between 1420 kg (gasoline ICEV) and 1640 kg (FCV).

The results of this cycle are only shown for the EU 2017 scenario because the sensitivity to the energy mix is relatively low (figure 6). In the EU 2050 scenario, the reduction in emissions ranges from 11% to 13% for all technologies. This effect was included in the cradle-to-grave cycle. Most GHG-100 emissions are produced in the manufacturing process of the mechanical components since they represent most of the mass of the vehicle (body, chassis, powerplant. . .). For HICE and FCV, which generate the most greenhouse gases, the increase in emissions is mainly due to mechanical

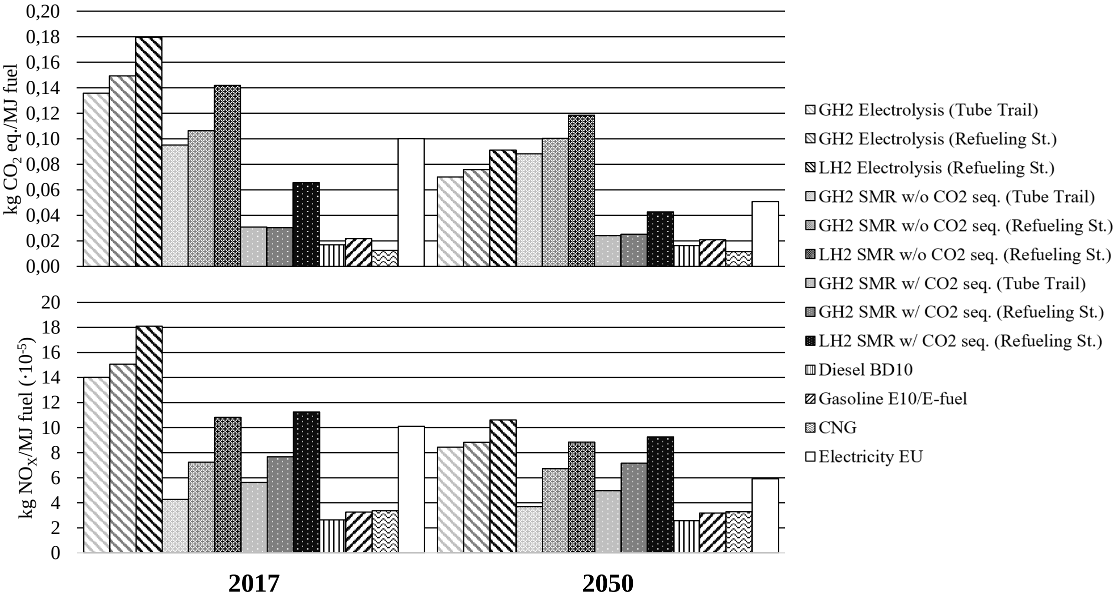

Figure 5: Fuel cycle comparison for H_2 and conventional fuels in terms of GHG-100 and NO_X.

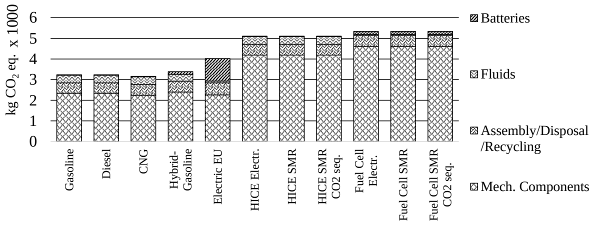

Figure 6: GHG-100 emissions in the vehicle production cycle.

components. In this case, the need for a carbon fiber reinforced type IV tank to store 700 bar of gaseous H_2 is the main factor that increases emissions. Among these two technologies, the FCV generates more GHG-100 because of the manufacturing of the fuel cell (102 kW), its corresponding balance of plant and the battery (34 kW) [30].

Emissions coming from batteries manufacturing are greater for the BEV since the Li-Ion batteries are bigger and require higher energy storage capacity than Ni-MH or lead-acid batteries, thus needing more materials. In contrast, ICEV have more emissions coming from fluids since they need engine oil to lubricate the reciprocating mechanism to reduce mechanical losses and increase the durability.

Even though alternative fuels and electricity for transportation may be interesting from the point of view of decentralizing emissions, they produce more pollution during the fuel and vehicle production cycles than conventional fuels. However, this issue is caused by the lack of development of these technologies and may be solved with time.

NO_X emissions present a similar trend as GHG-100 because they are produced from the electricity usage and high-temperature processes where CO_2 is also emitted.

5.3. Cradle-to-grave cycle

The cradle-to-grave cycle assessment presented in this section includes the fuel production, vehicle production, and operation cycles. In order to get the absolute value of emissions in the fuel production and operation cycles, it is necessary to set a life duration. In this case, life or usage was set to 150000 km. This value is realistic for current ICEV. However, it may be too high for BEV where batteries degrade over time. This value is used anyway because this issue could be solved by 2050 and not all the ICEV reach 150000 km.

5.3.1. GHG-100 emissions

Once the emissions coming from each cycle are put together, it is possible to realize that each part is significant depending on the technology or scenario considered. For example, in figure 7 the GHG-100 emissions in the vehicle production cycle for a FCV

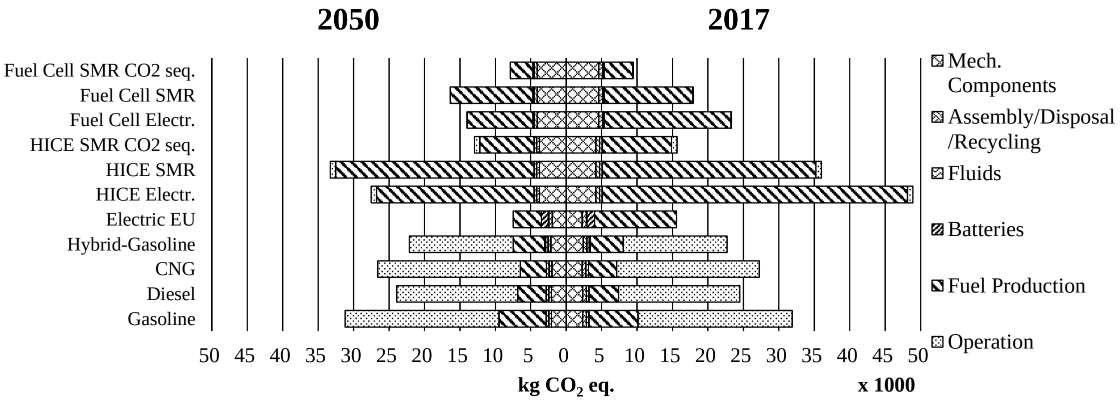

Figure 7: Cradle-to-grave cycle, GHG-100 emissions.

are higher than the operation and fuel production cycle if H_2 is produced from methane through SMR with CO_2 sequestration. In contrast, for conventional ICEV and HEV, the operation cycle is the most significant emissions-wise while vehicle production represents around 10% of the total life emissions. Due to the current trend towards more electrical propulsion in the automotive sector, it is possible that in the future the efforts in reducing emissions are shifted towards vehicle manufacturing.

For H_2-fuelled vehicles and BEV, most of the emissions come from the fuel production cycle. The effect of H_2O in the exhaust of FCV is almost negligible. In contrast, its effect on HICEV is noticeable. Particularly, in the case of a HICEV with H_2 produced from SMR with CO_2 sequestration, where it represents 5% of the total GHG-100 emissions. The noticeable difference in the emissions during the fuel production cycle between HICEV and FCV when the production technology is the same is due to the lower fuel consumption of FCV since less fuel is required for the same usage (Table 2).

According to the results of greenhouse emissions in the EU 2017 scenario (figure 7), the interest of using HICE or FC technologies is strongly dependent on the production strategy used. In the short-term, HICEVs are competitive against fossil fuels only if H_2 is produced through SMR with CO_2 sequestration. However, if H_2 is produced from electrolysis with energy from the energy mix, the total emissions double those of a Diesel car during the whole life.

Regarding FCV, in the short-term, they are already competitive, with any production technology, against fossil-fuelled vehicles. If electrolysis is used, there is not a big benefit of using FCV. By combining FCV with SMR and CO_2 sequestration, current FCV could produce less than two-thirds of the emissions of an BEV during the whole life.

In sight of the GHG-100 emissions in the EU 2017 scenario, the short-term strategy towards the H_2 economy should necessarily include the spreading and development of SMR with CO_2 sequestration to produce H_2. Concerning the powerplant selection, FCs have the advantage of lower fuel consumption and the drawback of higher cost, which forbids their extensive usage, while HICEs have higher fuel consumption but can be easily integrated into the society due to their lower cost as a competitive option against BEVs.

As expected, the change to a more-renewable energy mix (from EU 2017 to EU 2050) in figure 7 affects more significantly the emissions of BEV and H_2 technologies with H_2 produced from electrolysis. With this production technology, FCV would generate half of gasoline ICEV GHG-100 emissions while HICEV would start to be competitive against conventional ICEV. The most beneficial strategy would still be producing H_2 with SMR and CO_2 sequestration. This means that the long-term strategy to move towards H_2-based transport should be based on SMR with CO_2 capture rather than electrolysis. In this case, FCV and BEV would generate approximately similar GHG-100 during the whole life due to the higher share of clean energy available for powering BEV.

5.3.2. NO_X emissions

NO_X emissions produced by each technology (figure 8) must also be accounted for to assess EU objectives and H_2 powerplants. In the EU 2017 scenario, the less pollutant option is again the FCV whose H_2 is produced through SMR. This difference is significant even when compared with BEV. In contrast, BEV pro-

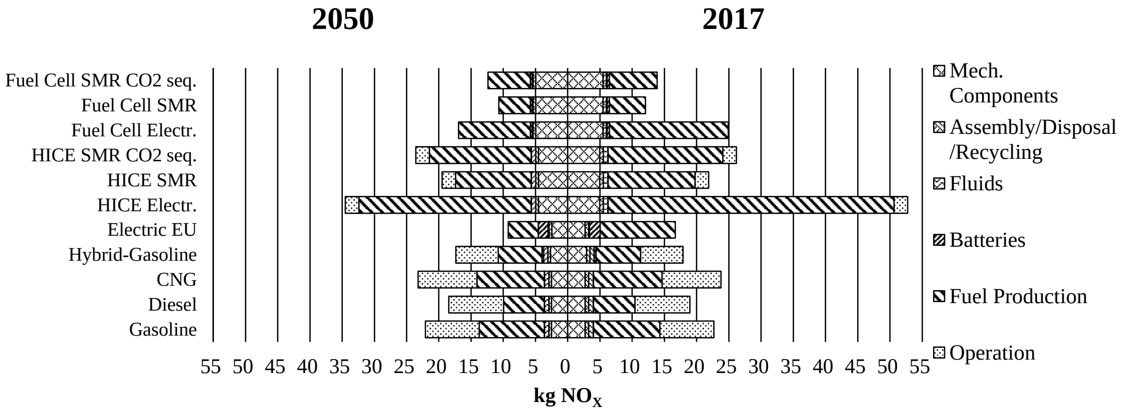

Figure 8: Cradle-to-grave cycle, NO_X emissions.

duce the lowest NO_X emissions in the EU 2050 scenario. The shift in the most favorable technology is due to the high sensitivity of BEV' emissions to the energy mix composition. In both scenarios, sequestering CO_2 in the central plants produce NO_X emissions due to the higher energy and resources consumption it implies. If H_2 is produced through electrolysis, FCV would produce NO_X emissions in the levels of conventional technologies in the short term. If EU objectives for 2050 are accomplished, FCV' NO_X emissions would be less than those produced by gasoline HEV or Diesel ICEV but still higher than BEV.

The amount of NO_X produced by HICEV during the whole life is always higher than any other technology, especially if electrolysis is used, no matter the energy mix scenario. This is because of the lower efficiency of HICE compared to FC and the high amount of NO_X produced per MJ of H_2 during the fuel production cycle. In this case, even though NO_X are low during the operation cycle and can be further reduced with the use of catalysts [31], this cycle only contributes to roughly 8% of the produced NO_X emissions. The sensitivity to the energy mix is the same as for the GHG-100 results. BEV, HICEV, and FCV whose H_2 has been produced through electrolysis show the biggest variation when the energy mix is modified.

If the EU strategy to shift towards H_2 and electric vehicles was purely based on NO_X emissions the approach would change from that based on GHG-100. In the short term, the most beneficial option would be to increase the amount of FCV drastically while keep increasing the amount of BEV. In the mid-term, BEV should be the predominant road transport for light-weight passenger cars. Finally, in the long-term, BEV, FCV, and HICEV with H_2 produced from electrolysis could coexist with an energy mix mainly based on renewable and nuclear energies.

5.3.3. Target consumption for HICE

HICE main limiting factor are the NO_X emissions produced during fuel production. In order to reduce them, the only option, apart from improving the fuel production efficiency or using catalysts in the production process, is to decrease its fuel consumption. This could be done by hybridizing the powerplant and/or by increasing the thermal efficiency optimizing the injection and combustion processes. This last option could be achieved by adopting several solutions, such as flexible engine hardware systems (direct injection system, variable valve actuation, variable compression ratio...) or advanced combustion concepts (highly diluted combustion). In this section, the target consumption of a HICE to match the NO_X emissions of an BEV during the whole life was estimated.

The NO_X emissions of an BEV during the whole life in the EU 2017 scenario are 16.7 kg NO_X. Considering the vehicle manufacturing emissions of a HICEV (6.3 kg NO_X), the NO_X emitted during the fuel production and operation cycles should be 10.4 kg NO_X. With a life of 150000 km, the target NO_X production rate would be $6.9\cdot10^{-5}$ kg NO_X/km to match BEV's total NO_X. From the data in figure 8, the estimated NO_X production rate of HICE using SMR with CO_2 sequestration (whose GHG-100 production is similar to that of an BEV) is $13.3\cdot10^{-5}$ kg NO_X/km. Assuming that the amount of NO_X is proportional to the fuel consumption, which is realistic if the engine is correctly calibrated and/or catalysts are used because most of the NO_X emissions come from the fuel production cycle, the fuel consumption should decrease by 48%, from 58.7 kWh/100 km to around 30 kWh/100 km. This value is hardly reachable in real driving with a PFI HICE even though H_2 increases the thermal efficiency due to its high reactivity and flame speed. However, the fuel consumption of state-of-the-art Diesel HEV is 3.3 l/100km (33 kWh/100km). Therefore, this consumption could only be expected (if reachable) with a DI HICE integrated into a serial hybrid vehicle architecture as the range extender, where the HICE is mostly operating at peak efficiency points and the smart energy management may improve the overall efficiency.

6. Conclusions

In this study the GHG-100 and NO_X emissions have been estimated for FCV, HICEV, BEV, gasoline HEV, and Diesel, gasoline and CNG ICEV considering a life span of 150000 km. The fuel production, vehicle manufacturing, and operation cycles were included in the LCA. The emissions were calculated based on the EU 2017 and EU 2050 energy mixes in order to assess the suitability of the current EU objectives to increase the renewable energy share in the energy mix to advance towards the H_2 economy. Electrolysis, SMR with and without CO_2 sequestration were considered to produce H_2.

Among the H_2 production strategies considered in this study, SMR with CO_2 sequestration was the best option to minimize GHG-100 while the option without CO_2 sequestration minimizes NO_X probably due to the extra resources and energy required to capture the CO_2. Therefore, the ideal production technology would be SMR with CO_2 sequestration with NO_X-reducing catalysts at the exhaust of the SMR plant. Transportation via tube trailer from central plants minimized the emissions because those produced by pre-treating H_2O locally at each refueling station outweighed those produced by the trailers transporting the H_2 tanks to the refueling stations. This production and transport strategies are the most optimum both in EU 2017 and EU 2050 scenarios because the renewable energy share in the energy mix is not high enough to make electrolysis less contaminant than SMR.

FCV with SMR and CO_2 sequestration produce lower GHG-100 emissions than any other propulsion technology in the EU 2017 scenario but slightly higher GHG-100 than BEV with the EU 2050 energy mix. Similarly, FCV with SMR without CO_2 sequestration produce the lowest NO_X in 2017 but BEV overcome them in the EU 2050 scenario. In none of the scenarios, H_2 produced from electrolysis produced both lower GHG-100 and NO_X than from SMR with CO_2 sequestration. However, in EU 2050, electrolysis might start to be competitive against fossil-fuelled ICEVs in both GHG-100 and NO_X. Therefore, EU renewable energy production objectives are not enough to produce all the H_2 from electrolysis. SMR with CO_2 sequestration should be used instead if these objectives are not redefined upwards.

Emissions produced by HICEV with SMR and CO_2 sequestration were superior in terms of GHG-100 and inferior in NO_X than fossil-fuelled technologies. Although if electrolysis was used, given the electricity mixes, using fossil fuels would produce much less GHG-100 and NO_X emissions than HICE. Using the most optimum H_2 technology, in order to match the emissions of HICEV and BEV in the EU 2017 scenario, it would be necessary to decrease the fuel consumption of HICE to around 30 kWh/100 km. This might be achievable if DI HICE were used in a hybrid range extender vehicle architecture.

Even though the effect of water as a greenhouse gas was included, its effect was almost negligible when using FCV, if HICEV are used its effect is noticeable. With HICEV, the H_2O effect on global warming might represent 5% of the total GHG-100 emissions if SMR with CO_2 sequestration is used to produce H_2.

This LCA study confirms how the optimum strategy to reduce GHG-100 and NO_X emissions depends on the energy mix. In the short-term, H_2 production through SMR with CO_2 strategy should be extended and FCV in the market increased through cost reduction. It would be recommendable to develop NO_X catalyst for SMR plants and thus introduce DI HICEV in the market whose total life cycle emissions are competitive against BEV. In the mid-term (EU 2050), FCV and BEV should coexist because of their complementary characteristics. H_2 should still be produced in SMR central plants with CO_2 sequestration. In the long term, when renewable energies compose most of the energy mix, electrolysis would produce fewer emissions than SMR and therefore producing all the H_2 through electrolysis would be plausible to reduce emissions. In this case, HICEV, FCV, and BEV could coexist, although FCV would probably dominate the market of H_2 technologies due to their lower fuel consumption.

Policy implications statement

With the study, the authors intended to elaborate recommendations to optimize the rate of decrease in emissions produced by the transport sector according to the

EU 2017 and 2050 scenarios. Promoting the purchase of such vehicles through actions such as tax reduction, focused on the most optimum technologies in the short, mid and long term, would probably minimize the GHG-100 and NO_X emissions in Europe. Additionally, measures are to be taken to gradually increase the renewable energy share in the European electricity mix. However, until the renewable energy share is enough, the road to H_2 economy should be based on H_2 production through SMR with CO_2 sequestration.

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